Ionization Yield of Protons in Nitrogen and Argon*

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(Received May 20, 1957)

The total ionization yield of protons in nitrogen and argon gases was studied for energies in the range of a few to 250 kev. A large cylindrical ionization chamber was used for this study. The chamber was alternately operated as a proportional counter and an ionization chamber to measure the rate with which the protons entered the chamber and the total ionization produced by them, respectively. The results indicated a straight-line relation between the ionization yield and proton energy for energies above 75 kev in nitrogen and above 25 kev in argon. Straight-line plots of the data had slopes corresponding to 36.6±0.7 ev per ion pair for nitrogen and 26.5±0.5 ev per ion pair for argon. For argon the straight-line plot intercepted the energy axis at 1.4±0.9 kev, indicating a slight ionization defect.

INTRODUCTION

IONIZATION of gases by charged atomic particles has been studied both experimentally and theoretically for many years, especially in regard to the relation between the energy of the particle and the total ionization produced. A review of the experimental results up to 1944 has been given by Gray,[1] and a more recent summary may be found by Bethe and Ashkin.[2] Massey and Burhop[3] have discussed the experimental results for very low-energy ions. Most investigations of the ionization yield have been concerned with the light nuclear particles produced by natural radioactive sources or by induced nuclear reactions. Consequently, the energies of the particles were generally above 0.5 Mev. The need for further measurements in the energy region below 500 kev was apparent. It was the purpose of the work reported herein to develop the apparatus and a technique for the measurement of the ionization yield of charged atomic particles in gases for energies in the range of a few to 250 kev. Preliminary studies were made of the ionization yield of protons in nitrogen and a more complete and precise study was made of protons in argon.

Ionization is one of the fundamental energy-loss processes for charged particles and it is in the low-energy region that the competing process of elastic scattering becomes significant. The total ionization produced has often been used as a means of determining the energy of a charged particle. Accordingly, the most practical aspect of ionization studies is the relation between the total ionization and the energy of the incident particle. The quantity of the greatest practical importance is the average energy per ion pair W which is the ratio of the total energy E of the incident particle and I, the total number of ion pairs produced:

$$W=E/I.$$

A theoretically more significant quantity is the energy W' per ion pair for a small change in particle energy:

$$W'=dE/dI.$$

At sufficiently high energies, W' approaches a constant value. In this energy region the energy of the incident particle may be expressed by the simple relation

$$E=E_0+W'I, \quad (v\gg v_0),$$

where v_0 is the velocity of the electron in the first Bohr orbit of the hydrogen atom, $e^2/\hbar$. The quantity E_0 is sometimes referred to as an ionization defect[4,5] and is a function of both the gas and the type of incident particle.

APPARATUS AND PROCEDURE

In order to investigate the low-velocity region over a continuous energy range, it was necessary to resort to an ion accelerator as a source of charged particles. The ion beams studied were produced by the kevatron, a 300-kv linear dc accelerator. The desired ion component of the kevatron beam was selected by magnetic analysis. The energy of the ion component was determined by means of electrostatic deflection. The resolving power of the electrostatic analyzer was adjusted to approximately 200.

A schematic drawing of the ionization chamber is shown in Fig. 1. The primary ions were admitted into the ionization chamber by means of an open window. Differential pumping was used between the ionization chamber and the high vacuum system of the electrostatic analyzer. This permitted the chamber to be operated at pressures up to 10 mm Hg. A liquid nitrogen cold baffle was installed in the differential pumping section to reduce the concentration of pump oil vapors in the vicinity of the window.

The chamber was cylindrical with a diameter of 10 inches and an over-all length of 41 inches. The stainless-

* Work was performed in the Ames Laboratory of the U. S. Atomic Energy Commission.
† Present address: University of Virginia, Charlottesville, Virginia.
[1] L. H. Gray, Proc. Cambridge Phil. Soc. **40**, 72 (1944).
[2] H. A. Bethe and J. Ashkin, in *Experimental Nuclear Physics*, edited by E. Segrè (John Wiley and Sons, Inc., New York, 1953), Vol. 1, p. 166.
[3] H. S. W. Massey and E. H. S. Burhop, *Electronic and Ionic Impact Phenomena* (Oxford University Press, Oxford, 1952).
[4] J. K. Knipp and R. C. Ling, Phys. Rev. **82**, 30 (1951).
[5] Knipp, Leachman, and Ling, Phys. Rev. **80**, 478 (1950).

