# Theory of the Origin and Relative Abundance Distribution of the Elements\*

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Ay, for 'twere absurd To think that nature in the earth bred gold Perfect i' the instant: something went before. There must be remote matter. The Alchemist, 1610, Ben Jonson.

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## I. INTRODUCTION

**M**ODERN astrophysical data and nuclear physics indicate an intimate connection between the origin and the relative abundances of nuclear species, and have within recent years provided the clues required for attempting an explanation. Astrophysical studies have shown, first of all, that one can attach a fairly definite age to the universe as we know it, second, that the relative abundances of nuclear species are, to a good approximation, universal quantities, and, third, that physical conditions near the "beginnings" of the universe as we now know it, or in the interiors of certain kinds of stars, may have been sufficiently severe to have permitted the nuclear processes required for forming all the nuclear species. In addition, nuclear physics indicates that there should be a generic relation between nuclear species, because all nuclei are composed of similar kinds of particles and there appear to be significant correlations between relative abundances and the systematic properties of nuclei. The problem of the origin of the elements is by no means a new one, and has almost always been considered an integral part of the origin, structure, and evolution of the universe.<sup>†</sup>

As reasonably good data on the observed universal relative abundance distribution of the elements have become available, several different theories to explain the origin of the distribution have been developed. In one of these theories the relative abundance of the elements is described as the result of a "frozen-in" thermodynamic equilibrium between atomic nuclei. The nature of this equilibrium distribution is principally determined by nuclear binding energies. In a second theory the abundances of the elements are considered as resulting from non-equilibrium processes, involving the formation of very light nuclei by thermonuclear processes and of the remaining nuclei essentially by only the successive capture of neutrons, with intervening  $\beta$ -disintegrations. In a third theory the light elements are pictured as formed by thermonuclear processes while the heavy elements are formed as a result of the fission of polyneutron complexes with subsequent  $\beta$ -disintegration and neutron evaporation of the fragments. Yet another theory involves the continuous creation of matter at an essentially undetectable rate, uniformly throughout the universe, or the sudden appearance in the universe of "drops" of nuclear material, of stellar dimensions. Obviously each of these theories involves a different type of cosmology.

Recently, discussions of several of the theories of the

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<sup>†</sup> It is interesting to compare modern studies of this subject with some of the early ideas. Until the time of Copernicus the universe was believed to be small, closed, and conceptually comfortable. The earth was the center of this universe, and all material bodies in it were supposed to be compounded of the four simple substances—air, earth, fire, and water. In very ancient times matter was conceived as made up of one basic substance such as water or wood. In this connection it may be noted that the word *hyle* derives from the Greek *hyle* meaning either wood or matter, and the now obsolete word *ylem*, derived from *hyle*, is defined as the primordial substance from which the elements were created. The word element was first used with its modern meaning by Robert Boyle, while the first scientific listing of the elements was made by Lavoisier in 1789.

origin and relative abundance of the elements have been given by Gamow [60],‡ Gamow and Critchfield [61], and ter Haar [72, 73]. These presentations are in various ways limited in scope. A review by ter Haar [73a] covering various topics including a section on the abundance of elements is scheduled for publication in this issue. The purpose of our review is to summarize in some detail the present status of the several theories and to discuss briefly their cosmological implications. It will become evident that there is as yet no single theory which does not suffer from some difficulties. For some of the work to be described, the nature of the basic assumptions must be questioned, while in other work it would appear that the results are at variance either with the abundance data or with generally accepted views of the structure and evolution of the universe.

Insofar as possible there has been included in this review the work of all investigators dealing with the origin and relative abundance of the elements from a theoretical point of view. Any omissions that occur arise solely from oversight rather than from a decision to omit mention of the work. Reference to or discussion of many of the papers concerned with the experimental methods and results of determining the relative abundances of the elements is not included, nor is there a detailed description of the systematics of nuclear properties and abundance, since this is not the principal purpose of this review. Differences between the notation in this review and that of original papers will be found, and arise because as uniform a notation as possible has been used throughout.

# II. THE OBSERVED RELATIVE ABUNDANCES OF THE ELEMENTS

## (a) The Data

The relative abundances of the elements in the universe have been studied in a variety of locales, including the atmospheres and interiors of stars, the dust, gas clouds and nebulae in interstellar space, other galaxies, and the various objects in the solar system. The first adequate and complete tabulation of modern universal relative abundances was that of Goldschmidt [65]¶ in 1938. His work also described in some detail the sources and the probable accuracy of the relative abundance data, the sources being principally analyses of meteoritic composition and stellar spectra. Since 1938, improvement in stellar spectral data, improved values for isotopic abundance ratios, and the great advances in the analysis and interpretation of meteoritic composition due to Brown have warranted a new tabulation. Such a tabulation has recently been made by Brown [30, 31, 102], while a discussion of the relative abundance data and a detailed description of the methods of measurement is in preparation by Page [120]. Brown's work includes new tables of stellar abundances of the lighter elements, meteoritic abundances, cosmic abundances of the elements, and cosmic abundances of nuclear species. There are many differences in detail from the earlier listings of Goldschmidt, but the major features of the abundance data are not altered.

That the so-called cosmic abundances given by either Brown or Goldschmidt are indeed cosmic is believed to be firmly founded.\*\* In general the agreement between the relative abundances determined in various regions of the universe is reasonably good. Of the few differences observed, most are explainable in terms of the present or past physical conditions in the locale involved. For example, meteoritic and terrestrial material differ from stellar material in the relative scarcity of the volatile elements, i.e., H, He, Ne, A, Kr, Xe, as well as of those elements which probably were contained in volatile compounds at the time of planet formation. These disparities are undoubtedly associated with the physical conditions accompanying the origin of the solar system [70, 71, 102]. Variations in the abundances of Li, Be, B, C, N as well as H, D, and He in various locales in the universe are almost certainly the result of the participation of these elements in the thermonuclear reactions responsible for energy production in the great majority of stars [19, 22]. Certain observed peculiarities in C and N abundances in hot Wolf-Rayet stars also arise from special features of the thermonuclear reactions in these stars [56]. Variations in isotopic abundance ratios have also been observed. The ratio He<sup>3</sup>/He<sup>4</sup> differs with locale on the earth presumably because of cosmic-ray neutrons. H<sup>1</sup>/H<sup>2</sup> ratios are different on earth than in the solar atmosphere, while C<sup>13</sup> is excessively abundant in the atmospheres of some cool stars. These last two deviations as yet are not explained. Recently, Thode [154] has reviewed the variations in terrestrial isotopic abundance ratios. In general these variations amount to at most several percent and would seem to be explained on the basis of the effect of nuclear mass in chemical reactions during the earth's long history.

The universal relative abundances of the nuclear species given by Brown are used throughout this review, with several additions and alterations, as the basic experimental data for comparison with theory. Since Brown does not give the abundances of nuclear species below oxygen, his elemental abundances were employed for the lighter nuclei including those for Li, Be, and B. Although there are uncertainties in isotopic abundance ratios for these light elements in the universe due to their participation in thermonuclear reactions, the appropriate element abundances were converted to

t Numbers enclosed in brackets signify references which will be found in the Bibliography at the end of this paper.

<sup>¶</sup> For a complete account of this subject through 1930, see the book by von Klüber [100].

<sup>||</sup> For a list of papers by Brown and collaborators, see reference [30].

<sup>\*\*</sup> For example, Unsöld [161], in discussing abundances in the atmosphere of the sun and  $\tau$ -Scorpii, has noted the remarkably similar constitution of these two astronomically unrelated stars.



FIG. 1. Relative abundance of nuclear species as a function of atomic weight according to the data of Brown [30]. The relative abundances are taken with respect to 10,000 atoms of silicon. Isobaric abundances have been added together. Below O<sup>16</sup> elemental abundances given by Brown were converted to isotopic abundances according to the isotopic abundance ratios given by Seaborg and Perlman [129]. Odd A and even A nuclei are denoted by  $\bigcirc$  and +, respectively. The positions of magic number nuclei are indicated on the abscissa.



FIG. 2. Relative abundance of elements as a function of atomic number according to the data of Brown [30]. The relative abundances are taken with respect to 10,000 atoms of silicon. Odd Z and even Z elements are denoted by O and +, respectively.

nuclear abundances by means of terrestrially observed isotopic abundance ratios [129]. Values of noble gas abundances in the universe as interpolated by Brown have been included while the radioactive nuclei have not. All the available data discussed are shown in Fig. 1, in which the logarithms of the relative abundances of the nuclear species in the universe, stated as the number of nuclei of a given atomic weight per 10,000 atoms of silicon, are plotted versus atomic weight. In this plot isobaric abundances have been added together. Since data are not available for a few nuclear species, some plotted points may eventually be shifted upward as new isobaric abundances are added. With regard to the precision of the data, Brown has stated that the relative abundance data are good to within a factor of about four in general, a factor of two being the more usual. In Fig. 2 the logarithms of the relative abundances of the elements, normalized as previously, have been plotted versus atomic number. In this case isotopic abundances have been added. This manner of presentation is included because some investigators have presented the abundance data in this form. However, most of the theories concerning relative abundances deal with the distribution as a function of atomic weight (i.e., the nucleon content) rather than of atomic number. Recently, the relative abundance data of Goldschmidt have been subjected to detailed examination by Jensen [89, 90], Suess [151, 152], and Jensen and Suess [93]. They have correlated relative abundances with mass number, isotopic number, atomic number, and with the parity of the nucleon number. The reader is referred to the original papers for discussions of many of the detailed features of the data and in particular for the correction factors which Suess has estimated as required on the basis of reasonableness and continuity to smooth the observed abundance data [152a]. Recently, Sonder [138] has also given an interesting presentation of the relative abundance data.

The most outstanding general feature of the dependence of relative abundance on atomic weight is the very rapid decrease of abundance with increasing atomic weight up to  $A \cong 100$  and the essential constancy for atomic weights greater than 100. In fact, the abundance data can be represented approximately by two straight lines in the regions A < 100 and A > 100[4]. This general behavior and the systematic details of the abundance data provide definite evidence for the close correlation between the properties of atomic nuclei on an atomic weight basis and their abundances. It seems reasonable to expect that a correct theory of the relative abundance of elements will duplicate, in first approximation, the approximately exponential decrease in abundance with increasing atomic weight up to  $A \cong 100$ , and the essential constancy of abundance for greater atomic weights. This principal feature of the abundance data was also evident in the work of Goldschmidt.<sup>††</sup> Many investigators have plotted relative abundance data on such a contracted abundance scale that the general dependence on atomic weight described is made to appear unimportant. Frequently, the practice of connecting plotted abundance points with lines also tends to obscure the behavior of the data since there are rather large fluctuations.

#### (b) Detailed Features of the Data

There are a number of detailed features of the relative abundance data which merit discussion.<sup>‡‡</sup> In presentations of abundance data the many members of radioactive families of nuclei between Pb and U are generally not considered, for the reason that the abundances observed for the parthenogenic progeny of the parent elements of the several radioactive series are in excellent agreement with the predictions of established statistical laws of radioactive disintegration [152]. Only the observed abundances of the long-lived parent elements, and, perhaps, the abundances of the progeny when originally formed by processes other than the disintegration of parent elements, require theoretical explanation. However, it may be noted that in a statistical approach to the problem of element formation in which

<sup>&</sup>lt;sup>††</sup> Compare Fig. 1 with Fig. 1 of reference [5] which presents a similar plot of Goldschmidt's data.

<sup>&</sup>lt;sup>‡‡</sup> Harkins [76] and [78], and Oddo [118], appear to have first pointed out that the features of the data on the relative abundances of the elements indicate that the universal abundances depend on nuclear rather than chemical properties.

individual nuclear processes are not specified in detail, the problem of explaining short-lived radioactive elements does not arise so long as it can be demonstrated that the necessary reversible reactions exist.

An examination of the data in Fig. 1 reveals rather large fluctuations in the relative abundances as compared to the general trend of the data. This is particularly noticeable among the very light elements. In the latter case, at least, there seems to be good reason to believe that the presently observed abundances may differ somewhat from an original distribution, i.e., the abundance distribution when the  $\cdot$  elements were formed. ¶¶ For example, all of the nuclear species below oxygen may participate strongly in thermonuclear reactions at elevated temperatures either in ordinary stars or in the presently observed abundances of some of the other light elements such as F<sup>19</sup> may also be affected by thermonuclear processes.

Perhaps the most striking irregularity in the abundance data is the high peak in the vicinity of iron. On the atomic weight plot, Fig. 1, it may be seen that this abundance peak lies roughly in the range A=53 to A=63 and is above the general trend of relative abundances in this vicinity by a factor of about 10<sup>4</sup>. On the plot of abundance versus atomic number this peak contains only several elements. It should also be noted that nuclei which might be regarded as having a completed shell structure on an  $\alpha$ -particle model exhibit rather large abundances, e.g., He, C, and O.

There are several datum points in Fig. 1 in the vicinity of A = 130 which lie well below the general trend. These are the abundances of certain Xe and Ba isobars; no abundance data have been given by Brown for Te whose isotopes, when included with the former isobars, would raise the datum points under discussion. Several of the strikingly low abundance data points in Goldschmidt's tabulation, notably Te, Ba, Os, and Re have been redetermined or removed from the tabulation by Brown and the redetermined points are now more nearly in line with the general behavior of the other heavy elements.

In the region of the heavy elements there are several abundance peaks on the atomic weight scale which deviate from the trend by a factor of about 10. These peaks are correlated in location with the atomic weights of nuclei of the magic number variety.  $\| \|$  Magic number nuclei, i.e., those containing 50 or 82 protons, or, 50, 82, and 126 neutrons, have been shown to exhibit

peculiar stability properties as the apparent result of some kind of completed nuclear shell structure. The regions of atomic weight in which the magic number nuclei are situated are indicated on the abscissa of Fig. 1. It may also be noticed that there are abundance peaks in the observational data which correspond quite closely to the two peaks in the mass yield curve for the fission of uranium and other nuclei [5, 67, 93].

The analysis of relative abundance data in terms of the systematic properties of atomic nuclei has led to a number of abundance rules which are of interest.\* As a result of the shell rule (the Pauli principle applying to neutrons and protons in the nuclear structure), nuclei of the even Z-even A type are both more numerous and more abundant than any of the other types. Nuclei of the odd Z-even A or even Z-odd A types are about equally numerous and abundant, while nuclei of the odd Z-odd A type are rare in number and scarce in nature. Consequently, odd Z nuclei have very few isotopes, and even Z nuclei have much larger numbers of isotopes. The small number of odd A isotopes among even Z nuclei, and the considerable variation of this number with Z, is illustrated in Table I [114].

The behavior with respect to shell completion is nicely correlated with the features of the nuclear energy surface, in which the minimal valley is occupied by even Z-even A nuclei, the hills on either side are populated by even Z-odd A and by odd Z-even A nuclei, while far up the hill is an occasional odd Z-odd A nucleus. Insofar as the general elemental abundances are concerned it is found that, on the average, even A elements are about ten times more abundant than neighboring odd A elements [77].† The variation of abundance with even and odd A is quite evident, particularly among the elements in the vicinity of the iron peak, in the large scale plot of the abundance data shown in Fig. 6.

The relative abundances of the isotopes of a given element show a much smaller spread among themselves than do the relative abundances of the elements. Among elements of even Z, one finds the relatively small abundance, as well as the already mentioned small number, of odd A isotopes. As a rule, for the even Z elements above  $_{34}$ Se the heaviest stable isotope is quite abundant [114]. Below  $_{34}$ Se, the trend is reversed, if anything. If one plots for even Z elements,

TABLE I. Percentage of odd A isotopes among even Z nuclei.

							and the second s	The second se
Range of $Z$	2	12	22	32	42	52	62	72
(even Z nuclei only)	to	ιo	10	10	to	10	to	10
	10	20	30	40	50	60	70	80
Average percentage of isotopes which have odd $A$	20	4	6	9	25	19	33	27

\* For example see Mattauch and Fluegge, reference [111].

 $<sup>\</sup>P\P$  In some theories to be described later the elements are pictured as being formed continually in special stellar models, so that the anomalous abundances of some of the light elements require special explanations.

 $<sup>\|</sup>$  || Atomic nuclei of this type have been discussed by a number of investigators. See, for example, references [15, 21, 44, 47-49, 69, 112, 117]. The following additional references deal with this subject: E. Bagge, Naturwiss. 35, 375 (1948); Haxel, Jensen, and Suess, Naturwiss. 35, 376 (1948) and Phys. Rev. 75, 1766 (1949); K. Way, Phys. Rev. 75, 1488 (1949); A. H. W. Aten, Jr., Science 110, 260 (1949); and E. Feenberg, Phys. Rev. 77, 771 (1950).

<sup>&</sup>lt;sup>†</sup> This rule was first enunciated by Harkins on the basis of the early abundance data given by I. Noddack and W. Noddack [116].

the relative even A isotopic abundances, a bell-shaped curve results which, as just pointed out, is skew toward the heavy isotopes for the heavy elements.

The spread in atomic weight among the isotopes of given elements decreases as the atomic number increases. In general, the lightest isotopes of a given element form a less regular sequence in abundance than do the heavier isotopes. The foregoing indicates that stable isotopes even lighter than those now known may exist in nature in undetectable amounts [113].

There are several indications of the possible existence of neutrons in large amounts during the process of element formation. Not only are neutron-rich isotopes more abundant, but there appears to be an isobaric abundance rule indicating a possible role for  $\beta$ -disintegrations in the processes leading to the observed element distribution. An examination of the abundance data shows that of 51 sets of stable isobars the isobar of lowest charge is more abundant in 44 cases [5, 33, 52], as would be expected if  $\beta$ -decay played a role.

The recent discovery of heavy particles in the primary cosmic radiation provides an interesting comparison with the universal relative abundance of the elements [28, 29, 29a, 53, 54]. In experiments with balloon-borne nuclear emulsions and cloud chambers, particles having a charge up to about Z=45 have been detected and the relative numbers of such particles determined as a function of Z. The abundance of the various heavy cosmic-ray particles as a function of Z is



FIG. 3. Observed frequency of heavy particles in the primary cosmic radiation as a function of atomic number, according to Freier, Lofgren, Ney, and Oppenheimer [53]. Compare with Fig. 2.

shown in Fig. 3.<sup>‡</sup> The decrease in abundance with Z is in general accord with the cosmic abundance data already discussed, although it would appear that there is an overabundance of the heavier elements. In this cosmic-ray work the ratio of protons to  $\alpha$ -particles is perhaps the best determined quantity. The value of four found for the H/He abundance ratio is in good agreement with, for example, the values 4.5 observed for the sun and 10 for universal elemental abundances. It is interesting to note in the details of the study [29] that the C, N, and O group of nuclei are quite abundant, that the high abundance of Fe is reflected in the cosmicray flux, and that Li, Be, and B have as yet been undetected, as might be expected from their relative scarcity in nature.

The fact that there appears to be an intimate relationship between some of the systematic features of the abundance data and the stability properties of atomic nuclei, and, in particular, the even-odd rules discussed, has led many investigators to study the possibility of element formation under equilibrium conditions. However, properties other than nuclear stability per se may also be correlated with the systematic features of the abundance data. Recently, a nonequilibrium theory of element formation has been developed which depends upon the behavior of the radiative capture cross sections of nuclei for neutrons. In any event, as pointed out by Gamow and Critchfield [61], any theory of the abundance of nuclei should reflect the stability properties of nuclei in the detailed variation of relative abundances from species to species.

## **III. EQUILIBRIUM THEORY**

# (a) Introduction

The fact that there is a connection between the relative abundances of the elements and the systematic stability properties of atomic nuclei was apparently first pointed out by Harkins [76] in 1917. Considerations of this kind led many investigators to attempt an explanation of the observed relative abundance data on the basis of an equilibrium theory. These theories depend primarily on the correlation between the abundances and the binding energies of nuclei. In order to illustrate this correlation, Fig. 4 has been prepared, in which the abundance data of Brown [30] are plotted *versus* binding energies tabulated by Rosenfeld [125]. Apart from the few elements such as Li, Be, and B, and the elements in the neighborhood of iron, a consistent trend is evident in this correlation plot [91].

The problem of calculating the equilibrium concentrations of various kinds of nuclear species can be treated by the well-known methods of thermodynamics and statistical mechanics. In general, the concentrations depend upon the temperature and density of the

<sup>10-15</sup> have been discriminated against by the method of data analysis.

assembly or system, as well as upon the nature of the constituents and the binding energies of the nuclei. Obviously it must be assumed that there exist all the nuclear reactions necessary to establish this equilibrium. Since the binding energies of atomic nuclei attain values of about 2000 Mev for the heaviest elements, the temperature and density must be correspondingly high in order that equilibrium be established in a reasonable length of time. The question of where and when the physical conditions necessary to explain the abundance problem may have existed, what constitutes a reasonable length of time for the establishment of equilibrium, and also the way the nuclear equilibrium might have been "frozen-in," are essentially cosmological problems. It has, in fact, been found that no single set of physical conditions would simultaneously give rise to the correct relative abundances of the elements over the entire range of atomic weight, and as a result it has been necessary to assume that the elements were formed at two distinct epochs in the evolution of the universe, or that they were formed in special stellar models [see Section III(b)5].

It is proposed to give in this Section an historical survey of the considerable amount of work done on equilibrium theories and a detailed account of the theories as they now shape up. This will involve a discussion of the basic equilibrium theory, refinements in the theory such as the inclusion of excited nuclear states, gravitational and electrostatic effects, and, finally, some of the cosmological questions which arise in the equilibrium treatment of the relative abundance problem.

## (b) Development of the Equilibrium Theory

# 1. Early Work

As early as 1922 Tolman [156] studied the thermodynamic equilibrium between hydrogen and helium and concluded that it was not possible to understand the observed H/He abundance ratio at temperatures less than 106°K. Some years later Suzuki [153] independently considered the H-He equilibrium in stellar interiors and concluded that the H/He abundance ratio might be understood only at temperatures of about 109°K. He also studied the thermal dissociation of nuclei into protons. electrons, and  $\alpha$ -particles at high temperatures. In 1931, Urey and Bradley [163] examined the possibility of equilibrium between the isotopes of a given element (Li, B, O, N, and C) in order to determine whether or not the observed relative abundances of the isotopes correspond to thermodynamic equilibrium under a single set of physical conditions. They concluded that the observed relative abundances of isotopes do not agree with the hypothesis of a thermodynamic equilibrium at any single temperature, a result which has not been changed by subsequent studies. In none of these early studies was the general problem of a thermodynamic equilibrium among all



FIG. 4. Logarithm of relative abundance versus binding energy. The former are the data of Brown [30], while the latter are taken from Rosenfeld [125].

nuclear species formulated from the point of view of obtaining theoretical relative abundances.

Perhaps the earliest formulations of an equilibrium theory for the relative abundance of the elements were those of Farkas and Harteck [45] and of Pokrowski [123]. Farkas and Harteck suggested that the equilibrium distribution of nuclei was established at a high temperature ( $\sim 10^{9}$ °K) and at densities of about 10<sup>5</sup> g/cm<sup>3</sup>, and was then frozen in by the cooling of the "stellar body" in which it was supposed to have been established. Under the assumption of the ideal gas laws for protons, electrons, and nuclei, the equilibrium concentrations were calculated using the law of mass action, as

$$\log[(C_p)^A (C_e)^{A-Z} (C_j)^{-1}] = -(\Delta m)_j c^2 (4.57T)^{-1} + 2.5(2A - Z - 1) \log T + a_j, \quad (1)$$

where  $C_p$ ,  $C_e$ , and  $C_j$ , are the concentrations of protons, electrons, and nuclei, A and (A-Z) are the numbers of protons and electrons which were considered at that time to make up the *j*th nucleus,  $(\Delta m)_j c^2$  is the mass defect of the *j*th nucleus, playing the role of the "heat of formation," and  $a_j$  is a quantity involving the entropy. They computed the equilibrium abundances of some of the light elements and obtained at least the same trend with increasing atomic weight as is shown by the observational data.

In a somewhat different approach to this problem Pokrowski showed by thermodynamic reasoning that for a nucleus of atomic weight A,

$$-(\Delta m)_j/A_j^2 = (aA_j)^{-1} \ln W_j + \text{constant}, \qquad (2)$$

where  $(\Delta m)_j$  is the mass defect in mass units,  $W_j$  is the thermodynamic probability for the particular nucleus under consideration, and a is a constant. By analogy with the classical Boltzmann probability law,  $S = k \ln W$  + constant, and assuming that  $W_j = \alpha_j A_j$ , where  $\alpha_j$  is

the abundance of the element of atomic weight  $A_{i}$ , Pokrowski finally arrived at the formula

$$\log(\alpha_{j}A_{j}) = a_{1}(\Delta m)_{j}/A_{j} + a_{2}A_{j} + a_{3}, \qquad (3)$$

where the a's are constants. An appropriate adjustment of the constants could yield an approximate agreement¶ for the light elements, since Eq. (3) is nearly of the form derived by the standard methods of statistical thermodynamics to be discussed. Pokrowski was apparently the first to point out the very large discrepancy between the observed relative abundances of the heavy elements and those calculated by a simple equilibrium theory. He therefore suggested that there must be some other kind of process involved in the building-up of the heavier elements, such as the possible breaking down of "very heavy nuclei."

The early work described is mainly of historical interest because the correct nuclear model was not known, mass defect and relative abundance data were inadequate, and the physical structure of a nuclear equilibrium theory was not adequately examined.

#### 2. Basic Theory

The detailed developments of equilibrium theories of relative abundance are based on one statistical approach or another, involving, for example, the classical Gibbs' grand canonical ensemble [64]\*\*, the method of Darwin and Fowler [51], or, the analog of Saha's ionization equation  $\lceil 127 \rceil$ , all of which are necessarily equivalent. In order to clarify the interpretation of some of the quantities involved in the various statements of the equilibrium theories, the Gibbs' and also the Darwin-Fowler methods are briefly sketched.

Consider a thermodynamic system made up of hindependent kinds of substances or components, which is "open" in the sense that the composition may be altered by the introduction or withdrawal of material. Any other substances in the system are regarded as somehow formed from these independent components. At equilibrium the particular methods of formation are of no concern provided there exist reactions connecting all the substances. One may characterize the system at equilibrium by its energy E, by the external coordinates,  $q_1, q_2, \dots, q_i$ , pertinent to the system, and by the number of moles,  $N_1$ ,  $N_2$ ,  $\cdots$ ,  $N_h$ , of each of the h different components contained in the system. If these quantities are taken as independent variables, then one may write for the change in entropy of the system, corresponding to a change from one equilibrium state to a neighboring state,

$$\delta S = \frac{\partial S}{\partial E} \delta E + \sum_{i} \frac{\partial S}{\partial q_{i}} \delta q_{i} + \sum_{h} \frac{\partial S}{\partial \mathbf{N}_{h}} \delta \mathbf{N}_{h}, \qquad (4)$$

and from the second law of thermodynamics, for a system of constant composition,

$$\delta S = \frac{1}{T} \delta E + \frac{1}{T} \sum_{i} Q_{i} \delta q_{i} + \sum_{h} \frac{\partial S}{\partial \mathbf{N}_{h}} \delta \mathbf{N}_{h}.$$
 (5)

In Eq. (5), the  $Q_i$  are generalized forces, conjugate to the coordinates  $q_i$ , and may be gravitational or other types of forces. One may define

$$\mu_r = -T \left( \frac{\partial S}{\partial \mathbf{N}_r} \right)_{E,q_i,\mathbf{N}_h} = \left( \frac{\partial E}{\partial \mathbf{N}_r} \right)_{S,q_i,\mathbf{N}_h}, \quad (6)$$

as the Gibbs' intrinsic potential for the *r*th component. For equilibrium with respect to the transfer of components from one system to another the  $\mu_r$  are constant from system to system. In reactions among the substances  $s_i$  there will be simultaneous changes in the number of moles of the substances,  $\Delta N_l$ , such that

$$\Delta \mathbf{N}_1 : \Delta \mathbf{N}_2 : \dots = k_1 : k_2 : \dots, \tag{7}$$

where the  $k_l$  are positive or negative integers. At equilibrium one hastt

$$\sum_{l} k_{l} \mu_{l} = 0, \qquad (8)$$

and a condition of this type will hold for every equilibrium reaction in the system.

It is convenient now to consider representative ensembles, called grand canonical ensembles, whose members can differ not only in state but also in the amounts of substances of the different kinds which they contain. Such an ensemble is required to treat equilibrium with respect to the transfer of both energy and matter between systems in the ensemble. A system in this ensemble is regarded as made up of h independent components or substances, and the number of such components in any one of the systems is designated by  $n_1, n_2, \dots, n_h$ . The probability of finding a member of the ensemble with a given composition, in an energy state  $E_j'$ , is given by

$$\mathbf{P}_{n_1,n_2,\cdots,n_h,E_j} = \exp\left[\left(\Omega + \sum_h \mu_h n_h - E_j'\right)/kT\right], \quad (9)$$

where  $\exp(\Omega/kT)$  is the density-in-phase, and  $\mu_h$  are the intrinsic potentials per particle of the substance h.

Applying the grand canonical ensemble to the problem of nuclear equilibrium, one may write

$$\mathbf{P}_{N,Z,E_j'} = \exp\left[\left(\Omega + \mu_n N + \mu_p Z - E_j'\right)/kT\right], \quad (9a)$$

in which each nucleus is considered as being made up of Z protons and N neutrons,  $E_j'$  includes binding energy as well as excitation energy, and  $\mu_n$  and  $\mu_p$  are the intrinsic potentials of neutron and proton, respectively, referred to rest mass as zero-point energy. It

<sup>¶</sup> We have not been able to reproduce the calculated abundances using the *a*'s given by Pokrowski in reference [123]. A rough agreement with Pokrowski's results is found if  $a_2 = -0.094$  instead -94 as stated. of -

<sup>||</sup> For an early comment on a possible reason for the low abundance of the heavy elements see Stone, reference [147]. \*\* See also Tolman, reference [158], and Klein, reference [98a].

<sup>&</sup>lt;sup>††</sup> See reference [64], Vol. I, pp. 144 and 331, for Gibbs' discussion of the effect of gravity and electrostatic forces on the equilibrium of heterogeneous substances.

can be shown, in the non-relativistic non-degenerate case, that

$$C_{j}(N, Z) = (2\pi m_{j}kT/h^{2})^{\frac{1}{2}} \Phi_{j}(N, Z)$$

$$\times \exp[(\mu_{n}N + \mu_{p}Z - E_{j})/kT], \quad (10)$$

where the  $C_j$  are the concentrations of particular nuclear species of mass  $m_j$ ,  $\Phi_j$  is the partition sum for the internal degrees of freedom, and  $E_j$  is the binding energy of this species, defined as

$$E_{j} = c^{2}(m_{j} - Nm_{n} - Zm_{p}).$$
(10a)

If only the ground state is considered, then Eq. (10) can be written as

$$C_{j}(N, Z) = (2i_{j}+1)(2\pi m_{j}kT/h^{2})^{\frac{3}{2}} \times \exp[(\mu_{n}N+\mu_{p}Z-E_{j})/kT], \quad (11)$$

where  $i_j$  is the nuclear spin quantum number. At equilibrium, for the proton-neutron-electron reaction, neglecting neutrinos, one has, from Eq. (8)

$$(\mu_n + m_n c^2) = (\mu_p + m_p c^2) + (\mu_{e^-} + m_e c^2), \qquad (12)$$

where  $\mu_{e^-}$  is the electron intrinsic potential. Similarly, the equilibrium condition for nuclei, neutrons, and protons, is

$$(\mu_j + m_j c^2) = N(\mu_n + m_n c^2) + Z(\mu_p + m_p c^2).$$
(13)

This formalism for examining the equilibrium problem has been employed and extended by Klein, Beskow, and Treffenberg [17, 18, 98a, 99] whose work is discussed later in detail. At that point it will be seen that a physically correct set of the  $C_j$  must involve the specification of electrical neutrality for the assembly [98a].

The application of the Darwin-Fowler method to the statistical equilibrium among nuclei has been elegantly presented by Sterne [143–146].‡‡ Consider an assembly consisting of the independent particles neutrons, protons, and electrons, whose masses are  $m_n$ ,  $m_p$ , and  $m_e$ , respectively, and whose total number  $D_n$ ,  $D_p$ , and  $D_e$  are constant, whether free or bound. The mean numbers of particles,  $\bar{X}$ , of the various kinds in the assembly is given by

$$\bar{X}_{e} = \lambda \frac{\partial}{\partial \lambda_{i}} \ln[1 + \lambda \exp(-\epsilon_{i}/kT)] = \lambda \frac{\partial \mathcal{O}_{e}}{\partial \lambda}, \quad (14a)$$

$$\bar{X}_{p} = \eta \frac{\partial}{\partial \eta} \sum_{k} \ln[1 + \eta \exp(-\epsilon_{k}/kT)] = \eta \frac{\partial \mathcal{O}_{p}}{\partial \eta}, \quad (14b)$$

$$\bar{X}_{n} = \zeta \frac{\partial}{\partial \zeta} \sum_{l} \ln[1 + \zeta \exp(-\epsilon_{l}/kT)] = \zeta \frac{\partial \mathcal{P}_{n}}{\partial \zeta}, \quad (14c)$$

and

$$\bar{X}_{j} = \chi_{j} \frac{\partial}{\partial \chi_{j}} \left\{ \pm \ln \left[ 1 \pm \chi_{j} \exp(-\epsilon_{m}/kT) \right] \right\} = \chi_{j} \frac{\partial \mathcal{O}_{j}}{\partial \chi_{j}}, \quad (14d)$$

<sup>‡‡</sup> Steinwedel and Jensen [142] have also given some of the formalism of the equilibrium problem in terms of the Darwin-Fowler method.

where j refers to a particular nuclear species, the  $\epsilon$ 's refer to the energy states, and the  $\mathcal{P}$ 's are the complete partition functions. The quantities  $\lambda$ ,  $\eta$ ,  $\zeta$ , and  $\chi_j$  appear in applying the method of steepest descents and satisfy the following relations:

 $\chi_j = \eta^Z \zeta^{A-Z} \exp(-E_j/kT),$ 

and

$$\zeta = \lambda \eta \exp\left[-c^2(m_n - m_p - m_e)/(kT)\right], \quad (15b)$$

in which  $E_j$  is the binding energy. Equations (15) must be satisfied at equilibrium and, as will be seen, are equivalent to Eqs. (12) and (13). One can regard  $E_j$  as the potential energy of the *j*th nucleus, where the zeropoint energy is that of the state in which the nucleus is completely dissociated into its constituent particles. The parameters in Eqs. (14) must be consistent with the conservation of particles, charge and energy, so that:

$$D_e = \bar{X}_e, \tag{16a}$$

$$D_p = \bar{X}_p + \sum_j \bar{X}_j Z_j, \tag{16b}$$

$$D_n = \bar{X}_n + \sum_j \bar{X}_j N_j, \qquad (16c)$$

and

and

$$E_T = \bar{E}_{rad} + \bar{E}_e + \bar{E}_p + \bar{E}_n + \sum_j \bar{E}_j,$$
 (16d)

where  $\bar{E}_{e}$ ,  $\bar{E}_{p}$ , and  $\bar{E}_{n}$  are the mean kinetic energies of the elementary particles,  $\bar{E}_{j}$  is the energy of the nucleus including its binding energy,  $\bar{E}_{rad}$  is the total energy contained in the radiation field, and  $E_{T}$  is the total energy not including rest mass. The condition for electrical neutrality requires that  $D_{e}=D_{p}$ , since positrons have not been considered. The density,  $\rho$ , of the system is given by

$$\rho V = m_e X_e + m_p \overline{X}_p + m_n \overline{X}_n + \sum_i (m_n N_j + m_p Z_j) \overline{X}_j + E_T / c^2, \quad (16e)$$

where V is the volume of the system. In actual studies that have been made of the equilibrium abundances, the density of matter is usually so high that one may neglect  $m_e \bar{X}_e$  and replace the last two terms in Eq. (16e) by  $\sum_j m_j \bar{X}_j$  with sufficient approximation. In the classical approximation where  $\chi_j \ll 1$  and  $kT \ll m_j c^2$ , the partition function for both even and odd nuclei is approximately

$$\mathcal{O}_j = \chi_j (2\pi m_j kT/h^2)^{\frac{3}{2}} V \Phi_j, \qquad (17)$$

where as before  $\Phi_j$  is the partition sum for the internal degrees of freedom. Thus, Eqs. (14) become

$$\bar{X}_{e} = \mathcal{O}_{e} = (2i_{e}+1)\lambda V (2\pi m_{e}kT/h^{2})^{\frac{3}{2}},$$
 (18a)

$$\bar{X}_{p} = \mathcal{O}_{p} = (2i_{p} + 1)\eta V (2\pi m_{p} kT/h^{2})^{\frac{3}{2}},$$
 (18b)

$$\bar{X}_n = \mathcal{O}_n = (2i_n + 1)\zeta V (2\pi m_n kT/h^2)^{\frac{3}{2}},$$
 (18c)

$$\bar{X}_j = \Theta_j,$$
 (18d)

(15a)

where the *i* are spin quantum numbers. These expressions may be shown to be equivalent to those obtained in the Gibbs' method as follows. Equations (18) can be rewritten with the aid of Eqs. (15), setting  $\bar{X}_j/V = C_j$ , as

$$C_p = (2i_p + 1)(2\pi m_p kT/h^2)^{\frac{3}{2}} \exp(\ln \eta),$$
 (19a)

$$C_n = (2i_n + 1)(2\pi m_n kT/h^2)^{\frac{1}{2}} \exp(\ln \zeta),$$
 (19b)

and

and

$$C_{j} = (2\pi m_{j}kT/h^{2})^{\frac{1}{2}} \Phi_{j}$$

$$\times \exp[N \ln\zeta + Z \ln\eta - E_{j}/(kT)]. \quad (19c)$$

Comparing Eqs. (19) with the Gibbs' formulation, one finds that

 $kT \ln \eta = \mu_p, \qquad (20a)$ 

$$kT\ln\zeta = \mu_n, \qquad (20b)$$

$$N(kT\ln\zeta) + Z(kT\ln\eta) = \mu_j, \qquad (20c)$$

$$kT \ln \lambda = \mu_{e^-}.$$
 (20d)

Writing Eqs. (15) in logarithmic form, and using Eqs. (20), one finds Eqs. (12) and (13), defining equilibrium in terms of the intrinsic potentials.  $\P\P$ 

In this development one can also include the effect of gravitational and electrostatic forces. All that is involved is effectively a redefinition of the Gibbs' potential to include, in addition to the intrinsic potential, the gravitational and electrostatic potentials for the particular kind of particle in question. Sterne [143] has also discussed degeneracy and relativistic effects on the formulation of the equilibrium problem. The partition functions are given by Sterne for non-degenerate statistics,  $\chi_j \ll 1$  (non-relativistic non-degenerate statistics,  $kT \ll m_j c^2$ , and, relativistic non-degenerate statistics,  $kT \gg m_j c^2$ ) and degenerate statistics,  $\chi_j \cong 1$  or  $\chi_i > 1$  (non-relativistic degenerate statistics,  $kT \ln \chi_i$  $\ll m_j c^2$ , and, relativistically degenerate statistics,  $kT \ln \chi_j$  $\gg m_i c^2$ ). For the case of non-relativistic non-degenerate statistics Sterne considered an hypothetical assembly, in the absence of fields, in which the density of matter is 10 g/cm<sup>3</sup> and in which there can exist radiation, electrons, protons, and the nuclei He<sup>4</sup>, O<sup>16</sup>, Fe<sup>56</sup>, and RaB<sup>214</sup>. He found that at  $T \leq 2 \times 10^{9}$  K the assembly was composed almost entirely of Fe (the element in the assembly with about the largest binding energy per nucleon),\* almost entirely of He<sup>4</sup> at  $T = 3 \times 10^{90}$  K, and of H<sup>1</sup> at  $T = 4 \times 10^{9}$  °K.

