THE STATUS OF THE γ-RAY CHANGE

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Abstract

It has been general to assume that the emission of γ-rays in radioactive disintegration is an effect subsidiary to the ejection of the α or β particle from the nucleus. Recently, however, the general experimental proof that the time interval concerned in this nuclear process is considerably greater than the time of relaxation of the inner extra-nuclear electrons—and, more particularly, an experiment by Jacobsen and its interpretation in terms of γ-ray disintegration constants as small as 10^5 sec^{-1}—have together resulted in a revision of opinion. In the present paper Jacobsen's interpretation is examined in more detail and is shown to imply that the emission of a quantum of γ radiation between the β particle disintegration of radium C and the α particle disintegration of radium C′ is an absolute necessity. This condition is attended with certain difficulties. In the course of the investigation it is shown that where α particles may be emitted either immediately following an earlier disintegration, or with the intermediate emission of any single member of a number of γ-ray quanta, then, as long as all the α particles possess identical velocities, the α activity of the immediate product of the former disintegration decays strictly according to the exponential law characterized by the disintegration constant of the α particle change.

In a second part of the paper an attempt is made to connect the complexity of the "normal" α radiation of thorium C and the emission of long range α particles from radium C and thorium C with the occurrence of certain lines in the γ-ray spectra of the elements in question.

Introduction

We know of three ways in which energy is spontaneously set free in the nuclear processes of radioactive disintegration. This available energy may appear as kinetic energy—possessed either by an expelled α-particle or by a β-particle—or it may be emitted in the form of radiant energy, as a quantum of γ radiation. Yet, from a broad general standpoint, these three modes do not appear to be of equal importance in nature. Considered merely in respect of quantity, α-particle kinetic energy represents roughly 85 percent, β-particle kinetic energy roughly seven percent and γ-ray energy the remaining eight percent of the total radiation from radioactive materials. But, in spite of this distinction, in the matter of fundamental importance, the α-particle change is not in a class by itself, with the β-and γ-ray changes subsidiary to it. Rather is it the γ-ray change which is peculiar, since in each case it is apparently incidental to some instance of particle disintegration, either α or β. Every atom of uranium, for example, is believed to undergo an

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invariable[1] sequence of α and β-particle disintegrations before passing over into an atom of uranium-lead, but concerning the various γ-ray quanta which may be emitted in the process there exists a wide range of possibility. Once γ-ray changes are included, the concept of the disintegration series loses all its simplicity.

The secondary nature of γ-ray emission was early recognised; it was only with the proof of the nuclear origin of the rays that the problem of their dependence became acute. More recently, with the spectral analysis of this type of radiation, and the establishment of systems of nuclear energy levels from which it is derived, the difficulties in the way of a satisfactory solution have, if anything, increased. At first the difficulty was concerned chiefly with the principle of conservation. If there was, in fact, a difference in the total energy radiated during the active lives of different atoms passing down the same disintegration series then it became difficult to escape the conclusion that for one and the same atomic species (uranium-lead, for example) different nuclei must possess different amounts of intrinsic energy—and that these different energy states must be permanent. A similar difficulty was encountered when the heterogeneity of β-particle velocities was finally established. Chalmers[2] has exhibited this difficulty in its most intractable form. For the present it is unlikely that in either case any solution will be found; it is very probable that these relatively small differences in internal energy-content as between otherwise identical nuclei may constitute a feature of common occurrence with the ordinary inactive elements. Some experiments of Rutherford and Chadwick[3] on artificial disintegration seem to admit of no other interpretation.

The Time Intervals Involved in γ-ray Emission

Apart from this question of conservation, however, and arising in most instances directly from the experimental work of Jacobsen,[4] there has recently been a growing concern with the more general problem of the status of the γ-ray change. Yet, even if Jacobsen's work be neglected, the crucial experiment of Ellis and Wooster,[5] proving that the order in time is invariably "β-particle emission$\rightarrow\gamma$ ray change," of itself is sufficient to lead to an important conclusion.[6] For the reorganization of the nucleus resulting in the emission of the γ-ray quantum is thereby shown to be slow compared with the time of relaxation of the extra-nuclear electrons of the K level. The work of Black[7] similarly shows that it is slow compared with the time of relaxa-

[1] The sequence is not strictly invariable, both for uranium X_1 and for radium C there are alternative modes of particle disintegration. For radium C, as for thorium C, the schematic representation of the "branching" is $B \xrightarrow{\beta} C \begin{cases} \xrightarrow{\beta} C' \xrightarrow{\alpha} \\ \xrightarrow{\alpha} C'' \xrightarrow{\beta} \end{cases}$ Reference to this diagram will make clearer subsequent discussions.

