On the Recoil of the Nucleus in Beta-Decay of Kr⁸⁸.

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The maximum energy and the average energy of the recoil atoms from Kr⁸⁸, which emits β - and probably also γ -rays, have been determined to 51.5 ± 2 ev and 29 ± 1 ev, respectively. The maximum recoil energy agrees closely with the maximum energy 2.4 Mev for the β -particles.

I. INTRODUCTION

 $\mathbf{S}_{\text{energy in } \beta\text{-transformations have been}}^{\text{EVERAL}}$ made.¹ When the radioactive substance is deposited on a surface, recoil efficiencies from 5 to 50 percent have been found; this shows that the emission of the recoil atoms is strongly influenced by surface effects. By the use of a radioactive gas surface effects are eliminated but, if the active atoms form part of polyatomic molecules, the recoil energy may be shared with the remaining atoms of the molecule with the result that the energy distribution is changed. Consequently, the active element must be a monoatomic gas at a pressure which is so low that the mean free path is large compared to the dimensions of the apparatus. Then the recoil atoms must necessarily be charged before they collide with the walls of the apparatus, and the recoil energy can be determined by a retarding electric field. If the daughter substance formed in the transformation is radioactive, the number of recoil atoms passing through the retarding field can be simply determined. These conditions, a radioactive monatomic gas with an active daughter substance, both with a suitable lifetime, are fulfilled only in very few cases; in fact, only two or three isotopes of krypton and xenon, formed by fission of uranium, can be used. Glasoe and Steigman² have found that the active deposit from the inert, active gases from fission of uranium consists, to a very high degree of purity, of Rb⁸⁸ if a sample of uranium is left for three hours after the irradiation with neutrons before the inert gases formed by fission are collected. The recoil phenomena for Kr⁸⁸ can thus be studied by using Rb⁸⁸ as a detector without any disturbances from the numerous other isotopes of Kr and Xe which are formed by the fission process. A few experiments have been carried out with Kr⁸⁹ and with Xe¹³⁸. In both cases the active deposits of Rb⁸⁹ and Cs¹³⁸, respectively, can only be obtained mixed with Rb⁸⁸ so that the interpretation of the results is more complicated and no definite results have yet been obtained.

II. THE MAXIMUM RECOIL ENERGY

The apparatus is shown in Fig. 1. The main part was a rectangular box B_1 made of sheet copper, one side of the box being closed by a brass wire gauze. The box was divided into a number of smaller partitions by means of crosswalls, the purpose of which was to limit the free path of the recoil atoms and thus to reduce the influence of the residual gases in the apparatus. The box with the wire gauze, electrically insulated, was placed inside a second box B_2 , also made from sheet copper. B_2 was kept at a suitable potential with respect to B_1 . The active deposit from the inert gases was collected on aluminium foils F_1 and F_2 attached to the inner side of B_2 . For measurements of the activity of the deposit F_1 and F_2 were removed from the apparatus and wrapped around a cylindrical counter. The difference between these activities was caused by recoil atoms starting from the interior of B_1 and having sufficient energy to surmount the potential difference between B_1 and B_2 . When the potential difference was varied in separate experiments, some indications of the energy distribution could be obtained; the limitations of

¹ K. Donat and K. Phillip, Zeits. f. Physik **45**, 512 (1927), A. I. Leipunski, Proc. Camb. Phil. Soc. **32**, 301 (1936); H. R. Crane and J. Halpern, Phys. Rev. **56**, 232 (1939); L. W. Alvarez, A. C. Helmholtz, and B. T. Wright, Phys. Rev. **60**, 160 (1941); James S. Allen, Phys. Rev. **61**, 692 (1942); and B. T. Wright, **71**, 839 (1947).

² G. N. Glasoe and J. Steigman, Phys. Rev. 58, 1 (1940).



FIG. 1. Diagram of apparatus.

the method will be discussed below. For the comparison of different experiments the amount of inert gas present in the apparatus, which varied somewhat from one experiment to another, had to be determined in some arbitrary unit. For this purpose the arrangement shown in the lower part of Fig. 1 was used. A circular brass disk D was placed in the bottom wall of a brass cylinder, electrically insulated, and kept at a potential of -220 volts relative to the cylinder. The activity of the disk, determined under standard conditions, was used as a measure of the total amount of Kr88 present. The constancy of the counters used was checked before and after each measurement by irradiation with a γ -ray source placed in a standard position.