Equilibrium distributions calculated on the basis of the analog to the Saha ionization equilibrium equation have been considered by several investigators. The formulation of the problem in this manner follows directly from the discussion already presented. Consider the equilibrium between neutrons, protons, and nuclei. Then, the equilibrium concentrations  $C_j(N, Z)$  and  $C_{j'}(N+\Delta N, Z+\Delta Z)$  can be found from Eq. (10) and written in the following form:

where  $i_0$  and  $m_0$  are the spin of an elementary particle and the unit of atomic mass, respectively. In this form the Gibbs' intrinsic potentials are replaced by the concentrations of neutrons and protons. The equilibrium concentration of a given nuclear species can be written conveniently in terms of the neutron and proton concentrations as follows [35, 79]:

$$\ln C_{j} = Z \ln C_{p} + (A - Z) \ln C_{n} + (A - 1) \ln [h^{3}/(2\pi kT)^{3/2}] + |E_{j}|/kT - (3/2)Z \ln C_{p} - (3/2)(A - Z) \ln C_{n} + (3/2) \ln m_{j},$$
(22)

or, using  $10^{9}$ °K as the unit of T, and mMU as the unit of energy,

$$\log C_{j} = 34.08 + (3/2) \log T_{9} + (3/2) \log A + (4.73/T_{9}) |E_{j}| + A [\log C_{n} - 34.08 - (3/2) \log T_{9}] - Z \log (C_{n}/C_{n}). \quad (23)$$

In all formulations of the equilibrium theory the predominant boundary condition on the determination of the distribution of abundances is the physical requirement of electrical neutrality for the assembly. For example, each specification of  $\mu_n$  and  $\mu_p$  for a given temperature defines an equilibrium distribution. However, for a given  $\mu_n$  (or  $\mu_p$ ) there is only one value of  $\mu_p$  (or  $\mu_n$ ) which gives electrical neutrality. The procedure involved in determining electrical neutrality is discussed later in this section.

One of the earliest detailed studies of the relative abundance of the elements was that of Chandrasekhar and Henrich  $\lceil 35 \rceil$  who extended the earlier qualitative considerations of von Weizsäcker [176]. Independently of Pokrowski [123], von Weizsäcker had pointed out the difficulty of explaining both light and heavy element equilibrium abundances under a single set of physical conditions. Lacking sufficiently accurate binding energy data, von Weizsäcker made no detailed calculations. He did, however, suggest that isotopic abundance ratios should be most accurately known, and, if the necessary binding energy data were available, should provide an interesting test of the equilibrium theory. This approach had already been made by Urey and Bradley [163] some years earlier and appears to have been generally overlooked. Such tests of the equilibrium theory by

 $<sup>\</sup>P$  The treatment of the equilibrium problem by Sterne [143] did not include the neutron-proton-electron equilibrium explicitly, and hence the equivalence of the conditions on the steepest descent parameters and the Gibbs' intrinsic potentials is not immediately evident.

 $<sup>\|</sup>$   $\|$  Lacking adequate mass defect data Sterne considered the proton-electron nuclear model rather than the proton-neutron model because of the relative simplicity of the former calculation.

<sup>\*</sup> M. Paul [121] suggested on the basis of oversimplified considerations that stars generate energy by going to the state where most of their mass is iron.

examination of isotopic abundance ratios also have been made by Chandrasekhar and Henrich [35], Singwi and Rai [133], van Albada [1], Jensen and Suess [92, 93], and Ubbelohde [160]. All of these investigators independently have used essentially the formalism developed by Urey and Bradley and their results, involving more modern nuclear masses, appear to confirm the conclusions of Urey and Bradley.

Chandrasekhar and Henrich applied Eq. (21) to five sets of isotopes (O, Ne, Mg, Si, and S), assuming that the reactions maintaining equilibrium between isotopes were those of neutron exchange. As indicated in Table II they found that while a mean temperature of several billion degrees was indicated for each set of isotopes in equilibrium, the required neutron concentrations varied over limits so wide as to preclude any conclusions concerning the freezing-in of a nuclear equilibrium under a single set of conditions. Nevertheless, they suggested that one should consider the entire observed abundance distribution of elements in determining the physical conditions of equilibrium. Singwi and Rai [133] have studied the isotope ratio problem in the same manner as Chandrasekhar and Henrich.

In his test of the equilibrium theory, van Albada used Eq. (21) in the following manner. Considering isotopes differing by  $\Delta N$  neutrons, one may write

$$\begin{array}{l} (C_{j'}/C_j)(C_n)^{-\Delta N} = (1/2)^{\Delta N} (\Phi_{j'}/\Phi_j) (2\pi m_0 kT/h^2)^{-(3/2)\Delta N} \\ \times [(A + \Delta N)/A]^{3/2} \exp[-(E_{j'} - E_j)/(kT)], \quad (24) \end{array}$$

or

$$\log C_n - (3/2) \log T = \mathbf{X} - \mathbf{Y}/T, \qquad (25a)$$

where

$$\begin{split} \mathbf{X} &= (3/2) \, \log(2\pi m_0 k/h^2) + \log 2 \\ &+ (\Delta N)^{-1} \{ \log(C_{j'}/C_j) - \log(\Phi_{j'}/\Phi_j) \\ &- (3/2) \, \log[(A + \Delta N)/A] \}, \end{split}$$
(25b) and

$$\mathbf{Y} = 0.434(-E_{j'} + E_j)/k. \tag{25c}$$

If one plots **Y** versus **X** for all pairs of isotopes for which binding energies and isotopic abundance ratios are known, then, if there is a single equilibrium condition for all pairs, the points should lie on a straight line whose slope gives the so-called freezing-in temperature and whose intercept on the **X** axis defines the neutron concentration. Such a plot was made by van Albada and the data were compared with the line defined by  $T=8\times10^{9}$ °K and  $\log C_n=29.30$ . In his opinion the scatter of the data was sufficient to preclude drawing the conclusion that the abundance distribution of nuclei corresponds to a nuclear equilibrium frozen in under one set of conditions. Singwi and Rai [133] as well as Jensen and Suess [92, 93] have constructed equivalent plots and arrived at the same conclusion.

Following a thermodynamic approach very similar to that of Urey and Bradley, Ubbelohde [160] has considered isotope equilibrium due to neutron absorption and emission only. One may relate the equilibrium constants of the system to the energy evolution and obtain for nuclei differing by one neutron,

$$\log[C_{j'}(Z, A+1)/C_{j}(Z, A)] = -4.7 \times 10^{12} (M_{j'} - M_{j})/T + \log(\Phi_{j'}/\Phi_{j}) + K_{e}, \quad (26)$$

in which

$$K_e = 2.303 \log[n] + 2.303 [-1.08 \times 10^{13} M_n/T + (3/2) - (3/2) \ln T] + a_1, \quad (26a)$$

the  $M_j$  are true mass numbers, and [n] is the neutron thermodynamic activity. In Eq. (26) the ratio of the thermodynamic activities of the two nuclear species has been taken with sufficient approximation as the ratio of the concentrations. The constant  $a_1$  depends on the neutron entropy as well as on universal constants. A plot of  $\log(C_{j'}/C_j)$  versus  $(M_{j'}-M_j)$  should again yield a straight line if the nuclear equilibrium corresponds to a single temperature. While Ubbelohde states that a freezing-in temperature of about  $10^{10^{\circ}}$ K is indicated, it is felt that the scatter of the data makes questionable the existence of a single set of equilibrium conditions and there does not seem to be any reason why his conclusion should differ from those of the many other investigators of this question.

In view of the difficulty of understanding the isotope ratios, Chandrasekhar and Henrich examined the equilibrium problem for two situations, namely, the equilibrium between protons, neutrons,  $\alpha$ -particles, electrons, and positrons, and in addition, the equilibrium between all kinds of nuclei, where abundances were studied as a function of atomic weight.<sup>†</sup> They obtained the physical conditions for an equilibrium calculation by considering the first situation. Those conditions under which protons and  $\alpha$ -particles were most abundant (thereby simplifying the problem of electrical neutrality) were then used to determine the abundances of the elements from O<sup>16</sup> to A<sup>40</sup>. Their best agreement was obtained with  $T=8\times10^{9}$  K,  $\log C_p=29.83$ ,  $\log C_n$ =29.3, and  $\log C_{\alpha}$ =30.3, or  $\rho$ =10<sup>7</sup> g/cm<sup>3</sup>. The general trend of the data up to about sulfur was reproduced, but beyond sulfur the agreement rapidly became poorer. Calculations were not carried past A = 40 for lack of adequate binding energy data.

A number of investigators have considered the problem of the formation of elements in studying the systematic properties of nuclei or the sources of stellar

TABLE II.<sup>a</sup> Equilibrium conditions for the isotopes of a given element.

Element	Т°К	$\log C_n$
O(16, 17, 18)	4.2×10 <sup>9</sup>	26.5
Ne(20, 21, 22)	$2.9 \times 10^{9}$	19.7
Mg(23, 25, 26)	$10.0 \times 10^{9}$	30.7
Si(28, 29, 30)	$12.9 \times 10^{9}$	31.2
S(32, 33, 34)	3.3×10 <sup>9</sup>	19.1

• For the masses and abundances used by Chandrasekhar and Henrich see reference [35].

 $\dagger$  The former phase of their work is considered in some detail in Section III(b)5.

energy. Guggenheimert suggested that since isotopic ratios are seemingly independent of the locale of observation, the elements must have been formed in equilibrium and consequently one should be able to compute nuclear binding energies from a knowledge of relative abundances. This latter point is also mentioned by Mattauch and Fluegge  $\lceil 111 \rceil$ . Discussions of the specific nuclear reactions of possible importance in the generation of energy in stellar interiors, and the formation of elements have been given by, among others, Atkinson [12], Atkinson and Houtermans [13], Bethe [19], Bethe and Critchfield [22], Steensholt [141], Strömgren [148], Walke [164–166], Wilson [180], and von Weiszäcker [176]. These and many other investigators have touched on the problem of element formation, in more or less detail, in considering stellar energy sources but a complete discussion of these studies would take us too far afield. However, it may be mentioned that Walke [164] suggested that in order to obtain sufficient abundances for the heavier elements, one must essentially abandon the equilibrium concept and consider a process of successive neutron captures by nuclei with intervening  $\beta$ -disintegrations to adjust the nuclear charge. Von Weizsäcker¶ pointed the way for much of the subsequent work on stellar energy sources and element formation. Failing to explain the synthesis of the elements beyond oxygen by proton reactions in stellar interiors he suggested that among the light element reactions there must exist one giving a continuing supply of neutrons for the synthesis of the heavier elements. A suggestion of this kind was also made by Döpel and Döpel [42]. The successful explanation of energy generation in main-sequence stars through the C-N cycle by Bethe, or the H-Hsequence of reactions by Bethe and Critchfield, demonstrated that elements beyond helium could not be continuously generated in anything like the observed abundances and the reactions found to be important did not include one providing a supply of neutrons.

Wataghin and his collaborators [104, 105, 155, 167-1747 have discussed various aspects of the calculation of the equilibrium abundances of the elements. For these calculations they have used an expression which is readily shown to be equivalent to those already discussed. One may rewrite Eq. (10) in logarithmic form, neglecting  $\Phi_j$ , and replacing  $E_j$ , the binding energy, by its equivalent in terms of mass differences, as

$$\log C_{j} = (3/2) \log(2\pi kT/h^{2}) + (3/2) \log m_{j} \\ + [(\log e)/(kT)] \{ (\mu_{n} + m_{n}c^{2})A + [(\mu_{p} - \mu_{n}) \\ - (m_{n} - m_{p})c^{2}]Z - m_{j}c^{2} \},$$
(27)

in which the m are in grams. If one sets

$$\alpha_{W} = -\left[(\mu_{p} - \mu_{n}) - (m_{n} - m_{p})c^{2}\right]\left[(\log e)/(kT)\right],$$
  

$$B_{W} = m_{0}c^{2}\left[(\log e)/(kT)\right],$$
(28)

and

$$D_W = (\mu_n + m_n c^2) [(\log e)/(kT)],$$

then one obtains

$$\log C_{j} = (3/2) \log(2\pi m_{0}kT/h^{2}) + (3/2) \log M_{j} + D_{W}A - \alpha_{W}Z - B_{W}M_{j}, \quad (29a)$$

or

$$\log(C_{j'}/C_j) = (3/2) \log(M_{j'} - M_j) + D_W(A' - A) -\alpha_W(Z' - Z) - B_W(M_{j'} - M_j), \quad (29b)$$

for absolute or relative concentrations, respectively. The  $M_i$  are true mass numbers (the mass numbers as usually tabulated less  $Zm_e$ , since measurements are usually given for neutral atoms) and  $m_j = m_0 M_j$ . Lattes and Wataghin [105] have applied Eq. (29b) to the detailed abundances between A = 16 and A = 40 and obtained general agreement for  $kT \cong 0.77$  Mev with physically reasonable values of  $\alpha_W$  and  $D_W$ . In the paper of de Toledo and Wataghin [155]\*\* it is reported that Eq. (29b) has been used successfully to represent the observed abundances over the entire range of atomic weights with sets of parameters such as (a), kT = 20Mev,  $\alpha_W = 1.16$ ,  $B_W = 20$ ,  $D_W = 20.385$ , or (b) kT = 40Mev,  $\alpha_W = 1.58$ ,  $B_W = 10$ ,  $D_W = 10.56$ . Since these results would seem to indicate that a simple equilibrium theory does provide an adequate explanation of the abundance data, they must be considered in more detail than has been indicated in the note of de Toledo and Wataghin. In particular the mass density and condition of electrical neutrality corresponding to the values of  $\alpha_W$ ,  $B_W$ , and  $D_W$  used must be examined. The present authors have calculated the absolute abundances of nuclei up to atomic weight 6, using the constants  $\alpha_W$ ,  $B_W$ ,  $D_W$ given by de Toledo and Wataghin in Eq. (29a), and find that the densities of matter corresponding to the sets (a) and (b) of constants are  $10^{15}$  and  $4 \times 10^{15}$  g/cm<sup>3</sup>, respectively. With all abundances added in, the densities would be even higher, perhaps by an order of magnitude, and would then be several orders of magnitude greater than nuclear density  $(2 \times 10^{14} \text{ g/cm}^3)$ .<sup>††</sup> In addition, calculation of the condition of electrical neutrality in these cases, using  $\mu_n$  and  $\mu_p$  determined from Eq. (28) in the manner previously described, indicates that the electron concentrations fall below those required for electrical neutrality by factors of 100 ln case (a) and 40 in case (b), respectively. It is evident from the densities computed that Eqs. (29) are not valid since they are based on classical statistics for

<sup>‡</sup> See Part II of reference [69]. ¶ For a discussion of von Weizsäcker's work see Chandrasekhar, " For a for a for a for the second of the se

a recent private communication which clarified portions of his work. We regret that it has not been possible to discuss further with Dr. Wataghin some of the questions raised in this review prior to its completion.

<sup>\*\*</sup> Note that in reference [155] the coefficient of (Z-Z') should have a minus sign.

tt The densities stated by de Toledo and Wataghin [155] would appear to be partial densities of protons; however, the proton abundance is less than the peak nuclear abundance in their distribution.



FIG. 5. Relative abundances as a function of atomic weight calculated for the light elements according to equilibrium theory with the parameters given by Klein, Beskow, and Treffenberg [99]. There is a computed point for A = 78 at -11.73 which is not indicated. The neutron concentration has not been included in the point for A = 1. The observed data, normalized to 10,000 atoms of silicon, are those of Brown [30].

nucleons and nuclei. Finally, at temperatures of the order of 20–40 Mev the effect of excited states, which has been neglected, might be expected to seriously alter the calculated equilibrium distribution.

Another treatment of the problem by Cherdyncev [36, 37] deals with the equilibrium of nuclei with  $\alpha$ -particles. Finding poor agreement for the heavier elements Cherdyncev considered an equilibrium between particles composed of four neutrons and nuclei composed entirely of neutrons, in a very dense neutron stellar core. In this case he obtained agreement with the observed trend in abundance with atomic weight.<sup>‡‡</sup> However, the possible existence of such particles must be questioned on physical grounds [see Section IV(e)].

Studies have also been made by Jensen and Suess [93] concerning the equilibrium abundances of the elements as a function of atomic weight. Their calculations involve the use of Eq. (21) and, of course, lead to the heavy element difficulty. The effect of excited states as well as the freezing-in problem have been discussed by these authors, and their work on these points is described later.

One of the most recent and complete calculations of the relative abundances of the elements on the basis of an equilibrium theory was made by Klein, Beskow, and Treffenberg [99]. Using the binding energy data tabulated by Mattauch and Fluegge [111] and ignoring the effects of spin multiplicity and excited nuclear states, they found the following values of the parameters in Eq. (11) to give the best representation of the observed abundances for the light elements:

$$\theta = kT = 1 \text{ Mev},$$
  

$$\mu_n = -7.6 \text{ Mev},$$
(30)

and

$$u_p = -11.6$$
 Mev.

The equilibrium abundances have been recomputed for

the stable nuclei by the authors with binding energies taken from Rosenfeld's tabulation [125] and with the values of  $\mu_n$ ,  $\mu_p$ , and  $\theta$  given in Eq. (30). The results for the light elements are compared in Fig. 5 with the observed abundances given by Brown [30]. The relative abundance of neutrons is not added to that computed for protons, as was done in the various calculations of Klein and collaborators. The general agreement with increasing atomic weight up to about A = 40 is satisfactory. However, there appear to be significant deviations between theory and observation with regard to the detailed variations of abundance as a function of atomic weight. This is particularly noticeable in that the abundance peak near iron is not reproduced by the theory. It is interesting to note that the computed abundances of nuclei with very high or very low values of the isotopic number  $\Delta = N - Z$  are, respectively, high or low. This might be expected at equilibrium with a high neutron concentration when the formation of neutron-rich nuclei would be favored. Klein, Beskow, and Treffenberg suggest that these nuclei in particular would undergo change in the freezing-in of the equilibrium. It is difficult to see why this should be the case, without a more detailed examination of the complex freezing-in problem [see Section III(c)]. Beyond  $A \cong 40$  the theory predicts much smaller abundances than are observed. In Fig. 6 theoretical abundances, computed as in Fig. 5, are again compared with the observed abundances, but on such a scale that the calculations for higher atomic weight can be shown. In this graph the observed abundances have been represented by two straight lines fit by least squares for  $1 \leq A \leq 100$  and  $100 \leq A \leq 238$ , in order to prevent confusion of observed and computed datum points. For the heavier elements, the computed abundances are extraordinarily small compared to the observational data. This difficulty of the simple equilibrium theory is well known, and has stimulated in-



FIG. 6. Logarithm of relative abundance versus atomic weight, calculated as in Fig. 5, compared over the entire range of atomic weight with observed abundances represented for clarity by two straight lines.

<sup>&</sup>lt;sup>‡‡</sup> See Part III of reference [36].

vestigators to consider various modifications in the basic equilibrium theory. It should be pointed out that this disagreement does not arise from the choice of particular parameters,  $\mu_n$ ,  $\mu_p$ , and  $\theta$ , but rather from the fact that the theory in the simple form thus far presented cannot simultaneously reproduce the observed abundances of light and heavy elements under physically reasonable conditions.

To illustrate qualitatively the nature of the simple equilibrium theory, let Eq. (11) be rewritten in logarithmic form, taking as a crude approximation for the light elements  $Z \cong a_1 A$  and  $E_j \cong -a_2 A$ , where  $a_1$  and  $a_2$  are constants. Then one obtains

$$\ln C_{j} = \left[ A/(kT) \right] \left[ (1-a_{1})\mu_{n} + a_{1}\mu_{p} + a_{2} \right] + (3/2) \ln m_{j} + (3/2) \ln(kT) + \text{constant.}$$
(31)

For a particular value of kT, say 1 Mev, one may fit this relation to the observed abundances in the region of low atomic weight and determine the values of  $\mu_n$ and  $\mu_p$ . It may be noted that, since the second term,  $(3/2) \ln m_j$ , is a slowly varying function of A, to this approximation

$$\ln C_i \propto A.$$
 (31a)

This result, also indicated by Jensen and Suess [93], demonstrates the essentially exponential decrease in computed abundances [57] evident in Fig. 6. The use of actual Z and  $E_j$  values makes the decrease even more rapid, particularly for the heavy elements. Clearly one could find values of  $\mu_n$ ,  $\mu_p$ , and  $\theta$  which would best fit the heavy elements, but the computed light element abundances under these conditions would then be scarce as compared to those observed. As already mentioned, the physical conditions required in fitting light and heavy element abundances are quite different. Beskow and Treffenberg [18] have pointed out that



FIG. 7. Electron concentration,  $C_{e^-}$ , as a function of the electron intrinsic potential,  $\mu_{e^-}$ , according to Fermi-Dirac statistics, Eq. (35), reproduced from the work of Klein, Beskow, and Treffenberg [18, 99]. This graph can also be used for computing positron concentrations.

for every choice of  $\mu_n$  only nuclei in a rather narrow range of atomic weights will be important and that the larger  $\mu_n$ , the larger the atomic weight of the predominant nuclei.

From the fit obtained to the light elements, Klein, Beskow, and Treffenberg have computed the density of matter as

$$\rho = \sum_{j} m_{j} C_{j}, \qquad (32)$$

where the concentrations of protons, neutrons, and nuclei are included in the summation. The density is determined in this particular case by  $C_n$ ,  $C_p$ , and  $C_\alpha$ , all other concentrations being negligible by comparison, and is found to be  $\sim 4 \times 10^8$  g/cm<sup>3</sup>. For the  $\mu_n$ ,  $\mu_p$ , and  $\theta$ taken, these authors have found, using Eq. (11), that

$$C_p/C_n = \exp[-(\mu_n - \mu_p)/(kT)] \cong 0.018.$$
 (33)

In calculating the electron concentration as required to check the validity of the  $\mu_n$ ,  $\mu_p$ , and  $\theta$  values insofar as electrical neutrality of the assembly is concerned, Klein, Beskow, and Treffenberg have considered the neutrino. Assuming equality of the electron and neutrino intrinsic potentials one may write that ¶¶

$$\mu_{e^{-\prime}} = (\frac{1}{2})(\mu_{n} + m_{n}c^{2}) - (\frac{1}{2})(\mu_{p} + m_{p}c^{2}), \qquad (34)$$

where

$$\mu_{e}-'=\mu_{e}-+m_{e}c^{2}.$$
 (34a)

The electron concentration can be calculated using Fermi-Dirac statistics from

$$C_{e^{-}} = (8\pi/h^3) \int_0^\infty \frac{p^2 dp}{\exp\{\theta^{-1} [E(p) - \mu_{e^{-}}]\} + 1}, \quad (35)$$

where

$$E(p) = c(m_e^2 c^2 + p^2)^{\frac{1}{2}}, \qquad (35a)$$

and p is the momentum of the electron. The evaluation of Eq. (35) by Klein, Beskow, and Treffenberg [18, 99] yields the plot of  $C_{e^-}$  versus  $\mu_{e^-}$  for  $\theta = 1$  Mev only, reproduced in Fig. 7.|||| For the values of  $\mu_n, \mu_p$ , and  $\theta$ discussed, they found  $C_{e^-}/C_a \cong 2.8$ , which indicates approximate electrical neutrality because helium is by far the most abundant element according to the theory (see Fig. 5).

It is clear from the foregoing discussion that while the simple equilibrium theory might be considered satisfactory for the light or heavy elements separately, it fails to yield a reasonable over-all explanation of the

#### $C_{e^{-}} = [8\pi/(3h^3c^3)][1 + \pi^2\theta^2/(\mu_{e^{-}}')^2](\mu_{e^{-}}')^3.$

For a development of this result see reference [109], p. 213. If  $\mu_e'$  is small or negative, Eq. (35) must be integrated numerically.

<sup>¶¶</sup> In the work of Klein, Beskow, and Treffenberg the electron intrinsic potential has been defined to include the electron rest mass. Note that their  $\mu$  and  $\lambda$  are referred to rest mass as a zero point while their  $\mu_n$  and  $\mu_p$  include the rest-mass energy.

point while their  $\mu_n$  and  $\mu_p$  include the rest-mass energy.  $\| \|$  As pointed out in reference [99], if  $\mu_{e^{-1}}$  is large compared to E(p) the electrons are degenerate and Eq. (32) may be integrated approximately to give

observed relative abundance data.\* However, Klein and his collaborators have successfully modified the simple equilibrium theory by taking into account degeneracy where required as well as gravitational effects in a special stellar model [see Section III(b)5], and have computed abundances over the entire range of atomic weights.

Steinwedel and Jensen [142] recently have examined in considerable detail the complete equilibrium problem, including relativistically degenerate statistics for the electron, electron-positron equilibrium, and the effect of the neutrino, but not the effect of a gravitational field.<sup>†</sup> They confirm, on the basis of their analysis, the earlier conclusion of Jensen and Suess [93] that the present element distribution must principally reflect non-equilibrium processes in an expanding and cooling universe rather than some initial equilibrium distribution. Steinwedel and Jensen also point out that the conditions required by an equilibrium theory are not compatible with the presently observed abundance distribution if one requires that the elements now are the result of freezing-in processes.

There have been suggested a variety of specific modifications of the simple theory in order to repair the difficulties noted and these are considered in the following sections.

# 3. Effect of Excited Nuclear States

In the calculations of equilibrium abundances thus far described, the effect of excited nuclear states has been neglected, although the spin multiplicity of the elementary particles has been included. Chandrasekhar and Henrich [35] have pointed out that since the density of excited nuclear levels is known to increase with increasing atomic weight, one should expect an increase in the relative abundances of the heavier elements if the excited states are taken into account. Quantitative calculations of abundances with excited states included have been made independently by Unsöld [162], Beskow and Treffenberg [17, 18], and Géhéniau, Prigogine, and Demeur [63]. The effect of excited states has also been discussed qualitatively

by Jensen and Suess [93] who concluded that there is little gain in the agreement of theory and data due to this factor.

In order to take into account the effect of excited states of nuclei one should include in the equation for the absolute concentration of a given kind of nucleus a factor,  $\Phi_j$ , representing the partition sum for all the internal degrees of freedom. Thus, in Eq. (10),  $\Phi_j$  is interpreted according to the nuclear model used. Unsöld [162] considered the rotational energy states for nuclei treated as rigid spheres, neglecting nuclear spin multiplicity as well as the vibrational energy states. The sum over the rotational states is

$$\Phi_{j}(\operatorname{rot}) = \sum_{r} \omega_{r} \exp(-\epsilon_{r}/kT) = \sum_{J=0}^{\infty} (2J+1)^{2} \\ \times \exp[-J(J+1)h^{2}/(8\pi^{2}I_{j}kT)], \quad (36)$$

where the moment of inertia  $I_j$  is given by

$$I_j = (2/5)m_j R_j^2. \tag{36a}$$

If the spin multiplicity is neglected, this sum is approximately

$$\Phi_j(\text{rot}) \cong \pi^{\frac{1}{2}} (8\pi^2 I_j k T/h^2)^{\frac{3}{2}}.$$
 (37)

Since  $m_j \propto R_j^3$  and  $R_j \propto A_j^{\frac{1}{2}}$ , it is clear that the partition sum is proportional to  $A^{5/2}$ . Unsöld found no material improvement in the agreement of computed and observed abundances resulting from the inclusion of rotational states.

In a continuation of their study of the equilibrium theory, Beskow and Treffenberg have considered both the vibrational and rotational energy states of nuclei. In their approximation the partition sum is taken to be

$$\Phi_{j}(N, Z) = \Phi_{j}(\operatorname{rot})\Phi_{j}(\operatorname{vib})$$
  
=  $\sum_{\sigma} \omega_{r} \exp(-\epsilon_{r}/kT) \sum_{\sigma} \omega_{v} \exp(-\epsilon_{v}/kT), \quad (38)$ 

where  $\omega_r$  and  $\omega_v$  are the statistical weights of the rotational and vibrational states, respectively. Treating the nucleus as a rigid sphere, the sum over rotational states was taken to be the same as that used by Unsöld. For the higher vibrational energy states, Beskow and Treffenberg have made use of the calculation of the calculation of the distribution of vibrational energy levels by Bohr and Kalckar [23], and by van Lier and Uhlenbeck [107]. The level density in this case is given by [17]

$$\rho_{\epsilon,A}d\epsilon = (48\epsilon^2)^{-\frac{1}{2}} \exp\{\pi [2\epsilon A/(3\epsilon_0)]^{\frac{1}{2}}\}d\epsilon, \qquad (39)$$

where  $\epsilon$ , the vibrational excitation energy, is given in Mev. The application of this result leads to the following approximate expression for the sum over the vibrational energy states:

$$\Phi_{j}(\text{vib}) = \sum_{v} \omega_{v} \exp(-\epsilon_{v}/kT)$$
$$= 1 + 6.93 \int_{x_{0}}^{\infty} x^{-1} \exp[-x + (x\delta)^{\frac{1}{2}}] dx, \quad (39a)$$

<sup>\*</sup> Demeur [39] purports to show that increasing the temperature of the equilibrium distribution will not sufficiently increase the concentration of the heavy elements. His conclusions are not understood since it is possible by a suitable choice of the density and temperature to make either the light or heavy elements predominantly abundant, see references [18, 79, and 143]. In fact, for a given temperature the higher the density the more the heavier elements are favored.

<sup>†</sup> In their paper, Steinwedel and Jensen present their results in the form of graphs showing: (a) the neutron concentration as a function of the neutron-proton concentration ratio at various temperatures; (b) total concentration of nucleons, free and bound, versus temperature for various neutron-proton concentration ratios, for various ratios of free to bound nucleons, and for the case where the *a*-particle abundance is half that of protons; and, (c), the same as (b) except that the curves for various neutronproton ratios are replaced by curves of constant entropy. In these graphs they have indicated the regions in which the heavier nuclei are predominantly abundant. While these calculations include neutrinos, they find neutrino effects to be small.

TABLE III. Values of the vibrational partition function at 1 Mev for various values of A.

A	50	100	150	200	250
$\log \Phi_i$	2.6	6.1	9.6	13.1	16.5
$\log \Phi_i'$	2.2	2.6	2.9	3.2	3.5

01100

where

$$x_0 = \pi^2 / (3\delta),$$
  
$$\delta = (2\pi^2 A k T) / (3\epsilon_0), \qquad (39b)$$

and where the energy difference between the lowest states has been assumed to vary as  $\epsilon_0/A$ . If kT=1 Mev, and  $\epsilon_0=10$  Mev, then  $\delta \cong 2A/3$ . Equation (39) shows the sum over the vibrational energy states to be an increasing function of atomic weight.

Taking the rotational and vibrational energy states into account, as above, and using the same values of  $\mu_n$ ,  $\mu_p$ , and  $\theta$  as in Eq. (30), Beskow and Treffenberg have recomputed the equilibrium concentrations of the elements. The region of agreement in general trend is extended to somewhat higher atomic weights (perhaps up to  $A\cong60$ ). In the region  $A\cong140$  abundances are increased by about  $10^{10}$  and for the heaviest elements this factor would be considerably larger, perhaps  $10^{20}$ . However, the abundances of the heavy elements are still by no means explained, since the factor required for the heavy elements is at least  $10^{100}$ . None of the deviations in detail, such as are evident in Fig. 5, are materially altered since the approximations taken for  $\Phi_j$  increase smoothly with increasing atomic weight.

Recently, ter Haar [74] has reinvestigated the evaluation of the nuclear vibrational partition function  $\Phi_j(\text{vib})$ , on the basis of an evaluation of level densities in nuclei by Wergeland.<sup>‡</sup> It is reported that Wergeland, using the liquid drop model of the nucleus, obtained for the level density,  $\rho_{\epsilon,A}$ , the following expression,

$$\rho_{\epsilon,A} d\epsilon = 1.14 A^{1/7} \epsilon^{-5/7} \exp(0.9A^{2/7} \epsilon^{4/7}) d\epsilon, \qquad (40)$$

where  $\epsilon$  is the excitation energy in Mev. D. ter Haar states that Eq. (40) gives better agreement with experimental data on level spacing than do the results of the work of Bohr and Kalckar, and van Lier and Uhlenbeck, although no experimental data are presented for verification. The vibrational partition sum is obtained, by use of Eq. (40), as

$$\Phi_{j}' = 1 + \int_{(\epsilon_{0}/2A)}^{\infty} \rho_{\epsilon,A} \exp[-\epsilon/(kT)] d\epsilon, \qquad (41)$$

# $\rho_{\epsilon, A} d\epsilon = 1.24 A^{-1/7} \epsilon^{-5/7} \exp(0.72 A^{2/7} \epsilon^{4/7}) d\epsilon$

where, as before,  $\epsilon_0 = 10$  Mev. In Table III,  $\Phi_j'$  is compared with  $\Phi_j$  from Eq. (39).¶ If  $\Phi_j'$  is more nearly correct than  $\Phi_j$ , then Beskow and Treffenberg have indeed overestimated the influence of excited states. Since the heavy element "catastrophe" is by no means repaired by the inclusion of excited states, this difference is unimportant in itself.\* However, as will be discussed in Section III(c), this difference may be very important both in considering an equilibrium in special stellar models [74] and in the subsequent freezing-in problem.