[2] Chalmers, Proc. Camb. Phil. Soc. **25**, 331 (1929).

[3] Rutherford and Chadwick, Proc. Camb. Phil. Soc. **25**, 186 (1929).

[4] Jacobsen, Nature **120**, 874 (1927).

[5] Ellis and Wooster, Proc. Camb. Phil. Soc. **22**, 844 (1925).

[6] Cf. Chalfin, Zeits. f. Physik **53**, 130 (1929).

[7] Black, Proc. Camb. Phil. Soc. **22**, 838 (1925).

tion of the L, M and N level electrons.[8] The inevitable nature of radioactive change, combined with its slowness, in many cases extreme, has long been recognised as the primary difficulty in the way of an explanation of particle disintegration in general;[9] here the same feature is seen to be characteristic of the γ-ray change also. Chalfin[6] has insisted that disintegration constants or transition probabilities have as real significance for γ-ray as for α or for β-particle processes. Now the range of value of these constants is very different in the two latter cases; the smallest α-particle disintegration constant is 10^{-18} sec^{-1}, the smallest β-particle disintegration constant 10^{-9} sec^{-1}.** There is at present no evidence to prove that γ-ray transition probabilities as small as 1 sec^{-1} do not exist, though in many cases they must be very much larger than this.

Jacobsen's experiments,[4] already referred to, have been interpreted as affording some indirect information concerning such transition probabilities. Making observations of the emission of α-particles from different regions in a beam of recoil atoms arising from the radium C β-particle disintegration, Jacobsen in effect showed that the α-particle activity of the subsequent product does not decay exponentially with the time, but instead rises from zero initially to a maximum and then decays, exactly as would be the case if the α-particle change were the second of two of nearly equal half period. Since the former process cannot involve the emission of particles, either charged or uncharged, it was suggested that it represents the general γ-ray emission which is known to take place intermediate in time between the β and α-particle changes. On this hypothesis the α-particle disintegration constant was estimated as 10^5 sec^{-1}, whilst a similar value was assigned for the effective γ-ray change.

The term "the general γ-ray emission" has been employed the more to emphasise the complexity of this radiation, "the effective γ-ray change" to stress the simplification which is made by comprehending these complex processes under a single constant of probability. For a complete explanation of the natural β-ray spectrum of radium C, γ-rays of nearly 50 different frequencies would be required. Suppose that partial transition probabilities $\lambda_1, \lambda_2, \cdots, \lambda_r, \cdots, \lambda_n$ be accorded to them, respectively, and let us investigate the problem more fully. At any time t let N be the number of atoms which have undergone the β-particle change only, $N_1, N_2, \cdots, N_r, \cdots, N_n$ the numbers which have undergone successive changes $\beta\gamma_1, \beta\gamma_2, \cdots, \beta\gamma_r, \cdots, \beta\gamma_n$, respectively, and $N\alpha_1, N\alpha_2, \cdots, N\alpha_r, \cdots, N\alpha_n$ the numbers which have undergone both β and α-particle changes with intermediate emission of $\gamma_1, \gamma_2, \cdots, \gamma_r, \cdots, \gamma_n$, respectively. For sake of simplicity we are assuming that not more than a single γ-ray quantum is emitted by each atom disintegrating. Furthermore, concerning the α-particle change, it is an experimental fact that the velocity of the particle ejected by any atom

[8] Rough estimates of these times are, for the K electrons, 10^{-15} sec, for the N electrons 10^{-10} sec.

[9] See, for example. Lindemann, Phil Mag. **30**, 560 (1915).