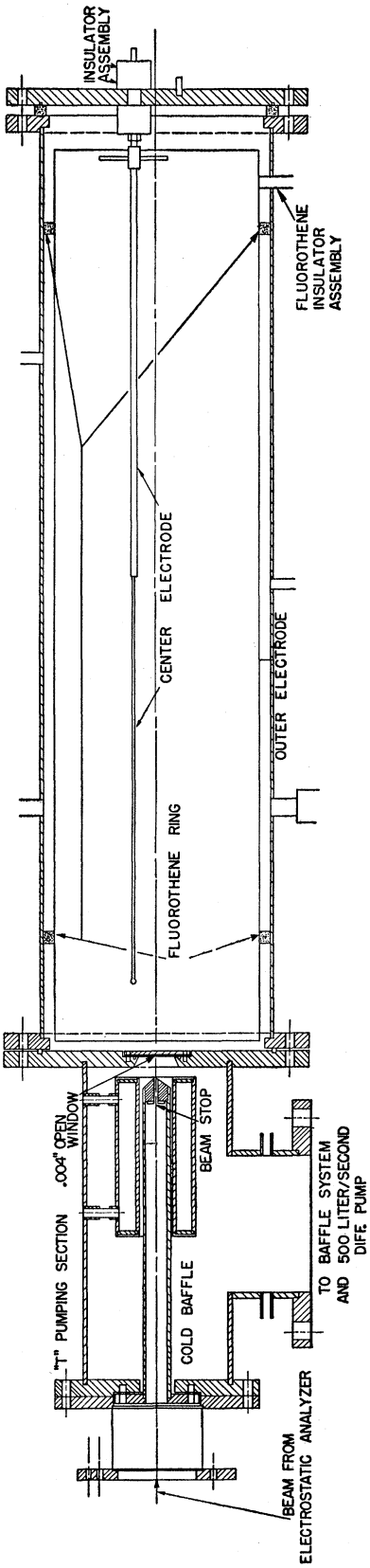

FIG. 1. Continuous-flow current ionization chamber.

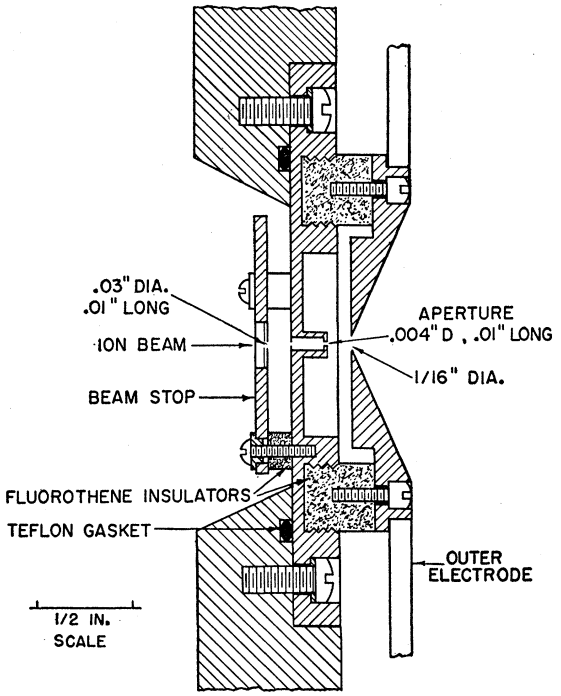

FIG. 2. Schematic diagram of the aperture assembly.

steel outer electrode was 9 inches in diameter and 40 inches long, being insulated from the chamber walls by fluorothene rings. An electrical connection was made to the outer electrode by way of a vacuum tight fluorothene feed-through insulator in the chamber wall. The center electrode was made of stainless steel, 0.125 inch in diameter with a 0.25-inch diameter sphere on the end. It was supported from the end plate by means of a fluorothene feed-through insulator. This electrode was located one inch off the axis of the chamber so it would not be directly in the path of the ion beam. The other end of the center electrode was 2.5 inches from the end of the outer electrode and was supported from the top of the outer electrode by means of a silk thread.