The factors required to repair the heavy element difficulty are indeed enormous. In order to repair the discrepancy Géhéniau, Prigogine, and Demeur [63] have suggested that one take

$$\Phi_j = (2i_j + 1)(A!)^2, \tag{42}$$

where A is the atomic weight. They suppose that  $(A !)^2$ represents the number of ways of realizing the fundamental state of the nucleus for a given value of the projection of the spin along an axis. Aside from the fact that the indistinguishability of individual particles in the nucleus, according to quantum theory, would not allow this interpretation of  $\Phi_j$ , the inclusion of this factor does not remove the difficulty. In fact their fit of calculated abundances to the heavy elements leads to a great relative overabundance of the light elements.

## 4. Degeneracy and Electrostatic Effects

As has been seen, the inclusion of excited nuclear states in the formulation of the equilibrium theory does not in itself lead to the necessary improvement in the calculated abundances of the heavy elements. Various investigators have suggested that the heavy elements require quite different physical conditions for their formation, certainly a higher density, and perhaps a higher temperature, than is required for the light elements. A higher temperature implies a higher reaction rate with charged particles and would favor larger equilibrium abundances for the heavier elements. Since the physical conditions required for light element formation are already severe, it was suggested by van Albada [1, 2] and by Hoyle [79, 80] that electron degeneracy and electrostatic effects be taken into account in discussing the formation of the heavy elements. Both investigators state that taking electron degeneracy into account lowers the Z/A and increases the A of the most stable nucleus (that nucleus having the maximum binding energy per nucleon), thereby indicating the improvement of the equilibrium theory for the heavy elements. Neither investigator has actually computed relative abundances according to these

<sup>&</sup>lt;sup>‡</sup> D. ter Haar [74] gives as a reference for the work of Wergeland, Fra Fysik. Verden, 223 (1945). Since we have not been able to obtain this journal it is not possible to discuss in what respects Wergeland's results differ from those of other work also based on a liquid drop model. However, we wish to point out that Devons, reference [40], p. 139, gives for the level density on the basis of a liquid drop model

where the units are as in Eq. (40) of the present paper. This expression is very similar but not identical to that attributed to Wergeland.

<sup>¶</sup> D. ter Haar [74] has recomputed  $\Phi_i$  and obtains good agreement with Beskow and Treffenberg [17].

<sup>\*</sup> In a private communication, G. Beskow has informed us of a preliminary calculation which shows that a small change in the neutron and proton chemical potentials will correct for an altered vibrational partition function without upsetting such auxiliary conditions as electrical neutrality.

modified physical conditions. However, as already mentioned, Beskow and Treffenberg [18] have perhaps explained the existence of the heavy elements in special stellar models by taking into account not only degeneracy effects but also the effect of the gravitational potential on the equilibrium. They have calculated abundances and their work is discussed in detail in the following section.

Let us consider that the electrons present in the equilibrium assembly are relativistically degenerate. The partition function for such electrons is given by [143]

$$\mathcal{O}_e = \left[ 2\pi V k^3 T^3 / (3h^3 c^3) \right] \left[ (\ln \lambda)^4 + 2\pi^2 (\ln \lambda)^2 + \cdots \right], \quad (43)$$

if  $kT \ln \lambda \gg m_e c^2$  and  $\lambda \gg 1$ . In Eq. (43) the various quantities are as defined previously in Section III(b)2. The mean number of these electrons,  $\bar{X}_{e}$ , can be obtained from  $\bar{X}_e = \lambda(\partial \mathcal{P}_e/\partial \lambda)$  and written in the form

$$\bar{X}_e = \left[ \frac{8\pi V k^3 T^3}{(3h^3c^3)} \right] \left[ (\ln\lambda)^3 + \pi^2 (\ln\lambda) + \cdots \right].$$
(44)

From Eq. (44) it follows that

$$\ln\lambda = \left[\frac{hc}{(2kT)}\right] \left[\frac{3\bar{X}_{e}}{(\pi V)}\right]^{\frac{1}{2}} - \left[\frac{2\pi^{2}kT}{(3hc)}\right] \left[\frac{3\bar{X}_{e}}{(\pi V)}\right]^{-\frac{1}{2}} + \cdots$$
(45)

The mean total energy  $\bar{E}_e$  of all the free relativistically degenerate electrons is

$$\bar{E}_{e} = kT(\partial \mathcal{O}_{e}/\partial T) = \left[2\pi V k^{4} T^{4}/(h^{3}c^{3})\right] \left[(\ln\lambda)^{4} + 2\pi^{2}(\ln\lambda)^{2} + \cdots\right], \quad (46)$$

or in terms of  $\bar{X}_{e}$ ,

$$\bar{E}_{e}/V = (\pi hc/8) [3\bar{X}_{e}/(\pi V)]^{4/3} \\
+ [\pi^{3}k^{2}T^{2}/(3hc)] [3\bar{X}_{e}/(\pi V)]^{2/3} + \cdots . \quad (46a)$$

If the electron concentration at a given temperature is sufficiently high, the second term in Eq. (46a) can be neglected.

According to Hoyle  $\lceil 79 \rceil$  electron degeneracy can be introduced into the term  $Z \log(C_n/C_p)$  in Eq. (23). From Eqs. (15) and (19), one obtains

$$C_n/C_p = \lambda (m_n/m_p)^{\frac{1}{2}} \exp[-c^2(m_n-m_p-m_e)/(kT)].$$
 (47)

Neglecting the second term on the right-hand side of Eq. (45),

$$\ln\lambda \cong [hc/(2kT)][3\bar{X}_{e}/(\pi V)]^{\frac{1}{2}}, \qquad (45a)$$

so that Eq. (47) becomes, with  $(3/2) \ln(m_n/m_p)$  taken equal to zero,

$$\ln(C_n/C_p) = [m_e c^2/(kT)]y, \qquad (48)$$

where

$$y = x - (m_n - m_p - m_e)/m_e,$$
 (48a)

and

$$x = [(3h^{3}C_{e^{-}})/(8\pi m_{e}^{3}c^{3})]^{\frac{1}{2}}.$$
 (48b)

In terms of T in units of  $10^9$  °K, Eq. (48) becomes

$$\log(C_n/C_p) = (4.73/T_9)(0.543y).$$
(48c)

Using this result in Eq. (23) one can solve for  $\log C_n$  as

$$\log C_{n} = 34.08 + (3/2)\log T_{9} - (4.73/T_{9})(|E_{j}|/A - 0.543yZ/A) + (1/A)[\log(AC_{j}) - 34.08 - (3/2)\log T_{9} - (5/2)\log A],$$
(49)

where  $E_j$  is in units of mMU. For a given A one may find the charge Z and the density at which the nucleus (Z, A) will be the most stable. If the temperature were sufficiently low this nuclear species would be the most abundant in an equilibrium theory. The most stable nucleus, according to Hoyle's discussion, is that for which

$$\partial E_j^* / \partial A = \partial E_j^* / \partial Z = 0,$$
 (50)

$$E_j^* = (|E_j| - 0.543yZ)/A.$$
 (50a)

Physically this result is presumably equivalent to modifying the binding energy per nucleon. Hoyle has employed the empirical binding energy formula given by Weizsäcker in the form:

$$|E_j|/A = 14.9 - 21[(A - 2Z)/A]^2 - 14.2A^{-1/3} - 0.625Z^2A^{-4/3}, \quad (51)$$

in which  $E_i$  is in mMU. From the conditions for an extremum one may obtain

$$Z_m = (A_m/2)(1 - 6.46 \times 10^{-3} y_m) \times (1 + 7.44 \times 10^{-3} A_m^3)^{-1}, \quad (52)$$
  
and

$$y_m = 154.80 - 1043.5A_m^{-\frac{1}{2}}(1 + 7.44 \times 10^{-3}A_m^{\frac{2}{3}}),$$
 (52a)

where  $Z_m$  and  $A_m$  make  $E_j^*$  a maximum for a given  $y_m$ . For a given  $A_m$  one may compute from Eq. (52) the corresponding  $y_m$ . Since from Eq. (48a) the corresponding x may be found and since x determines the electron concentration  $C_{e^-}$  from Eq. (48b), one may compute the density of matter excluding neutrons,  $\rho$ , providing again that the most stable nuclear species is also predominantly abundant. If this is the case, then,

$$C_{e} \simeq (Z_{m} \rho) / (A_{m} m_{p}), \qquad (53)$$

which fixes  $\rho$ . This procedure has been used in preparing Table IV for various values of  $A_m$ .\*\* As may be seen from Table IV, the atomic weight of the most stable nuclear species shifts to higher values as the density increases. These nuclei are quite neutron-rich,

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Am	$Z_m$	$E_{j}^{*}(\max)$	$\log \rho$
75	29.2	4.94	10.23
110	35.4	1.09	11.25
130	38.4	-0.16	11.52
160	42.6	-1.44	11.78
180	45.2	-2.06	11.91
240	52.2	-3.27	12.16

|| See reference [79].

\*\* The values given in Table IV differ slightly from those given by Hoyle in reference [79], Table XI, because the constant term in the first of his Eqs. (48) should be 6.1 rather than 5.1.

and

where

A	ZA	Ζ	logp	Log <i>p</i> corrected
60	27.1	26.9	6.69	6.69
80	35.3	32.0	10.00	9.98
100	43.3	36.6	10.67	10.63
120	51.1	40.8	11.03	10.91
140	58.6	44.9	11.26	11.07
160	65.8	48.6	11.45	11.20
180	73.0	52.3	11.57	11.28
200	80.0	55.8	11.68	11.34
220	86.8	59.1	11.77	11.37
240	93.6	62.5	11.84	11.39

TABLE V.

as might be expected from the fact that at these densities the neutron concentration is high. The physical interpretation of  $E_j^*$  is not clear. This is particularly so when one attempts to determine whether or not the neutrons present at the highest densities, where  $E_j^*$ is negative, are degenerate. The application of the usual criteria<sup>††</sup> would indicate that, for all densities in Table IV where  $E_j^*$  is negative, one has appreciable neutron degeneracy. If this is the case, then it is necessary to discuss the equilibrium problem with degenerate neutrons as well as degenerate electrons.

A somewhat different approach to the problem of the existence of the heavy elements is that of van Albada [1, 2]. His approach is a more physical one, and is given here with some modifications which it is hoped will clarify the development. Let us consider the energy per nucleon in an assembly consisting of one nuclear species (Z, A) and of electrons. It is assumed that the electrons are relativistically degenerate, that electrical neutrality is maintained locally, and that the temperature is constant and extremely low. It is necessary to consider the energy per nucleon because it is tacitly assumed that the number of nucleons is conserved. The mean energy per nucleon may be considered as being made up of mass energy, thermal energy, energy of the degenerate electrons, and energy associated with electron interactions. The mass energy per nucleon may be written, in mMU, as

$$\mathcal{E}_m = 1000 + f,\tag{54}$$

where f is the packing fraction, in mMU, of the nucleus (Z, A) under consideration. van Albada has obtained an improved version of the semi-empirical packing fraction formula of Bohr and Wheeler [24], developed in their study of nuclear fission. The improved formula for f is given, in mMU, by:

$$f = 83[(A - 2Z)/(2A)]^{2} + 0.8[(A - 2Z)/(2A)] -6.65 + 0.63Z^{2}A^{-4/3} + 15A^{-1/3}, \quad (55)$$

which includes terms for nucleon interaction, Coulomb energy of the nucleus, and surface tension. From Eq. (46a), the energy per unit volume,  $\mathcal{E}_d'$ , of the assembly

of relativistically degenerate electrons is given for high density and low temperature in mMU by

$$\mathcal{E}_{d} \cong (\pi hc/8) (3C_{e^{-}}/\pi)^{4/3} [10^{3}/(m_{0}c^{2})],$$
 (56)

where  $C_{e^-}$  is the electron concentration and  $m_0$  is the unit of atomic mass. The energy of the degenerate electrons, per nucleon, in mMU, is

$$\mathcal{E}_d = [m_j/(A\rho)] \mathcal{E}_d', \qquad (56a)$$

where  $\rho$  is the density of matter in the assembly, and  $m_j$  is the mass of the nucleus (Z, A). If one assumes local electrical neutrality, then,

$$C_{e^{-}} = ZC_{j} = Z(\rho/m_{j}) = Z\rho [Am_{0}(1+10^{-3}f)]^{-1}, \quad (57)$$

so that, in mMU per nucleon,

$$\mathcal{E}_{d} = (\pi hc/8)(10^{3}/c^{2})(3/\pi)^{4/3}(Z/A)^{4/3}\rho^{1/3}m_{0}^{-4/3} \times (1+10^{-3}f)^{-1/3} \quad (58)$$

$$=q\rho^{1/3}(Z/A)^{4/3}(1+10^{-3}f)^{-1/3},$$
(58a)

where

$$q = (\pi hc/8)(10^3/c^2)(3/\pi)^{4/3}m_0^{-4/3} = 4.15 \times 10^{-3}.$$
 (58b)

If the electrons are uniformly distributed, then there are Z electrons in a sphere of radius  $r_Z$ , where

$$(4\pi/3)C_{e}-r_{Z}^{3}=Z.$$
 (59)

Considering the nucleus to be a point charge, one finds the energy per unit volume,  $\mathcal{S}_{e'}$ , due to nucleus-electron attraction and mutual electron repulsion may be written, in mMU, as

$$\mathcal{E}_{e}' = [10^{3}/(m_{0}c^{2})][-(3Z^{2}e^{2})/(2r_{Z}) + (3Z^{2}e^{2})/(5r_{Z})]C_{j}. \quad (60)$$

Using Eq. (57), one may obtain

$$\mathcal{E}_{e}' = - (9/10) Z^{2} e^{2} [10^{3}/(m_{0}c^{2})] \times (4\pi/3)^{1/3} m_{0}^{-4/3} \rho^{4/3} A^{-4/3} (1+10^{-3}f)^{-4/3}.$$
(60a)

Again per nucleon this energy can be written in mMU as

$$\mathcal{E}_e = \left[A m_0 (1 + 10^{-3} f) / (A \rho)\right] \mathcal{E}_e' \tag{61}$$

$$= -p\rho^{1/3}Z^2A^{-4/3}(1+10^{-3}f)^{-1/3}, \qquad (61a)$$

where

$$p = (9/10)(10^3/c^2)e^2(4\pi/3)^{1/3}m_0^{-4/3} = 1.91 \times 10^{-5}.$$
 (61b)

The inclusion of the term  $\mathcal{E}_e$  implies a sufficiently high density such that the electrons are confined to a small region surrounding the nuclei, and the attraction between nuclei and electrons may be regarded as essentially reducing the electrostatic term in the packing fraction formula. The ordinary binding between electrons and nuclei is implicitly contained in the packing fraction f. Energy due to the interaction between nuclei as well as between nuclei and more than the surrounding Z electrons is neglected.<sup>‡‡</sup> Combining all the

<sup>&</sup>lt;sup>††</sup> The criterion employed is that for weak degeneracy, such as is given in reference [109]. The extent of the degeneracy is given by  $\rho_n h^3 / [2^{\frac{1}{2}}m_n(2\pi m_n kT)^{\frac{3}{2}}]$  as compared to unity.

<sup>&</sup>lt;sup>‡‡</sup> As pointed out to us by Professor O. Klein, it would seem that the conditions here are more like those in a crystal than an

various contributions, one finds that the total energy per nucleon in this assembly, expressed in mMU, can be given by the following expression, provided thermal energy may be neglected:  $\P$ 

$$\begin{split} &\mathcal{E} = 10^3 + 83 [(A - 2Z)/(2A)]^2 \\ &+ 0.8 [(A - 2Z)/(2A)] - 6.65 + 0.63Z^2 A^{-4/3} \\ &+ 15A^{-1/3} - p\rho^{1/3}Z^2 A^{-4/3} (1 + 10^{-3}f)^{-1/3} \\ &+ q\rho^{1/3} (Z/A)^{4/3} (1 + 10^{-3}f)^{-1/3}. \end{split}$$

If  $\mathcal{E}_{\epsilon}$  and  $\mathcal{E}_{d}$  are sufficiently small, one may neglect  $(10^{-3}/3)f$  as compared to unity in Eq. (62).

Inspection of Eq. (62) indicates that  $\mathcal{E}$  is a function of Z, A, and the density  $\rho$ . From  $d\mathcal{E}=0$  one may determine the values of A and Z for the nucleus which would lead to the lowest specific energy of the assembly at a given density. From combinations of  $\partial \mathcal{E}/\partial Z$  $=\partial \mathcal{E}/\partial A=0$ , the following equations can be derived:

 $A^2/Z^2 = 8.4 \times 10^{-2} B_o A_1$ 

and

$$A/Z = 1.98 + 1.52 \times 10^{-2} B_{\rho} A^{\frac{3}{2}}$$

where

$$B_{\rho} = 1 - 3.03 \times 10^{-5} \rho^{\frac{1}{3}}.$$
 (63c)

 $+2.89 \times 10^{-5} (B_{\rho} \rho A)^{\frac{1}{2}},$  (63b)

These equations have been solved by van Albada and his results are given in Table V, in which the first two columns give the atomic weight and the "normal" charge  $Z_A$ . The latter quantity has been calculated from the packing fraction formula given in Eq. (55) and smoothed with respect to Z for a given A. The remaining columns are interpreted as follows. A nuclear species of atomic weight A will provide the lowest specific energy content at a density  $\rho$ , when its charge is reduced to the value Z. It will be recalled that  $\rho$  refers to the density of this particular nuclear species and does not include neutrons, and that if thermal energies are small compared to  $\mathcal{E}$ , the most stable nuclear species will also be the most abundant.

The question still remains as to whether it has been a sufficient approximation in the foregoing development to neglect the presence of neutrons. To examine this point van Albada has considered the energy per nucleon in an assembly consisting of one kind of nucleus, degenerate electrons and free neutrons at  $T \cong 0$ °K. Let  $\phi_0$  be the fraction of nucleons bound in nuclei and  $(1-\phi_0)$  the fraction of nucleons (neutrons) which are free. Then the energy per bound nucleon, in mMU, will be

$$\phi_0[(10^3 + f) + \phi_0^{\frac{1}{3}}(\mathcal{E}_e + \mathcal{E}_d)], \tag{64}$$

where the  $\phi_0^{\dagger}$  arises from the conversion to partial density in the expressions for  $\mathcal{E}_d$  and  $\mathcal{E}_e$  which contain  $\rho^{\dagger}$ . The additional energy due to free neutrons is, in mMU,

$$(1-\phi_0)(10^3+f_n),$$
 (65)

where  $f_n$  is the packing fraction of the neutron. In this mixed assembly the energy per nucleon can then be written as

$$\mathcal{E}' = \phi_0 f + \phi_0 (\phi_0^{\frac{1}{3}} \mathcal{E}_d + \phi_0^{\frac{1}{3}} \mathcal{E}_e) + (1 - \phi_0) f_n.$$
(66)

Differentiation with respect to Z, A, and  $\phi_0$  yields

$$\phi_0 \frac{\partial f}{\partial Z} + \phi_0^{4/3} \frac{\partial}{\partial Z} (\mathcal{E}_d + \mathcal{E}_e) = 0, \qquad (67a)$$

$$\phi_0 \frac{\partial f}{\partial A} + \phi_0^{4/3} \frac{\partial}{\partial A} (\mathcal{E}_d + \mathcal{E}_e) = 0, \qquad (67b)$$

and

(63a)

$$f + (4/3)\phi_0^{\frac{1}{3}}(\mathcal{E}_d + \mathcal{E}_e) - f_n = 0.$$
 (67c)

Free neutrons will become important at densities above a value which satisfies Eq. (67c) for  $\phi_0 = 1$ . This critical density,  $\rho_c$ , according to van Albada, is given by  $\log \rho_c$ = 11.70. Above this density free neutrons will be formed rather than heavier nuclei. Hence from Table V one finds that nuclei above  $A \cong 200$  cannot be explained. G. B. van Albada suggests circumvention of this difficulty by the addition of a nuclear volume correction to the packing fraction of the form

$$\Delta f = -166[(A - 2Z)/(2A)]^4 + [20A^{-1/3} - 0.42Z^2A^{-4/3}][(A - 2Z)/(2A)]^2. \quad (68)$$

Physically this correction arises from the fact that, if in a given nucleus the protons are converted to neutrons, the newly formed neutrons will have to occupy higher quantum states which will cause an increase in the nuclear volume in neutron-rich nuclei. The correction  $\Delta f$  is so taken as to change each term in f which contains nuclear volume and gives twice the nuclear volume for an all-neutron nucleus. Incorporating  $\Delta f$ into the foregoing development does not materially change the Z calculated for a given A but it does reduce the density at which the nucleus (Z, A) is most stable. In fact, the tabulated values of the corrected densities in Table V show that the density is reduced below the critical value even for an A = 240. It has been suggested by van Albada that one need not depend entirely upon this correction because one can consider the formation of the heaviest nuclei by fusion. As one increases the density, the stability maximum with respect to atomic weight shifts to higher atomic weight and formation of heavy elements by fusion becomes energetically possible. It is well known that it is ener-

ionic solution, and it is difficult to see how to apply the considerations of van Albada without a closer analysis of the statistics of the medium along the lines of the Debye theory of the solid state. Such an analysis is being considered at the present time by G. Beskow.

<sup>¶¶</sup> It should be noted that the implication in van Albada's work that Eq. (62), apart from the term 10<sup>3</sup>, is a modified packing fraction is misleading in that the result is valid for  $T \cong 0^{\circ}$  K only, where there would be only one nuclear species present. At higher temperatures where thermal energy cannot be neglected this result would not lead to the correct abundance distribution. Note that at any finite temperature all nuclear species will be present and it is not clear how one would divide the energy of the assembly among the nuclei.

getically possible to form the most stable nuclei both by fission of heavier nuclei and fusion of lighter nuclei. However, the reaction rates are exceedingly low except, under special circumstances, for the neutroninduced fission of uranium and other very heavy nuclei, and the synthesis of light elements from hydrogen [61].

The work of van Albada, in the opinion of the authors, may not yet explain the possible existence of the heaviest elements in an equilibrium assembly for the following reasons. In order to form the elements in equilibrium in any reasonable length of time one requires temperatures in excess of  $\sim 10^9$  °K, as shown by Hoyle [79]. Above this temperature nuclei will not be degenerate even at the densities considered by van Albada. Electron degeneracy is present at these densities up to  $T \cong 10^{11}$  °K and it appears that there will be a considerable concentration of neutrons which are at least partially degenerate. It seems to us again that one must examine heavy element abundances according to an equilibrium theory using appropriate statistics, and it is interesting to note that Lacroute [103] has given qualitative arguments that the inclusion of free neutrons makes difficult the explanation of the existence of the heavy elements unless neutron degeneracy is taken into account. Finally, one would expect at high temperatures that neutron-induced fission of the very heavy elements would be as important as the fusion process discussed by van Albada.

While neither Hoyle nor van Albada has attempted to compute the relative abundances of the nuclei, taking into account the effects of electron degeneracy, they have considered the types of stars in which element formation might take place. In this connection they also consider the freezing-in of the equilibrium. This phase of their work is described later.



FIG. 8. Equilibrium concentrations at various temperatures of electrons, positrons, protons, and  $\alpha$ -particles as a function of neutron concentration in an assembly consisting of these particles only, according to Chandrasekhar and Henrich [35].

# 5. Gravitational Effects and Stellar Models

As mentioned earlier, it is difficult to understand how both light and heavy elements can be explained as resulting from a nuclear equilibrium at a given set of values of density and temperature. The question therefore arises as to where and when the physical conditions required for element formation in equilibrium might have existed. The prestellar state of the universe is generally pictured as consisting of some kind of hot, dense material which expands and cools very rapidly from some "initial" state. Chandrasekhar and Henrich [35] have suggested the existence of nuclear equilibrium in the prestellar state since current theories of stellar interiors seem to preclude the formation of elements heavier than helium in the known types of stars. Their picture of prestellar element formation is as follows. At the early high  $\rho$  and T the heavy elements might have been formed. The expansion and cooling of the universe may have frozen in a few parts per million of the heavy elements. In a subsequent equilibrium the lighter elements may have been established in their present relative abundances. When the temperature fell below about  $4 \times 10^9$  °K, the nuclear reactions supporting the equilibrium ceased. At still lower temperatures the very light element abundances adjusted according to the kinds of non-equilibrium reactions which now go on in stellar interiors. This qualitative picture must be questioned since it is difficult to see how during the prestellar phase there would be a sufficiently long time for equilibrium to be established at any high  $\rho$  and T. There have been no calculations made along these lines, although Steinwedel and Jensen [142] have considered the problem qualitatively. Rather, various investigators have examined the possible formation of elements at equilibrium in special kinds of stars. Both Hoyle [79] and van Albada [2] have considered stars whose interior densities are sufficiently high to introduce electron degeneracy, which, as seen earlier, might help to explain the existence of the heavy elements. In addition to effects of electron degeneracy it might be expected that the gravitational potential would affect the nuclear equilibrium in stars. This latter problem was recently considered by Beskow and Treffenberg [18], Klein [97, 98b], and Wataghin [174].

The problem of element formation is considered by Beskow and Treffenberg to involve some kind of star in which the proper equilibrium conditions for lighter and heavier nuclei could be realized simultaneously in different spherical shells. The star is assumed to be isothermal and all of their calculations are made at kT=1 Mev.|||| This particular temperature was selected because it was satisfactory for the formation of the light elements, and it was hoped that gravitational

 $<sup>\|</sup>$  ||We have been informed by Professor Klein (private communication) that these calculations are being repeated for other values of kT.

effects would be sufficient to correct the heavy element difficulty. It was supposed that after a sufficient time for equilibrium to have been established a very sudden explosion or expansion mixed and froze in the various elements. However, the latter problem has not been examined quantitatively by Beskow and Treffenberg.

Their approach to the problem of nuclear equilibrium in a stellar model involves the determination of the chemical potentials for the neutron and proton as a function of position in the model. A knowledge of the local intrinsic potentials enables a calculation to be made of the local abundance distribution of the elements according to the Gibbs' formulation, in which the intrinsic potentials must be modified for an equilibrium system in a field.\* If  $\phi_{\varrho}$  and  $\phi_{e}$  are the gravitational and electrostatic potentials, respectively, then the condition for equilibrium may be written as

$$\mu_n + m_n \phi_g = \text{constant}, \tag{69a}$$

for the neutron, and

$$\mu_p + m_p \phi_q + e \phi_e = \text{constant}, \tag{69b}$$

for the proton. If one assumes spherical symmetry in the star, he finds the gravitational potential,  $\phi_{g}(r)$ , is related to the total mass density,  $\rho(r)$ , i.e., the density of all matter and radiation according to Poisson's equation

$$\nabla^2 \phi_g = \frac{1}{r} \frac{d}{dr} \left( r^2 \frac{d\phi_g}{dr} \right) = 4\pi G \rho, \tag{70}$$

where G is the gravitational constant. Substitution of  $\phi_{q}$  from Eq. (69a) into Eq. (70) gives the following relationship between  $\mu_{n}$ ,  $\rho$ , and r:

$$\frac{d^2\mu_n}{dr^2} + \frac{2}{r}\frac{d\mu_n}{dr} + 4\pi m_n G\rho = 0.$$
(71)

An equivalent equation for  $\mu_p$  is not obtained from the Poisson equation for the electrostatic potential by Beskow and Treffenberg, but rather it is assumed that the condition of electrical neutrality is satisfied locally [98a]. This condition leads to a relationship between  $\mu_p$  and  $\mu_n$  but, of course, does not take into account automatically the electrostatic effects considered by van Albada [1, 2]. In addition, one needs a relationship between  $\mu_n$  and the total mass density  $\rho$  in order to determine each as a function of radial position.

In order to clarify some of the details involved in finding  $\mu_n$  as a function of  $\mu_p$ , and in particular to illustrate the behavior of the concentrations of electrons and positrons, let us consider the equilibrium between protons, neutrons,  $\alpha$ -particles, electrons, and positrons, following the treatment of Chandrasekhar and Henrich [35].<sup>‡</sup> The fundamental reactions maintaining equilibrium in general are taken to be

$$_{1}H^{1}+e^{-} \rightleftharpoons_{0}n^{1}$$
 (72a)

$$e^+ + e^+ \rightleftharpoons \text{photons},$$
 (72b)

and  

$$_{Z}X^{A} + (\Delta Z)(_{1}H^{1}) + (\Delta N)(_{0}n^{1}) \rightleftharpoons_{Z'}X^{A'},$$
 (72c)  
where

$$A' = A + (\Delta N) + (\Delta Z), \quad Z' = Z + (\Delta Z).$$
 (72d)

It is assumed that the role of the neutrino may be neglected. The condition that the assembly be electrically neutral is

$$C_{e^{-}} = \sum' Z_j C_j (A, Z) + C_{e^{+}}, \tag{73}$$

where  $\sum'$  designates summation over all nuclei, and  $C_{e^-}$  and  $C_{e^+}$  are the electron and positron concentrations, respectively. The examination of these reactions for a given density and temperature involves a simultaneous system of equations of high order. However, in the case under consideration, this problem is considerably simplified. The equation governing neutron, proton, and electron concentrations, if one assumes no degeneracy, is

$$C_{p}C_{e}-C_{n}^{-1} = 2(2\pi m_{e}kT)^{\frac{1}{2}}h^{-3}\exp[-c^{2}(m_{p}-m_{n})/(kT)],$$

$$(T \leq 4 \times 10^{9} \text{ }^{\circ}\text{K}), \quad (74a)$$

$$= 16\pi[kT/(hc)]^{3}\exp[-c^{2}(m_{p}-m_{n})/(kT)],$$

$$(T \geq 4 \times 10^{9} \text{ }^{\circ}\text{K}), \quad (74b)$$

according to whether the electrons obey non-relativistic or relativistic statistics. Similarly the electron and positron concentrations must also satisfy

$$C_{e}-C_{e} = 4(2\pi m_{e}kT)^{\frac{1}{2}}h^{-3}\exp[-2m_{e}c^{2}/(kT)],$$

$$(T \leq 4 \times 10^{9} \text{ °K}), \quad (75a)$$

$$= 256\pi^{2}[kT/(hc)]^{6}\exp[-2m_{e}c^{2}/(kT)],$$

$$(T \geq 4 \times 10^{9} \text{ °K}), \quad (75b)$$

again depending on the type of statistics. The concentration of  $\alpha$ -particles is given by

$$(C_p)^2 (C_n)^2 (C_\alpha)^{-1} = 2(2\pi m_0 kT)^{9/2} h^{-9} \exp\left[-c^2 (2m_n + 2m_p - m_\alpha)/(kT)\right],$$
(76)

and, finally,

$$C_{e-} = C_{e+} + C_p + 2C_{\alpha}.$$
 (77)

<sup>\*</sup> The relativistic generalization of the Gibbs' condition for equilibrium in a gravitational field has been given recently by O. Klein [98a, 98b]

<sup>†</sup> Wataghin [174] suggests that gravitational fields cannot influence the nuclear equilibrium. It is true that for a given gravitational potential,  $\phi_0$ , the equilibrium is essentially independent of the value of  $\phi_0$  so long as  $\phi_0/c^2 \ll 1$  and  $\phi_0$  is constant over the system. However, the Gibbs' condition for equilibrium in a gravitational field, viz.,  $\mu + m\phi_0 = \text{constant}$ , implies that, for an isothermal star in which there is nuclear equilibrium, as  $\phi_0$ varies with position in the star, the intrinsic potentials  $\mu$  must vary accordingly. Now the distribution of abundances depends strongly on the values of the  $\mu$ 's involved and hence the equilibrium distribution will vary with position in the star. Depending on the stellar model, and hence on the form of  $\phi_0(r)$ , one would have various relative abundance distributions as functions of the radial position and of the model.

<sup>&</sup>lt;sup>‡</sup> Steinwedel and Jensen [142] have examined this particular equilibrium problem using relativistically degenerate statistics for the electron, so that their results are applicable to higher densities. Singwi and Rai [133] have also considered electron degeneracy in this connection.

Chandrasekhar and Henrich have solved Eqs. (74)–(77) simultaneously for various neutron concentrations and temperatures by trial and error. These results are given in Fig. 8 which shows that for low neutron concentrations the material density is almost entirely due to electron-positron pairs. As the concentration of protons and  $\alpha$ -particles becomes appreciable,  $C_{e^-}$  begins to increase so as to satisfy the condition of electrical neutrality, while the positron concentration  $C_{e^+}$  decreases sharply. The extension of the curves for  $C_{\alpha}$  to high values is physically incorrect because other nuclei will form at high neutron concentrations, but the extensions are included to illustrate the form of the solutions  $\lceil 142 \rceil$ .

The procedure followed by Beskow and Treffenberg in finding  $\mu_n$  as a function of  $\mu_p$  is essentially that given, except in that all nuclear species are taken into account in the condition of electrical neutrality. The relationships between the intrinsic potentials of the various elementary particles corresponding to the equilibrium reactions described by Eqs. (72a) and (72b), may be written as:

and

$$\mu_n' = \mu_p' + \mu_{e^-}' \tag{78a}$$

$$\mu_{e^{-}} + \mu_{e^{+}} = 0, \tag{78b}$$

where since particles are not conserved  $\mu_n' = \mu_n + m_n c^2$ ,  $\mu_p' = \mu_p + m_p c^2$ , and  $\mu_e' = \mu_e + m_e c^2$ . In Eq. (78a) the intrinsic potential of the neutrino is neglected because, as Beskow and Treffenberg point out, while the electrons in the assembly are strongly affected by a gravitational field, the neutrinos are not. If, as pointed out by Klein [98a], the neutrino and antineutrino are identical then it follows that the neutrino intrinsic potential is zero. In Eq. (78b) the intrinsic potential for photons is taken as zero since the photon concentration at equilibrium is a function of temperature only and the equilibrium is considered in an isothermal star. It follows from Eqs. (78) that¶

$$\mu_{e'} = -\mu_{e'} = -\mu_{p'} = \mu_n - \mu_p + (m_n - m_p)c^2. \quad (78c)$$

The values of  $\mu_n$  and  $\mu_p$  must simultaneously satisfy Eqs. (35), (73), and (78c). The electron and positron concentrations can be determined as functions of their intrinsic potentials by the appropriate solutions of Eq. (35) which applies to either electrons or positrons if the corresponding  $\mu_e$  is used. This use of Eq. (35) automatically accounts for possible electron degeneracy. In order to carry out the calculation of  $\mu_n$  as a function of  $\mu_p$  it is necessary to evaluate  $\sum' Z_j C_j$  for particular sets of values of  $\mu_n$  and  $\mu_p$ . Beskow and Treffenberg, using the semi-empirical formula for the binding energy per nucleon of Mattauch and Fluegge [111] obtain the following approximation, where  $C_j$ , in the form given by Eq. (10), has been taken including the factors for spin multiplicity and excited states:

$$\sum' C_j Z_j \cong (3.20 \times 10^{-6} \pi m_0)^{\frac{1}{2}} h^{-3} \pi^{\frac{1}{2}} a_0$$

$$\times \int_{0}^{\infty} b_{0}^{-\frac{1}{2}} A^{3} \exp[\gamma_{0} + (a_{0}^{2}/b_{0})A] dA, \quad (79)$$

where

$$a_0 = 41.05 - (\mu_n - \mu_p)/2$$
 (79a)

(79b)

$$b_0 = 82.10 + 0.602A^{\frac{2}{3}}$$

and

$$\gamma_0 = (\mu_n - 5.72)A - 15.04A^{\frac{2}{3}}, \quad (A \le 100), \tag{79c}$$

$$=3.4+(\mu_n-5.68)A-15.40A^{\frac{2}{3}}, (A \ge 100).$$
 (79d)

Equations (79) are valid only for kT=1 Mev. The evaluation of Eqs. (79) implies that one has taken into account for a given value of A all values of Z consistent with the empirical binding energy formula. The contribution of the less stable nuclei to the sum is quite small. The plot of  $\mu_n$  versus  $\mu_p$  obtained by Beskow and Treffenberg is not reproduced here, since it is useful only as a step in their calculations. Likewise, other figures used only in the derivation of their final result have not been included.