The metal parts of the apparatus were placed in a Pyrex tube with a brass flange and lid. The uranium used for the production of the radioactive Kr and Xe was placed in a glass bulb which was connected to the main part of the apparatus by a long glass tube, so that the uranium could be brought in between the coils of the cyclotron magnet. A stopcock (H) and a U-tube were placed in the glass tube. The uranium was in an emanating form obtained by precipitating a mixture of UO₂(NO₃)₂ and FeCl₃ with surplus ammonia. In the state in which the uranium was used in the experiments, it gave off large amounts of water vapor and other gases when placed under vacuum. When the U-tube was cooled by liquid air most of these gases were condensed, the residual pressure being from 10^{-4} to 10^{-3} mm of mercury.

The procedure during an experiment was as follows. After the apparatus had been evacuated, the stopcock H was closed, and the uranium irradiated by slow neutrons for about half an hour. Three hours after the irradiation, the stopcock H was opened, the U-tube being immersed in liquid air. The active deposit was collected for about 30 minutes, and the activity of the foils F_1 and F_2 and of the disk D was then measured.

In Fig. 2 the ratio I_2/I_1 of the activities of F_2 and F_1 is plotted against the retarding voltage V. It is seen that at 51.5 volts and upwards I_2 and I_1 are equal, i.e., no recoil nuclei can escape from inside B_1 so that they are able to reach the foil F_2 . From this it may be concluded that the maximum recoil energy $E_R(\max)$ is equal to 51.5 ± 2 ev.

In Fig. 3 is shown the difference $I_2 - I_1$ as a function of V, giving an indication of the energy distribution of the recoil atoms. However, the fact that the recoil atoms pass through the retarding field in all directions has a considerable influence on the results, and the corrections which should be applied to the curve on Fig. 3 to get the actual energy distribution can hardly be calculated unambiguously. The retarding field between the wire gauze and the collecting foil may to a first approximation be considered as homogeneous. An ion starting from the interior of B_1 with kinetic energy E in a direction making an angle θ with the direction of the retarding field will, during its passage through the field, move in a parabolic orbit and will reach the collecting foil only if $E \cdot \cos^2 \theta \ge V$. If the ions starting from the interior of the box all had the same energy, the measurements would show an energy distribution from E=0 to the actual energy of the particles. The field at the edges of B_1 and around the wires of the gauze is too complicated for an exact calculation to be carried out, but it is clear that a particle which starts in B_1 and which does not follow a line of force in the field, will not reach F_2 when V is equal to the energy E of the particle but when V is somewhat lower. The resolving power as regards energy

distribution is thus rather poor, with the result that the number of recoil atoms with small energy is overstimated; the maximum value of the recoil energy is not affected by the geometry of the apparatus.

III. THE AVERAGE RECOIL ENERGY

More definite information about the energy distribution can be obtained from a determination of the average value of the energy. As will be shown now, the average energy can be determined when the active deposit from the gas is collected in a homogeneous electric field.

The apparatus used is shown in Fig. 4. It consists of a plane-parallel condenser placed in a vacuum-tight metal box with a lid tightened with a rubber ring (shown in black in the figure). One of the plates was insulated from the box and placed at a potential of + V volts with respect to the box. This plate and the bottom of the box formed the condenser. The central part of the condenser plates where the field is homogeneous could be removed and placed under counters. The ratio of the intensities on these two plates N_1/N_2 gives a determination of the average recoil energy when V fulfils the condition

$$V > E_R(\max). \tag{1}$$

The number of disintegrations which take place within limits dx (see Fig. 5) and with a recoil momentum in a direction within limits $d\theta$ where θ is the angle between the recoil momentum and the direction perpendicular to the plates and with a recoil energy within limits dE_R is given by

$$N \cdot dx/a \cdot \frac{1}{2} \sin\theta d\theta \cdot P(E_R) dE_R \tag{2}$$

where N is the total number of disintegrations, a is the distance between the plates (equal to 1 cm) and $P(E_R)dE_R$ is the probability that E_R lies within limits dE_R .



FIG. 2. Ratio of activities I_2/I_1 , of the plates F_2 and F_1 as a function of the retarding voltage.



FIG. 3. Difference of the activities $I_2 - I_1$ as a function of the retarding voltage.