The open window consisted of an aperture 0.004 inch in diameter. The aperture assembly is shown in Fig. 2. The end of the outer electrode was about 0.125 inch from the aperture plate and insulated from it.

The gas from the supply tanks passed through a dry-ice cold-trap and was admitted into the chamber by means of a thermal valve. In the case of argon, the gas entered the chamber through a hot calcium purifier operated at 300 to 350 degrees C. An automatic pressure regulating device[6] was developed to maintain a given pressure in the chamber over the long periods required for the measurements.

The negative ions produced in the chamber were collected on the outer electrode by operating the center electrode at a negative potential. This ionization was measured as a current with a vibrating-reed electrome-

[6] Lowry, Osher, and Miller, Rev. Sci. Instr. **27**, 309 (1956).

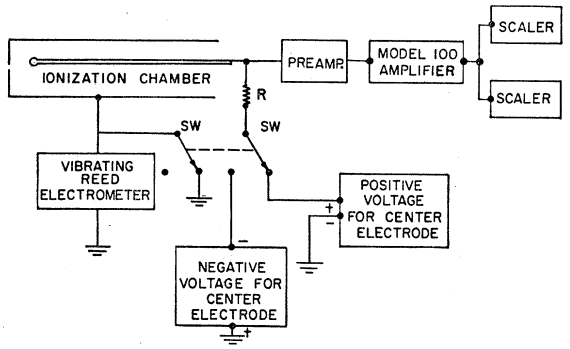

FIG. 3. Block diagram of apparatus for ionization-yield measurements.

ter. The outer electrode was made slightly positive with respect to the chamber walls by inserting a small battery between the input of the electrometer and the outer electrode. In this way the ionization between the outer electrode and the aperture was collected and included in the current measurements. The ionization currents were generally in the range of 10^{-13} to 10^{-12} ampere. The input time constant for the electrometer when connected to the outer electrode was about 70 seconds.

The rate at which the primary ions entered the chamber was determined by alternately operating the chamber as a proportional counter. In this way each primary particle entering the chamber was detected and counted. The pulses which originated in the chamber were amplified with a minimal noise preamplifier[7,8] followed by a Los Alamos Model 100 amplifier[9] with the input adapted to delay-line clipping. The total gain of the amplifiers was approximately 10^5. The pulses from the amplifiers were recorded by two scalers operated in parallel. Their resolving times were about one microsecond. The total resolving times for counting were 7 to 10 microseconds for nitrogen gas and 15 to 20 microseconds for argon. Counting rates of 1000 to 3000 counts/sec were used for nitrogen and 500 to 2000 counts/sec for argon.

Figure 3 is a block diagram of the measuring apparatus. In the sequence of measurement, the voltage on the center electrode was first adjusted to a negative value which collected all of the ionization but gave no multiplication. The outer electrode was then ungrounded and the electrometer was allowed to record for 400 seconds. The outer electrode was again grounded and the center electrode voltage was adjusted to a positive value which operated the chamber as a proportional counter. The primary ions were counted for 100 seconds.

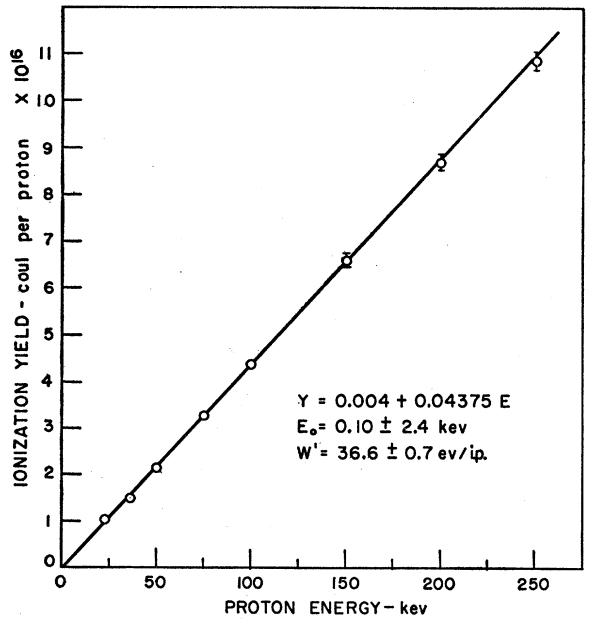

FIG. 4. Ionization yield of protons in nitrogen.