Having found  $\mu_n$  as a function of  $\mu_p$ , it is necessary to find either  $\mu_n$  or  $\mu_p$  as a function of the total mass density,  $\rho$ . Rather than compute the density of nuclei,  $\rho_m$ , from  $\rho_m = \sum' m_j C_j \cong m_0 \sum' A_j C_j$  by methods similar to those used to obtain  $\mu_n$  and  $\mu_p$ , Beskow and Treffenberg have approximated  $\rho_m$  as

$$\rho_m \cong (m_0 A/Z) \sum' C_j Z_j = (m_0 \bar{A}/\bar{Z}) (C_{e^-} - C_{e^+}), \quad (80)$$

where  $\overline{A}$  and  $\overline{Z}$  refer to the predominantly abundant nucleus for a given  $\mu_n$ . They have found this most abundant species by solving  $(\partial C_j/\partial Z)_{\overline{A}} = 0$  which, since

$$C_{j} = (3.2 \times 10^{-6} \pi m_{0})^{\frac{1}{2}} h^{-3} A^{\frac{1}{2}} \times \exp[\gamma_{0} + 2a_{0}Z - b_{0}(Z^{2}/A)], \quad (81)$$

gives

$$\bar{Z} = (a_0/b_0)\bar{A} 
= \begin{bmatrix} 1 - 1.22 \times 10^{-2} (\mu_n - \mu_p) \end{bmatrix} 
\times (2 + 1.46 \times 10^{-2} A^{\frac{3}{2}})^{-1} \bar{A}. \quad (82)$$

These equations are also valid only for kT=1 Mev. In their integration of Eq. (79), Beskow and Treffenberg state that for a given value of  $\mu_n$  only a rather narrow range of A values contributes to the sum. Knowing these predominant  $A(=\bar{A})$  values one may for a given  $\mu_n$  fix  $\bar{Z}$  and thus the density,  $\rho_m$ . For the case where  $\bar{A}$ 

$$\Phi_{i} = \Sigma_{r} \Sigma_{v} = \exp(0.14A + 0.36A^{3}), \quad A \leq 100 \\ = \exp(3.4 + 0.184A), \quad A \geq 100$$

 $<sup>\</sup>P$  Compare this result with Eq. (34) in which case it was assumed that the intrinsic potentials of electron and neutrino are equal.

<sup>||</sup> The product of summations over rotational and vibrational states has been approximated by

The evaluation of this product would require modification if, as discussed in Section III(b)3, Wergeland's result for the density of nuclear vibrational states is a better approximation. There are several typographical errors in sign in the detailed development of  $\Sigma' Z_i C_i$  in reference [18]. However, the final result given there is correct.

corresponds to the light nuclei, the known Z is used.\*\* In the calculation of the contribution of photons, electrons, positrons, and neutrinos to the total mass density, the energy density plays the principal role, whereas for neutrons, protons, and nuclei, only rest mass energy is considered.\*\*\*

The next step is to determine  $\mu_n$ ,  $\mu_p$ , and  $\rho$  as functions of r, the radial distance from the center of the gravitating body. Klein<sup>††</sup> has pointed out that this calculation may be described in the following manner. Having the equilibrium condition  $\mu_n + m_n \phi_g = \text{constant}$ , one may write instead of Poisson's equation the following:

$$\frac{d\mu_n(r)}{dr} = -\frac{GM(r)m_n}{r^2} \left[ = -m_n \frac{d\phi_g(r)}{dr} \right], \quad (83a)$$

and

$$dM(r)/dr = 4\pi r^2 \rho(\mu_n). \tag{83b}$$

Given initial values of  $\mu_n$  and M(r) at a given  $r_0$ , these equations may be integrated to yield the values of these quantities for  $r > r_0$ . Beskow and Treffenberg have assumed a model in which there is a central core of radius  $r_0$ , mean total mass density  $\bar{\rho}_0$ , and intrinsic potential  $(\mu_n)_0$ . Fixing these quantities, they have taken small increments in  $\mu_n$  which yield corresponding new values of  $\rho$  from the relationship between  $\mu_n$  and  $\rho$ , and of r as follows. A sphere of radius  $r_1$  slightly larger than  $r_0$  will have a mass

$$M_{1} = M_{0} + \int_{r_{0}}^{r_{1}} 4\pi r^{2} \rho dr,$$
  

$$\cong (4\pi/3) [r_{0}^{3}(\bar{\rho}_{0} - \bar{\rho}_{0,1}) + r_{1}^{3} \bar{\rho}_{0,1}], \qquad (84)$$

where  $\bar{\rho}_{0,1}$  is the mean mass density in the spherical shell between  $r_0$  and  $r_1$ . From Eqs. (83) one can obtain,

concentrations. \*\*\* The contribution to the total mass density of the gravitating mass of photons, electrons, positrons and neutrinos can be written  $\rho = (u+3P)/c^2$ , where u is the energy density and P is the partial pressure [157]. For photons and neutrinos the rest mass is taken as zero, so that P=u/3 and  $\rho=2u/c^2$ . Since for radiation  $u=aT^4$ , kT=1 Mev gives  $\rho$  (radiation) = 3.06 × 10<sup>5</sup> g/cm<sup>3</sup> throughout the stellar model. For neutrinos, the chemical potential is zero, and Fermi-Dirac statistics give, with  $E=c\rho$ ,

$$u(\text{neutrinos}) = (8\pi/h^3) \int^{\infty} E p^2 [\exp(E/kT) + 1]^{-1} dp$$
$$= (7/8)u(\text{photons}),$$

so that  $\rho$ (neutrinos) = 2.68×10<sup>5</sup> g/cm<sup>3</sup>. The same value would result for antineutrinos. The factor 7/8 arises because photons obey Bose-Einstein statistics. For electrons the energy density is given by

$$u(\text{electrons}) = \int_0^\infty E(dC_{e^-}/dp)dp$$

where  $C_{e^-}$  is given by Eq. (35) in this paper. The electron partial pressure is given by  $(1/3) \int_0^\infty (p/E) (\partial u/\partial p) (\partial E/\partial p) dp$ , where the u is that for electrons. It will be recalled that  $C_{e^-}$  varies with  $\mu_{e^-}$  and therefore with  $\mu_n$  and  $\mu_p$ . Similar calculations are used for positrons.

*†*<sup>†</sup> Private communication.

making appropriate approximations,

 $(r_1/r_0)^3 + a(r_1/r_0) - b = 0,$  (83c)

where

$$a = 2(\bar{\rho}_0/\bar{\rho}_{0,1}) - [3/(2\pi m_n G)] \{ [(\mu_n)_0 - (\mu_n)_1]/(r_0^2 \bar{\rho}_{0,1}) \} - 3, \quad (83d)$$
  
and

$$b = 2[(\bar{\rho}_0/\bar{\rho}_{0,1}) - 1].$$
 (83e)

In these equations one picks a  $(\mu_n)_1$  and finds the corresponding  $\bar{\rho}_{0,1}$  from the previously discussed rela-



FIG. 9. The total number of nuclei of various atomic weights in the stellar models of Beskow and Treffenberg [18], compared with the observed abundance distribution of Brown [30]. The material inside the stellar core of radius  $r_0$  is not included but neutrons exterior to the core are included as part of the proton abundance. The observed data have been adjusted so as to yield  $1M_{\odot}$ , following Beskow and Treffenberg.

tionship between  $\mu_n$  and  $\rho$  and then one computes  $r_1$ . Successive application of this procedure enabled Beskow and Treffenberg to find both  $\mu_n$  and  $\rho$  as functions of r, which then fixes the model.

An examination of Eq. (79) shows that for  $\mu_n > 5.68$ the sum  $\sum' Z_j C_j$  diverges and Beskow and Treffenberg take the corresponding mean mass density of the closepacked core to be nuclear density in the limiting case. They have considered models with such cores of nuclear density having various radii  $r_0$ , ranging from  $10^5-10^6$ cm. They have, in addition, studied the case for a less dense core,  $(\mu_n)_0=3.0$ , with  $r_0=10^5$  cm. Given the intrinsic potentials  $\mu_n$  and  $\mu_p$  as functions of r, one may calculate the relative abundances of the elements in each spherical shell. Then, integration of the concentration of each species over the stellar model yields the absolute abundances of the elements in this model. In

<sup>\*\*</sup> Although it is not stated in the papers of Beskow and Treffenberg, Professor Klein has informed us that degenerate statistics were employed for determining the neutron and proton concentrations.

Model	$r_0 \times 10^{-5}$ cm Core $\bar{\rho}_0$ , g/cm <sup>3</sup> Core ( $\mu_n$ ) <sub>0</sub> , Mev	$1 \\ 5 \times 10^{12} \\ 3.0$	3 1.4×10 <sup>14</sup> 5.68	5 1.4×10¼ 5.68	5.5 1.4×10 <sup>14</sup> 5.68	6 1.4×10 <sup>14</sup> 5.68	7 1.4×10 <sup>14</sup> 5.68	$10 \\ 1.4 \times 10^{14} \\ 5.68$
Mass of core, in	$M_{\odot}$	1.0×10-5	7.9×10-*	3.7×10 <sup></sup> ²	4.9×10-2	6.3×10 <sup>-2</sup>	1.0×10-1	2.9×10 <sup>-1</sup>
Total mass, in J	$M_{\odot}$	39	39	39	39	39	15	0.31

TABLE VI. Masses of cores and total masses for various models.

the integration the core of radius  $r_0$  is not included, the neutron and proton concentrations are added, and the "super-heavy" nuclei which the theory predicts are also excluded. In Fig. 9 the total number of certain nuclei in the various models are plotted versus atomic weight and compared with the relative abundance data of Brown [30]. Calculations were reported only for A = 1, 4, 12, 30, 60, 90, 120, 180, 240, and 300. In order to make the plotted comparison in the same manner as in the original paper of Beskow and Treffenberg, Brown's data were multiplied by a factor such that the abundance of hydrogen corresponds to a mass of about  $1.3 \times 10^{33}$  g, approximately the mass of the sun. The results of Beskow and Treffenberg lie above Brown's data at least for the reason that most of the models exceeded one sun mass. The masses of the cores and total masses of the models are given in Table VI. It is clear from Fig. 9 that if the computed abundances are regarded as relative, the plots for the various models with  $r_0 < 6 \times 10^5$  cm could be adjusted to yield fairly good agreement with observation. However, whatever adjustment is made there still remains a relative deficiency in the abundance of the very light elements as compared to the heavy elements.

Beskow and Treffenberg have not discussed the problem of the "explosion" of these stellar models or the freezing-in of the equilibrium distribution in any detail. They indicate that models with  $r_0 = 5$  and  $5.5 \times 10^5$  cm, which give the best agreement with observation, have too much hydrogen and heavy nuclei in absolute amounts, and suggest that heavy element fission as well as the escape of protons in the explosion may repair the supposed difficulty.<sup>‡‡</sup> Actually it would appear that there is relatively a deficiency of the very light elements, perhaps even including hydrogen. Furthermore, if in fact the observed abundance distribution of Brown is universal, it is difficult to see how repairing an abundance distribution in a local region can do anything but spoil it elsewhere. Nevertheless, it should be pointed out that the work of Beskow and Treffenberg gives for the first time reasonable agreement between the calculated equilibrium abundances and those observed.

In a discussion of the properties of their stellar models, Beskow and Treffenberg indicate that it is implied that matter was collected into stars embedded in a sea of radiation corresponding to a temperature of 1 Mev while equilibrium was established. In a subsequent explosion large remnants may have been left from which present stars might be derived. These remnants would be distributed in size and possess differing final constitutions.

These ideas are based on Klein's [97] more recent discussion of the cosmological implications of the star models of Beskow and Treffenberg. One of the principal astrophysical problems in this connection is how these "prestellar stars" might tie in with a static universe filled with matter and radiation of the densities required by Beskow and Treffenberg for their equilibrium problem. Such a universe would be extremely small in size and mass. Thus, a closed Einstein universe of mean density  $\rho \cong 10^6$  g/cm<sup>3</sup>(kT \cong 1 Mev) has a radius of  $4 \times 10^{10}$  cm and a mass of  $10^6 M_{\odot}$ . On the other hand, a homogeneous expanding universe large enough to include the observable regions would expand so rapidly as to make thermal equilibrium untenable, and would require the examination of non-equilibrium processes for the formation of the elements. Klein has suggested that in order to overcome these difficulties and retain the successes of the equilibrium theory of the formation of the elements in the situation examined by Beskow and Treffenberg, one might assume that a radiation field was originally concentrated around each star. This radiation would consist of photons, electronpositron pairs, and neutrinos. As Klein has pointed out, such a radiation field, with dimensions large as compared to those of the embedded star, is required in order that an equilibrium be possible. Otherwise, the escape of neutrinos from a star of reasonable size might seriously affect the establishment of a nuclear equilibrium. Klein has obtained a spherically symmetric solution of the Einstein equations corresponding to a mass of radiation held together by its own gravitation. The solution yields the following relations for the radiation density and temperature in this radiation star, for large r:

$$\rho_r = 3(2\kappa c^2)^{-1} r^{-2} = 8 \times 10^{26} r^{-2} \text{ g/cm}^3, \qquad (85a)$$

$$T = 3.16 \times 10^{15} r^{-\frac{1}{2}} \,^{\circ} \mathrm{K},$$
 (85b)

where r is the distance from the origin in cm, and  $\kappa = 8\pi G/c^4$  is the Einstein gravitational constant. For  $r = 2.6 \times 10^{10}$  cm, one has  $kT \cong 1$  Mev (or  $\rho_r \cong 10^6$  g/cm<sup>3</sup>).

and

<sup>&</sup>lt;sup>‡‡</sup> The implication of this discussion is that hydrogen should be more abundant in interstellar space than in stars. According to Strömgren [149] the assumption of a standard stellar composition for interstellar matter leads to a not unreasonable density for this matter. However, according to [150] and to Dr. L. Spitzer, Jr., private communication, the possibility that hydrogen is as much as ten times more abundant in interstellar space than in stars cannot be excluded at the present time.

Thus, at a distance r of the order of the radii of the Beskow-Treffenberg models, the temperature has the 1-Mev value required by these investigators. Near the origin the solution for the temperature is

$$(T/T_0)^4 = (p/p_0) = 1 - 2s + (13/5)s^2 - (18/7)s^3 + (356/175)s^4 - \cdots,$$
 (86)

where p is the pressure,  $p_0$  and  $T_0$  the values at r=0, and

$$s = \kappa p_0 r^2. \tag{86a}$$

This result shows that the temperature is sensibly constant over a region comparable in size to the size of the kind of stars required by Beskow and Treffenberg. Klein further points out that the inclusion of matter in this body of radiation makes little difference in the solution so long as the gravitational potential of the matter within the distribution is small compared to  $c^2$ . This is the case in all the models considered by Beskow and Treffenberg. Thus, a star of  $\sim 40 M_{\odot}$ , with a central temperature of  $\sim 1$  Mev, embedded in radiation will be essentially isothermal and provide the locale for establishing the equilibrium distribution of the elements.¶¶ Unfortunately, the solution obtained by Klein gives a radiation star of infinite extent and an infinite radiation mass, so that there would exist only one material star of about  $40M_{\odot}$ . Thus, there would be no "outside" region into which the radiation might escape and allow the rapid explosion of the embedded material star. However, Klein suggests that it would be interesting to consider static solutions of the Einstein equations corresponding to many individual stars embedded in a radiation field, i.e., a galactic system of stars, as well as a further solution to the problem which may give a suitable representation of a supergalactic system. It is also pointed out that a static solution can be only an approximation to the state of the universe when the elements were formed, since conditions have changed markedly since that time, and it is expected that nonstatic solutions might provide the instability required. An originally weak radiation current in this non-static model should gradually increase as the gravitational forces holding the field together decrease. The last stage of this process would involve the sudden escape of the radiation and lead to a violent explosion of the embedded star. Such solutions might provide insight into the "beginnings" of the expanding universe. It should be emphasized that Klein and his collaborators visualize the equilibrium formation of the elements once and for all at a time in the "beginnings" of the universe. III This point of view is quite different from that of other investigators to be discussed who construct special stars in which the equilibrium element forming processes may take place even now.

Other types of stellar models have been considered for the formation of the elements by Cherdyncev [36], Hoyle  $\lceil 79 \rceil$ , and van Albada  $\lceil 2 \rceil$ . It will be recalled that the latter investigator found it necessary to assume densities of the order of  $2.5 \times 10^{11}$  g/cm<sup>3</sup> in order to explain the existence of elements with  $A \cong 240$ . G. B. van Albada has shown that the interior of ordinary white dwarfs do not provide the conditions required to explain heavy elements. What apparently is required is a kind of white dwarf in which there is a non-uniform rotation whose velocity increases toward the center. This proposed model provides a degenerate core in which the heavy elements might exist and further provides rotational instability as the star evolves. Presumably some kind of explosion ensues which he identifies with a supernova outburst. This mechanism would distribute heavy elements in space in a nonhomogeneous manner depending on the nature of the freezing-in of the equilibrium, and would leave a remnant white dwarf. It is suggested that the light elements might form in stars with hot, condensed, but nondegenerate cores. These stars are tentatively identified by van Albada as red giants. However, no mechanism is provided for the distribution of the lighter elements in space. It should be pointed out that if the elements are formed in stars then one should expect differences in the relative abundances of the nuclear species between parts of a galaxy and between galactic systems.

In connection with stellar models, it will be recalled that Cherdyncev [36] considered the formation of the elements as resulting from an equilibrium between neutron nuclei and four-neutron particles. He suggested that one would require a dense neutron-core star whose temperature and density are about 4 Mev and 0.03 nuclear density, respectively, which explodes and distributes this mixture of neutrons and neutronnuclei in space, Subsequent  $\beta$ -decay is discussed as the mechanism leading to the stable nuclei.

Perhaps the most detailed discussion of the kinds of stars in which one might expect the equilibrium formation of the elements has been given by Hoyle [79, 80]. He has considered stars which have exhausted their hydrogen so that radiation from nuclear energy production no longer counterbalances gravitational forces. Such stars will begin to contract and the central temperature and density will rise. If these stars rotate, then at some stage of the contraction the stars become rotationally unstable. Hoyle has made a classical calculation of the values, at the onset of rotational instability, of the central temperature, density and of the radius of a star which has collapsed through a series of homologous configurations. In Table VII is shown the effect of the initial rotational velocity on the characteristics of the collapsing star. This calculation indicates that in such a collapsing star the central temperature and density attain values suitable for nuclear

 $<sup>\</sup>P$  One should take into account the fact that the embedded stars are not strictly isothermal and that they must be in mechanical equilibrium for so long as is required to establish a nuclear equilibrium.

 $<sup>\|</sup>$   $\|$  Jordan [95] suggests that the results of Klein and collaborators provide confirmatory evidence for his theory of stellar origin based on a cosmology in which the universal constants are functions of time.



FIG. 10. The evolutionary track, CDEFG, of the central material of a contracting star of  $5M_{\odot}$  in the temperature-density plane, according to Hoyle [79]. The  $(T, \rho)$  plane is divided by the curve AEFB into regions in which the predominantly abundant nuclei are those of helium or heavy elements.

reactions at appreciable rates prior to the onset of rotational instability. As Hoyle points out, thermonuclear reactions with protons at appreciable rates require temperatures of the order of 109-1010 °K.† These stars presumably may be identified as O-, B-, and A-type stars whose lifetime according to the Bethe energy production cycle is a factor of ten less than the age of the universe. Hoyle's detailed study of such stars involves the following procedure. It is assumed that the temperature and density at the center of the collapsing star are related through the equations of statistical equilibrium between nuclei, neutrons, protons, and electrons. The effect of electron degeneracy at high densities can be taken into account in the manner discussed in Section III (b)4. The mass of the particular star is introduced as a parameter through the neutron concentration by considering the portion of the gravitational energy released in the collapse which is absorbed in the production of free neutrons. As the star contracts, the composition of its interior will change but will have a definite equilibrium distribution at each Tand  $\rho$  so long as the latter quantities vary sufficiently slowly.

From the equations of statistical equilibrium Hoyle has shown that the composition of an equilibrium mixture consists of essentially all helium or of all heavy elements (A > 50) when the temperature is greater or less than  $T_9'$  given by

$$T_{9}' = \frac{18.92[E_{j}(Z, A)/A - E_{\alpha}/4 + 0.543y(0.5 - Z/A)]}{10.3 + (3/2)\log T_{9}' + (5/2)\log 4 - \log \rho},$$
(87)

where  $T_{9}'$  is in units 10<sup>9</sup> °K, the density  $\rho$  does not include neutrons, and the other quantities are as defined in Section III(b)4. For a given  $\rho$  and T one finds Z and A by maximizing  $(E_i - 0.543yZ)/A$  as discussed earlier, and these values of  $E_j$ , Z, and A are used in Eq. (87). According to this relationship, the  $(T, \rho)$ plane shown in Fig. 10 is divided by the curve AEFB into a heavy element region and a region in which the material is composed almost entirely of helium. In Fig. 10 is also shown the "track" of a contracting star, CDEFG, calculated by Hoyle for the interior of a star of  $5M_{\odot}$ . The star initially at C is composed almost entirely of helium. When it reaches D the temperature is high enough for nuclear reactions to begin and between D and E the element distribution is approximately as shown in Table VIII. Hoyle suggests that the exothermic nature of the reactions along the track between the points D and E provides sufficient radiation pressure to maintain the star for perhaps 10<sup>6</sup> years. However, at E the situation changes and from E to G the collapse is essentially one of free fall, requiring perhaps a hundred seconds. As the density rises in the heavy element zone, particular heavy elements will be most abundant according to Table IV, Section III(b)4. The point G represents the breakdown of the classical equations of statistical equilibrium for this particular star. The initial rotational velocity of the star is not considered in obtaining the track but determines the point on the track at which the star becomes rotationally unstable. If the angular momentum is large, instability sets in before the material at the center of the star reaches the point E and one has essentially only the elements in the first half of the periodic table. On the other hand, if the angular momentum is small the star will evolve into the heavy element zone. These unstable stars explode and distribute their material in space leaving a white dwarf remnant. Since the density and temperature vary in the star with radial position, the composition of the ejected material will be more complicated than described. Hoyle identifies novae

TABLE VII. Effects of initial rotational velocity.

Mass=10M Initial radius=3.5>	Initial central density = $3 \text{ g/cm}^3$ Initial central temperature = $3.5 \times 10^7 ^{\circ}\text{K}$						
Initial rotational velocity (km/sec.)	1	5	10	15	20	40	100
Radius at onset of instability (cm)	9.16×10 <sup>5</sup>	2.29×107	9.16×10 <sup>7</sup>	$2.06 \times 10^{8}$	3.68×10 <sup>8</sup>	$1.47 \times 10^{9}$	9.16×10 <sup>9</sup>
Central temperature (°K)	$>4\times10^{9}$	$>4\times10^{9}$	$>4\times10^{9}$	$>4\times10^{9}$	$>4 \times 10^{9}$	$>4\times10^{9}$	$1.34 \times 10^{9}$
Central density (g/cm <sup>3</sup> )	1.67×1017	1.07×1013	1.67×10 <sup>11</sup>	1.47×1010	2.59×10 <sup>9</sup>	4.07×107	1.67×105

<sup>†</sup> It should be noted that in equilibrium processes very high densities favor the formation of heavier elements, while at temperatures higher than those stated, competing process leading to nuclear evaporation would become important and favor lighter elements.

TABLE VIII.

*****								
Element	He	0	Si	Fe	Cu	Kr	Sn	Pb
$\log C_i$	27.2	20.2	23.7	28	26.7	18.8	-3.3	-222

and supernovae with stars which become unstable before or after the point E, respectively, and suggests that the rate of occurrence of novae and supernovae may be sufficient to account for the various amounts of elements in the universe at the present time.\*

Qualitatively it would appear that Hoyle's mechanism of collapsing stars might account for the relative abundances of the elements. However, there are a number of points in Hoyle's discussion which may require further examination. It does not seem to have been shown that equilibrium might be established at any values of  $\rho$  and T after the beginning of the rapid collapse. Despite the detailed examination of the collapsing model no calculation has been made of the actual universal abundance distribution which might be expected. It would probably prove necessary to take into account gravitational effects in computing the equilibrium distribution as did Beskow and Treffenberg [18]. A question raised by Johnson<sup>†</sup> concerns the existence of heavy elements in stars which still have large amounts of hydrogen. Hoyle has answered this point by suggesting that these stars acquire heavy elements by sweeping them up from interstellar space once his heavy element mechanism has started in other stars in the universe. Presumably the necessary elements for the C-N cycle would also be swept up. From the available published material it seems to us that the beginnings of Hoyle's stellar mechanism need clarification as does the effect of the freezing-in of the equilibrium on the final composition.

# (c) Discussion

From an historical point of view it was natural to describe the origin and relative abundances of the elements as resulting from a frozen-in nuclear equilibrium. In principle one need not know the detailed reactions involved in such an equilibrium as long as it is reasonable to assume that there existed reactions connecting all elements and that there was available a sufficiently long time for equilibrium to be established. Actually, one need only know the binding energies of the various nuclear species. It would seem to be desirable for a theory to explain the building of elements in the early prestellar state of the universe and have done with it. Except possibly for a small percentage of special kinds of stars there do not appear to have been places in the universe where a prestellar distribution of elements might have undergone appreciable change. If, in fact, stars have been formed by the condensation of interstellar material according, say, to the Whipple-Spitzer hypothesis [139, 177], then there should be an identity of composition except for the very light elements involved in reactions giving stellar energy. Nevertheless, as has been seen the expanding prestellar state has been rejected by most investigators as a locale for nuclear equilibrium because of the rapid changes that occur in the physical conditions according to current cosmological theories. The suggestion of Chandrasekhar and Henrich [35] that an equilibrium might have been established and in small part might have survived an expansion and cooling of the universe seems difficult to demonstrate and has also been rejected by most of the proponents of equilibrium theories. One is then left with the problem of explaining element synthesis in the small percentage of stars which are unstable, or in "prestellar" stars. If indeed there is element synthesis in these stars one has to explain the source of the explosion in some models, the mechanism of the ejection of central material in other models, as well as the survival of the equilibrium during the outburst required for the distribution of the matter in the universe. This latter is still the principal problem, although Beskow and Treffenberg, Cherdyncev, Hoyle, van Albada and others assume that the outburst will be sufficiently "violent" to freeze-in an established equilibrium.

The assumption that an equilibrium distribution can be frozen in without serious change requires a more detailed examination. There are two extremes within which the freezing in problem must lie. One of these extremes would be the expansion and cooling of the stellar material through a series of quasi-equilibrium states. In this case the equilibrium distribution would continually readjust and it is important to note that the heavy element concentrations would be most seriously affected. It will be recalled that the inclusion of excited states increased the equilibrium concentrations of the heavier elements by a factor of at least 10<sup>3</sup> and perhaps more than 10<sup>10</sup>. This factor would disappear rapidly with a decrease in temperature and would in itself lead to a very different abundance distribution. In the other extreme the expansion and cooling would have to be so rapid as to stop all nuclear reactions essentially instantaneously, since at the densities and temperatures of the equilibrium considered reaction rates are very high. The actual situation would lie between these extremes and it is difficult to see exactly what changes would result in the initial distribution of elements, since it would be necessary to consider in detail all the reactions in this non-equilibrium process, perhaps including reactions involving nuclei in excited states. In particular, in all the exploding stellar models considered there would be a very high concentration of neutrons whose capture probabilities would increase as the material cooled, so that the distribution of elements would be radically changed, certainly at the densities involved. The radioactive decay of the neutron

<sup>\*</sup> Hoyle [80] suggests that the Crab Nebula, the supernova of 1054 A.D., had, prior to its outburst, a central temperature in excess of that required for his theory.

<sup>&</sup>lt;sup>†</sup> See the discussion section of reference [80].

does not help in this connection since the neutron lifetime is too long, and as will be seen later, neutroncapture reactions give important changes in element abundances even at densities of the order of  $10^{-8}$  g/cm<sup>3</sup>, whereas here the densities initially are perhaps 10<sup>20</sup> times as large. Furthermore, as has been seen, it is necessary to consider extremely neutron-rich nuclei in the equilibrium distribution in order to explain the existence of the heavy elements. Subsequent to an explosion these nuclei will at least undergo  $\beta$ -decay as suggested by all the authors discussed. In addition, however, many of these nuclei will be unstable with respect to particle emission.<sup>‡</sup> This effect will also change the distribution. It would appear that a complete solution of the problem of the formation of elements should involve serious consideration of non-equilibrium processes either in the prestellar state or in exploding stellar models.

Jensen and Suess [93] have also considered qualitatively the problem of the freezing-in of an equilibrium distribution in some detail. They have expressed the opinion that while the presently observed abundance distribution of the elements seems to reflect in some measure an original thermodynamic equilibrium, this distribution must have suffered considerable change both in its over-all features and in detail due to nonequilibrium processes during the freezing-in. They suggest that as the temperature drops in an expanding medium, proton reactions quickly become unimportant, the originally high neutron concentration will lead to large numbers of neutron reactions, and the heavy elements will be built up at the expense of the light elements. The present distribution should then reflect such neutron reactions and the associated  $\beta$ -disintegrations. This point of view raises the question of specifying an original equilibrium distribution which, as the result of non-equilibrium processes during the freezingin, would have evolved into the presently observed distribution. This is indeed a difficult problem and, as will be seen later, it is apparently not necessary to consider non-equilibrium processes as developing from an equilibrium state as the initial condition.

The equilibrium theory has represented the observed relative abundances of the light elements principally in trend and moderately well in detail. Certain deviations such as the low abundances of Li, Be, and B require special explanation. If the elements were formed in "prestellar" stars there would have been an opportunity for thermonuclear reactions to modify the "original" distribution. On the other hand, the situation is not so clear in theories of continual element formation in special stars. The distribution of heavy elements has been qualitatively explained by the introduction of special stellar models. In particular, one approach to this problem has been the introduction of the effect of gravitation on the equilibrium, while other approaches

have involved electrostatic effects and electron degeneracy. Clearly a satisfactory theory of element synthesis in stars should include all such effects, as well as the possible degeneracy of matter other than electrons. The physical conditions described by some investigators would seem to imply neutron and proton degeneracy, which has not always been taken into account. The possibly important role of neutrinos in stars does not seem to have been adequately covered either except perhaps by Klein in his radiation star model where neutrino equilibrium is implied.¶ For the physical conditions in the interiors of the special models described by some of these authors, one might expect a large amount of energy to escape via neutrinos so that the existence of thermodynamic equilibrium may perhaps be questioned. The argument is given by Hoyle [79]that this energy loss is small compared with the energy involved in the nuclear reactions and may therefore be neglected. However, Hoyle has also stated that the assembly in his stellar models is not in thermodynamic equilibrium because neutrinos escape and reactions involving neutrinos therefore are not thermodynamically reversible. In this connection he states that an equilibrium calculation is an adequate approximation to what is at best a steady-state problem. The validity of this assumption is not clear to us from his work.

Difficulties of the kind mentioned and in particular the freezing-in problem have led to the development of non-equilibrium theories of element formation, unattractive as they may be in requiring the discussion of specific nuclear reactions. A description of the nonequilibrium theories that have been developed constitutes the major portion of the remainder of this review.

#### IV. NON-EQUILIBRIUM THEORY

## (a) Introduction

The difficulties in explaining the origin and relative abundances of the elements according to an equilibrium theory have led to the examination of non-equilibrium processes. In addition, there are a number of indications that element formation probably took place in an early prestellar state of the universe, in which it is difficult to conceive of the existence for a sufficiently long time of the physical conditions required for an equilibrium among nuclei. There is a good deal of self-consistent evidence that the age of the universe is of the order of several billion years, including, among other things, the abundance ratios in radioactive families of elements, the dynamics of stellar clusters, and the expansion of the universe [25, 110].

The very existence of radioactive nuclei implies that

<sup>&</sup>lt;sup>‡</sup> The work of Mayer and Teller [114] and of Smart [134, 136] discussed later is pertinent to this point.

<sup>¶</sup> See, for example, the discussion of the possible role of neutrinos in stellar interiors by Cernuschi [32] and by Gamow and Schoenberg [62].

<sup>||</sup> For Tolman's very interesting discussion of this point see reference [159]. A paper by Saha and Nagchowdhury [128] is also of interest in this connection.

the elements have not existed in their present form for all time [4, 61]. There is of course the possibility that elements are formed continually, but this ignores the interesting agreement obtained between the apparent age of the elements and the universe as computed by a variety of independent methods. The observed uraniumlead abundance ratio and the measured half-life of uranium are consistent with each other and with the fact that the uranium half-life is of the order of the age of the universe. If one assumes that radioactive isotopes were equally abundant when formed, then from their presently observed relative abundances one may compute the epoch at which they were formed. For those sets of radioactive isotopes having measurable abundances now, this calculation consistently gives several billion years for the epoch of formation. In Table IX are listed several sets of naturally radioactive isotopes together with their isotopic abundances and half-lives. It is clear that elements whose half-lives are of the order of 10<sup>9</sup> years or greater are still quite abundant, while isotopes with short half-lives are scarce. Using the law of radioactive decay, one finds that K<sup>40</sup> would have been as abundant as  $K^{39}$  about  $1.0 \times 10^9$ years ago while the time of formation computed from  $U^{235}$  and  $U^{238}$  is about  $4.2 \times 10^9$  years. All of the evidence seems to indicate that there was an "event" of some kind several billion years ago which possibly included the formation of elements in essentially their presently observed relative abundances. Since, as already discussed in Section III, the conditions necessary for element formation do not appear to exist generally now nor do they appear to have existed in the known types of stars, one must examine the prestellar state, "unpleasant" as it may be to consider, to determine whether a non-equilibrium process can be satisfactorily developed. The fact that there appears to be a remarkable uniformity in the abundance of elements throughout the universe also favors consideration of the concept that the elements were formed in the prestellar state at one time uniformly throughout the universe. If this is the case then a non-equilibrium theory of element formation would be intimately connected with cosmology.

There has recently been developed a non-equilibrium theory of the formation and relative abundance distribution of the elements which involves neutron-capture reactions primarily. The importance of neutron-capture reactions has been recognized particularly for the heavy elements by Walke [164], Gamow [55], von Weiszäcker [176], Jensen and Suess [93] and Frank [52]. All but the last two of these investigators were concerned with equilibrium reactions furnishing stellar energy. Walke, for example, in order to explain the building-up of heavy elements, suggested a series of successive neutron captures by nuclei with intervening  $\beta$ -decay to adjust the charge. He pictured this as proceeding in a star where some reaction furnished a neutron source. Gamow [55] and von Weiszäcker [176] independently sug-

Isotope	Isotopic abundance in percent	Activity	Half-life in years
K <sup>39</sup>	93.38	None	
K <sup>40</sup>	0.012	β	$2.4 \times 10^{8}$
K41	6.61	None	
Sm <sup>144</sup>	3.16	None	
Sm147	15.07	None	
Sm <sup>148</sup>	11.27	α	$1.4 \times 10^{11}$
Sm149	13.84	None	
Sm <sup>150</sup>	7.47	None	
Sm <sup>152</sup>	26.63	None	
Sm154	22.53	None	
Rb <sup>85</sup>	72.8	None	
$\mathrm{Rb}^{87}$	27.2	β	6.3×1010
Th <sup>230</sup>		ά	8.3×10 <sup>4</sup>
$\mathrm{Th}^{232}$	100.0	α	$1.39 \times 10^{10}$
$U^{235}$	0.719	α	$7.07 \times 10^{8}$
$U^{238}$	99.274	α	$4.51 \times 10^{9}$

TABLE IX. Natural radioactive isotopes.

gested nearly the same concept. Frank suggested that the building of heavy elements by successive neutron captures with intervening  $\beta$ -decay would be a natural sequel to the thermodynamic equilibrium indicated for the light elements, although it is not stated whether heavy element formation would involve equilibrium or non-equilibrium reactions, nor is a locale mentioned. However, the possibly important role of neutron capture in a non-equilibrium process in the prestellar epoch was first suggested by Gamow [57], while Jensen and Suess [93] discussed the same question in connection with the freezing-in of a thermodynamic equilibrium. Recognizing the difficulty of applying equilibrium concepts to the prestellar state of the expanding universe, Gamow suggested a non-equilibrium process of neutron aggregation in which the final atomic species were subsequently formed by  $\beta$ -emission. It was also suggested that the longer time required to form the heavier complexes might explain the decrease in relative abundance with increasing atomic weight while the competition between neutron decay and radiative capture in the coagulation process may have led to the present high abundance of hydrogen.

These ideas have been developed into a non-equilibrium theory of element origin and formation by Alpher, Gamow, Herman, and Smart as well as by Fermi and Turkevich, whose work is discussed in the sections following.