** If we neglect the β-ray bodies potassium and rubidium.

is independent of the γ-ray quantum just previously emitted. We must therefore assume that a single α-particle disintegration constant, $\lambda\alpha$, applies uniformly throughout. Now suppose that in a small fraction of the cases no γ ray is emitted between the two particle disintegrations.[10] Since the α-particle emitted in such a case is indistinguishable from any other, $\lambda\alpha$ must apply here also. Writing $N\alpha$ for the number of atoms which at the time t have undergone β- and α-particle changes in this way, in immediate succession, the complete equations of radioactive decay may be written[11]

$$\frac{dN}{dt} = -\left(\lambda\alpha + \sum_1^n \lambda r\right)N, \qquad \frac{dN\alpha}{dt} = \lambda\alpha N,$$

$$\cdots\cdots\cdots\cdots \qquad \cdots\cdots$$

$$\frac{dN_r}{dt} = \lambda r N - \lambda\alpha N r, \qquad \frac{dN\alpha_r}{dt} = \lambda\alpha N r,$$

$$\cdots\cdots\cdots\cdots \qquad \cdots\cdots$$

From these equations we obtain

$$N + N\alpha + \sum_1^n Nr + \sum_1^n N\alpha_r = N_0.$$

Taking as initial conditions $N = N_0$, and writing $\lambda\alpha + \sum_1^n \lambda r = \lambda$, the relevant solutions become

$$N = N_0 e^{-\lambda t},$$

$$\cdots\cdots\cdots$$

$$Nr = \frac{\lambda_r N_0}{\lambda - \lambda\alpha}(e^{-\lambda\alpha t} - e^{-\lambda t}),$$

$$\cdots\cdots\cdots$$

If A is the total α-particle activity of the preparation at the time t, then

$$A = \frac{dN\alpha}{dt} + \sum_1^n \frac{dN\alpha_r}{dt},$$

or

$$A = \lambda\alpha N_0 e^{-\lambda t} + \frac{\lambda\alpha N_0}{\lambda - \lambda\alpha}(e^{-\lambda\alpha t} - e^{-\lambda t}) \sum_1^n \lambda r,$$

$$= \lambda\alpha N_0 e^{-\lambda\alpha t}.$$

[10] With some γ-ray bodies it is almost certainly the case that on the average less than one quantum is emitted per atom disintegrating—but actually this is not so with radium C.

[11] There is implicit here the assumption that the radium C β-particle disintegration does not result in sets of atoms effectively "earmarked" to emit various γ-rays, after times statistically determined by the relevant disintegration constants λr. In such circumstances, with a source consisting of atoms collected immediately after the β-particle change, the quality of the γ-rays would vary with time. We can only assume that this is not the case, since the possibility of a direct experimental investigation seems very remote.

This result[12] indicates that, in spite of the occurrence of γ-ray emission, the quanta of which may be characterised by any number of different disintegration constants, yet, as long as no atom in the course of disintegration emits more than one quantum of radiation, and always provided that some atoms emit none at all, then the α-particle activity of the preparation decays accurately according to the exponential law determined by the disintegration constant of the α-particle change.

Such circumstances as we have assumed, therefore, cannot conspire to the result which Jacobsen found experimentally. However, the conditions with radium C are more complex than we have considered. Probably in this case, on the average, between one and two quanta of γ-radiation are emitted per atom disintegrating. Whilst this does not necessarily invalidate the supposition that in some instances the actual number is zero, it is incompatible with the supposition that this number is never greater than unity. Yet, as long as we retain the former permissible condition, it may be shown that the α-particle activity, starting from some value initially different from zero, decreases at first, whilst its rise to a maximum at a later time is entirely dependent upon the relative values of $\lambda\alpha$ and the various constants λr. If, therefore, Jacobsen's experiments are reliable in the more minute particulars it must be conceded that his suggested explanation stands or falls with the decision for or against the absolute necessity for γ-ray emission in all cases following the β-particle disintegration of radium C. It will be obvious that there is at least ample scope for an alternative hypothesis—as well as an urgent need for the development of other means of experimental attack upon this difficult problem. However, until fresh experiments have been performed it would be rash to make any further speculations.