TABLE I. Ionization yield of protons in nitrogen.

Proton energy kev	$10^{16} \times Y$ coul per proton	W ev/ion pair	Estimated uncertainty percent
23	1.016	36.3	3.9
36	1.494	38.6	2.7
50	2.137	37.5	3.0
75	3.270	36.7	1.8
100	4.378	36.6	1.8
150	6.59	36.5	1.9
200	8.70	36.8	1.7
250	10.75	36.9	1.8

The ionization yield Y (coulombs per primary ion) was computed by the equation

$$Y = \text{Ionization current/count rate},$$

and the average energy per ion pair

$$W = E \times 1.6 \times 10^{-19}/Y \text{(ev/ion pair)},$$

where E was the energy of the primary particles in electron volts.

One disadvantage of the above technique was that the ionization current and the primary ion rate were not measured simultaneously. Hence, any variation in the incident beam current during the period of counting resulted in an error in the ionization yield determination. To reduce the effect of fluctuations in the primary beam current, the sequence of measurements summarized above was repeated 10 to 25 times and averaged for each ionization yield determination.

ERROR AND ADJUSTMENT OF DATA

The uncertainties in the measurements were estimated as follows: primary ion energy, $\pm 1.0\%$; ioni-

[7] K. Enslein and B. Brainerd, Rev. Sci. Instr. 24, 916 (1953).
[8] R. L. Chase, Rev. Sci. Instr. 25, 1219 (1954).
[9] W. C. Elmore and M. Sands, *Electronics* (McGraw-Hill Book Company, Inc., New York, 1949).

TABLE II. Ionization yield of 50-kev protons for various outer electrode potentials.

Chamber pressure mm Hg	Outer electrode potential volts (+)	Center electrode potential volts (−)	$10^{16}\times Y$ coul per proton	W ev/ion pair
4.46	0	137	2.784	28.77±0.37
4.46	1.5	137	2.869	27.92±0.28
4.46	3.0	137	2.869	27.92±0.36
6.50	0	137	2.724	29.41±0.39
6.50	1.5	137	2.922	27.41±0.29
6.50	3.0	223	2.924	27.39±0.37
8.79	1.5	223	2.865	27.96±0.26
8.79	3.0	223	2.876	27.85±0.30
8.79	4.5	223	2.892	27.70±0.27

zation current, ±1.0%; particle counting, ±0.5%. It was necessary to make correction for the energy lost by the primary ions in the aperture assembly. This loss was of the order of 0.015 kev per mm Hg of chamber pressure. Uncertainties in the counting rate included the correction for resolving time of counting. Except for the errors in the absolute calibration factors, the over-all uncertainties in the final results were reduced by averaging several independent determinations. The over-all uncertainties in the final values for the ionization yield of protons in argon ranged from ±1.5 to ±2.0%.

RESULTS

Nitrogen

A preliminary investigation of the ionization yield of protons in nitrogen was made to check the feasibility of the experimental method and the performance of the apparatus. Nitrogen was used so that gas purity would not be too important. The chamber was not completely vacuum tight and consequently the results given for nitrogen in Table I are actually for a gas mixture with a composition somewhere between pure nitrogen and air. The values of the ionization yield given in the second column are plotted in Fig. 4. The straight line represents a least-squares plot of the points at 75, 100, 150, and 200 kev. The slope of this line corresponds to 36.6±0.7 ev per ion pair. Systematic errors related to gas composition, saturation, and aperture losses may have been much larger than the random uncertainties specified for this value of W' and, consequently, the total uncertainty might well be increased to as much as ±10%.

TABLE III. Ionization yield of protons in argon.