## (b) The Prestellar State of the Expanding Universe

The problem of element formation in the prestellar state requires an estimate to be made of the physical conditions during this epoch. Some information can be obtained from our present knowledge of the universe as interpreted with the aid of relativistic cosmology [9]. The portion of the universe which has been studied in any detail is some  $5 \times 10^8$  light years in extent. If one accepts the observed red-shift of galaxies as indicating a true expansion, and this at present appears to be the only physically reasonable interpretation, the region observed is a fair sample of the universe. There are

three main features of this observed region, namely, large-scale homogeneity and isotropy, a smeared-out density of matter of the order of  $10^{-30}$  g/cm<sup>3</sup>, and a remarkably consistent relation between an apparent recession velocity of the galaxies and their separation  $\lceil 83 \rceil$ . The homogeneity and isotropy is large scale in the sense that there appears to be no preferred direction in space and the distribution of galaxies in space is observed to be essentially random. At the present time the mean density, 10-30 g/cm3, results from averaging the matter concentrated in galaxies alone, uniformly through space. Stebbins and Whiftord [140] have recently stated that intergalactic material may be present in sufficient quantity to raise the mean density from  $10^{-30}$  g/cm<sup>3</sup> for galactic material alone to  $10^{-28}$  g/cm<sup>3</sup> for all material. However, a change of this magnitude in the mean density does not materially alter the picture of the early state of the universe.\* The apparent recession velocity,  $v_r$ , of the galaxies as revealed by the Doppler shift is given by

$$v_r = Hl, \tag{88}$$

where H, Hubble's constant, is  $1.8 \times 10^{-17}$  (cm/sec.)/cm, and l is proper distance. The interpretation of Eq. (88) depends on accepting the concept that the red-shift does indeed imply recession. One may note that if the expansion of the universe has always been as described by the foregoing equation the apparent age of the universe would be about  $0.56 \times 10^{17}$  sec.  $(1.8 \times 10^9)$ years). It has not been conclusively demonstrated that the assumption of homogeneity is correct. For example, Omer [119] has recently developed a non-homogeneous cosmological model which satisfactorily represents many of the observed features of the universe. However, it does not appear that this model can be extrapolated back in time to the early prestellar epoch. The correct cosmology may involve homogeneity in the early epoch and inhomogeneity now. It would not appear to be too unreasonable, in examining the prestellar state, to extrapolate back in time with a homogeneous model. In the following discussion the general non-static homogeneous cosmological model, exhibiting spatial isotropy, is considered.

As is well known [157], the line element for such a model may be written as

$$ds^{2} = -e^{g(t)} [1 + r^{2}/(4R_{0}^{2})]^{2} (dr^{2} + r^{2}d\theta^{2} + r^{2}\sin^{2}\theta d\phi^{2}) + dt^{2}, \quad (89)$$

where r,  $\theta$ ,  $\phi$  are spherical polar coordinates, g(t) is an undetermined function of time, and  $R_0$  is a constant which determines the radius of curvature of the space. The quantity  $R_0$  may be real, imaginary, or infinite. Equation (89) is in the usual relativistic units. Let it

be supposed that the material in the universe consists of a homogeneous and isotropic mixture of non-interconverting radiation and matter which behaves as a perfect fluid. The relativistic energy equation may then be written, in relativistic units, as

$$(d/dt)e^{\frac{1}{2}g(t)} = \pm [(8\pi/3)\rho e^{g(t)} - R_0^{-2}]^{\frac{1}{2}},$$
 (90)

in which  $\rho$  is the density of mass, and the cosmological constant,  $\Lambda$ , is taken equal to zero. The radius of curvature, R, is given by

$$R/R_0 = l/l_0 = e^{\frac{1}{2}g(t)}, \qquad (90a)$$

where l is any proper distance, and  $l_0$ , the unit of length, together with  $R_0$ , the unit of radius of curvature, must be determined from the boundary conditions for Eq. (90). It should be noted that solutions of Eq. (90) will involve  $(l/l_0)$  and not l alone. The density of mass,  $\rho$ , which determines the geometry of the space includes the density of matter,  $\rho_m$ , and radiation,  $\rho_r$ , as well as density of kinetic energy. In the situations to be discussed the density of kinetic energy may be neglected. If matter is to be conserved,

$$\rho_m l^3 = \alpha = \text{constant.} \tag{91a}$$

If the universal expansion is adiabatic, so that the temperature, T, varies as  $l^{-1}$ , then for blackbody radiation,

$$\rho_r l^4 = \mathfrak{B} = \text{constant.}$$
 (91b)

In models of this type, energy is not conserved. Equations (91a) and (91b) may also be written as

$$\rho_r \rho_m^{-4/3} = \text{constant}, \tag{91c}$$

a condition which must hold throughout the expansion. Substituting Eqs. (91a) and (91b) in Eq. (90) and converting to c.g.s. units, leads to

$$dl/dt = + \left[ (8\pi G/3) (\alpha l^{-3} + \alpha l^{-4}) l^2 - c^2 l_0^2 / R_0^2 \right]^{\frac{1}{2}}, \quad (92)$$

where the positive sign indicates expansion and c and G are the velocity of light and the gravitational constant, respectively. Equation (92) can be integrated and the result given in the following form [9], where  $L=l/l_0$  is the running variable:

$$t = K_{1} + K_{2}^{-1} (\gamma \rho_{r''} + \gamma \rho_{m''} L + K_{2} L^{2})^{\frac{1}{2}} - [\gamma \rho_{m''} / (2K_{2}^{\frac{3}{2}})] \ln[(\gamma \rho_{r''} + \gamma \rho_{m''} L + K_{2} L^{2})^{\frac{1}{2}} + K_{2}^{\frac{1}{2}} L + \gamma \rho_{m''} / (2K_{2}^{\frac{1}{2}})], \quad (93)$$

where

and

$$K_{1} = \left[ \gamma \rho_{m''} / (2K_{2}^{\frac{3}{2}}) \right] \ln \left[ (\gamma \rho_{r''})^{\frac{1}{2}} + \gamma \rho_{m''} / (2K_{2}^{\frac{3}{2}}) \right] - (\gamma \rho_{r''} / K_{2}^{2})^{\frac{3}{2}}, \gamma = 8\pi G/3,$$
(93a)

 $K_2 = c^2 / |R_0^2|$ .

The quantities 
$$\rho_{m''}$$
 and  $\rho_{r''}$  are the densities of matter  
and radiation when  $L=1$ , and should not be confused  
with running variables. In order to evaluate the inte-  
gration constant one must specify the units  $R_0$  and  $l_0$ .

<sup>\*</sup>According to a more recent statement by A. E. Whitford [see Astron. J. 54, 138 (1949)], the suggestion of intergalactic material should perhaps now be abandoned. The possible presence of such material was based on the reddening of the light from distant elliptical galaxies, whereas spiral galaxies show little change of color with distance.

Equation (90) indicates that  $R_0$  can be determined if  $[l^{-1}(dl/dt)]l = l_0$ ,  $\rho_m$ , and  $\rho_r$  are specified at any given time. The first of these quantities is the expansion rate of space as determined by Hubble [83] and is equal to  $1.8 \times 10^{-17}$  sec.<sup>-1</sup>. The quantity  $\rho_m$  at the present epoch is taken as  $\rho_{m^{\prime\prime}}\!=\!10^{-30}~g/cm^3\!,$  and if we assume  $\rho_{m^{\prime\prime}}$  $\gg \rho_{r''}$ , then one may evaluate  $R_0$  and  $K_2$ . In doing this one takes L=1,  $l=l_0=10^{10}$  cm, i.e.,  $l_0$  is the side of a cube containing one gram of matter now, and finds  $R_0 = 1.7 \times 10^{27} (-1)^{\frac{1}{2}}$  cm and  $K_2 = 3.2 \times 10^{-34}$  sec.<sup>-2</sup>. For convenience it has been assumed that L=0 at t=0 but Eq. (93) will be employed in such a manner that this singularity is of no consequence. Clearly in utilizing Eq. (93) one must choose  $\rho_{m'}$  and  $\rho_{r'}$ , the densities at an earlier epoch, together with a corresponding value of L in such a way as to obtain the presently observed densities.

Approximate forms of Eq. (93) valid for early t will be of particular interest in studying the non-equilibrium problem in the prestellar state. For an expanding universe containing both matter and radiation, Eq. (93) may be written as  $\lceil 9 \rceil$ 

$$t = (4\gamma \rho_{r''})^{-\frac{1}{2}} L^2 + \left[ \rho_{m''} / (6\gamma^{\frac{1}{2}} \rho_{r''}^{\frac{1}{2}}) \right] L^3 + (4\gamma \rho_{r''})^{-\frac{1}{2}} \left\{ \left[ 3\gamma \rho_{m''}^2 / (4\rho_{r''}) \right] - K_2 \right\} L^4 + \cdots, \quad (94)$$

where the condition for early *t* may be written

$$L\{(\rho_{m''}/\rho_{r''}) + [K_2 L/(\gamma \rho_{r''})]\} < 1.$$
(94a)

There are two other approximate forms of interest, namely, for the cases of a universe consisting of radiation only or of matter only, which are as follows. For radiation only,

$$t = (4\gamma \rho_{r''})^{-\frac{1}{2}} L^{2} [1 - K_{2} (4\gamma \rho_{r''})^{-1} L^{2} + K_{2}^{2} (4\gamma \rho_{r''})^{-2} L^{4} - \cdots ]$$
  
$$= (4\gamma)^{-\frac{1}{2}} \rho_{r}^{-\frac{1}{2}} [1 - K_{2} (4\gamma \rho_{r''})^{\frac{1}{2}} - \rho_{r}^{-\frac{1}{2}} + 2K_{2}^{2} (4\gamma \rho_{r''})^{\frac{1}{2}} - \rho_{r}^{-1} - \cdots ], \quad (95)$$

and for matter only,

$$t = (2/3)(\gamma \rho_{m''})^{-\frac{1}{2}} L^{\frac{1}{2}} [1 - (3/10)K_{2}(\gamma \rho_{m''})^{-1}L + (9/56)K_{2}^{2}(\gamma \rho_{m''})^{-2}L^{2} - \cdots ]$$
  
= (2/3) $\gamma^{-\frac{1}{2}} \rho_{m}^{-\frac{1}{2}} [1 - (3/10)K_{2}(\gamma \rho_{m''})^{\frac{3}{2}} - \rho_{m}^{-\frac{1}{2}} + (9/56)K_{2}^{2}(\gamma \rho_{m''})^{\frac{3}{2}} - \rho_{m}^{-\frac{3}{2}} - \cdots ], \quad (96)$ 

where  $\rho_{r''}$  and  $\rho_{m''}$  are as defined in Eqs. (93). Clearly, for sufficiently early t one has for radiation only,

$$\rho_r = (4\gamma)^{-1} t^{-2} = 4.48 \times 10^5 t^{-2} \text{ g/cm}^3,$$
 (97)

while for a matter universe only,

$$\rho_m = [4/(9\gamma)]t^{-2} = 7.94 \times 10^5 t^{-2} \text{ g/cm}^3.$$
 (98)

In a radiation universe expanding adiabatically, the temperature  $T \propto l^{-1}$ , so that from Eq. (91b)

$$T = (c^2/a_r)^{\frac{1}{4}} \rho_r^{-\frac{1}{4}}, \tag{99}$$

where the radiation density constant  $a_r = 7.57 \times 10^{-15}$  erg cm<sup>-3</sup> deg.<sup>-4</sup>, and for early *t*,

$$T = 1.52 \times 10^{10} t^{-\frac{1}{2}} \text{K}.$$
 (100)

In a universe in which the expansion is controlled by radiation, i.e.,  $\rho_r \gg \rho_m$ , it can be shown that the matter density will vary as follows for early t:

$$\boldsymbol{\rho}_m = (4\gamma)^{-\frac{3}{4}} \rho_{m''} \rho_{r''}^{-\frac{3}{4}} t^{-\frac{3}{2}}.$$
 (101)

This involves the assumption of a temperature equilibrium between matter and radiation.

The validity of the relationships  $\rho = \rho(t)$  is questionable for very early times, i.e., in the vicinity of the singularity at t=0, when the energy of light quanta was comparable to the rest mass energy of elementary particles.\*\* Einstein [43] has pointed out that there is a difficulty at very early times because of the separate treatment of the metric field (gravitation) and electromagnetic fields and matter in the theory of relativity. For large densities of field and of matter the field equations and even the field variables which enter into them will have no real significance. In the application to be discussed the "beginning" does not enter the problem and hence the difficulty is avoided.

In order to utilize the foregoing development, it is necessary to specify  $\rho_{m''}$  and  $\rho_{r''}$ . While it appears that one need specify the matter and radiation densities at the present time only, because of Eq. (91c) this is equivalent to specifying these densities at any other epoch, and in particular at the epoch of element formation.

#### (c) Neutron-Capture Cross Sections

There are several alternative explanations for the apparent correlation of the relative abundances of atomic nuclei with their systematic properties. As has been seen, the correlation with nuclear binding energies forms the principal basis of the equilibrium theory. On the other hand, the non-equilibrium neutron-capture theory of the relative abundance distribution of the elements to be described is derived from a correlation of the radiative-capture cross sections of nuclei for neutrons with their relative abundances. The essence of this relationship is that whereas the relative abundances decrease approximately exponentially with increasing atomic weight up to  $A \cong 100$ , the neutroncapture cross sections increase exponentially up to  $A \cong 100$ . For atomic weights greater than 100 the relative abundances and cross sections remain about constant.<sup>††</sup>

The neutron-capture cross sections of atomic nuclei vary with the incoming neutron energy. In formulating a neutron-capture theory of element formation it is necessary to take into account this energy variation and to simplify the problem, select a mean energy corresponding to the physical conditions during the formation process. One is assisted in the latter choice

<sup>\*\*</sup> For example, the production of electron-positron pairs would have persisted perhaps until kT dropped to about 1 Mev and might alter the very early cosmological picture.

<sup>&</sup>lt;sup>††</sup> A theoretical discussion of this general behavior of the capture cross sections of the elements has been given by Feshbach, Peaslee, and Weisskopf [50].

by the fact that the relative abundance data do not reflect the enormous resonances which are observed in the cross sections of certain nuclei up to neutron energies of the order of  $10^3-10^4$  ev. Furthermore, it is clear that the average thermal energy of the nuclei being formed could not have exceeded the mean binding energy per nucleon without dissociation resulting. Thus one is led to assume that the temperature during the element forming process must have corresponded to mean thermal energies for the nuclei of the order of 0.1 Mev. The possibility cannot be excluded that the neutronrich unstable nuclei, which may have existed during the formation process, led to peaks in the presently observed abundance data because of resonances. However, as will be seen, it is reasonable to suppose that the nuclear species involved in the neutron-capture process were not too different from the stable nuclei.

The most important evidence for the correlation of relative abundances with neutron-capture cross sections results from the recent work of Hughes and collaborators [84-86, 132].‡‡ Capture cross sections were measured for a number of nuclear species utilizing unmoderated fission neutrons from a pile. The effective energy of the neutrons was about 1 Mev. The cross sections measured were actually an average over the spectrum of fission neutron energies. The fact that cross sections vary about as 1/E for all elements near 1 Mev enabled the conversion of measured cross sections corresponding to the known fission spectrum to capture cross sections at 1 Mev. The data of Hughes, Spatz,



FIG. 11. Logarithm of the observed neutron-capture cross section at 1 Mev versus atomic weight. These data are:  $\bigcirc$ , Hughes and collaborators [84-86];  $\diamond$ , Dementi and Timoshuk [38]; +, Griffiths [68];  $\times$ , Halban and Kowarski [75];  $\square$ , Mescheryakov [115]; and  $\bullet$ , Los Alamos [86]. All the data were adjusted to Hughes' point for Au<sup>197</sup> denoted as the fitting point. The point at A = 48 with a downward arrow is an upper limit to the cross section. The very low points at A > 100 are near magic number nuclei. Data for magic number nuclei are not included. The straight lines were fitted to Hughes' data [see Eq. (102)].

and Goldstein [86] and of Hughes and Sherman [85] are given in Fig. 11 where the logarithm of capture cross section at 1 Mev is plotted *versus* atomic weight. It may be noted that the radiative-capture cross sections at 1 Mev are almost negligible compared with the total cross sections at the same energy. The latter are of the order of 1-10 barns and are practically independent of atomic weight [66].

The data of Hughes and collaborators appear to be the most reliable in that the neutron energy was well determined. However, several other investigators have determined capture cross sections in experiments in which neutron energies were not so well defined. The relative cross sections measured for different nuclear species at the particular neutron energies used in the experiments, however, should be reliable. If the cross sections measured in these experiments are multiplied by a constant to bring them into agreement with the known absolute cross section of a particular nuclear species, then one can use these data together with those of Hughes and collaborators. Adjusting to Au<sup>197</sup> as the standard, as suggested by Hughes, Spatz, and Goldstein [86], the data¶¶ of Dementi and Timoshuk [38], Griffiths [68], Halban and Kowarski [75], Mescheryakov [115], and Los Alamos, || || have been plotted together with Hughes' data in Fig. 11. Apart from the magic number nuclei, which have been omitted in plotting this collection of cross-section data, the capture cross sections exhibit a remarkably regular dependence on atomic weight. The main features of this dependence are well represented by the two straight lines shown in Fig. 11. These lines were adjusted to fit the cross-section data of Hughes and collaborators separately for atomic weight A < 100 and A > 100. On the basis of the crosssection data, the division at A = 100 seemed most reasonable. Thus, for neutron energies of 1 Mev the capture cross sections of the elements are approximately represented by [5]

$$\log \sigma = 0.03A - 4.00, A < 100,$$
 (102a)

and

$$\log \sigma = -1.00, A > 100,$$
 (102b)

where  $\sigma$  is in barns.

In view of the experimental difficulty of obtaining

¶¶ In a recent paper by Allen, Bishop, Demers, and Halban [3], ratios of cross sections at 220 and 950 kev are given for a number of different nuclei. Since it was not possible to reduce these data in the manner described, they are not included in Fig. 11. The data of Fields, Russell, and Wattenberg [86] are also not included because they do not give the cross section of  $Au^{19}$  which was used as the fitting point. The neutron cross-section data obtained at Los Alamos with a Van de Graaff accelerator were not suitable for inclusion in this comparison because the cross-section measurements for the various nuclear species were made at the same energies in too few cases. Only those data of Dementi and Timoshuk which were given in reference [86] have been included here.

|| || The cross-section data from Los Alamos were read from graphs kindly supplied by Dr. Hughes and adjusted according to the procedure outlined. These data resulted from photo-neutron measurements and appropriate credit to the investigators was given by Hughes who reported their unpublished results.

<sup>&</sup>lt;sup>‡‡</sup> We are grateful to Dr. Hughes for furnishing to us some additional cross-section values measured since the publication of reference [86].



FIG. 12. Logarithm of observed neutron-capture cross section at 1 Mev versus neutron content plotted according to the key given in Fig. 11. Magic number nuclei, 50, 82, and 126 neutrons, are included.

capture cross-section data, the scatter evident in Fig. 11 is quite small. In Hughes' data, the most accurate of those shown, the measured cross sections lie within a factor of about two from the straight lines. The data of other investigators scatter somewhat more but this might have been expected since the neutron energy and flux in these cases are not as well defined as in Hughes' measurements.

In the neutron-capture theory to be described, Eqs. (102) are taken as defining the neutron-capture cross sections of the elements at 1 Mev. This approximation ignores such detailed features as the variation of cross section with even and odd A, the small cross sections associated with the magic number nuclei, and the more complicated behavior of the cross sections of the very light elements. In addition, Hughes\* has recently found evidence that nuclei near the magic numbers have quite small cross sections. Note in Fig. 11 the low cross sections for Ce<sup>142</sup> (84 neutrons), Tl<sup>203</sup>, Tl<sup>204</sup>, Tl<sup>205</sup> (122, 123, and 124 neutrons, respectively).

The investigation of nuclear mass defects [21] indicates that even numbers of neutrons and protons lead to nuclei of higher net binding energy than do odd numbers of either. As a result of the Pauli principle, a pair of neutrons or protons plays much the same role in nuclear structure as closed shells in atomic structure. Thus, a third neutron would have to reside in a higher state and consequently be less strongly bound than a pair of neutrons. In his paper on the semi-empirical theory of the nuclear energy surface, Feenberg [46] deduced an expression for the excitation energies of intermediate nuclei formed by the capture of neutrons whose energies are small compared to the additional binding energy. His computations indicate that, because of the two-particle "shell" structure, and because of the relative displacement of equivalent proton and neutron levels arising from the electrostatic repulsion between protons, nuclei of even charge and odd atomic weight



FIG. 13. Correlation plot of observed neutron-capture cross sections at 1 Mev and observed universal relative abundances. The data are those of Hughes and collaborators [84-86], and Brown [30]. The straight line shown was fitted by least squares.

should exhibit larger radiative-capture cross sections than neighboring nuclei. For example, this behavior is exhibited by the isotopes of mercury [87]. Unfortunately, the even-odd variation is not apparent in the available cross-section data because of their scatter and because there are practically no instances of data for nuclei of successive atomic weights. The question of the even-odd variation and its specific interpretation in a non-equilibrium theory has been discussed by Smart [136] [see Section IV(d)3].

The available data on the cross sections of nuclei having 50, 82, or 126 neutrons, or 50 or 82 protons, were not included in Fig. 11. As first pointed out to the authors by Wigner and Way,<sup>†</sup> the apparent existence of some type of completed shell structure in nuclei of this type leads to abnormally small capture cross sections compared to neighboring nuclei. Discussions of these magic number nuclei have been given by a number of investigators [see Section II(b)]. To demonstrate this detail in the behavior of the capture cross sections of the elements, cross-section data at neutron energies of 1 Mev for all species<sup>‡</sup> are plotted against the neutron content of the nuclei in Fig. 12. A similar plot has not been made against proton content because the only available cross-section datum point is 82Pb<sup>208</sup> which also contains 126 neutrons.

The general behavior of the neutron-capture cross sections as a function of atomic weight as approximately represented by Eqs. (102) has been used by Alpher and Herman [5, 8] in the formulation of a neutron-capture theory of element formation. Detailed consideration has been given to the formation of the very light elements by Fermi and Turkevich¶ who have taken into account the actual cross-section variations and reactions other than neutron-capture in a non-equi-

\* Private communication.

<sup>†</sup> Private communication.

<sup>&</sup>lt;sup>‡</sup> It is pertinent to note that Hughes, Spatz, and Goldstein [86] interpret the regular increase of cross section with atomic number among the 82-neutron group of nuclei as arising from a decrease in stability as protons are added.

<sup>¶</sup> Private communication.

librium process. This work is discussed in Section IV(d)2.

The correlation between neutron-capture cross section and relative abundance is illustrated in Fig. 13 where the data of Hughes and collaborators at 1 Mev (excluding magic or near magic number nuclei) have been plotted against the relative abundance of the particular nuclei as given by Brown [30]. Thus, it is evident that there is in general an inverse correlation between capture cross section and relative abundance. This correlation, considered together with the fact that magic number nuclei are more abundant than neighboring nuclei and that even A nuclei are more abundant than odd A nuclei, suggests the possibility that the elements were formed by a neutron-capture process. If the various atomic nuclei were indeed formed by the successive capture of neutrons, one might expect to find evidence for the role of  $\beta$ -disintegrations in establishing the final distribution of stable nuclei from neutron-rich isobars. Given an unstable neutron-rich nucleus,  $\beta$ -disintegrations should stop when a nucleus is reached having the lowest possible charge consistent with stability. Therefore, it is pertinent to note that the observational data on relative abundance indicates that the most abundant of a group of isobars is generally the one with lowest charge [see Section II(b)]. This result lends credence to the idea that the abundance distribution with respect to atomic number was established in final form by  $\beta$ -decay processes.

The principal assumption involved in the use of the cross-section data presented above in a simple neutroncapture theory is that the cross-section values are strictly a function of the atomic weight and do not vary appreciably with neutron content. This assumption as well as several other points connected with the use of these data are more fully discussed later.

#### (d) Development of the Non-Equilibrium Theory

## 1. Neutron-Capture Theory

As has been seen, there is considerable evidence that the neutron-capture cross sections of the elements may have played an important role in the process by which the elements were formed. There is also evidence indicating that the element-forming process may have taken place in the "beginnings" of the evolution of the expanding universe. While the rapid changes in physical conditions with time vitiate the expanding universe as a locale for the equilibrium formation of the elements, the dilution of the material by the expansion would be expected to act as one of the controlling mechanisms in a non-equilibrium theory. The process of element formation is conceived as an integral part of the early stage of the expanding universe and the following

qualitative picture has been suggested. Very shortly after the start of the universal expansion the material in the universe consisted of neutrons only. It is assumed that the matter density was sufficiently low to permit the decay of neutrons. On the other hand, it is supposed that for some time after the start of the expansion the temperature was sufficiently high to prevent the building up of nuclei in appreciable quantities, because of thermal dissociation and photo-disintegration processes. After some time, when the universe had expanded and cooled sufficiently, it is suggested that deuterons would be formed by the capture of neutrons by protons, and that successive neutron captures would build the heavier nuclei. It is assumed throughout the development of the neutron-capture theory that it is not possible to have nuclei built of neutrons only. The time between successive neutron captures is assumed to be sufficiently long to allow adequate charge adjustment by  $\beta$ -decay of the nuclei being formed. Consequently, the density of matter must be sufficiently low so as to provide for  $\beta$ -decay. Otherwise,  $\beta$ -decay rates would be the controlling factor and very neutron-rich nuclei would have to be considered in computing capture probabilities. Actually it would appear that the density was such that  $\beta$ -decay and neutron capture were of about equal importance. The neutron-capture rate depends upon the capture cross sections of the elements as well as upon the density and temperature of the reacting material. The relative amounts of the various nuclei formed will depend upon the density and temperature assigned for the process, the rate of the dilution of matter caused by the universal expansion, and the rate at which the neutron concentration decreases by virtue of neutron  $\beta$ -decay. As will be seen, a neutroncapture theory can be developed which requires only the specification of the density of matter and of radiation at a given epoch in the expanding universe. Qualitatively it may be seen that a very low matter density will yield principally hydrogen as the end product, whereas a very high density will lead to an overabundance of the heaviest elements. The reduction of the neutron concentration as a result of both capture and decay, as well as the reduction of reaction rates as matter dilutes in the expansion, would terminate the element-building process in a time of the order of several neutron lifetimes.

In the general formulation of the neutron-capture theory [8] it is assumed that radiative capture of neutrons is the only important particle reaction, and that thermal dissociation and photo-disintegration may be neglected. Except possibly for the very lightest elements neutron-capture reactions would certainly predominate by virtue of the lack of a Coulomb barrier.

Let V be any finite volume element in the universe,  $\mathfrak{N}_j$  the number of nuclei of species j in that volume, where j is the number of nucleons, and let  $C_j$  be given by  $C_j = \mathfrak{N}_j/V$ . Assuming that the composition of the

<sup>||</sup> There are no cross-section data for the very light elements or for the elements in the vicinity of the iron peak. Inclusion of these data together with those of magic number nuclei would undoubtedly complicate the correlation.

universe was homogeneous, one may write

$$\frac{dC_j}{dt} = \frac{1}{V} \frac{d\mathfrak{N}_j}{dt} - \frac{C_j}{V} \frac{dV}{dt}.$$
(103)

It has already been seen that in a universe consisting of a homogeneous, isotropic, perfect fluid the density of matter,  $\rho_m$ , varies during the early stages of the universal expansion according to

$$\rho_m \propto t^{-\psi}.\tag{104}$$

If the universe contains matter only, the exponent  $\psi = 2$ , according to Eq. (98), whereas if the matter is but a trace in a universe whose expansion is controlled by radiation, then according to Eq. (101),  $\psi = 3/2$ . Since  $\rho_m = \mathfrak{M}/V$ , where  $\mathfrak{M}$  is the mass of the matter in the volume V, it follows that

$$\frac{1}{\rho_m} \frac{d\rho_m}{dt} = -\frac{1}{V} \frac{dV}{dt} = -\frac{\psi}{t}.$$
 (105)

Substitution of Eq. (105) into Eq. (103) yields

$$\frac{dC_j}{dt} = \frac{1}{V} \frac{d\mathfrak{N}_j}{dt} - \frac{\psi C_j}{t}.$$
 (106)

The rates of change of the total number of neutrons,  $\mathfrak{N}_n$ , protons,  $\mathfrak{N}_1$ , and nuclei of species j,  $\mathfrak{N}_j$ , in the volume V, are given by

$$d\mathfrak{N}_n/dt = -\lambda\mathfrak{N}_n - \sum_{j=1}^J p_j C_n \mathfrak{N}_j, \qquad (107a)$$

$$d\mathfrak{N}_1/dt = +\lambda\mathfrak{N}_n - p_1 C_n \mathfrak{N}_1, \qquad (107b)$$

and, in general,

and

$$d\mathfrak{N}_j/dt = p_{j-1}C_n\mathfrak{N}_{j-1} - p_jC_n\mathfrak{N}_j, \qquad (107c)$$

where  $\lambda$  is the neutron decay constant,  $p_j$  is the effective neutron-capture volume swept out per second by nuclei of species j, and J is the total number of nuclear species differing only in atomic weight A. If one sets  $\tau = \lambda t$ , substitutes Eqs. (107) into Eq. (106), and normalizes the particle concentrations  $C_n$ ,  $C_1$ , and  $C_j$  with respect to the nucleon concentration  $C_0$  at time  $t=t_0$ , the following expressions result:

$$d\xi_n/d\tau = -\left[1 + (\psi/\tau)\right]\xi_n - \sum_{j=1}^J P_j \xi_n \xi_j, \quad (108a)$$

$$d\xi_1/d\tau = \xi_n - P_1\xi_n\xi_1 - (\psi\xi_1/\tau), \qquad (108b)$$

and, for the remaining nuclear species,

$$d\xi_{j}/d\tau = P_{j-1}\xi_{n}\xi_{j-1} - P_{j}\xi_{n}\xi_{j} - (\psi\xi_{j}/\tau), \quad (108c)$$
 where

$$\xi_j = C_j / C_0 \tag{108d}$$

$$P_j = p_j C_0 / \lambda. \tag{108e}$$

It is evident that Eqs. (108) state that the neutron concentration is decreased by decay and capture, the proton concentration is increased by neutron decay and decreased by the formation of deuterons, the concentration of species j is increased as a result of neutron capture by the species (j-1) and decreased as it captures neutrons, and, finally, all concentrations decrease according to the universal expansion.

Equations (108) may also be written in terms of concentrations by weight,  $x_j$ , given by

$$x_j = m_j C_j / \rho_m, \qquad (109)$$

where  $m_j$  is the mass in grams of the *j*th nuclear species. In terms of the  $x_j$ , Eqs. (108) become

$$dx_n/d\tau = -x_n - \sum_{j=1}^{J} [p_j \rho_m/(\lambda m_j)] x_n x_j, \quad (110a)$$

$$dx_1/d\tau = x_n - \left[ p_1 \rho_m / (\lambda m_1) \right] x_n x_1, \qquad (110b)$$

and, for the *j*th species, j > 1,

$$dx_{j}/d\tau = (m_{j}/m_{n}) \{ [p_{j-1}\rho_{m}/(\lambda m_{j-1})] x_{n}x_{j-1} \\ - [p_{j}\rho_{m}/(\lambda m_{j})] x_{n}x_{j} \}, \quad (110c)$$

where to a sufficient approximation one may take  $m_j/m_n=j$ . It can be seen readily that the general equation for the *j*th species, Eq. (110c), in terms of particle concentration can be written as<sup>\*</sup>

$$d\eta_j/dz = P_{j-1}\eta_{j-1} - P_j\eta_j, \qquad (111)$$

$$\xi_j = \eta_j \tau^{-\psi} \tag{111a}$$

$$z = \int_{\tau_0}^{\tau} \xi_n d\tau.$$
 (111b)

In terms of concentration by weight Eq. (110c) becomes

$$dx_{j}/dy = (p_{j-1}/m_{j-1})x_{j-1} - (p_{j}/m_{j})x_{j}, \qquad (112)$$

where

where

and

$$y = (j/\lambda) \int_{\tau_0}^{\tau} \rho_m x_n d\tau.$$
 (112a)

The integrands in Eqs. (111b) and (112a) are singular at  $\tau=0$  so that one must take  $\tau_0>0$ . This, as already mentioned, implies the choice of an initial time at which a neutron-capture process would start. To be sure, it is not physically correct to speak of a definite starting time for the process. In the early stages of the expansion there are competing processes, i.e., photodisintegration and thermal dissociation, whose effects decrease approximately exponentially with time and become unimportant so that the process of successive neutron captures quickly becomes predominant. Clearly the inclusion of all the competing processes in a single

<sup>\*</sup> If all the reactions in Eq. (108) were of the same order it would then be possible to completely eliminate the explicit dependence on  $\tau$  and determine ratios of concentrations independently of the nature of the expansion. However, this is not the case.

mathematical formulation imposes serious computational difficulties. This is so because it then becomes necessary to solve the growth equations simultaneously for all j. However, as will be seen later it is possible and quite reasonable to select a finite starting time such that processes other than neutron capture need not be considered, at least in a first approximation.

In any approximation the solutions of, say, Eqs. (108) requires the specification of the initial concentrations of the various nuclear species as well as the coefficients  $P_j = p_j C_0 / \lambda$ . In order to calculate  $p_j$ , assume first that the medium may be treated as an ideal gas. It will be seen that this is a sufficient approximation since the density and temperature during the elementforming process are found to be of the order of  $10^{-6}$ - $10^{-8}$ g/cm<sup>3</sup> and 10<sup>9</sup> K, respectively. It shall also be assumed that reaction rates are sufficiently high so that the particles involved may be considered to be in thermal equilibrium at any instant of time. The reasonableness of this assumption may be seen from a comparison of the process rate for a given nuclear species with the rate of dilution due to the expansion. From the kinetic theory of gases it follows that the number of neutrons captured per second per unit volume by nuclei of atomic weight A(=j) with collision energies in the range dE at E may be written for a given temperature as follows:

$$Bm_n {}^{\dagger}C_n C_j [(m_n + m_j)/(m_n m_j)] {}^{\dagger}\sigma(j, E)E \\ \times \exp[-E/(kT)] dE, \quad (113)$$

where

$$B = [8/(\pi m_n)]^{\frac{1}{2}}(kT)^{-\frac{3}{2}}, \qquad (113a)$$

and  $\sigma(j, E)$  is the neutron-capture cross section. Approximating  $m_j/m_n \cong j$ , the total number of transmutations of nuclei from atomic weight j to (j+1) per unit volume per second is given by

$$\mathbf{N}_{T}(j) = BC_{n}C_{j}[(1+j)/j]^{\frac{1}{2}}$$
$$\times \int_{0}^{\infty} \sigma(j, E)E \exp[-E/(kT)]dE. \quad (114)$$

From Eq. (114) it is clear that for a given T, since  $N_T(j) = p_j C_n C_j$ , one has

$$p_j = B[(1+j)/j]^{\frac{1}{2}} \int_0^\infty \sigma(j, E) E \exp[-E/(kT)] dE. \quad (115)$$

An examination of the experimental data on capture cross sections led to Eqs. (102), relating the cross section to the atomic weight at neutron energies of 1 Mev. The energy dependence of capture cross sections [179] has been found to be  $\sigma \propto E^{-\frac{1}{2}}$  for low and medium energies, and  $\sigma \propto E^{-1}$  for high energies (up to Mev energies). However, if kT in Eq. (115) is of the order 0.1 Mev, then the main contribution to the integral from  $\sigma$  will be in this region, and it should be a sufficient approximation to use  $\sigma \propto E^{-\frac{1}{2}}$ . Rewriting Eqs. (102) to include the energy dependence as  $E^{-\frac{1}{2}}$ , one obtains

$$\log(\sigma E^{\frac{1}{2}}) \cong -30.886 \pm 0.03j, j < 100,$$
 (116a)

$$\log(\sigma E^{\frac{1}{2}}) \cong 27.886, \quad j > 100,$$
 (116b)

if  $\sigma$  is in cm<sup>2</sup> and E is in ergs. Inserting these expressions in Eq. (115), and integrating, it follows that

$$p_j = 1.4 \times 10^{-19+0.03} [(1+j)/j]^{\frac{1}{2}} \text{ sec.}^{-1}, j < 100, (117a)$$

and

and

$$p_j = 1.4 \times 10^{-16} \text{ sec.}^{-1}, j > 100,$$
 (117b)

where in Eq. (117b) (1+j)/j has been replaced by unity with sufficient accuracy. Had the dependence  $\sigma \propto E^{-1}$  been used, the values of  $p_j$  given in Eqs. (117) would have differed by a multiplicative factor  $(\pi kT/4)^{-\frac{1}{2}}$ where kT is in Mev. Let us examine the possible effect of this multiplicative factor. If one assumes that the capture process started at  $kT \cong 0.1$  Mev, and is essentially over when  $kT \cong 0.03$  Mev (this corresponds to a time change from about 170 to 2000 sec., i.e., several neutron half-lives), then the  $p_i$  calculated using  $\sigma \propto E^{-1}$ would vary from  $\sim \frac{1}{3}$  to  $\sim \frac{1}{2}$  of the values given by Eq. (117). In view of the lack of precision of the crosssection measurements, since the cross-section dependence does not involve the same power of E for all Ebelow 1 Mey, and, further, since the energy dependence of  $\sigma$  varies somewhat with *i*, the approximation represented by Eqs. (117) seems reasonable.