Energy Considerations—Abnormal Particle Effects

There is a further aspect of the problem of the status of the γ-ray change which is best considered in its relation to a distinction which has already been drawn. Particle disintegration in general fits well with the scheme of the disintegration series; the inclusion of γ-radiation, on the other hand, plays havoc with its basic simplicity. Nevertheless, in certain cases, even as regards particle disintegration, anomalies occur—and the question arises, may not these anomalies be in some way connected with peculiarities of the associated γ-ray changes? The most pronounced abnormalities characterise the disintegration of the C bodies in all three series—there is the well-known dual disintegration of radium C, thorium C, and actinium C, there is the emission of groups of long range α- particles of rare occurrence by radium C and thorium C or their subsequent short-lived products,[13] and, finally, there is the additional complexity recently revealed in the "normal" α-particle emission of thorium C by the work of Rosenblum.[14] These two latter features, at least, possess

[12] Soddy, Phil. Mag. **18,** 739 (1909), in connection with the characteristics of hypothetical "rayless" changes, reached a result mathematically merely a special case of the above ($n=1$).

[13] See Nimmo and Feather, Proc. Roy. Soc. **122A,** 668 (1929).

[14] Rosenblum, Comptes rendus **188,** 1401 (1929).

numerical characteristics which might be made the basis of tentative hypotheses.

It is interesting to calculate the energy differences as between the common and the uncommon modes of disintegration in the different cases. For the satellite lines in the α-particle spectrum of thorium C this may be done directly. Rosenblum's results indicate groups of particles of energy respectively 0.4×10^5 electron volts greater, and 3.0 and 4.7×10^5 electron volts less, than the normal. For the groups of long range α-particles, where the immediate data consist of ranges, the calculation is very much less certain. To employ the simple Geiger law, as was done by Bates and Rogers,[15] may lead to grave error. In the following table Laurence's[16] correction curve has been used. This curve, showing the departure from the simple law as a function of the range, has been linearly extrapolated. Energy differences are probably correct in all cases, at least to 3 percent.

Product	α particle range* cm	Energy (electron volts $\times 10^{-6}$)	Energy difference (electron volts $\times 10^{-6}$)
Radium C	6.96	7.69	—
	8.1	8.45	0.76
	9.16	9.10	1.41
	10.0**	9.62	1.93
	11.0	10.20	2.51
Thorium C	4.78	6.02	−2.75
	8.62	8.77	—
	9.90	9.56	0.79
	11.70	10.59	1.82

* "Extrapolated ionization range" in air at 15°C and 760 mm.
** Rather more doubtful than the rest.

Let us first consider the results of Rosenblum's experiment. There are three possible explanations: the effect may be extra-nuclear, or, if intra-nuclear, then either primary or secondary. The magnitude of the energy differences and the sharpness of the satellite lines [17] almost certainly rules out the first explanation. If the effect were a primary intra-nuclear phenomenon then either we must assume that the disintegration of an atom of thorium C is "governed" by five partial disintegration constants, $\lambda\beta$, $\lambda\alpha_1$, $\cdots$, $\lambda\alpha_4$, corresponding to the β-particle mode and the four α particle modes, or we must regard thorium C as a mixed element each atom of which has in some way become "earmarked" to undergo one mode of disintegration rather than another. In the first case the Geiger-Nuttall, or some equivalent relation, would indicate that the frequencies of the different α-particle modes should be uniquely related to the respective energies, the group of highest energy being the most abundant—and, in the second case, on the basis of a similar argument, we should not expect the four α-particle groups to decay with the

[15] Bates and Rogers, Proc. Roy. Soc. **105A,** 97 (1924).

[16] Laurence, Proc. Roy. Soc. **122A,** 543 (1929).

[17] The writer has had the opportunity of examining a reproduction of an original photograph.

same period. Periods roughly in the ratios 2/3: 1:60: 550 might be anticipated. Neither conclusion will bear the test of experiment: the most intense group is not that of highest energy, whilst, if tested over an interval of 30 hours against the α-particles of thorium C′, the α-particle emission of thorium C exhibits a simple exponential decay.[18]