Proton energy kev	$10^{16}\times$ionization yield coul/proton	Average energy per ion pair ev
25	1.455±0.026	27.52±0.49
50	2.879±0.044	27.82±0.43
100	5.957±0.114	26.89±0.51
150	8.923±0.145	26.93±0.44
200	12.04 ±0.18	26.61±0.40
250	15.26 ±0.25	26.25±0.43

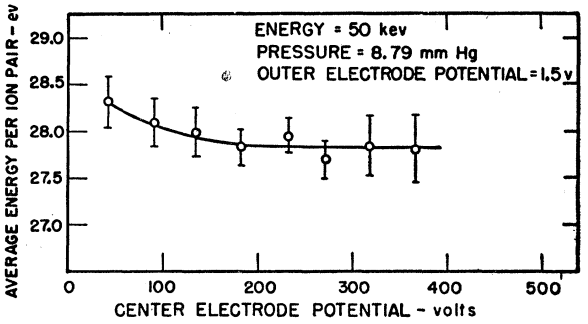

FIG. 5. Saturation curve for 50-kev protons in argon.

Argon

A more precise study was made of the ionization yield of protons in argon. The characteristics of the chamber were studied in detail. For a given energy the ionization yield was measured as a function of the center electrode potential to ascertain that all ionization produced in the chamber was being collected. These saturation studies were made for 50- and 200-kev protons. A typical saturation curve is displayed in Fig. 5.

The collection of the ionization in the gap between the aperture and the outer electrode was also investigated. This study was made at low energies where the ionization produced in this gap was a significant portion of the total ionization. The ionization yield of 50-kev protons was measured for several different values of outer electrode potential. The results for three different pressures are shown in column four of Table II.

It was also necessary to establish that no primary ions were lost to the electrodes. This was accomplished by measuring the ionization yield as a function of the pressure. The results of these investigations are illustrated by the curves in Fig. 6. This as well as the other results will be discussed in the next section.

The final values for the ionization yield of protons in argon are given in Table III and generally are the weighted average of several determinations. For each energy studied, both the ionization yield and average

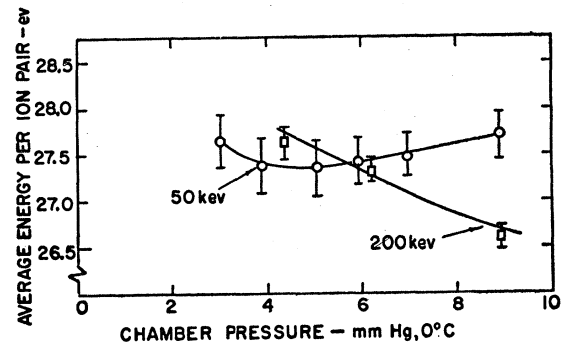

FIG. 6. Average energy per ion pair for 50- and 200-kev protons in argon as a function of chamber pressure.

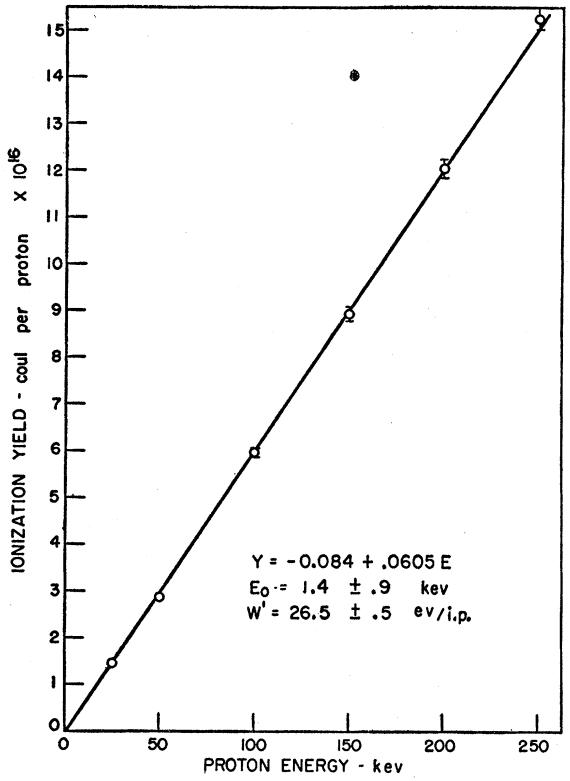

FIG. 7. Ionization yield of protons in argon.

energy per ion pair are given. The ionization yields are plotted in Fig. 7. The straight line represents a weighted least-squares plot of the data and has a slope corresponding to 26.5±0.5 ev/ion pair. The least-squares plot intercepts the energy axis at 1.4±0.9 kev, indicating a slight ionization defect for protons.