The neutron-capture process has been examined quantitatively in several successively improved approximations.\*\* Perhaps the simplest of these [4-6] involved the assumptions, first, that the effect of the universal expansion could be neglected, and, second, that the concentrations of neutrons and protons were constant during the process. This implies a constant rate of deuteron formation. With regard to the first of these assumptions, examination of Eq. (101) shows that if the process went on during the time between  $\sim 100$  and  $\sim$ 1000 sec., the decrease in matter density was between one and two orders of magnitude. The second assumption is indeed a crude one, ignoring as it does the decay of neutrons as well as the neutrons and protons used in building the elements. The latter effect must be relatively small since the preponderance of the final material is hydrogen. As a result of the foregoing discussion, Eqs. (107) may be written approximately as

$$\frac{1}{C_n} \frac{dC_j}{dt} = p_{j-1}C_{j-1} - p_jC_j, \quad j > 1.$$
(118)

<sup>\*\*</sup> It is interesting to note (see reference [9]) that the correlation between neutron-capture cross section,  $\sigma$ , and relative abundance,  $\alpha$ , shown in Fig. 13 may be expressed as  $\log \sigma = a_1 \log \alpha + a_2$ , where the *a*'s are constants. Combining this result with Eq. (102), one can derive relationships of the form  $\log \alpha = a_3A + a_4$ , A < 100, and  $\log \alpha = \text{constant}$ , A > 100, which give two straight lines approximately representing the observed abundances.

Range of atomic weights	$\overline{j}$	$p(\overline{j})$ cm <sup>3</sup> /sec.
1- 20	10.5	0.16×10 <sup>-19</sup>
21-40	30.5	0.63×10 <sup>-19</sup>
41-60	50.5	0.25×10 <sup>-18</sup>
61-80	70.5	$0.98 \times 10^{-18}$
81-100	90.5	$0.39 \times 10^{-18}$
101-120	110.5)	
121-140	130.5	
141-160	150.5	
161-180	170.5	$0.70 \times 10^{-17}$
181-200	190.5	
201-220	210.5	
221-240	230.5	
	,	

According to Bateman [16],<sup>††</sup> the solutions of Eqs. (118) subject to the conditions,  $C_n = \text{constant}$ ,  $C_1 = \text{constant}$ ,  $C_j(t=0)=0$ , are given by

$$C_2/C_1 = (p_1/p_2)[1 - \exp(-p_2C_n t)],$$
 (119a)

$$C_{3}/C_{1} = (p_{1}/p_{3}) \{1 - p_{2}p_{3}[(p_{2}p_{3} - p_{2}^{2})^{-1} \exp(-p_{2}C_{n}t) + (p_{2}p_{3} - p_{3}^{2})^{-1} \exp(-p_{3}C_{n}t)]\}, \quad (119b)$$

etc., where there is obviously the restriction that no two p's may be equal. Since both  $C_n$  and  $C_1$  are taken as constant and since one is interested only in relative values of the  $C_j$ , one may replace  $C_j/C_1$  by  $C_j/C_n$ . Equations (119) prove difficult to evaluate with any degree of accuracy in practice and are not valid for j > 100. Consequently, Eqs. (118) were integrated numerically. In order to reduce the number of equations to be solved, and it is clear that equations of this type can be solved successively, the nuclear species can be grouped. In calculating the  $p_j$  it has been found that, since the ratio  $p_{j+1}/p_j$  is very nearly unity, one obtains approximately the same value of a mean p(j) for a group of as many as twenty species, whether one considers the arithmetic, geometric, or harmonic mean. The method of grouping may be described as follows. Consider two adjacent groups containing q and q'species, respectively. The average  $p_i$  for what is considered to be the representative element of the group qis clearly

$$\bar{p}_j = (\sum p_j)/q, \qquad (120)$$

where the sum is taken over the group q. However, it requires (q+q')/2 successive neutron captures, on the average, to go from the representative element of the group q to that of the group q'. Hence, the effective neutron-capture volume swept out per second by the representative "species" of the group q, going into the group q', may be written

$$p(j) = \bar{p}_j / [(q+q')/2].$$
 (121)

This is not the only way of looking at the average p for a group. However, an examination of the several

ways indicates that the resulting p(j) is not very critical to the procedure used, and in this kind of calculation differences of less than an order of magnitude can be neglected.

In the approximation under discussion, the grouping and the effective p(j) are shown in Table X.<sup>‡‡</sup> Solutions obtained with the values of p(j) in Table X are shown in Fig. 14, where  $\log[C(j)/C_n]$  is plotted versus  $\log(C_n t)$ . It is clear from Fig. 14 that one has in this case a steady-state problem and the steady-state values of  $C(j)/C_n$  are inversely proportional to their respective p(j), as may be seen from Eqs. (119) for large  $C_n t$ . The calculated steady-state abundances are not particularly significant because physically one does not have a steady state, the calculated abundances do not agree with those observed, and processes such as fission, which would completely modify the theory in a steadystate approximation, have been ignored. The values of  $C(\mathbf{j})/C_n$  at any  $C_n t$  are the relative abundances of the grouped species. In this approximation one must select a value of  $C_n t$  such that the abundances correspond to those observed. Physically this implies termination of the process by neutron decay and expansion, factors not included in this approximation. The set of computed relative abundances corresponding to  $\log(C_n t)$ =17.91, i.e.,  $C_n t = 0.81 \times 10^{18}$  sec./cm<sup>3</sup>, was selected as giving the best fit. A curve drawn through the grouped abundances is compared in Fig. 15 with the universal



FIG. 14. Relative concentrations as functions of time in the neutron-capture theory approximation with no neutron decay or universal expansion, according to Alpher [5]. The  $\bar{j}$  denote the atomic weights of the representative elements of the groups of elements for which the calculations were made. The neutron concentration,  $C_n$ , was taken as constant for the process.

<sup>&</sup>lt;sup>††</sup> A discussion of equations of this type has been given by Rubinson [126], who has considered the problem of nuclei subjected to neutron capture as well as to decay transformations in a pile with constant neutron flux.

 $<sup>\</sup>ddagger$  In reference [5], a p(j) is given for j=1 which is incorrect. However, the results in that reference are unaffected since this quantity is not involved in the solutions.



FIG. 15. Comparison of relative abundances computed according to the neutron-capture theory approximation with no neutron decay or universal expansion, Alpher [5], with the observed abundance data of Brown [30], normalized with respect to 10,000 atoms of silicon. The best fit is with curve II,  $C_n t=0.81$  $\times 10^{18}$  sec./cm<sup>3</sup>. The other curves, I,  $C_n t=1.3 \times 10^{18}$  sec./cm<sup>3</sup> and III,  $C_n t=0.51 \times 10^{18}$  sec./cm<sup>3</sup> illustrate the sensitivity of the fit. The steady-state curve represents the abundances resulting from the neutron-capture process in this approximation having continued for an indefinite period.

relative abundance data. In Fig. 15 are also shown computed abundances for other values of  $C_n t$  to illustrate the sensitivity of the solutions to the cut-off value  $(C_n t)_c$ . It is possible to interpret crudely the value at the cut-off,  $(C_n t)_c = 0.81 \times 10^{18} \text{ sec./cm}^3$ , in the following manner. While  $C_n = \text{constant}$  was used in the calculation, one may describe  $(C_n t)_c$  as

$$(C_n t)_c \cong \int_{t_0}^{t_c} C_n(t) dt, \qquad (122)$$

from which it is possible, as shown by Alpher [5], to estimate an average density of matter during the process as well as a starting time for the process. Even though now superceded by better approximations to the neutron-capture theory [8], the results in this approximation indicate that the explicit inclusion of the universal expansion should involve the behavior of a radiation universe containing a relatively small amount of matter.

In the next approximation [8], the decay of the neutron has been taken into account explicitly while the universal expansion was again not considered. For this case Eqs. (108) can be written in the following form:

$$\begin{aligned} d\xi_n/d\tau &= -\xi_n - P_1\xi_n\xi_1 - P_2\xi_n\xi_2 - P_3\xi_n\xi_3 - P_4\xi_n\xi_4, \\ d\xi_1/d\tau &= \xi_n - P_1\xi_n\xi_1, \\ d\xi_2/d\tau &= P_1\xi_n\xi_1 - P_2\xi_n\xi_2, \\ d\xi_3/d\tau &= P_2\xi_n\xi_2 - P_3\xi_n\xi_3, \\ d\xi_4/d\tau &= P_3\xi_n\xi_3 - P_4\xi_n\xi_4, \end{aligned}$$
(123)

$$d\xi_j/d\tau = P_{j-1}\xi_n\xi_{j-1} - P_j\xi_n\xi_j, \quad j > 4,$$

where it has been assumed that neglecting the summation term  $\sum P_j \xi_n \xi_j$  for j > 4 in the neutron equation does not materially affect the remainder of the computation because of the almost exponential decrease in relative abundance with increasing atomic weight. The first five of Eqs. (123) were solved simultaneously and the remaining equations were grouped in a manner similar to that described earlier and solved successively. In Table XI the group sizes are indicated, and the  $P_j$  and  $\Pi(j)$  given according to

and

$$P_j = p_j C_0 / \lambda, \quad j \leq 4, \tag{124a}$$

$$\Pi(j) = [q(q+q')/2]^{-1} \Sigma' P_j, \qquad (124b)$$

where  $\sum'$  designates summation over the group of size q, and q' is the number of nuclear species in the succeeding group. It was found by trial and error that these values of the coefficients gave the best fit to the relative abundance data and this choice corresponds to a value of  $C_0/\lambda = 2.36 \times 10^{19}$  sec./cm<sup>3</sup>. As initial conditions for the integration it was assumed that the capture process started at  $\tau=0$ , with  $\xi_n=1$  and  $\xi_j=0$  for all j.

TABLE XI.ª

ī	$\Pi(\bar{j})$	$j = \overline{j}$ $1$ $2$ $3$ $4$ $[\xi(\overline{j})/\xi_1]_{\max}$	<i>P</i> <sub>1</sub> 5.01 4.62 4.72 4.88	ī	$[\xi(\bar{j})/\xi_1]_{max} \\ 1.000 \\ 0.782 \\ 0.442 \\ 0.204 \\ \Pi(\bar{j}) $	[{(j)/{{{\xi}_1}}]_{mux}}
+ 7	1 16	2 56 × 10-9		70	07.0	1 21 × 10-10
10	1.10	3.30 × 10 -		14	97.0	7.25 (10-1)
12	1.00	3.32 X 10 °		11	157	7.35 X 10 "
17	2.21	$3.40 \times 10^{-4}$		82	193	$4.38 \times 10^{-11}$
22	3.11	$3.94 \times 10^{-5}$		87	273	$2.72 \times 10^{-11}$
27	4.37	$5.39 \times 10^{-6}$		92	385	$1.76 \times 10^{-11}$
32	6.16	$8.75 \times 10^{-7}$		104.5	134	$1.63 \times 10^{-11}$
37	8 70	$1.69 \times 10^{-7}$		124.5	134	$1.20 \times 10^{-11}$
42	123	3 89 × 10-8		144.5	134	$9.17 \times 10^{-12}$
47	17.3	1.06 × 10-8		164.5	134	$600 \times 10^{-12}$
52	24.4	$3.41 \times 10^{-9}$		194.5	134	$5.28 \times 10^{-12}$
52	24.4	1.27×10-9		204 5	124	5.20 10
57	34.4	1.27 × 10 *		204.5	154	
62	48.6	$5.34 \times 10^{-10}$		224.5	134	
67	68.7	$2.53 \times 10^{-10}$		244.5	134	

\* The index  $\overline{j}$  designates the center element of the group of five or twenty species for which it is the representative element. Entries are not made for  $\overline{j} > 184.5$  because linear extrapolation was possible on a semilogarithmic plot.

In this approximation  $\tau_0 = 0$  is considered as the starting time of the process and is not the starting time of the expansion. On the latter time scale  $\tau_0$  would have a value of the order of  $10^2-10^3$  sec. The results of the integration have been plotted in Fig. 16 where the logarithms of the relative abundances are given versus  $\tau = \lambda t$ . It may be seen that the individual  $\xi(j)$  reach limiting values  $[\xi(j)]_{\max}$  when  $\tau$  has reached a value of  $\sim$ 1.2. These limiting values are given with respect to the limiting value of  $\xi_1$  in Table XI. The attainment of a limiting value may be understood since the neutrons are lost to decay and capture. ¶¶ As the atomic weight increases the rate at which the limiting value is attained becomes smaller. However, the error involved in taking  $\tau = 1.21$  as the limiting value does not exceed several percent even for the elements of highest j. Values of  $[\xi(j)]_{\max}$  taken at  $\tau = 1.21$  from Fig. 16 are, after appropriate normalization, the relative particle concentrations or relative abundances per 10,000 atoms of silicon as computed by the neutron-capture theory in this approximation. These limiting values are plotted against atomic weight, A = j, in Fig. 17 and compared with Brown's [30] abundance data. The individually computed points lie on the smooth curve shown and have not been indicated in order to avoid confusion with the observational data. The agreement between theory and observation is excellent in terms of the principal trend of the abundance data. The reproduction of any of the detailed features of the data should not be expected because the capture cross-section data were smoothed and none of the detailed stability properties of specific nuclei were included in the formulation.

The quantity  $C_0/\lambda$  is the only arbitrary parameter in this approximation of the neutron-capture theory. Thus, it is possible to determine  $C_0$ , the initial particle concentration, and hence the density of matter at the start of the process, by assigning a value for the neutron decay constant  $\lambda$ . The most recently reported measurement of the neutron half-life by Robson [124] gave a minimum of 9 and a maximum of 18 min. as compared to the theoretical value according to the Fermi theory of  $\beta$ -decay of ~15 min. [20]. Using  $\lambda \cong 10^{-3}$  sec.<sup>-1</sup>, one obtains|| ||  $C_0=2.36\times 10^{16}$  cm<sup>-3</sup>, which corresponds to an average density of matter for the process of  $\rho_m=4$  $\times 10^{-8}$  g/cm<sup>3</sup>. While it has not been necessary to specify explicitly the temperature for the process, it is reasonable, as has already been discussed, to suppose that a temperature  $kT \cong 0.1$  Mev must have prevailed. The value of  $\rho_m$  obtained in this approximation should be considered as a lower limit because, first, the introduction into the calculations of small cross sections for certain nuclear species would depress the computed abundances and require a higher value of  $C_0$ , and second, taking the universal expansion into account would also require a higher value of  $C_0$  to overcome the dilution of the ylem. It is the nature of the solutions of Eqs. (123) that too high a value of  $C_0$  leads to computed abundances for the heavy elements that are too high with respect to hydrogen, and vice versa.

Had the universe in early times consisted of matter only, then a  $\rho_m \cong 4 \times 10^{-8}$  g/cm<sup>3</sup> would have been attained, according to Eq. (98), at a time very long as compared with the neutron lifetime. With this state of affairs it would be difficult to suppose a sufficient concentration of neutrons to carry on the neutroncapture process. Thus, the starting time of the process would have to be taken as less than the neutron lifetime. Comparing  $\rho_m$  with the density of radiation corresponding to a kT of about 0.1 Mev ( $\sim 10^{9}$ °K), one sees that the early stages must have corresponded to a radiation universe,  $\rho_r \gg \rho_m$ , including a trace of matter. With this model, according to Eq. (97), a kT of 0.1 Mev  $(\rho_r \cong 10 \text{ g/cm}^3)$  would be reached at about 200 sec., and, of course, one would have essentially the initial supply of neutrons at this time. This kind of information,



FIG. 16. Logarithm of the relative abundances versus  $\tau = \lambda t$ ( $\lambda =$  neutron decay constant) in the neutron-capture theory approximation including neutron decay but not the universal expansion, according to Alpher and Herman [8]. The numbers on the righthand side denote the nuclear species and the representative elements of the groups of elements considered. The curve labeled *n* shows the neutron concentration.

<sup>¶¶</sup> The tabulated values of  $[\xi(j)/\xi_1]_{\max}$  differ slightly from those previously reported, reference [8], because of a small numerical error in the transition in group size from j=4 to j=7. The effect of correcting this error was to displace upward the computed abundances for  $j \leq 4$  by 0.08 on a logarithmic scale. This small shift has been made in Figs. 16 and 17 but is hardly evident. Inclusion of the neutrons remaining at  $\tau = 1.21$  with the protons in the normalization would also alter the limiting values by a small factor.

by a smart actor.  $\|\|$  In reference [8] the density in this approximation was stated as being  $\rho_m \cong 5 \times 10^{-9}$  g/cm<sup>3</sup>. However, in computing this  $\rho_m$ ,  $\lambda$  was taken as (1/1800) sec.<sup>-1</sup> (see reference [137]), and, in addition, there was an error of a factor of four in the  $p_i$  used in computing  $C_0$ . This accounts for the increase in  $\rho_m$  reported here by a factor of about 7.

namely, the specification of the densities of matter and radiation at a specific and early epoch in the expanding universe is precisely what is required for studying, in any detail, the properties and consequences of this cosmological model.

Some calculations have been made recently by Alpher and Herman of the neutron-capture process including the universal expansion, as described by Eqs. (108). Because of the singular nature of the term  $(\psi \xi_j / \tau)$ , it is necessary in this approximation to start the integration at a finite time  $\tau_0$ . The boundary conditions for this integration have been selected as follows. In accordance with the discussion concerning the process temperature given earlier, the time corresponding to a temperature of  $1.0 \times 10^9$ °K in a radiation universe has been selected as the starting time for the formation process. This time,  $t_0 \cong 230$  sec., from Eq. (100), leads to a  $\tau_0 \cong 0.128$ if one takes  $\lambda^{-1} \cong 1800$  sec.\* Assuming neutrons only



FIG. 17. Comparison of theoretical with observed relative abundances *versus* atomic weight. The theoretical abundances are calculated from the neutron-capture theory approximation including neutron decay but not the universal expansion, according to Alpher and Herman [8]. The observed data are those given by Brown [30]. The theoretical curve corresponds to a matter density of  $4 \times 10^{-8}$  g/cm<sup>3</sup> at the start of the elementforming process provided the neutron decay constant  $\lambda = 10^{-3}$ sec.<sup>-1</sup>.

\* These calculations were made by the authors prior to the publication of the result of Robeson, reference [124], and this value of  $\lambda$  was believed reasonable. Had one used  $\lambda^{-1} \cong 10^3$  sec., then  $\tau_0 \cong 0.23$  and the selected initial values of  $\xi_n$  and  $\xi_p$  would

at  $\tau=0$ , and also assuming free neutron decay thereafter, one has at  $\tau_0=0.128$ ,  $(\xi_n)_0=0.88$ , and  $(\xi_p)_0=0.12$ . The concentrations of all other species were taken to be zero at the starting time.

It should be re-emphasized that this choice of a specific starting time is arbitrary in the sense that capture processes did not suddenly begin but rather became progressively more important as competing processes declined in the cooling material. In this connection it is interesting to discuss the behavior of one of these competing processes, namely, photodisintegration, following a treatment given by Smart.<sup>†</sup> From Planck's formula for blackbody radiation one may write for the number of photons per unit volume,  $N_{h\nu}$ , having an energy greater than  $B_0$ .

$$N_{h\nu} = 1.3 \times 10^{31} kT B_0^2 \exp[-B_0/(kT)],$$
 (125)

if kT and  $B_0$  are given in Mev, and  $B_0/kT \gg 1$ . The probability of a  $(\gamma, n)$  reaction is

$$\lambda_{\gamma} = N_{h\nu} \sigma(\gamma, n) c \text{ sec.}^{-1}, \qquad (126)$$

where c is the velocity of light and  $\sigma(\gamma, n)$  is the cross section for the  $(\gamma, n)$  reaction. The exponential factor in Eq. (126) is so important that the variation of  $\sigma(\gamma, n)$  with energy may be replaced by an average value for an order of magnitude calculation. According to Smart, one may take

$$\sigma_{Av}(\boldsymbol{\gamma}, \boldsymbol{n}) \cong 3 \times 10^{-27} A^{\frac{2}{3}} \text{ cm}^2.$$
(127)

In Eq. (127) A is the atomic weight and the nuclear surface presented to a high energy photon is approximately proportional to  $A^{\frac{2}{3}}$ . Assuming that the temperature varies in accordance with Eq. (100), for a radiation universe, one may calculate the variation of  $\lambda_{\gamma}$ with time in the early stages of the expansion for a nucleus of given A, where  $B_0$  is the threshold energy for photoemission of a neutron. The result of such a calculation for A = 125 and  $B_0 = 6$ , 8, and 10 Mev is shown in Fig. 18, where for convenience of interpretation kT is also shown. This plot illustrates the extremely rapid decline with time of this competing photodisintegration process. It is evident that after several hundred seconds the probability for photoemission of neutrons can be neglected by comparison to, say, the capture probability for the nuclear species under discussion which is of the order of unity at  $kT \cong 0.1$  Mev. Thus a starting time of several hundred seconds for the neutron-capture process is reasonable at least in terms of this one kind of competing process.

The integration of Eqs. (108) was carried out by solving simultaneously the first five equations with J=4, i.e., neutrons used in forming nuclei with atomic weight greater than 4 were neglected in the growth

have been somewhat different. The principal result of this would be to necessitate a slightly different initial density of matter in order to achieve the desired fit to the abundance data.

**<sup>†</sup>** We are indebted to Dr. J. S. Smart for communicating these results to us prior to publication and for his kindness in permitting us to use the material in this review.



FIG. 18. Logarithm of the  $(\gamma, n)$  reaction probability,  $\lambda_{\gamma}$ , versus log *t* for the nucleus of A = 125 for various binding energies of the last neutron, according to Smart (unpublished). The graph shows the decrease in  $\lambda_{\gamma}$  as the temperature, *T*, decreases in an expanding universe controlled by radiation [Eq. (100)].

equation for neutrons. Recognizing that this case including the universal expansion would require a higher initial density of matter than in the static case, five starting densities were considered. The solution which appears to be most promising is that for which the coefficients  $P_j$  in Eqs. (108) were taken as 100 times those given in Table XI. This corresponds to an initial density  $\rho_m \cong 5 \times 10^{-7}$  or  $4 \times 10^{-6}$  g/cm<sup>3</sup> depending on whether one takes  $\lambda^{-1} = 1800$  or 1000 sec., with corresponding starting times  $t_0 \cong 230$  or 130 sec. The solutions of these five simultaneous equations for five initial densities were obtained on an IBM relay calculator in the range  $0.128 \le \tau \le 1.17$ . In Fig. 19 the results for the case  $100P_j$  discussed are plotted as  $\log \xi_j$  versus  $\log \tau$ . A comparison of these solutions with those of the static case given in Fig. 16 shows the marked effect of the expansion in that the relative concentrations quickly go through a peak and by  $\tau \cong 1.2$  all the concentrations are decreasing as  $\tau^{-\frac{3}{2}}$ , i.e., the universal expansion is by then the only term of any importance. It will be seen that the curves for j=2-4 are essentially parallel by this time and the relative concentrations cannot change thereafter. The neutron concentration, on the other hand, continues to decrease mainly according to both decay and expansion and it is clear that the sum  $\xi_n + \xi_p$ is approximately parallel to the other curves. The relative concentrations of species j=1, 2, 3, and 4calculated in this approximation, with coefficients in Eq. (108) taken as 100 times those in Table XI, are in good agreement with those obtained in the static case which led to the fit over the entire range of atomic weight shown in Fig. 17. Calculations are in progress for species of higher atomic weights and it may be expected that a satisfactory fit to the observed data might result.

To be sure, calculating the relative abundances of the very light elements in the manner described possibly represents a poor approximation to the state of affairs for these light elements. The specific neutron-capture cross sections have not been used for these very light elements although the smoothed equation for  $\sigma$  is not too far in general from the observed values. More important is the fact that there are reactions other than neutron capture among the light elements which should be taken into account. Recently, Fermi and Turkevich have considered in detail all the nuclear reactions involved in the formation of the elements through helium in an expanding universe. This work is described in detail in Section IV(d)2.

# 2. The Formation of Light Nuclei

A preliminary study of the non-equilibrium formation of light nuclei, taking into account specific reactions and the actual cross sections, was first carried out by Gamow [59, 61]. He considered the building up of deuterons by neutron-proton capture. This process is described by Eqs. (108a) and (108b), taking only the first term in the summation in the former equation. Replacing  $p_1$  by  $\sigma_1 v$ , where  $\sigma_1$  is the absolute cross section for deuteron formation [20], setting  $\rho_m = \rho_0 t^{-\frac{1}{3}}$ [see Eq. (101)], and replacing all temperature dependent quantities by their equivalent time-dependent forms, one obtains for the concentrations by weight of neutrons and protons [7, 9],

$$dx_n/d\tau = -x_n - \alpha_G x_n x_H/\tau, \qquad (128a)$$

$$dx_H/d\tau = +x_n - \alpha_G x_n x_H/\tau, \qquad (128b)$$

where,

$$\alpha_{G} = \left[ \frac{(2^{9/4} \pi^{5/4} G^{1/4} a_{r}^{1/4} e^{2\hbar})}{(3^{1/4} m_{0}^{9/2} c^{11/2} k)} \right] \\ \times \left( |\mathbf{u}_{H}| + |\mathbf{u}_{n}| \right)^{2} (\epsilon^{1/2} + \epsilon_{0}^{1/2}) \epsilon^{3/2} \rho_{0}. \quad (128c)$$

In Eq. (128c), G is the gravitational constant,  $a_r$  the radiation density constant,  $m_0$  the unit of atomic mass,



FIG. 19. Relative abundance as a function of time  $(\tau = \lambda t)$  in the neutron-capture theory approximation including neutron decay and the universal expansion, according to Alpher and Herman (unpublished). These curves are solutions of Eqs. (108), J=4, with the density of matter at the start of the elementforming process  $\rho_m = 4 \times 10^{-6}$  g/cm<sup>3</sup>. The neutron decay constant is taken as  $\lambda = 10^{-3}$  sec.<sup>-1</sup>. The ordinate is actually log  $\xi_i$ , where according to Eq. (108d) the  $\xi_i$  are concentrations normalized with respect to the concentration of nucleons at the start of the element-forming process. Hence the effect of the universal expansion is evident in the solution.

 $\mathbf{y}_n$  and  $\mathbf{y}_H$  the magnetic moments of neutron and proton in nuclear magnetons, and  $\epsilon$  and  $\epsilon_0$  are the binding energies of the singlet and virtual triplet states of the deuteron. Equations (128) have been integrated with  $x_n(\tau=0)=1$ ,  $x_H(\tau=0)=0$ , and with the condition that the final concentration by weight of protons be 0.5, since hydrogen constitutes about 50 percent by weight of all matter. To obtain this final condition [7] one must take  $\alpha_G=1$ . This integration yields for the density of matter at 1 sec.,  $4.8 \times 10^{-4}$  g/cm<sup>3</sup>, which is in moderately good agreement with the matter densities obtained in other non-equilibrium calculations.

The non-equilibrium formation of the very light elements in an expanding universe has been examined in greater detail by Fermi and Turkevich.<sup>‡</sup> All thermonuclear reactions which are less endothermic than the disintegration of the deuteron and which can go on between neutrons (N), protons (H), deuterons (D), tritons (T), He<sup>3</sup>, and He<sup>4</sup> were considered, as well as the radioactive decay of the neutron and triton. The cosmological model chosen was that of a radiation universe containing a relatively small quantity of matter, for which the dependence of temperature on time is given by Eq. (100), namely,

$$T = 1.52 \times 10^{10} t^{-\frac{1}{2}}$$
 °K.

In this model, density or particle concentration varies as  $t^{-\frac{1}{2}}$ , as shown by Eq. (101). Fermi and Turkevich have assumed that the nucleon concentration was  $10^{21}$ cm<sup>-3</sup> at 1 sec., so that the nucleon concentration at any t is

$$C_{\rm nuc} = 10^{21} t^{-\frac{3}{2}} \,{\rm cm}^{-3}.\tag{129}$$

This corresponds to an assumed matter density of  $\sim 1.7 \times 10^{-3}$  g/cm<sup>3</sup> at t=1 sec., or  $\sim 5 \times 10^{-7}$  g/cm<sup>3</sup> at t=230 sec. [Compare Section IV(d)1.]

The 28 reactions considered in detail are listed in Table XII. Examination of the reaction rates for the nuclear processes listed confirmed that until  $t \cong 300$  sec. the only event of any importance was neutron decay. The high temperature prevents the formation of an appreciable concentration of deuterons, and the nuclei past the deuteron must form through the deuteron, since at the density and temperature under consideration, many-body processes should not be important. A starting time of 300 sec. was therefore selected for the calculation. The initial relative concentrations of

TABLE XII. Reaction rates. [The quantities  $a_1$  and  $a_2$  are defined in Eq. (130),  $T_8$  is the temperature in units of 10<sup>8</sup> °K,  $T_8 = 152t^{-1/2}$  from Eq. (100), and  $q_0 = 10^{21}$  sec.<sup>3/2</sup> cm<sup>-3</sup>.]

No.	Reaction	Specific reaction rates	Term in rate equations, $\mathcal{R}'$ [See Eq. (132)]
1	$N=H+e^{-}$	$10^{-3}$ sec. <sup>-1</sup>	10 <sup>-3</sup> <b>x</b> <sub>N</sub>
2	$N+H=D+h\nu$	$6.6 \times 10^{-20} \text{ sec.}^{-1}$	$6.6 \times 10^{-20} q_0 \mathbf{x_N x_H} t^{-3/2}$
3	$N+D=T+h\nu$	$2.0 \times 10^{-22}$ sec. <sup>-1</sup>	$2.0 \times 10^{-22} q_0 \mathbf{x_N x_D} t^{-3/2}$
4	N+D=N+N+H	Negligible (see reaction 18)	0
5	$N + He^3 = He^4 + h\nu$	$10^{-21}$ sec. <sup>-1</sup> (estimated)	$10^{-21}q_0 \mathbf{x_N x_{He3}} t^{-3/2}$
6	$N+He^3=T+H$	$1.5 \times 10^{-15} \text{ sec.}^{-1}$	$1.5 \times 10^{-15} q_0 \mathbf{x}_N \mathbf{x}_{He^3} t^{-3/2}$
7	$\mathbf{H} + \mathbf{H} = \mathbf{D} + e^+$	$a_1 = 2 \times 10^{-39}; a_2 = 3.16$	$7.0 \times 10^{-41} q_0 (\mathbf{x_H})^2 t^{-7/6} 10^{-0.592 t^{1/6}}$
8	$H+D=He^3+h\nu$	$a_1 = 8.6 \times 10^{-21}; a_2 = 3.48$	$3.0 \times 10^{-22} q_0 \mathbf{x_H x_D} t^{-7/6} 10^{-0.652 t^{1/6}}$
9	H+D=H+H+N	Negligible (see reaction 18)	0
10	$H+T=He^4+h\nu$	$a_1 = 1.5 \times 10^{-19}; a_2 = 3.62$	$5.3 \times 10^{-21} q_0 \mathbf{x_H x_T} t^{-7/6} 10^{-0.678 t^{1/6}}$
11	$H+T=He^{3}+N$	$1.5 \times 10^{-15} \times 10^{-36.8/T_8}$ sec. <sup>-1</sup>	$1.5 \times 10^{-15} q_0 \mathbf{x_H} \mathbf{x_T} t^{-3/2} 10^{-0.242 t^{1/2}}$
12	$D+D=He^4+h\nu$	$a_1 = 3.07 \times 10^{-19}; a_2 = 3.99$	$1.08 \times 10^{-20} q_0(\mathbf{x}_D)^2 t^{-7/6} 10^{-0.747 t^{1/6}}$
13	$D+D=He^3+N$	$a_1 = 3.0 \times 10^{-15}; a_2 = 3.99$	$1.1 \times 10^{-16} q_0(\mathbf{x}_D)^2 t^{-7/6} 10^{-0.747 t^{1/6}}$
14	D+D=H+T	$a_1 = 3.0 \times 10^{-15}; a_2 = 3.99$	$1.1 \times 10^{-16} q_0(\mathbf{x}_D)^2 t^{-7/6} 10^{-0.747 t^{1/6}}$
15	$D+T=He^4+N$	$a_1 = 5.0 \times 10^{-13}; a_2 = 4.24$	$1.8 \times 10^{-14} q_0 \mathbf{x}_D \mathbf{x}_T t^{-7/6} 10^{-0.794 t^{1/6}}$
16	$D+He^3=He^4+H$	$a_1 = 1.5 \times 10^{-12}; a_2 = 6.72$	$5.3 \times 10^{-14} q_0 \mathbf{x_D x_{He3}} t^{-7/6} 10^{-1.259 t^{1/6}}$
17	$D+He^4=Li^6+h\nu$	$a_1 = 1.4 \times 10^{-21}; a_2 = 6.96$	$4.9 \times 10^{-23} q_0 \mathbf{x_D x_{He4}} t^{-7/6} 10^{-1.304 t^{1/6}}$
18ª	$D+h\nu=H+N$	$5.9 \times 10^{12} T_8^{3/2} 10^{-110/T_8} \text{ sec.}^{-1}$	$1.1 \times 10^{+16} \mathbf{x}_{D} t^{-3/4} 10^{-0.723 t^{1/2}}$
19	$T = He^3 + e^-$	$1.8 \times 10^{-9} \text{ sec.}^{-1}$	$1.8 \times 10^{-9} x_{T}$
20	$T+T=He^{4}+N+N$	$a_1 = 2.6 \times 10^{-13}; a_2 = 4.57$	$9.1 \times 10^{-15} q_0 (\mathbf{x_T})^2 t^{-7/6} 10^{-0.856 t^{1/6}}$
21	$T+T=He^{6}+h\nu$	$a_1 = 2.6 \times 10^{-19}; a_2 = 4.57$	$9.1 \times 10^{-21} q_0(\mathbf{x_T})^2 t^{-7/6} 10^{-0.856 t^{1/6}}$
22	$T+He^3=He^4+N+H$	$a_1 = 1.5 \times 10^{-12}; a_2 = 7.24$	$5.3 \times 10^{-14} q_0 \mathbf{x_T x_{He3}} t^{-7/6} 10^{-1.356 t^{1/6}}$
23	$T+He^3=He^4+D$	$a_1 = 1.0 \times 10^{-13}; a_2 = 7.24$	$3.5 \times 10^{-15} q_0 \mathbf{x_T x_{He}} t^{-7/6} 10^{-1.356 t^{1/6}}$
24	$T+He^3=Li^6+h\nu$	$a_1 = 3.1 \times 10^{-18}; a_2 = 7.24$	$1.1 \times 10^{-19} q_0 \mathbf{x_T x_{He3}} t^{-7/6} 10^{-1.356 t^{1/6}}$
25	$T + He^4 = Li^7 + h\nu$	$a_1 = 5.5 \times 10^{-19}; a_2 = 7.56$	$1.9 \times 10^{-20} q_0 \mathbf{x_T x_{He}} t^{-7/6} 10^{-1.416 t^{1/6}}$
26	$\mathrm{He^3} + \mathrm{He^3} = \mathrm{Be^6} + h\nu$	$a_1 = 1.4 \times 10^{-17}; a_0 = 11.49$	$4.9 \times 10^{-19} q_0 (\mathbf{x}_{\mathbf{He}3})^2 t^{-7/6} 10^{-2.151 t^{1/6}}$
27	$He^3+He^3=He^4+H+H$	$a_1 = 1.4 \times 10^{-11}; a_2 = 11.49$	$4.9 \times 10^{-13} q_0 (\mathbf{x_{He^3}})^2 t^{-7/6} 10^{-2.151 t^{1/6}}$
28	$\mathrm{He}^{3} + \mathrm{He}^{4} = \mathrm{Be}^{7} + h\nu$	$a_1 = 1.7 \times 10^{-19}; a_2 = 12.01$	$6.0 \times 10^{-21} q_0 \mathbf{x_{He^3} x_{He^4}} t^{-7/6} 10^{-2.250 \iota^{1/6}}$

\* The photon concentration is included in the constant.