The recoil particles move in parabolic orbits and only those particles for which

$$E_R \cdot \cos^2 \theta \ge V x/a \text{ and } 0 \le \theta < \pi/2$$
 (3)

can reach plate 1. The number of recoil particles reaching plate 1 can be determined by an integration of (2) with the limits of integration determined by (1) and (3). One finds

$$N_{1} = N/(2a) \cdot \int_{0}^{E_{R}(\max)} \int_{0}^{\pi/2} \int_{0}^{E_{R}a \cdot \cos^{2}\theta/V} dx$$
$$\cdot \sin\theta d\theta \cdot P(E_{R}) dE_{R} \quad (4)$$

$$N\langle E_R \rangle/6V$$
 (5)

where $\langle E_R \rangle$ is the average recoil energy.

All the other particles go to plate 2 and we have

$$N_2 = N(1 - \langle E_R \rangle / (6V)). \tag{6}$$

From (5) and (6) one gets

$$\langle E_R \rangle = 6 V \cdot (N_1/N_2) / (1 + N_1/N_2).$$
 (7)

Thus it is seen that the ratio N_1/N_2 gives a determination of $\langle E_R \rangle$. The experimental results are given in Table I.

It is seen that the value found for $\langle E_R \rangle$ does not depend on the retarding potential V in the range 52 to 220 volts, and further, that at low pressures the result is independent of the pressure; there is, however, a definite effect at much higher pressures as shown in the table. From the results which are not affected by the pressure one finds $\langle E_R \rangle = 29 \pm 1$ ev.

The pressure dependence is in qualitative agreement with the results from experiments with potassium ions,³ where a mean free path of about 20 to 30 cm at a pressure of 10^{-3} mm of mercury has been found. Thus the mean free

³ Otto Schmidt, Ann. d. Physik 21, 241 (1934).



FIG. 4. Diagram of apparatus used to measure average recoil energy.

path has been large compared to the dimensions of the apparatus in all the low pressure measurements, and collisions between the recoil atoms and gas molecules at the low pressures used are so rare that the result is not affected within the limits of error given.

As a common result from all the experiments it was further found that neutral recoil atoms are not present to any measureable extent. Neutral recoil atoms might be produced if the recoil atoms could be reflected from the walls, the reflected atoms almost certainly being neutral. The presence of neutral Rb atoms would involve a dependence of the measured average energy on the retarding potential, but as mentioned above no such dependence was observed.

Finally it may be mentioned that it is possible to find the average momentum by comparing the activity on the plates of a condensor placed in a homogeneous magnetic field which is parallel to the plane of the condensor with the activity on the plates of a similar condensor without magnetic field. However, no such measurements have been performed.

IV. THE MAXIMUM β-ENERGY

The maximum β -energy has been determined by absorption. Fifteen hours after an irradiation the only inert gas in the fission products which has a β -energy higher than 1 Mev is Kr⁸⁸. Thus, the absorption curve for absorbers thicker than 0.34 mg/cm² of aluminum obtained with a mixture of the inert gases belongs to Kr⁸⁸. The

 TABLE I. Experimentally determined values of the average recoil energy.

V volts	Pressure mm Hg	$\langle E_R angle angle { m ev}$
52	5.10-4	28±2
52	3.10-1	30 ± 1
52	0.1	18 ± 1
65	10-8	29 ± 1
220	3.10-3	28 ± 2



influence of the very high energy β -particles from Rb⁸⁸ has been eliminated by measuring for each absorber the growth of activity from pure gas to equilibrium with Rb⁸⁸. When it is known that this growth is due to a 17.8 minutes half-life and that the equilibrium state decays with a half-life of 2.65 hours it is possible to estimate the activity from the pure gas with fairly great accuracy.

The absorption curve has been compared with the absorption curves for RaE, UX₂, RaC, and Cl³⁸ for absorbers thicker than 0.34 mg/cm² of aluminum and the maximum energy was found equal to 2.4 Mev in agreement with the measurements of Weil.⁴

V. Y-RAYS

The determination of a possible γ -radiation from Kr⁸⁸ is complicated by the presence of Kr⁸³, Kr⁸⁵, Kr⁸⁷, Xe¹³⁵, and Rb⁸⁸ all of which are known to emit γ -rays. The decay of the γ -radiation from the mixture of the inert gases in equilibrium with Rb⁸⁸ can easily be determined, but an analysis of such a complex decay is hardly possible. Unfortunately, Kr⁸⁸ can only be obtained by fission and the only possible method for obtaining Kr⁸⁸ in a pure state is to apply a mass separation to the gas mixture obtained by fission. With the neutron source at our disposal the intensity is too small for this purpose, and for the present this important question has to be left open.