DISCUSSION

The chamber was filled through the hot calcium purifier from a tank of argon supplied by the Matheson Company and reported to have a purity of 99.9%. The rise times of the proportional counter pulses were much longer for argon than for nitrogen. An appreciable nitrogen or carbon dioxide contamination in the argon gas would have noticeably decreased these rise times. However, upon filling the chamber with new gas, there was no detectable change in the rise time of the pulses, the multiplication, or ionization yield. On the basis of these observations it was concluded that the purity of the argon gas was at least 99.0%.

The cylindrical geometry and highly nonuniform electrical fields in the chamber made it difficult to saturate. The degree of difficulty depended upon the spatial distribution of the ionization. The spatial distribution of the ionization produced in a gas by a collimated beam of heavy ions has been studied by Evans, Stier, and Barnett,[10] and by Cook, Jones, and Jorgensen.[11] The results of the former investigation indicated that the attenuation of the beam of ions was approximately exponential in the axial direction and approximately Gaussian in the lateral direction. The lateral spread of the ionization increased with energy, causing a greater portion of the ionization to be produced near the outer electrode. In this extended region the fields were weak and, consequently, the region was difficult to saturate. Thus, in the present experiment it was more difficult to saturate the chamber for 200-kev protons than for 50 kev.

The data in Table II indicated that an outer electrode potential of 1.5 to 3.0 volts was adequate to collect all of the ionization produced in the gap between the aperture and outer electrode. It was assumed, therefore, that the correction for loss in the aperture itself was the only one necessary for the energy of the protons. This correction was at most only 0.13 kev.

Some of the primary ions were lost to the center electrodes and at sufficiently low pressures some were lost to the outer electrode. This loss to the electrodes decreased as the pressure was increased, which accounted for the negative slope of the curves in Fig. 6. However, at the maximum permissible operating pressure of 10 mm Hg, the average energy per ion pair for 200-kev ions continued to decrease as indicated by Fig. 6 and, hence, there may still have been significant energy loss to the outer electrode. The primary energy lost to the center electrode was approximated by means of an empirical equation[10] for the ionization distribution in the chamber. The constants of the equations were evaluated from the ionization density contours for protons in argon published by Cook *et al.*[11] It was possible to establish 0.8% of the total energy as an upper limit for the energy lost to the center electrode by 250-kev protons in argon at a pressure of 9 mm Hg. The energy loss was less at lower energies.

The pressure curve in Fig. 6 for 50-kev protons seems to indicate the presence of a second effect whereby the apparent ionization yield decreased as the pressure was increased. The variations were not outside of the uncertainties in the data but were, in general, reproducible. It may be possible for such a pressure anomaly to exist due to emission of electrons from the electrodes. Generally, it is expected that the emission from the center electrode of a cylindrical ionization chamber will be less than from the outer electrode because of the difference in area. However, in the present experiment, the density of excited and ionized atoms was much greater in the vicinity of the center electrode. As the pressure was decreased, the lateral dimension of the ionization envelope expanded and surrounded the center electrode with a greater density of excited and ionized atoms. This could have increased the emission of secondary electrons which would have been

[10] Evans, Stier, and Barnett, Phys. Rev. **90**, 825 (1953).

[11] Cook, Jones, and Jorgensen, Atomic Energy Commission Report AECU-2467, 1952 (unpublished).

detected as an increase in the ionization yield. Such an effect might account for some of the differences in measurements of ionization yields by fast (electron collection) and by slow (total ion collection) chambers.

Since the velocity of a 200-kev proton is about the same as that of an 800-kev alpha particle, it is to be expected that the average energy per ion pair should be the same for the two. Investigations[12,13] of the ionization yield of alpha particles in argon for energies above 1 Mev have indicated the existence of a straight-line relation between the ionization yield and the energy. The slope of this linear function corresponded to 26.4 ev/ion pair. The present data for protons in argon seem to fit a straight line and the value obtained for W', 26.5 ev/ion pair, is in good agreement with the above results for alpha particles. The value of 1.4 kev for E_0 is consistent with Jesse's[12] prediction that the ionization defect, if present, should be small.