‡ We are indebted to Drs. E. Fermi and A. Turkevich for their cooperation and communication of unpublished results. The authors take the responsibility for the correctness of this transcription and interpretation of their work.

neutrons and protons selected, namely, 0.70 and 0.30, respectively, correspond approximately to those resulting from free neutron decay starting at t=0 sec. One assumes neutrons only to begin with, and a neutron decay constant  $\lambda = 10^{-3}$  sec.<sup>-1</sup>.

The specific rates taken for each of the reactions are also listed in Table XII, and were obtained as follows. The neutron and triton decay constants were taken as  $10^{-3}$  sec.<sup>-1</sup> and  $1.8 \times 10^{-9}$  sec.<sup>-1</sup>, respectively, in accordance with experiment. Of the three neutroncapture reactions, (2), (3), and (5), specific reaction rate constants consistent with experiment were assigned to reactions (2) and (3), while a constant was estimated for reaction (5). In all three cases the rate constants are independent of temperature and therefore of time. Many of the reactions in Table XII are of the form

$$X+X' \rightarrow Y+Y'+$$
energy,

where X and X' are heavy charged particles, while Y and Y' are either heavy charged particles or a gammaray plus a heavy charged particle. The specific rate constant for such thermonuclear reactions may be obtained from the usual expression for thermonuclear reaction rates [61], as¶

$$\mathcal{K} = a_1 T_8^{-\frac{2}{3}} 10^{-a_2 T_8^{-\frac{1}{3}}} \,\mathrm{cm}^3/\mathrm{sec.},\tag{130}$$

where  $T_8$  is the temperature in units of 10<sup>8</sup> °K,  $a_1$ , which depends mainly on the reaction probability after penetration, is given by

$$a_{1} = \left[ (4\hbar\Gamma r_{0}^{2}a_{2}^{2}\ln^{2}10)/(3^{5/2}m_{r}e^{2}Z_{1}Z_{2}) \right] \\ \times \exp(32m_{r}e^{2}r_{0}Z_{1}Z_{2}/\hbar^{2})^{1/2}, \quad (130a)$$

while  $a_2$ , which depends on the height of the potential barrier for the reaction may be written

$$a_2 = 3 \times 10^{-8/3} (\log e) [(\pi^2 m_r e^4 Z_1^2 Z_2^2) / (2\hbar^2 k)]^{1/3}.$$
 (130b)

In the above,  $A_1$ ,  $A_2$ ,  $Z_1$ ,  $Z_2$ , are the atomic weights and numbers of the reacting nuclei,  $m_r$ , the reduced mass, is given by  $m_r = m_1 m_2 (m_1 + m_2)^{-1}$  g, the combined radius  $r_0 \cong 1.6 \times 10^{13} (A_1 + A_2)^{\frac{1}{3}}$  cm, and  $\Gamma/\hbar$  is the probability per second for the reaction after penetration of the barrier. Fermi and Turkevich have used for  $\Gamma$  the values given by Bethe [19, 61] or values obtained by procedures consistent with those used by Bethe. As is well known, in the absence of resonances,

TABLE XIII. Relative abundances of the light nuclei.

	Computed	Observed
$H^1$	1.00	1.00
$H^2$	$1.3 \times 10^{-2}$	2×10-4
He <sup>3</sup>	2.6×10-4	10-7
He <sup>4</sup>	$1.5 \times 10^{-1}$	10-1

¶ In reference [61], p. 266, the quantity N there is incorrectly given insofar as stated dimensions are concerned. It is  $(g \text{ sec.})^{-1}$  rather than  $(\text{cm}^3 \text{ sec.})^{-1}$  as stated.



FIG. 20. Relative abundance of the very light elements as a function of time according to the non-equilibrium formulation of Fermi and Turkevich (unpublished). The nucleon concentration was taken to be  $10^{21}$  cm<sup>-3</sup> at t=1 sec. in an expanding universe controlled by radiation. The relative abundances are the ratios of the number of nuclei of a given species in a volume V to the total number of nucleons in that volume. Since both quantities vary with the universal expansion in the same way the effect of the expansion is not evident.

 $\Gamma$  is about 10<sup>6</sup> times smaller for radiative-capture reactions than for reactions with particle emission only.

The specific reaction rates can be written in terms of the time as variable instead of the temperature since, from Eq. (100),  $T_8 = 152t^{-\frac{1}{2}}$ . In the last column in Table XII terms are given corresponding to the specific reactions as they would appear in rate equations involving concentrations of nuclei by weight,  $x_j$ . From Eqs. (110) it is clear that these terms have the form

$$\Re = \mathcal{K}m_j(m_j'm_j'')^{-1}\rho_m x_j' x_j'' \text{ sec.}^{-1}, \qquad (131)$$

or, using Eq. (101),

$$\mathfrak{R} = \mathcal{K}m_{j}(m_{j}'m_{j}'')^{-1}q_{0}m_{0}t^{-\frac{3}{2}}x_{j}'x_{j}'' \operatorname{sec.}^{-1}, \quad (131a)$$

where  $m_0$  is the mass of a nucleon,  $q_0 = 10^{21}$  sec.<sup>3</sup> cm<sup>-3</sup>, and the  $\mathcal{K}$  correspond to the  $p_j$  previously discussed. This form denotes the contribution to  $dx_j/dt$  arising from the reaction between species j' and j'' leading to species j. For example, in the case of reaction (10) in Table XII, namely,  $H+T=\mathrm{He}^4+h\nu$ ,

$$\mathcal{K}_{10} = 1.5 \times 10^{-19} T_8^{-\frac{3}{2}} 10^{-3.62T_8^{-\frac{1}{2}}},$$

and the term denoted by Eq. (131a) is as given in the third column of Table XII for this reaction. Fermi and Turkevich have found that many of the reactions listed in Table XII can be neglected to a sufficient approximation. The reactions retained are evident from the following set of equations whose simultaneous solutions were obtained by Fermi and Turkevich through numerical integration. Denoting the term in Table XII corresponding to a given reaction by  $\mathfrak{R}'$ , one has

$$d\mathbf{x}_{\rm N}/dt = -\mathfrak{R}_{1}' - \mathfrak{R}_{2}' + \mathfrak{R}_{15}', \qquad (132a)$$

$$d\mathbf{x}_{\rm H}/dt = + \mathcal{R}_1' + \mathcal{R}_{14}' + \mathcal{R}_{18}' - \mathcal{R}_2', \qquad (132b)$$

$$d\mathbf{x}_{\rm D}/dt = + \Re_2' - \Re_{12}' - \Re_{13}' - \Re_{14}' - \Re_{15}',$$
 (132c)

$$d\mathbf{x}_{\rm T}/dt = + \Re_{14}' - \Re_{15}', \qquad (132d)$$

and

$$d\mathbf{x}_{\mathrm{He}^{4}}/dt = + \Re_{15}',$$
 (132e)

where  $\mathbf{x}_j = x_j/j$  and j is the atomic weight. The quantities  $\mathfrak{R}'$  are related to the  $\mathfrak{R}$  in Eq. (131a) by

$$\mathfrak{R}' = \left[ j/(j'j'') \right] \mathfrak{R}, \qquad (132f)$$

in which j is the atomic weight of the particular nuclear species whose rate of change is being considered and where j' and j'' are the atomic weights of the two reacting species, respectively. An equation for He<sup>3</sup> is not included for the reason that to a sufficient approximation the concentration  $\mathbf{x}_{\text{He}^3}$  is maintained at a steady state by the fast reactions (6) and (13), the other reactions involving this species being unimportant. From  $d\mathbf{x}_{\text{He}^3}/dt=0$ , Fermi and Turkevich have found that to within a factor of  $\sim 3$ ,

# $x_{He^3}x_D^{-2} \cong 2 \times 10^{-2}$

in which it is assumed that  $\mathbf{x}_n$  is constant. The factor of 3 arises from the rather small time dependence of the rates in the steady state equations. While tritium balance is maintained almost as well yielding,  $x_T x_D^{-1}$  $\cong 10^{-2}$ , the tritium reactions were considered in detail. The results of the integration of Eqs. (132), subject to the initial conditions already discussed, are given in Fig. 20 where the relative concentrations particle-wise are plotted versus the time for the various nuclear species. As can be seen in Table XIII, the computed relative abundances (at t = 2000 sec.) may be considered as in agreement with those observed for these species. in view of the fact that very light element abundances are not well known and would vary in different locales because of the participation of these species in thermonuclear reactions after the element-forming epoch. The computed relative abundances of H<sup>3</sup> and He<sup>3</sup> have been added together because of the radioactivity of the former. The observed relative abundances are obtained from Brown (Table III of reference [30]) and corrected for isotopic abundance ratios [129].

Fermi and Turkevich have also examined the problem of forming the light elements heavier than He<sup>4</sup>. The only reactions involved are capture reactions since there are no exothermic reactions giving heavy particles. Reactions (17), (21), and (25) are of this kind, with (17) and (21) giving Li<sup>6</sup>, the latter by  $\beta$ decay of He<sup>6</sup>, while (25) yields Li<sup>7</sup>. Under the conditions discussed, and assuming no resonances, these reactions are very slow and lead to an insufficient amount of material, about 10<sup>-7</sup> by weight, past He<sup>4</sup>. The existence of a resonance in reaction (25) might repair this difficulty. In this case Fermi and Turkevich have considered how close a resonance would have to be in order to get an appreciable conversion of He<sup>4</sup> and H<sup>3</sup> to Li7. A detailed examination of this reaction under the conditions discussed indicates that a resonance would have to be at about 400 kev or closer in order to convert any appreciable amount of the material into Li<sup>7</sup>. At the present time the first observed level is at about 4 Mev. Turkevich has also considered this reaction (25) under a different set of initial conditions. namely, a nucleon concentration of 10<sup>23</sup> instead of 10<sup>21</sup> cm<sup>-3</sup>, and a neutron-proton ratio of 6 to 1, both at t=1 sec. Non-resonance processes with these initial conditions lead to  $3 \times 10^{-4}$  by weight of Li<sup>7</sup>, which is much closer to what is required and it is concluded that even a resonance closer than 1 Mev would be interesting. To the best of the authors' knowledge this reaction has not been studied directly. Another possibility first proposed by Wigner || for building appreciable amounts of elements past He<sup>4</sup> involves the idea of a "seed" nucleus. An example of an exothermic chain reaction involving a seed nucleus, studied by Turkevich, is

# $_{6}C^{10}+_{1}H^{3}\rightarrow_{3}Li^{6}+_{4}Be^{7}+_{2}Mev.$

If the two product nuclei would again build up to C<sup>10</sup>, then one has a method of forming appreciable amounts of nuclei past the gap. However, since C<sup>10</sup> is neutron deficient it is difficult to see, in this particular case, how it could be re-formed from the product nuclei. As pointed out by Gamow [60] there may exist other possible reactions of this type in which the "seed" nucleus would have a neutron rather than a proton excess. The difficulty of finding a scheme to bridge the non-existent nuclei at A = 5 and 8 is discussed again later. In any event, neutron capture should be the predominant reaction at the temperatures considered and should certainly be responsible for building the elements past the first eight or ten.

Another question of interest in connection with the non-equilibrium formation of elements is the possible participation of some of the light elements in thermonuclear reactions in the expanding universe after the completion of the initial element forming process. As has been seen in previous discussion, the temperature after the element forming process was still quite high so that thermonuclear reactions between light nuclei and protons could go on. This problem has been studied

<sup>||</sup> See reference [60],

by Alpher, Herman, and Gamow [10] in the following manner. An examination of the relative abundance data suggests that the abundances of certain of the light elements such as Li, Be, and B have been markedly decreased since the "original" formation process. It is to be noted that these elements have relatively large cross sections for proton reactions [19].

It can be shown that for certain of the light elements the difference between the present relative abundance and the abundance computed according to the neutroncapture theory [10] is consistent with known thermonuclear reaction rates and with cosmological information furnished by the theory. Making use of Eq. (130) one may write for the number of thermonuclear reactions per gram of matter per second,

$$r_{\rm th} = \mathcal{K} x_j x_p (m_j m_p)^{-1} \rho_m(t), \qquad (133)$$

where  $\rho_m$  is the density of matter,  $x_j$  and  $x_p$  the concentrations by weight of the reacting species j and protons, of mass  $m_j$  and  $m_p$ , respectively. In a manner analogous to that used in obtaining Eq. (110c) one finds

$$d[\ln x_j]/dt = -r_{\rm th}m_j/x_j. \tag{134}$$

In this equation  $\rho_m$  and T are taken to be defined as functions of time according to Eqs. (101) and (100). Assuming the concentration by weight of protons to be a constant,  $x_p \cong 0.5$ , one obtains for the ratio,  $\alpha_R$ , of the observed relative abundance to that computed according to the neutron-capture theory,

 $\ln \alpha_R = B_1 [I(t_0) - I(t_P)],$ 

in which

and

$$B_1 = (152)^{-\frac{3}{2}} a_1(m_j m_p)^{-1} \rho_0, \qquad (135a)$$

(135)

$$I(t) = t^{-1/6} \exp(-B_2 t^{1/6}) + B_2 E i (-B_2 t^{1/6}) \quad (135b)$$

$$B_2 = (152)^{-\frac{1}{3}} (\ln 10) a_2. \tag{135c}$$

The quantity  $\rho_0$  is the matter density at t=1 sec.  $(\rho_m = \rho_0 t^{-\frac{3}{2}})$  and  $a_1$  and  $a_2$  are as given in Eqs. (130). One can find  $\log \alpha_R$  as the difference between the logarithms of the observed and the computed relative abundances directly from the datum points and curve in Fig. 17. The time  $t_0$  represents the time at which the proton reactions became important while  $t_P$  refers to the present epoch. The reaction probabilities  $\Gamma$  in  $a_1$ are tabulated by Bethe [19], and Gamow and Critchfield [61] for the reactions of interest.

Applying Eqs. (135) to the reactions of Li, Be, and B with protons, one finds that if  $t_0$  is of the order of 10<sup>3</sup> sec. then the present relative scarcity of these elements can be explained. This value of  $t_0$  is consistent with the time estimated for the cessation of neutroncapture processes. The same analysis applied to other light elements such as F<sup>19</sup>, which are now scarce, yields similar results since these elements have high cross sections for proton reactions. On the other hand, those light elements which have relatively low cross sections for proton reactions are now found to lie approximately on the computed abundance curve. For example, in the cases of C and N there is no appreciable depletion. In fact, taking  $t_0 \cong 10^3$  sec. for N<sup>14</sup> leads to depletion due to thermonuclear reactions of only one part per million up to the present epoch. In the foregoing treatment it has been assumed that the time dependences of  $\rho_m$  and T given by Eqs. (101) and (100) are a sufficient approximation for the problem since the main part of the depletion occurs during the period when these expressions are valid.

# 3. Effects of Nuclear Stability

In the theory of the neutron-capture process presented thus far, it has been assumed that the time between successive neutron captures was sufficiently long to allow any necessary adjustment of charge by  $\beta$ -decay. Clearly the validity of this assumption depends upon the density of the reacting material and upon the  $\beta$ -decay rates of the nuclei participating in the process. Smart [134, 136] has recently examined this problem in detail. One may consider the effect of  $\beta$ -decay on the neutron-capture process in three situations differentiated according to the density of matter. In the case of very low density, the time between successive neutron captures will in general be long enough to allow  $\beta$ -decay between captures for all species, and the capture reaction rates will control the formation processes. The nuclei involved will be stable or have one excess neutron. For a very high density, on the other hand, the reacting, nuclei would probably have as high a neutron excess as is consistent with neutron binding, and the rate of growth of particular species would be principally determined by their  $\beta$ -decay rates in these neutron-rich states. Finally, at intermediate densities one would expect a competition between neutron capture and  $\beta$ -decay processes.

The case of low density can be dismissed readily. Clearly it is meaningless to speak of a non-equilibrium process of successive neutron captures if the mean time between successive captures is appreciable as compared with the neutron lifetime. It can be shown that the upper limit to this low density case is of the order of  $10^{-11}$  g/cm<sup>3</sup>. If one equates the neutron lifetime, ~1000 sec., to the mean time between captures, then one has  $1000 = m_0(\rho_l \sigma v)^{-1}$ , where  $m_0$  is the nucleon mass,  $\rho_l$  the limiting density,  $\sigma$  the neutron-capture cross section for a particular species, and v the relative velocity, ~4×10<sup>8</sup> cm/sec. for 0.1 Mev. Taking  $\sigma \cong 10^{-24}$  cm<sup>2</sup>

TABLE XIV. Decay constants for neutron-saturated nuclei.

A	$(T_{\zeta})_m$	$W_{\beta}(\operatorname{in} m_{e}c^{2})$	$\gamma(Z, W_{\beta})$	$\lambda_{\beta}$ (sec. <sup>-1</sup> )
50	10.8	17.3	1.37	6.9
64	13.3	16.5	1.55	7.4
100	19.9	16.3	1.93	13.5
125	24.5	14.7	2.32	11.8
180	34.5	11.6	3.48	7.6

TABLE XV. Capture and  $\beta$ -decay probabilities for A < 40.<sup>a</sup>

Nucleus	2 <i>T</i> ζ	$W_{\beta}(Mev)$	α,	E <sub>E</sub> (Mev)	λ <sub>β</sub>	λα
N <sup>16</sup>	2	7.4	1.1	7.2	444	
O18			1.1	5.1		38.0
O17	-		1.2	8.9		670
O18			1.3	5.2		42
O19			1.4	8.6		657
O <sup>20</sup>	4	3.2	1.5	4.5	15.7	27.7
O <sup>21</sup>	5	5.7	1.6	7.1	209	289
O <sup>22</sup>	6	5.3	1.6	3.5	251	
$F^{22}$	4	5.8	1.75	7.9	262	537
$F^{23}$	5	4.6	1.95	5.0	230	60.9
Ne <sup>23</sup>	3	4.6	1.95	9.4		1430
Ne <sup>24</sup>	4	2.0	2.1	5.4		97
Ne <sup>25</sup>	5	3.8	2.25	7.3		466
Ne <sup>26</sup>	6	3.4	2.4	5.0	27	75
Ne <sup>27</sup>	7	5.6	2.6	6.9	386	405
Ne <sup>28</sup>	8	3.7	2.85	4.0	55	29
Na <sup>28</sup>	6	4.7	2.85	7.5		675
Na <sup>29</sup>	7	5.1	3.1	4.6	242	
Mg <sup>29</sup>	5	3.3	3.1	8.5		1376
Mg <sup>30</sup>	6	2.6	3.4	5.7		205
$Mg^{31}$	7	4.0	3.7	7.6		938
$Mg^{32}$	8	4.0	4.0	3.3	82	
Al <sup>32</sup>	6	4.5	4.0	8.3		1576
Al <sup>33</sup>	7	5.0	4.3	4.3	218	63
Si <sup>33</sup>	5	2.3	4.3	9.0		2540
Si <sup>34</sup>	6	2.0	4.6	5.1		160
Si <sup>35</sup>	7	4.2	5.1	6.7		688
Si <sup>36</sup>	8	4.4	5.5	4.5	132	101
$P^{37}$	7	3.9	6.0	5.2		228
$\mathbf{P^{38}}$	8	5.3	6.5	6.8		942

• The  $\lambda$ 's are given in arbitrary units but fixed relative to each other to give a best fit to the relative abundance data. Where  $\lambda_{\beta}$  has not been given, the values are negligible compared with  $\lambda_{C}$  and vice versa.

(heavy nuclei at 0.1 Mev), one obtains  $\rho_i \cong 10^{-11} - 10^{-12}$  g/cm<sup>3</sup>.

To examine the formation process at very high densities, it is necessary to determine the  $\beta$ -decay rates of nuclei having a limiting neutron excess, i.e., a nuclear composition such that the next neutron will not be bound. Smart, using a nuclear model based on the Wigner theory of the symmetric Hamiltonian [61, 178], has computed the maximum isotopic spin  $(T_f)_m$ = (A-2Z)/2, i.e., the isotopic spin for a nucleus of maximum neutron content, as a function of atomic weight. In order to accomplish this he has solved the equation  $(\partial E_B/\partial A)_Z = 0$ , where  $E_B$ , the binding energy of the nucleus, is obtained by means of the Wigner theory. The results are quite accurately approximated by

$$(T_{\zeta})_m = 0.183A + 1.6,$$
 (136)

which is approximately equivalent to the statement that nuclei containing about 70 percent neutrons are at the limit of stability against neutron emission. This result has also been obtained by Mayer and Teller [113, 114], who have computed the maximum neutron content by using the semi-empirical binding energy formula due to Bohr and Wheeler, as modified by van Albada [2] [see Section III(b)4]. The solution of  $(\partial E_B/\partial A)_Z=0$ , where  $E_B$  is determined from the packing fraction, f, given by Eq. (55), yields a relationship between  $(T_5)_m$  and A which is nearly the same as that obtained by Smart in Eq. (136). This result might have been expected since both the Wigner and the Bohr-Wheeler formulas use empirically determined constants and for heavy nuclei contain essentially the same terms.

It is next required to calculate the  $\beta$ -decay rate for nuclei at the stability limit, i.e., for  $T_{\xi} = (T_{\xi})_m$ . If the energy available for  $\beta$ -decay,  $W_{\beta}$  (neglecting the difference in kinetic energy between initial and final nuclei) is large compared with  $m_ec^2$ , then the  $\beta$ -decay constant may be written [61, 101]

$$\lambda_{\beta} = (30u_0)^{-1} |\mathbf{M}|^2 \gamma(Z, W_{\beta}) W_{\beta^5}, \qquad (137)$$

where  $W_B$  is explicitly given as the sum of the change in potential energy due to the mutual repulsion of protons, the change in the potential energy due to nuclear forces, the neutron-proton mass difference and the electron rest mass. In Eq. (137),  $W_{\beta}$  is in units of  $m_e c^2$ ,  $\gamma(Z, W_{\beta})$  is a function depending on the effect of the Coulomb field on the electron emission which has been tabulated by Konopinski [101],  $u_0$  is a constant whose value is approximately 8800 sec., and  $|\mathbf{M}|$  is the matrix element for the  $\beta$ -transition. Equation (137) must be summed over all excited levels of the final nucleus to obtain the total decay constant. Smart has assumed that there is always one final state for which the matrix element  $|\mathbf{M}|$  is by far the largest, and for this state he has taken the spin-isotopic spin part of the matrix element to be  $2(T_{\zeta})_m$ , while for the orbital part he has taken 1/25. The final expression for the decay probability of the  $(T_{\zeta})_m$  nuclei is obtained as

$$\lambda_{\beta} = (375 \dot{u}_0)^{-1} (T_{\zeta})_m \gamma(Z, W_{\beta}) W_{\beta}^5, \quad W_{\beta} \gg m_e c^2.$$
(138)

In Table XIV are given decay constants calculated from Eq. (138) for neutron-saturated nuclei [136]. Smart's calculation of  $W_{\beta}$  involves the nuclear model based on Wigner's theory of the symmetric Hamiltonian. Table XIV indicates the interesting result that the mean  $\beta$ -lifetime for neutron-saturated nuclei is about 0.1 sec. and is practically independent of atomic weight, at least for A > 50. The results in Table XIV have not been carried below A = 50 for these neutronsaturated nuclei because of uncertainties in the evaluation of  $E_B$  from the Wigner theory.

In the high density case, the relative abundances would be controlled by the  $\beta$ -decay rates of the nuclei involved, and one would expect that, after a sufficiently long time (clearly this would have to be less than the  $\beta$ -lifetime of the neutron), the relative abundances of various nuclei would be inversely proportional to their  $\beta$ -decay constants at the neutron stability limit. However, this gives essentially constant abundance at least for A > 50, in contradiction to the universal relative abundance data. If, on the other hand, the process terminated before "radioactive saturation" was attained, the elements with A > 50 would show a rapid decrease in abundance with increasing A, again in contradiction to the observed abundance data. Hence, one must conclude that the element forming process went on at a density such that  $\beta$ -decay rates did not in themselves control the process.

It is possible to estimate the density above which  $\beta$ -decay rates would be the controlling factor. For this estimate it is required to know the neutron-capture cross sections of neutron-rich nuclei. The capture cross-section data discussed in Section IV(c) are for stable nuclei, and will, of course, be smaller for nuclei in which the binding energy of the captured neutron is reduced. Smart has estimated the reduction in the capture cross section in the following way. One may write the cross section for radiative capture of a neutron as

$$\sigma(n,\gamma) = \sigma_n * \overline{\Gamma}_r / (\overline{\Gamma}_r + \overline{\Gamma}_n), \qquad (139)$$

if the two competing processes are capture and scattering. The cross section for formation of the compound nucleus is  $\sigma_n^*$ , while  $\overline{\Gamma}_r$  is the average radiation width and  $\overline{\Gamma}_n$  the average neutron width of the excited state. The quantities  $\sigma_n^*$  and  $\overline{\Gamma}_n$  are averaged over different angular momentum states. At 0.1 Mev inelastic scattering can be neglected so that the formation of the compound nucleus and neutron emission are inverse processes. According to Weisskopf [175],  $\sigma_n^* \propto \overline{\Gamma}_n$  under such conditions, and, if  $\overline{\Gamma}_n \gg \overline{\Gamma}_r$ , then

$$\sigma(n,\gamma) \propto \overline{\Gamma}_r. \tag{140}$$

Since, as indicated by Weisskopf, the radiation width varies as  $E_{E^5}$  for transitions to the ground state, and  $E_E^6$  if transitions to excited states are appreciable,  $\sigma(n,\gamma)$  has about the same energy dependence as  $\lambda_{\beta}$ . The quantity  $E_E$  is, of course, the excitation energy, i.e., the neutron kinetic energy plus the energy of binding of the capture neutron. Smart has compared capture cross-section values such as are discussed in Section IV(c) with values based on the  $E_{E^5}$  dependence for neutron-rich nuclei, using the Wigner theory of the symmetric Hamiltonian for  $E_E$ . He has found that for heavy nuclei capture cross sections should be reduced by about a factor of 1000, while for light nuclei a factor of 10 to 100 is indicated. These factors may be understood as follows. For heavy nuclei, the excitation energy in the stable case is of the order of 8 Mev, while in the neutron-rich case the excitation is of the order of 2 Mev, i.e.,  $\sim 1$  Mev neutron kinetic energy and  $\sim 1$  Mev binding energy. The factor  $1000 \cong (8/2)^5$ . Similar considerations yield a factor of  $\sim 10$  to  $\sim 100$  for the light elements.

A similar result has been indicated by Wigner.\*\* As already mentioned, the capture cross sections of neutron-rich isotopes should be less than those of stable isotopes of the same element because of the decreased excitation energy in the neutron-rich case of the compound nucleus formed by the addition of a neutron. This decreased excitation energy causes an increased level spacing D. The average absorption cross section can be written as follows  $\lceil 179 \rceil$ 

$$\sigma_{kv} = 1800 E_k^{-\frac{1}{2}} (f_W^{-1} + 4.4 \times 10^{-4} D E_k^{\frac{1}{2}} / \Gamma_r)^{-1} \times (1 + A^{\frac{1}{2}} E_k^{\frac{1}{2}} / 3100)^2 \text{ barns,} \quad (141)$$

in which  $f_W \cong 1$ , A is the atomic weight,  $E_k$  is the kinetic energy of the incident neutron in ev, and  $\Gamma_r$  the radiation width in ev. One is interested in nuclei whose lifetime for  $\beta$ -decay is of the order of 0.1 sec. For such nuclei the  $\beta$ -decay energy is roughly 10–15 Mev. In a region in which the  $\beta$ -decay energy is  $W_{\beta}$ , the nuclear excitation energy is decreased by about  $W_{\beta}/2$ . Making use of the Weisskopf expression for the level spacing D, namely [20],

$$D = 10^{6} \exp(-2E_{E^{\frac{1}{2}}}) \exp(\operatorname{light nuclei}), \quad (142a)$$

and

$$D = 10^5 \exp(-4E_E^{\frac{1}{2}}) ev(heavy nuclei),$$
 (142b)

where the excitation energy  $E_E$  is in Mev, one can make an estimate of the change in D between the stable nucleus and its isobar having a  $\beta$ -lifetime of about 0.1 sec. For light and heavy nuclei, taking 8 Mev for the excitation energy in the stable case, the factors of increase in level spacing are roughly 25 and 600, respectively. If  $4.4 \times 10^{-4} DE_k {}^{\frac{1}{2}} \Gamma_r^{-1}$  is large as compared to  $f_W$ , and if  $\Gamma_r$  does not change with the neutron enrichment, then  $\sigma_{Av} \propto D^{-1}$ , and the decreases in  $\sigma_{Av}$  due to neutron enrichment are also by factors of 25 and 600 for the light and heavy nuclei, respectively. It must be emphasized that these considerations are quite approximate but nevertheless are interesting when compared with Smart's results, particularly in view of the different assumptions concerning the variation with excitation energy of the level spacing and the radiation width.



FIG. 21. Relative abundances of the light elements versus atomic weight according to the steady-state formation chain of Smart [134]. The observed data are those of Brown [30], normalized to 10,000 atoms of silicon. Smart's calculations are fitted at Si<sup>28</sup>.

<sup>\*\*</sup> We are indebted to Professor E. P. Wigner for these considerations.

In view of the foregoing one may compute the lower limit to the high density case. Taking the  $\beta$ -decay lifetime for neutron-saturated nuclei as 0.1 sec. and using one-hundredth the observed cross sections for light nuclei at 0.1 Mev, one finds the lower density limit to be of the order of  $10^{-2}$  g/cm<sup>3</sup>.

In the case of intermediate densities, i.e.,  $10^{-2} - 10^{-11}$ g/cm<sup>3</sup>, there will be some degree of competition between neutron capture and  $\beta$ -decay in the control of the formation process. It would appear to be necessary to consider in detail the capture and  $\beta$ -decay probabilities for individual nuclear species, and consider various densities to find under which condition these competing processes would lead to the observed relative abundances. Each of these probabilities depends upon the energy differences between neighboring nuclei, so that a detailed knowledge of the nuclear energy surface is required. Smart has considered the building up of successively heavier nuclei as represented by a path traced along the floor and slopes of the Heisenberg energy valley, in which the allowed moves are  $\Delta A = 1$ ,  $\Delta N = 1$  for neutron capture, and  $\Delta A = 0$ ,  $\Delta Z = 1$  for  $\beta$ -decay. A reasonable picture involves nuclei which capture neutrons successively, becoming less and less stable to  $\beta$ -decay, and which reach a point where  $\lambda_{\beta} > \lambda_{C}$ , so that they then decay and move down toward the floor of the Heisenberg valley. As they move toward the valley, the binding energy of the last neutron increases,  $\lambda_{C}$  becomes larger than  $\lambda_{\beta}$  and the nuclei again move up the slope of the Heisenberg valley. The detailed examination by Smart [134] of such formation processes indicates that in general the formation path is such that  $\lambda$ (capture) $\cong \lambda(\beta$ -decay).

As seen earlier, the capture cross section varies approximately as  $E_E^5$ , where  $E_E$  is the excitation energy, and Smart writes

$$\lambda_C \propto f_S \alpha_S E_E^5. \tag{143}$$

The quantity  $\alpha_s$  describes the variation of capture cross section with atomic weight, and is taken to be, within a constant factor, the smoothed capture crosssection function of A given by Eq. (102), while the factor  $f_s$  depends on the matter density. The  $\beta$ -decay probabilities and energies can be determined as already described. Smart [134] has made a chart of isotopes, with  $W_{\beta}$  and  $E_E$  computed for each species, and has traced a formation path for the light elements. He finds that the path must go through N<sup>16</sup>, since there is no reasonable detour. He has then examined in detail the subsequent formation chain among the light nuclei. The result of this work is shown in Table XV. The values of  $W_{\beta}$ ,  $E_E$ , and  $T_{\zeta}$  are given in Table XV for the nuclei in the chain. The factor  $\alpha_s$  in Eq. (143) is also listed. The  $\beta$ -decay probability  $\lambda_{\beta}$  has been computed according to Eq. (138) and is listed in arbitrary units. The capture probability  $\lambda_c$  was computed according to Eq. (143) with a factor of proportionality (including  $f_s$ ) arbitrarily adjusted so as to yield best agreement between observed and computed relative abundances. The relative abundances have been computed in the following manner, which represents an approximate solution to the extremely complicated growth equations.

Smart  $\lceil 134 \rceil$  has assumed that the formation process reached a steady state determined by the density of matter and by the competition between  $\beta$ -decay and the radiative capture of neutrons. Clearly the latter process depends on density whereas the former does not. In the event that for a given nuclear species  $\lambda_c > \lambda_{\beta}$ then in the steady state the relative abundance of this species is inversely proportional to  $\lambda_c$ . If, on the other hand, for a given type of nucleus  $\lambda_{\beta} > \lambda_{C}$ , then these nuclei will  $\beta$ -decay on the average before capturing a neutron. If for the resulting nucleus  $\lambda_{c} > \lambda_{\beta}$ , the relative abundances of the original nuclei are taken proportional to  $(1/\lambda_{\beta}) + (1/\lambda_{C})$ , while if it takes more than one  $\beta$ -decay to reach a nucleus with a higher capture than  $\beta$ -decay probability, then the abundance of the original nuclei is taken proportional to  $[1/\lambda_{\beta}(1)]$  $+ [1/\lambda_{\beta}(2)] + \cdots + [1/\lambda_{\beta}(i-1)] + [1/\lambda_{c}(i)]$ . This procedure is very nearly equivalent to the statement that each nuclear species has a probability  $(\lambda_{\theta} + \lambda_{C})$  sec.<sup>-1</sup> of changing into some other species, so that its relative abundance in the steady state is proportional to  $(\lambda_{\beta}+\lambda_{C})^{-1}$ . Smart has computed relative abundances in the former manner using the values of  $\lambda_{\beta}$  and  $\lambda_{C}$  given in Table XV and his results are compared with Browns' data in Fig. 21. Isobaric abundances have been added and theory and observation fitted at A = 28. The agreement in detail is better than that obtained by equilibrium theory (see Fig. 5), in that not only is there agreement in trend but also most of the computed abundances follow the detailed variations in the observed abundance data. The density of matter corresponding to the adjustment of the  $\lambda_c$  relative to the  $\lambda_{\beta}$  to obtain a fit may be determined from the fact that the average neutron-capture cross section for these light elements is about 10<sup>-4</sup> barn according to the average variation in the binding of the last neutron between the Heisenberg valley and the formation path. Assuming  $kT \cong 0.1$  Mev during the process, one obtains a matter density of about  $10^{-6}$  g/cm<sup>3</sup>. It is interesting to note that this value of the density is in agreement with that obtained in the simple neutron-capture theory discussed earlier. Since the formation path would appear to have been that in which  $\lambda_{\beta} \cong \lambda_{c}$ , a theory involving  $\lambda_c$  only should predict the general trend one would obtain using both  $\lambda_{\beta}$  and  $\lambda_{C}$ .<sup>††</sup> It is reasonable to suppose that the effect of ignoring  $\beta$ -decay becomes less important as one goes toward the heavy elements. However, even here one should expect that  $\lambda_{\beta}$  would

<sup>††</sup> One can formulate a non-equilibrium theory which includes not only neutron decay and the universal expansion but also the competing  $\beta$ -decay process. This involves a growth equation for each species (Z, A) including terms of the type  $\lambda_{\beta}\xi$ . Furthermore, one should take into account the specific  $\sigma_c(n, \gamma)$  and  $\lambda_{\beta}$  for each species (Z, A).

be involved in the detailed features of the computed abundances.<sup>‡‡</sup>

An interesting result of Smart's work [136] is that it makes reasonable the observed even-odd variation in abundance. Consider the behavior of the energy available for  $\beta$ -decay,  $W_{\beta}$ , and the binding energy of the last neutron,  $E_B$ .