VI. DISCUSSION

The maximum β -energy corresponding to the maximum recoil energy of 51.5 ± 2 ev is for Kr⁸⁸ 2.43 \pm 0.06 Mev in good agreement with the value obtained from absorption measurements. From this agreement is may be concluded that transformations with the energy 2.43 Mev lead to the ground state of Rb⁸⁸.

If all the disintegration processes led to the

⁴ G. L. Weil, Phys. Rev. 60, 167 (1941).

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ground state of Rb⁸⁸, the value for the average energy might be used to distinguish between various assumptions regarding the angular distribution of the electron and the neutrino. Should, however, β -decay to some intermediate state, followed by γ -radiation, occur, a correction must be applied to the measured value of the average energy to obtain the value corresponding to a simple β -spectrum. This correction may change the average recoil energy to

such an extent that with our present lack of knowledge about the level scheme of the transformation, such comparisons would be inconclusive.

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Relaxation Effects in Nuclear Magnetic Resonance Absorption*

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The exchange of energy between a system of nuclear spins immersed in a strong magnetic field, and the heat reservoir consisting of the other degrees of freedom (the "lattice") of the substance containing the magnetic nuclei, serves to bring the spin system into equilibrium at a finite temperature. In this condition the system can absorb energy from an applied radiofrequency field. With the absorption of energy, however, the spin temperature tends to rise and the rate of absorption to decrease. Through this "saturation" effect, and in some cases by a more direct method, the spin-lattice relaxation time T_1 can be measured. The interaction among the magnetic nuclei, with which a characteristic time T_2' is associated, contributes to the width of the absorption line. Both interactions have been studied in a variety of substances, but with the emphasis on liquids containing hydrogen.

Magnetic resonance absorption is observed by means of a radiofrequency bridge; the magnetic field at the sample is modulated at a low frequency. A detailed analysis of the method by which T_1 is derived from saturation experiments is given. Relaxation times observed range from 10^{-4} to 10^2 seconds. In liquids T_1 ordinarily decreases with increasing viscosity, in some cases reaching a minimum value after which it increases with further increase in viscosity. The line width meanwhile increases monotonically from an extremely small value toward a value determined by the spin-spin interaction in the rigid lattice.

I. INTRODUCTION

TN nuclear magnetic resonance absorption, energy is transferred from a radiofrequency

The effect of paramagnetic ions in solution upon the proton relaxation time and line width has been investigated. The relaxation time and line width in ice have been measured at various temperatures.

The results can be explained by a theory which takes into account the effect of the thermal motion of the magnetic nuclei upon the spin-spin interaction. The local magnetic field produced at one nucleus by neighboring magnetic nuclei, or even by electronic magnetic moments of paramagnetic ions, is spread out into a spectrum extending to frequencies of the order of $1/\tau_c$, where τ_c is a correlation time associated with the local Brownian motion and closely related to the characteristic time which occurs in Debye's theory of polar liquids. If the nuclear Larmor frequency ω is much less than $1/\tau_c$, the perturbations caused by the local field nearly average out, T_1 is inversely proportional to τ_c , and the width of the resonance line, in frequency, is about $1/T_1$. A similar situation is found in hydrogen gas where τ_c is the time between collisions. In very viscous liquids and in some solids where $\omega \tau_c > 1$, a quite different behavior is predicted, and observed. Values of τ_e for ice, inferred from nuclear relaxation measurements, correlate well with dielectric dispersion data.

Formulas useful in estimating the detectability of magnetic resonance absorption in various cases are derived in the appendix.

circuit to a system of nuclear spins immersed in a magnetic field, H_0 , as a result of transitions among the energy levels of the spin system. For each of N non-interacting spins, characterized in the usual way by $I\hbar$ and μ , the maximum z components of angular momentum and magnetic moment, respectively, there are 2I+1

^{*} A brief account of this work has appeared in Nature

 <sup>160, 475 (1947).
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