It can be seen in Table III that the average energy per ion pair increases as the energy decreases. This variation can be interpreted as indicating that the energy-loss processes competing with ionization are becoming more significant at lower energies.

[12] Jesse, Forstat, and Sadauskis, Phys. Rev. **77**, 782 (1950).
[13] W. P. Jesse and J. Sadauskis, Phys. Rev. **90**, 1120 (1953).

PHYSICAL REVIEW VOLUME 109, NUMBER 3 FEBRUARY 1, 1958

Relaxation Effects for Coupled Nuclear Spins*†

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(Received May 13, 1957)

Nuclear relaxation effects in molecules containing several coupled spins are treated by a generalization of the method of Wangsness and Bloch. Variations in line shape and structure for two coupled spin-$\frac{1}{2}$ nuclei as various parameters are changed from one extreme to another are derived and plotted graphically. The calculation is continued to obtain an expression for the line shapes in the double-resonance experiment of Bloom and Shoolery. It is next shown that under certain circumstances the concept of "semimacroscopic magnetic moments" enables one to write the quantum-mechanical equations of motion in the form of coupled Bloch phenomological equations. In addition, a discussion of internal relaxation and the Overhauser effect is given.

I. INTRODUCTION

WANGSNESS and Bloch[1] have derived an equation which gives the dynamical behavior of an ensemble of nuclear spins that interact with their molecular surroundings but not with each other. The essential feature of this calculation is that the molecular surroundings are assigned the properties of an infinite heat bath. The action of the molecular surroundings on the spin system influences the behavior of the spin system, but the action of the spin system on the molecular surroundings is assumed to leave the state of the molecular surroundings unchanged. The immediate molecular surroundings may be considered to be in thermal contact with an infinite heat reservoir at some temperature T. Any change in the state of the molecular surroundings brought about by contact with the spin system will, in a short period of time, be canceled by a much stronger coupling to the heat reservoir.[2] The molecular surroundings will be in thermal equilibrium, and thus in a Boltzmann distribution, at this temperature T.[2]

As is well known, line structure may arise from spin-spin interactions between nuclei of a polyatomic molecule in a liquid.[3–7] The form of this interaction was proposed independently by Gutowsky, McCall, and Slichter[5] and by Hahn and Maxwell.[6] The mechanism giving rise to the interaction was pointed out by Ramsey and Purcell[8] and a more detailed treatment has been given by Ramsey.[9] The methods of Wangsness and Bloch will now be used to obtain equations from which one may determine the dynamical behavior of an ensemble formed of systems of interacting spins.[10]

* This research was done while the author held a National Science Foundation Predoctoral Fellowship.
† Based on a dissertation submitted to Stanford University in partial fulfillment of the requirements for the Ph.D. degree in physics (1956).
‡ Now at Varian Associates, Palo Alto, California.
[1] R. K. Wangsness and F. Bloch, Phys. Rev. **89**, 728 (1953).
[2] For additional comments on this point, see U. Fano [Phys. Rev. **96**, 869 (1954)].
[3] W. G. Proctor and F. C. Yu, Phys. Rev. **81**, 20 (1951).
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[5] Gutowsky, McCall, and Slichter, Phys. Rev. **84**, 589 (1951).
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[8] N. F. Ramsey and E. M. Purcell, Phys. Rev. **85**, 143 (1952).
[9] N. F. Ramsey, Phys. Rev. **91**, 303 (1953).
[10] The work reported here is a generalization of the original Wangsness-Bloch theory. A somewhat different generalization has been developed by F. Bloch [Phys. Rev. **102**, 104 (1956)]. The relation between these generalizations is as follows: (a) The work presented here is concerned with spin systems in which the spin-spin interactions may be considered small compared to differences between unperturbed energy levels. In the treatment of Bloch, this is not necessary. (b) The relaxation terms are here calculated through the use of correlation functions. In this approach the conditions for validity of the treatment appear in somewhat different, although largely equivalent, form. (c) It is shown that