A	$W_{oldsymbol{eta}}$
Odd	Decreases monotonically with increasing $Z$
Even	Decreases with increasing $Z$ but is larger for
	odd than for even $Z$
Ζ	$E_B$
Odd	Decreases with increasing $A$ , larger for odd $A$
	than even $A$
Even	Decreases with increasing $A$ , larger for even $A$
	than odd $A$

Since it would appear that formation goes on with  $\lambda_{\beta} \cong \lambda_C$  and that the respective probabilities for the species (Z, A) depend on  $W_{\beta}(Z, A)$  and  $E_B(Z, A+1)$ , the favored processes for various kinds of nuclei should be in general as follows:

A	Ζ	Most probable event
Odd	Odd	$\beta$ -decay
Odd	Even	Capture
Even	Odd	No choice
Even	Even	No choice

Clearly the tendency is to form nuclei with even Z. The tendency to form even A nuclei can be seen from the variation of  $E_B$  with A for even Z nuclei. The magic number nuclei are of particular interest here in that these nuclei, which are quite abundant compared with neighboring elements, should be theoretically favored in abundance not only by their small neutron-capture cross sections but also by the fact that less stable isobars should decay to the magic numbers and accululate.

#### 4. Special Problems

While the non-equilibrium theory of element formation described has been generally successful in explaining the general trend of the relative abundance data as well as some of the detailed features, there are nevertheless certain difficulties. Perhaps the most serious of these is the fact that there do not appear to exist nuclei of atomic weights 5 and 8. Under the physical conditions which apply for non-equilibrium theory calculations one would not expect to find the many-body reactions which would provide the simplest mechanism for bridging these gaps. It may be mentioned that in equilibrium theories the high densities involved obviate this problem. In addition to the gaps at A=5 and 8, there are difficulties at A=10 and 14 as discussed by Smart [136]. The nuclei Be<sup>10</sup> and C<sup>14</sup> both have halflives for  $\beta$ -decay in excess of 10<sup>3</sup> years so that neutron capture would be expected to be important. However, neutron capture by these nuclei leads to Be<sup>11</sup> and C<sup>15</sup> which are unstable with respect to neutron emission. Smart has also determined that from A = 15 onward there do not appear to be any breaks in the formation chain which cannot be satisfactorily by-passed.

As already discussed, Fermi and Turkevich have examined the gap at A=5 in particular and were not able to find a forward-going reaction that would provide a sufficient quantity of the heavier nuclei. Thus, if one begins a non-equilibrium element-forming process at low densities with nucleons only, there does not appear as yet any simple means of bridging the gap. However, a small amount of material does leak through the gap and if the density of matter for the process is raised, this amount would be increased. It is suggested that a density increase together with the introduction of a "cycling" process might yield a sufficient quantity of heavy elements. A cycling process of this kind might involve the fission of very heavy nuclei which would feed the formation process with nuclei past the troublesome gaps. These "seed" nuclei would build up the quantity of heavy elements at the expense of neutrons principally. There are as yet no quantitative calculations regarding this question. Such a calculation would require the theoretical determination of the fission yield of heavy neutron-rich nuclei, such as might be formed in the neutron-capture process. A highly speculative way of providing "seed" nuclei to bridge the gaps is intimately connected with the nature of the "explosion" of the primordial material. It has been tacitly assumed throughout that in this explosion the material expanded into individual nucleons and that nuclei formed of neutrons only could not exist. However, one might consider the possibility that mixed with the nucleon gas there were some aggregates of neutrons, however short-lived they may be with respect to dissociation. These "droplets" would perhaps be distributed in size and because of neutron emission together with rapid  $\beta$ -decay might have yielded some nuclei suitable for a formation chain past the gaps.

In any event, if there is a neutron-capture formation chain, fission of the heaviest elements will serve to terminate the increase in atomic weight and the fission fragments would be distributed over the mass yield spectrum. Fission may not become important until one reaches nuclei beyond A = 238 for the reason that the nuclei formed must have been, in general, deficient in proton content. These extremely heavy nuclei would undergo spontaneous or neutron-induced fission after sufficient  $\beta$ -decay. Another possibility for terminating the formation of heavy nuclei would be the  $(n,\alpha)$  reaction, which again because of the reduced charge of these nuclei would be expected to become important for nuclei well past A = 238. It is interesting to note that the mass yield curves [61, 67] for the fission of

<sup>&</sup>lt;sup>‡‡</sup> In this connection it should be noted that the calculation of isotopic abundances in a non-equilibrium theory would require knowledge of neutron capture and  $\beta$ -decay probabilities for all nuclear species in the detailed formation chain. In addition, the relative effect of such secondary processes as photoemission of neutrons, etc., on the isotopes of a given element would have to be considered.

nuclei such as  $U^{236}$  and  $U^{239}$  have peaks near  $A \cong 95$ and 140, which may be compared with peaks in the abundance data observed in these regions of atomic weight.

Another of the difficulties in the neutron-capture theory is the existence of "shielded" isobars. If nuclei are formed with a neutron excess, then in general for a given atomic weight one would perhaps expect to find only those nuclei with the lowest Z necessary for stability. However, one does find in nature isobars which cannot be formed from elements containing 2, 4,  $\cdots$  less protons. Most sets of isobars are of the form  $zX^A$  and  $z_{+2}X^A$ , and it is the latter or shielded isobar which apparently cannot be reached by  $\beta$ -decay. Smart [135] has suggested that the shielded isobars result from  $(\gamma, n)$  reactions. He has examined 55 sets of even Z-even A isobars of which in 41 cases a  $(\gamma, n)$  reaction with a stable odd nucleus could have yielded the shielded isobar. In the remaining 14 cases the shielded isobar could have resulted from two successive  $(\gamma, n)$  reactions. In support of this suggestion Smart has examined the abundances of shielded isobars and of parent nuclei for the  $(\gamma, n)$  reaction. Let  $\alpha_i$  and  $\alpha_f$  be the abundances of the parent element and shielded isobar, respectively. Then  $(\alpha_i + \alpha_j)$  would have been the abundance of the parent element as formed by neutron capture only. The quantity  $\alpha_f/(\alpha_i + \alpha_f)$  would be the fraction converted into the shielded isobar. Computing this fraction for the 41 cases mentioned, one finds the average value to be about 40 times larger in the situation where the initial nucleus has an odd neutron content than for an even neutron content. This is in line with the fact that the binding of the last neutron is weaker in odd Nnuclei and consequently at a given temperature the  $(\gamma, n)$  reaction probability would be higher. It is significant that the  $(\gamma, n)$  mechanism does not predict any shielded isobars which are not observed. Smart [135] has examined the  $(\gamma, n)$  reaction quantitatively and finds that at  $kT \cong 0.1$  MeV its probability is somewhat less than the probabilities of  $\beta$ -decay or neutron capture, but is sufficiently large to yield the desired effects. This question of shielded isobars is of particular interest in the work of Mayer and Teller [114] to be discussed.\*

In connection with  $(\gamma, n)$  reactions it is interesting to compare the proportionality, recently pointed out by Jensen [94], between the  $(\gamma, n)$  cross section and isotopic number for Z < 40 with the observation that for Z < 40 in most cases the lightest stable isotope for a given element is predominantly abundant. The striking proportionality between  $\sigma(\gamma, n)$  and  $T_{\rm f}$  is no longer evident for Z > 40, and it should be noted that among the heavier elements the predominantly abundant isotopes are skewed toward higher atomic weight. Mayer and Teller [114] also point out that among the light elements the lightest isotopes are predominantly abundant. They explain this behavior as arising from a building up process consisting of the addition of protons to already existing nuclei. This implies that for a given Z the most proton-rich isotope will, of course, be the lightest and in a process of proton captures, the most abundant.

An outstanding feature of the relative abundance data is the existence of a large peak in the vicinity of iron. The existence of this peak has yet to be explained by any of the theories of element formation. The fact that the elements near iron are most stable makes the explanation of this peak according to some kind of equilibrium theory quite attractive. However, this explanation has not as yet been satisfactorily made although one should carefully examine the apparent abundance peak at A = 60 in several of the stellarmodel solutions of Beskow and Treffenberg  $\lceil 18 \rceil$  (see Fig. 9). In a non-equilibrium theory it would seem reasonable to suppose that thermal dissociation might reduce the abundances for the elements below iron since the rate of thermal dissociation is an exponential function of the binding energy per nucleon, which energy falls off rapidly with decreasing A below  $A \cong 56$  [11]. While thermal dissociation has been assumed to be negligible in the non-equilibrium theory discussed, whatever dissociation there was would have tended to emphasize in abundance the elements near iron. On the heavy element side of the iron peak whatever photodisintegration processes there were would also have tended toward emphasis of this peak since cross sections for  $(\gamma, n)$  reactions are observed to rise rather sharply for atomic weights greater than about 60 [27, 122]. Perlman and Friedlander [122] have recently found that the yields of  $(\gamma, n)$  reactions appear to be about an order of magnitude larger for  $Z \ge 29$  than for lower Z. The transition is quite abrupt and corresponds very closely to the position of the transition between the iron abundance peak and the rest of the abundance curve (see Fig. 2). This effect is related to that reported by Jensen [94] since there is definite jump in the isotopic spin at Z=29 for the nuclei considered by these investigators. However, these correspondences must be examined in light of the fact that the  $\gamma$ -ray energies used by Perlman and Friedlander were 50 and 100 Mev.

Finally, the question of abundance peaks in the neighborhood of the magic number nuclei should be mentioned again. Qualitatively the neutron-capture theory would seem to predict high abundance for these nuclei of low capture cross section. Some rough calculations indicate that an element of small  $\sigma(n, \gamma)$  does indeed pile up but the abundances of the succeeding elements are greatly depressed [8]. The depression in relative abundance is so large that the picture is undoubtedly not this simple. It has been suggested that [8] while certain elements with small neutron-capture cross sections may have piled up, the succeeding elements may have resulted mostly from neutron capture

<sup>\*</sup> In a paper being prepared for publication Dr. J. S. Smart discusses the probably greater effect of capture  $\gamma$ -rays than the  $\gamma$ -rays in the blackbody distribution in forming shielded isobars.

by isobaric nuclei of more normal cross section. A more accurate description of the reasons for the correspondence between the observed abundance peaks and the regions of magic number nuclei, particularly since the reacting nuclei may be neutron-rich, would appear to require consideration of detailed formation chains.

The special problems discussed have at best received qualitative consideration here. Unfortunately, in the present state of the neutron-capture theory, there are as yet no quantitative discussions to decide these questions unequivocally.

# (e) The Polyneutron Fission Theory

The work of Cherdyncev  $\lceil 36 \rceil$  on the origin of the elements is, strictly speaking, an equilibrium theory (see Section III). However, his work is particularly interesting in that it described for the first time the properties of a "nucleus" of stellar dimensions, and it should perhaps be considered together with the work of Mayer and Teller [113, 114]. These nuclei are considered to be made up essentially of neutrons. Cherdyncev arrives at the result that such nuclei would be unstable against fission, and that the nuclear forces are strongly modified by gravitation. As mentioned earlier, he has examined the equilibrium distribution in such a "stellar" nucleus of pure neutron-nuclei in the range of ordinary atomic weights. To obtain the required equilibrium distribution, Cherdyncev uses a temperature of about 4 Mev and a density of 0.03 times nuclear density. One must question the validity of an analysis based on the existence and individuality of such neutron-nuclei in what is essentially a large body of nuclear fluid. Cherdyncev supposes that "stellar" nuclei whose "atomic weight" exceeds  $\sim 10^{-5}$ - $10^{-6}M_{\odot}$  should explode quite rapidly, that the equilibrium distribution of abundances with respect to atomic weight will be preserved, and that the common nuclei result from the  $\beta$ -decay of the purely neutron fragment-nuclei. While Cherdyncev recognizes the very approximate nature of his freezing-in considerations, it is difficult to see why the distribution in atomic weight would be preserved at all, particularly in view of the work of Smart and of Mayer and Teller. While  $\beta$ -decay would tend to stabilize such nuclei, the neutron-nuclei will certainly evaporate neutrons, and the distribution in atomic weight will be altered at least in this regard. In addition, the freezing-in problems already discussed in Section III must be even more carefully examined in this picture.

A very different approach to the problem of the origin of the elements is that taken by Mayer and Teller [113, 114]. Noting the differences between light and heavy elements in over-all abundance and in isotopic abundance ratios, they suggest that the light elements were built up by thermonuclear processes, involving principally protons. They emphasize, on the other hand, that the heavy elements, Z > 34, must have formed under conditions involving high neutron concentrations. This conclusion results from the observation that for the heavy elements the heavy isotopes are much more abundant than the lightest isotopes. If neutrons are not involved one must resort to charged particle reactions and quite high temperatures, and it would be difficult to understand the observed characteristics of heavy nuclei.

Mayer and Teller have examined the hypothesis that the heavy nuclei are fragments resulting from the break-up of a "cold" nuclear fluid consisting primarily of neutrons. The break-up of the primordial nuclear fluid is assumed to be similar to fission processes, except that the source nuclei may be much heavier than those now known. Because of their high charge these nuclei break up, yielding highly excited fragments with large



FIG. 22. Isotopic abundances calculated according to the break-up hypothesis by Mayer and Teller [114]. The observed isotopic abundances are those given by Seaborg and Perlman [129].



FIG. 23. The time dependence in the expanding universe of proper distance, L, the densities of matter and radiation,  $\rho_m$ , and  $\rho_r$ , and the temperature, T, for the physical conditions given by Eq. (146), according to Alpher and Herman [9].

neutron excess. These fragments in turn should undergo neutron evaporation and eventually  $\beta$ -decay, and become, in this theory, the present heavy nuclei. The difference between "cold" nuclear fluid and a compressed hot nucleon gas, the latter being involved in the non-equilibrium theory previously discussed, lies in the specification, according to Gamow and Critchfield [61], of a so-called critical temperature of the nuclear fluid. Mayer and Teller have assumed that the temperature was sufficiently low so that the nuclear fluid was essentially a liquid.

Without considering at this point a polyneutron model to serve as the source for the break-up process, one can examine the possible consequences of such a break-up in terms of the isotopic abundance ratios which might result. The neutron content of a fragment of charge Z is not fixed, but the average value of N should depend on the properties of the polyneutron prior to the break-up. Mayer and Teller assume that the ratio N/Z does not vary widely, and that in the break-up process the probability of finding a fragment with various internal energies can be represented by a Gaussian distribution. These fragments will lose neutrons successively by evaporation until finally there will not be sufficient excitation energy to evaporate another neutron and the process terminates. The probability of the process terminating at a definite isotope with neutron number N is taken to be

$$F(Z)[E_B(N, Z) - E_B(N-1, Z)] \exp\{-[E_B(N, Z) - E_B(N_0, Z)]/s_T^2\}, \quad (144)$$

where  $E_B(N, Z)$  is the binding energy of an isotope containing N neutrons,  $E_B(N_0, Z)$  is the binding energy of the nucleus for which the probability is a maximum,  $s_T = s_T(Z)$  is the isotope spread for a given Z, and F(Z)is a normalization factor. The factor  $[E_B(N, Z) - E_B(N-1, Z)]$  is, of course, the binding energy of the



FIG. 24. The time dependence in the expanding universe of proper distance, L, the densities of matter and radiation,  $\rho_m$ , and  $\rho_r$ , and the temperature, T, for the physical conditions given by Eq. (148), according to Alpher and Herman [9].

last neutron. Mayer and Teller have examined Eq. (144) for several sets of isotopes, using for the mass of an atom the semi-empirical formula [2, 24, 114]

$$M = A - 0.00081Z - 0.00611A + 0.014A^{\dagger} + 0.083[(A/2) - Z]^2 A^{-1} + 0.000627Z^2 A^{-1} + \delta, \quad (145)$$

where  $\delta = 0$  for odd A, and  $\delta = \mp 0.036A^{-\frac{3}{4}}$  for even Aeven Z and even A-odd Z, respectively. This expression for M does not take into account stability fluctuations such as occur for the magic number nuclei. Consequently, Mayer and Teller restricted their attention to the region  $62 \leq Z \leq 78$  in which the influence of shell structure is not important. From Eq. (145) one may calculate the value of Z for a given A for which the binding energy is a minimum. The points (Z, A) so determined define a stability line, and the observed asymmetric isotopic abundance distributions require that the maximum of the Gaussian distribution lies at higher N then that on the stability line for a given Z. In making their calculations they have assumed that if (N, Z) lies on the stability line, the quantity  $[E_B(N, Z) - E_B(N_0, Z)]$  is a constant whose value they have taken as 0.03569 mass unit. The spread  $s_T = 0.02415$  mass unit was so chosen that the abundance of the lightest isotopes best fit the observed values. The results of their calculations are shown in Fig. 22 where they are compared with observed isotopic abundances as tabulated by Seaborg and Perlman [129]. The agreement is quite good. This calculation appears to give and even exaggerate the variations of abundance with A which have been attributed to the "shielded" elements. Mayer and Teller suggest that these abundance calculations can be improved in several respects. First, one should take into account, in determining  $E_B$ , the nuclear shell structure. Second, one should include the effect of the observation, in studies of nuclear fission, that in some cases  $\beta$ -decay can be

followed by further neutron evaporation when this evaporation leads to a "magic number" nucleus. Finally, one should account for the possibility of the recapture of evaporated neutrons. Such neutron capture would affect the final abundance distribution according to the neutron-capture cross sections.

According to Mayer and Teller, the even-odd abundance variation may be understood as resulting from the fact that for large neutron excess in the original nuclear fluid, structural units similar to  $\alpha$ -particles would be favored over the much less stable triton configurations. This would be reflected in a predominance of even Z fragments in the break-up process.

The polyneutron model considered by Mayer and Teller as a starting point for the break-up process is assumed to be a configuration of nuclear fluid, which cannot spontaneously disintegrate into free nucleons and whose mass is less than that of a star. The latter qualification avoids the necessity of discussing the effects of gravitation and general relativity. However,  $\beta$ -decay can go on, with two results. The product protons will combine and form principally  $\alpha$ -particle configurations, while the electrons, whose escape would soon make  $\beta$ -decay impossible, can remain in so large a "nucleus" and neutralize the positive charges. Considerations of the stability of tritons and  $\alpha$ -particles indicate that when the electron density reaches a value such that the zero-point energy of the electrons is the same as the energy release in triton formation,  $\beta$ -decay will cease. One then has a thin atmosphere of electrons, whose thickness is about  $6 \times 10^{-11}$  cm. This electron atmosphere is shown to reduce, and, in fact, render negative the surface tension forces of the polyneutron, because electrostatic repulsion between the electrons will exceed the surface tension due to nuclear forces. Examination of the surface stability properties of the polyneutron indicates that droplets of fluid will break off and their dimensions will be of the order of the thickness of the electron atmosphere. Teller and Mayer have shown that the charges of these droplets should spread over the whole range of known atomic numbers and well beyond. In the present state of this theory, no estimate has been made of the relative abundances of nuclear species to be expected. However, the proposed polyneutron model leads to a break-up process and is apparently consistent with the proposed scheme for predicting isotopic abundance ratios.

An interesting application of the work of Mayer and Teller might be to the freezing-in problem confronting equilibrium theories of element formation. If in the "explosion" of configurations in which there is an equilibrium distribution of nuclei, there appear fragments of very neutron-rich material one might apply these ideas in examining the behavior and final form of these fragments. As a theory of element formation *per se* the work of Mayer and Teller involves certain difficulties [61]. The role of a "cold" primordial nuclear fluid either in the initial stages of the expanding universe or in any kind of stellar structure is not clear. Furthermore, it would appear that completely different mechanisms would be required for the formation of light and heavy nuclei. These mechanisms would have to be examined to see if they are consistent with one another and if they lead to the observed abundance distribution of elements. In any event the break-up hypothesis should be examined further if only for the reason that it predicts quite successfully the observed isotopic abundance distributions among the heavy elements.

#### (f) Element Formation and Cosmology

The explanation of the observed relative abundance distribution of the elements is of interest not only in understanding the nature of the distribution in terms of nuclear properties but also because in this way one can examine the physical conditions which must have prevailed in the locale of element origin. The latter information provides a link between the element forming process and cosmology or stellar structure. In the case of equilibrium theories of element abundance it has been seen that there are essentially two points of view, namely, formation in some kind of prestellar body or continuously in special types of stars [see Section III(b)5]. In the former case Klein [97] has considered the cosmological implications of prestellar bodies but as yet no conclusions can be drawn as to the nature of the non-static solutions. With regard to element formation in special types of stars the cosmological model is involved insofar as these stars must fit into any general theory of stellar structure and evolution.

In the case of the neutron-capture theory it has been seen that the calculation of theoretical abundances involves the specification of a particular cosmological model. It is of interest to consider the possible information that results from the specification in this theory of physical conditions at a given epoch. It will be recalled that each of the several attacks on the non-equilibrium capture theory of element formation has yielded values of the temperature and matter density during the period of element formation. In general the physical conditions obtained are consistent with one another.

Alpher and Herman [9] have examined the time dependence of the temperature, of the densities of matter and radiation, and of the proper distance in the expanding universe under the following conditions. On the basis of the neutron-capture theory in the expanding universe it appears reasonable to take at t=670 sec. after the start of the expansion

$$\rho_{m'} \cong 10^{-6} \text{ g/cm}^3, \\
\rho_{r'} \cong 1 \text{ g/cm}^3(T \cong 0.6 \times 10^{9^{\circ}} \text{K}).$$
(146a)

In order to examine the behavior of the various quantities up to the present epoch it is necessary to specify, say, the density of matter now,  $\rho_{m''}$ , and for this the following value has been taken

$$\rho_{m''} = 10^{-30} \text{ g/cm}^3.$$
 (146b)

From Eq. (91c) one can then calculate  $\rho_{r''}$ , the present density of radiation (the residual radiation density from the expansion alone), as

$$\rho_{r''} = 10^{-32} \text{ g/cm}^3.$$
 (146c)

This value of  $\rho_{r''}$  corresponds to a temperature now of about 5°K. Using the proper distance l and  $l_0$  as already defined [see Section IV(b)], one may determine the constants  $\alpha$  and  $\alpha$  in Eq. (91). With the densities given in Eq. (146),  $\alpha = 1$  g and  $\alpha = 10^8$  g cm. These values of  $\alpha$  and  $\beta$  fix the dependence of  $\rho_m$  and  $\rho_r$  on time through  $L(=l/l_0)$ . The time dependence of L may be found from Eq. (93) while the time dependences of  $\rho_m$ ,  $\rho_r$ , and T may be found from Eqs. (91) and (99). The result of this calculation is given in Fig. 23 where it may be noted that all the quantities plotted bear simple relationships with time to within one or two orders of magnitude of the time when the universal expansion changed from one controlled by gravitation to one of free escape. This transition, which depends on the relative magnitude of the two terms in Eq. (92), occurs in this case in the region 1013-1014 sec. Furthermore, this transition is such that earlier than the transition,  $\rho_r > \rho_m$ , while later  $\rho_m > \rho_r$ . After this transition the plotted quantities again become simple functions of the time and it may be shown, on the basis of the discussion in Section IV(b), that one has for large t the following approximate relationships:

> $L = K_2^{\frac{1}{2}}t,$   $\rho_m = (\rho_{m''}/K_2^{\frac{1}{2}})t^{-3},$  $\rho_r = (\rho_{r''}/K_2^{2})t^{-4},$

(147)

and

$$T = [(c^2 \rho_{r''})/(a_r K_2^2)]t^{-1},$$

where the notation is that of Section IV(b). These results are somewhat altered if one takes the cosmological constant  $\Lambda$  as being different from zero. In particular, it can be shown ¶¶ that with the physical conditions already discussed, one obtains  $\Lambda = 8.6 \times 10^{-34}$ sec.<sup>-2</sup> if the present epoch is taken as  $t_P = 10^{17}$  sec. The corresponding unit of radius of curvature, and therefore the radius of curvature at the present epoch [see Eq. (90a)], is found to be  $5 \times 10^{27} (-1)^{\frac{1}{2}}$  cm. In any event the physical conditions discussed appear to indicate that the universe is in a freely expanding state and is of the open hyperbolic type. In order to study the sensitivity of the time dependences of L,  $\rho_m$ ,  $\rho_r$ , and T with respect to the choice of physical conditions at a given epoch, the following additional set of density values have been considered:

 $\begin{array}{l} \rho_{m'} \cong 1.78 \times 10^{-4} \text{ g/cm}^3, \\ \rho_{r'} \cong 1 \text{ g/cm}^3, \\ \rho_{m''} \cong 10^{-30} \text{ g/cm}^3, \end{array}$ (148)

and

$$\rho_{r''} \cong 10^{-35} \text{ g/cm}^3(T_{\text{now}} \cong 1^\circ \text{K}).$$

The results of the calculations using Eq. (148) are shown in Fig. 24. The significant difference between this and the previous case considered is that the transition from a controlled to a free universal expansion occurs at about  $t=10^{10}$  sec.

The specification of the physical conditions in the expanding universe as functions of time should make it possible to examine a variety of cosmological problems, including, for example, the origin of cosmic radiation, the formation of molecules, and the condensation of galaxies. If it should prove that cosmic radiation is a universal phenomenon then its characteristics should perhaps be connected with the original elementforming process. II II It seems feasible also to examine the possible origin and abundance distribution of molecules and perhaps grains in the prestellar phase, since one now has information concerning the temperature, density, and abundance distribution of nuclei in the expanding universe. This study should prove in some respects to be formally similar to the non-equilibrium theory of element formation. As the temperature in the universe drops below the dissociation energies of molecules, nuclei which have previously captured some electrons would form the various kinds of molecular species. Their relative abundances should be related to the original nuclear distribution and to the probability for formation of the particular molecular species. Qualitatively, at least, the most abundant molecules might be expected, to be simple combinations of H, C, N, O, and Fe. Clearly, calculations of this kind compared with observed molecular abundances in the solar system would have an interesting bearing on the nature of the formation of the planets [102].†

While it is not feasible in this review to give anything in the way of a survey of cosmological consequences, it is of interest to discuss briefly an example of the kind of study that one can make using the cosmological model whose constants are based on the neutron-capture theory of element formation. Recently, Gamow [58] suggested that the galaxies may have condensed at the transition time, i.e., the time when  $\rho_r$  became equal to  $\rho_m$ . At this time,  $t_c$ , when matter begins to take over the principal role, the previously homogeneous material might break-up into separate bodies which subsequently separate due to the expansion. As a rough approximation to a condition for condensation, Gamow [59-61]has applied the Jeans' principle of gravitational instability [88, 130]. This criterion which gives the diameter,  $D_G$ , of a condensation may be written in the following form

$$D_{G^2} = (5\pi kT_c) / (3Gm_0\rho_{m,c}), \qquad (149a)$$

|| || For example, see Lemaître [106] for some interesting views in this connection. However, with the cosmological model considered in the present paper it seems unlikely that primordial cosmic radiation could have survived through the early epoch of the expanding universe when mass density was quite high.

<sup>†</sup> The problem of molecular formation and the physics of cosmic grains in interstellar space recently has received much attention [See, for example, F. Cernuschi, Astrophys. J. 105, 241 (1947)]. A discussion of these problems would take us too far afield.

 $<sup>\</sup>P$  The authors' calculations concerning the cosmological constant are given by Gamow [60].

where  $T_c$  and  $\rho_{m,c}$  are taken at the time of transition,  $t_c$ , when  $\rho_{m,c} = \rho_{r,c}$ . Then the mass of the condensation is

$$M_G \cong \rho_{m,c} D_G^3. \tag{149b}$$

With the set of density conditions as given in Eq. (146) one obtains for  $D_G$  and  $M_G$  the values  $2 \times 10^3$  light years and  $4 \times 10^7 M_{\odot}$ , respectively, which are in moderate agreement with the observed average values  $\lceil 9 \rceil$ . At the transition time,  $t_c \cong 10^7$  years,  $\rho_{m,c} \cong 10^{-24}$  g/cm<sup>3</sup> and  $T_c \cong 6 \times 10^{20}$  K. If the Jeans' criterion was satisfied, the separation between condensations of diameter  $D_{G}$ would also have been of the order of  $D_G$ . This separation distance would then have increased because of the expansion, whereas the condensations themselves would not have expanded. From the time variation of proper distance one may compute the present separation of condensations to be about 10<sup>6</sup> light years in agreement with observed separations. For the set of densities given in Eq. (148), on the other hand, one obtains  $D_G \cong 1$  light year,  $M_G \cong 3 \times 10^5 M_{\odot}$ ,  $t_c \cong 6 \times 10^2$  years,  $\rho_{m,c} \cong 10^{-15}$  g/cm<sup>3</sup>, and  $T_c \cong 10^{5^{\circ}}$ K. Clearly the calculation is very sensitive to the choice of densities. It may be pointed out that one should not use for these calculations extrapolations of the approximate equations for T,  $\rho_m$ , and  $\rho_r$  as functions of time which are valid only for early t [7, 9, 59]. This would lead to a  $t_c$  greater than the present age of the universe.

The classical Jeans' stability criterion is not actually valid [9, 60, 131] here, since it does not take into account the possible effects of the universal expansion, the presence of radiation, the low matter density, and relativistic effects due to the large-scale phenomena involved. Lifshitz [108] has recently investigated gravitational instability in non-static isotropic models of the universe and has shown that arbitrarily small perturbations of the field and of the distribution of matter either decrease or grow so slowly as to be unimportant in the formation of condensations. This problem has also been examined by Gamow. Metropolis. Teller, and Ulam [60] who arrived at the same conclusion. However, the effects of radiation were not considered in either case nor were finite perturbations. The latter authors have recently modified this condensation problem by taking into account the effects of radiation  $\lceil 60 \rceil$ . They have shown that in the expansion of the universe the difference in the specific heats of radiation and of matter would result in the temperature of matter lagging the temperature of radiation according to

$$(T_r - T_m)/T_r = 10^{-12} t$$
(years). (150)

At the time  $t=10^7$  years, when galaxies are presumed to have formed, the lag is about  $0.01^{\circ}$ K. This small temperature difference nevertheless gives rise to a strong interaction between particles which may be described as a "shadow-casting" effect in analogy to radiation pressure phenomena. These forces vary with distance in the same way as gravitational forces and should assist in overcoming the expansion in the formation of condensations. Quantitative calculations are in progress concerning the possibility of galactic formation involving these "mock-gravity" forces.\* It would appear from their work that an increase in the value found for the density of matter in the universe at the time of element formation would help to understand the possibility of forming condensations. This required density increase is consistent with the apparent need for a higher density to bridge the gaps at A=5 and 8 in the non-equilibrium theory of element formation [see Section IV(d)2].

Certain recent developments in cosmological theory are perhaps intimately connected with the problem of the origin and relative abundances of the elements since they involve the intriguing possibility of matter creation [109a]. Perhaps the earliest suggestion of this kind was that of Jeans [88] who conjectured that in the centers of galaxies there might be singular points through which matter entered the universe. More recently Dirac [41] hypothesized the time variation of many of the universal constants and in particular pointed out that a decrease in the constant of gravitation with time would require an increase in the amount of matter throughout the universe. Jordan [96] has developed a cosmological theory involving time-dependent universal constants and has suggested that not only does matter somehow enter the universe continuously with time but that the matter enters as "drops" containing about the same number of elementary particles as in presently observed giant stars  $(50M_{\odot})$ . In Jordan's theory the conservation of energy is preserved. Bondi and Gold [26], and Hoyle [81, 82] consider stationary universes showing expansion properties associated with the continuous creation of matter everywhere at small rates. Hoyle suggests that this matter may enter the universe in the form of neutrons which decay into protons and yield the predominant abundance observed for hydrogen. All of these ideas obviously are pertinent to the question of the origin and relative abundances of the elements and their further development should prove quite interesting.

#### (g) Discussion

The neutron-capture theory of element formation predicts the general trend of the observed relative abundance data. This degree of success has been obtained with a theory which involves the specification of essentially only one free parameter, namely, the density of matter during the formation process. However, the non-equilibrium theory is seen to be intimately connected with the choice of a particular cosmological model. Failing the successful demonstration of the survival of an equilibrium distribution of elements through a freezing-in process, it seems to us at present

<sup>\*</sup> Gamow (private communication) informs us that these additional forces are apparently not yet sufficient to provide a mechanism for condensation.

more reasonable to suppose that the elements were formed in the prestellar state of the universe. This would seem to require a non-equilibrium theory of element formation.

The inclusion of the stability properties of atomic nuclei would appear to explain moderately well some of the detailed features of the abundance data. The detailed nature of the formation processes for the very light elements has been examined and found to be consistent with the picture proposed but this study leads to the principal difficulty in this theory which is as yet unresolved. This difficulty concerns the nonexistence of nuclei of atomic weights 5 and 8. A somewhat similar difficulty exists in connection with the magic number nuclei in that the formation process must by-pass these nuclei of abnormally low neutron-capture cross section. The quantitative demonstration that the shielded elements are indeed the result of  $(\gamma, n)$  processes has yet to be made. The theory in general is in a quite approximate state at the present time and it remains to be seen whether detailed calculations, which would appear to be extremely difficult, will satisfactorily explain the detailed features of the abundance data.

#### V. SUMMARY

The following brief summary is presented in order to emphasize what the authors believe to be the successes and the major difficulties of the several theories of the origin and relative abundance distribution of the elements which have been discussed.

Equilibrium theory applied to an assembly at a single density and temperature does not reproduce the observed relative abundance distribution. However, general agreement with the observed distribution can be obtained by considering an equilibrium assembly in a spatial configuration in which the density and perhaps the temperature vary with position. The specification of these physical conditions then defines the nature of the configuration or stellar model. Such an approach has led to the concept of element formation in an isothermal material prestellar body embedded in a sea of radiation. In another approach it is considered that the elements are formed in dehydrogenized, collapsing, rotating stars, where it is supposed that the conditions attained in the stellar interior give rise principally to elements in a certain narrow range of atomic weights. The entire abundance distribution is then presumed to be the consequence of the existence of a variety of such collapsing stars with differing initial physical conditions. Such stars have been identified with novae and supernovae. One of the principal difficulties with an equilibrium theory lies in the fact that no mechanism has yet been proposed for the explosion of the "prestellar" radiation stars, while in the case of novae it is generally believed that chiefly surface material is ejected in the outburst. In this latter case, therefore, it is not clear how the elements formed in the interior would be distributed throughout space. Circulation in the collapsing

novae would not appear to improve the situation since the equilibrium distribution would shift with radial position in the star. In the case of supernovae, which have been suggested as the source for the heaviest elements, the explosion may be of such nature as to disrupt the entire star and perhaps leave no remnant. In general, however, it is as yet not possible to understand how any nuclear equilibrium that might have been established could survive in anything like its original form through the changing physical conditions associated with an explosion required to distribute material homogeneously throughout the universe. A specific difficulty in an equilibrium theory lies in the lack of a significant correlation between the computed and observed detailed variations in abundance from element to element. Finally, the observed isotopic abundance ratios do not seem to be compatible with an original nuclear equilibrium.

A non-equilibrium theory of element formation requires the specification of detailed nuclear processes, principally successive radiative capture of neutrons with intervening  $\beta$ -disintegration. Since there appears to be good reason to suppose that the elements were formed in the prestellar state of the universe, it is also necessary to specify a cosmological model. The general trend of the observed relative abundance distribution has been successfully reproduced by a non-equilibrium theory. The details of the abundance distribution in most respects appear to be understandable at least qualitatively and in some cases semi-quantitatively. The major difficulty faced by this theory is the nonexistence of nuclei at atomic weights 5 and 8 with the consequence that a formation chain through the lightest elements has not yet been constructed. There is at present no quantitative demonstration that such detailed features as the iron peak, the magic number peaks, the shielded isobars, and isotopic abundances are consistent with the non-equilibrium theory that has been formulated. However, this theory is flexible in the sense that the many secondary phenomena not yet included in the theory are of such nature as to indicate the successful prediction of the detailed features of the data which now lack quantitative explanation. It appears to us that another attractive feature of the non-equilibrium theory is that it fits in with the generally accepted picture of the early stages and present structure of a homogeneous, isotropic, expanding universe.

The polyneutron fission theory has had considerable success in predicting relative isotopic abundances. However, with this theory and with theories of "matter creation" the supposed initial conditions seem improbable to us at the present time. It is not yet possible to say whether these approaches can lead to a consistent theory of the origin and relative abundance distribution of the elements.

The authors have attempted in this review to bring together the rather large amount of work that has been done on the problem of the origin and relative abundance distribution of the elements. It should be clear from the discussions given that this subject is far from being in a settled state and it is hoped that this study will serve to point up many of the interesting problems that remain.

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