There remains only the possibility, on the basis of present ideas, that the effect is both nuclear and also secondary in character, that is, that the small energy differences found outside the atom are due, not to the fact that the different groups of α-particles have originated in fundamentally distinct quantum levels within the nucleus, but rather to some internal process which has changed the intrinsic energy of the disintegration particle in one nucleus and not in another. We are as yet almost entirely ignorant of the mechanism of γ-ray emission. There is the suggestion that it is probably concerned with α-particle transitions;[19] it may, in fact, be just such a process as that for which we are here in search. The second most prominent line in the γ-ray spectrum of thorium B corresponds to a quantum of energy 3.02×10^5 electron volts. Some of the atoms of thorium C which undergo α transformation must have emitted this γ-ray.[20] Now Rosenblum has shown that in a number of cases the α-particle is ejected with 3.0×10^5 electron volts energy less than the normal. In a similar manner we know that the β-particle disintegration of thorium C′′ follows the α-particle disintegration in all cases—and in a certain fraction of these the γ-ray of energy 0.408×10^5 electron volts is finally emitted. Now there are cases in which the thorium C α-particle has already carried away 0.4×10^5 electron volts excess energy. It may be that in each case we are here concerned merely with numerical coincidences, but it is tempting to believe that more than this is involved. Nevertheless, in respect of the third satellite line, corresponding to 4.7×10^5 electron volts energy defect, nothing as promising can be found; thorium *B* possesses no γ-rays of this energy, although there is, in fact, a weak line, (number 28) in the natural β-ray spectrum of thorium C′′[21] which corresponds to a γ-ray of energy 4.79×10^5 electron volts.

In the γ-ray spectrum of radium C—we are concerned now with the long range α-particles—there are prominent lines corresponding to quanta of energy 0.773 and 1.426 million electron volts, respectively, and individual lines in the natural β-ray spectrum[22] (E49 and E57) suggest weak γ-rays of energy 1.95 and 2.53 million electron volts, respectively. These four energy values are in good agreement with the first four entries in the last column of our table. If this agreement be significant it relates the most prominent group of

[18] Marsden and Barratt, Proc. Phys. Soc. Lond. **24,** 50 (1911).

[19] Kuhn, Zeits. f. Physik **44,** 32 (1927). Phil. Mag. **8,** 625 (1929). In the second paper additional evidence is advanced for assigning transition probabilities smaller than 10^{10} sec^{-1} to certain γ-ray processes.

[20] It is to be supposed that an equivalent fraction of those undergoing the β transformation will likewise have emitted the γ-ray in question.

[21] Black, Proc. Roy. Soc. **109A,** 166 (1925).

[22] Ellis, Proc. Camb. Phil. Soc. **22,** 369 (1924).

long range α-particles with the γ-ray of 1.426×10^6 electron volts energy,[23] and this appears f om measurements of internal (extra-nuclear) absorption to be the most intense in the spectrum.[24] Again, however, it may be that we are concerned merely with cases of numerical coincidence. In addition there might seem to be an *a priori* objection to the reality of the supposed connection, for, whilst the emission of a long range α-particle is an extremely rare occurrence, the emission of the more abundant γ-rays is relatively frequent. Yet the objection is not entirely cogent. Ordinarily it must be assumed that the γ-ray emission and the α-transition are consecutive processes, but, in a small fraction of the cases, when the latter event occurs very shortly after the preceding β-particle change, it may be possible that the total energy of the two processes should appear as kinetic energy associated with the α-particle.[25]

In the parallel case of the long range α-particles from thorium C difficulties at once appear. In the first place, since energy differences refer to energies in excess of the value characteristic of the α-particle disintegration of thorium C′, we ought logically to restrict our investigations to that branch of the series. Now there is no known γ-ray definitely attributable to thorium C itself, and the γ-radiation, if any, of thorium C′ must be very small in amount. There is, therefore, no material for comparisions such as we have previously made. Since, however, the chance of a very small time interval between β and α-particle changes is presumably much greater in the case thorium C→thorium C′ than in the corresponding case in the radium series, it may be that a very weak γ-radiation from thorium C would be sufficient to explain the effect. Finally, we should notice in passing that a moderately strong γ-ray, supposedly due to thorium C″, possesses exactly the energy (0.79×10^6 electron volts) representing the difference between the energies of the 9.9 cm and the normal 8.6 cm α-particle groups. Yet, for the characteristic energy of the most abundant of all the groups of long range α-particles—that of 11.7 cm range from thorium C—it is not possible to find any simple numerical relationship of the type that we have been discussing.

[23] Also it arbitrarily attributes the particles to an atom of atomic number 84. There is no definite experimental evidence on this point.

[24] It is now known from measurements of the excited spectra that the true intensity is, in fact, much smaller than this; cf. discussion on the structure of atomic nuclei, Proc. Roy. Soc. **123A,** 385 (1929).

[25] Cf. Nimmo and Feather, reference 13 p. 685.