ON THE ISOMERISM OF Zn^{69}.

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ABSTRACT.

The isomers of radioactive Zn^{69} produced by neutron and deuteron bombardment has been studied. An accurate determination of the 13·8 hr. γ-ray energy has been made by using recoil electrons and the value found is 450 kev. Internal conversion of the γ-ray has been observed and the K, X-rays from zinc has been used to estimate the internal conversion coefficient at ·06. The relative capture cross-sections of the two isomeric levels has been estimated for slow, fast and resonance neutrons.

The nuclear isomerism of the radioactive zinc isotope Zn^{69} has been well known for some time. The two radioactive decay periods associated with this isotope are 57 minutes and 13·8 hours, both having, as far as detectable, the same β-ray spectra. The 57 minute period is associated with a continuous β-ray spectrum only, with absorption end point at 0·41 gm./cm.^{2} of aluminium ($E_m = 1{\cdot}0$ mev. from Feather's relation) without any detectable intensity of γ-rays. The 13·8 hour period is associated with β- and γ-rays. The γ-ray absorption in lead gives a half value thickness of 3·6 gm./cm.^{2} ($E = 0{\cdot}47$ mev.) and the β-rays follow the same absorption curve as that of the 57 minute period. The results suggest, as pointed out by Kennedy *et al.* (1939) that the 13·8 hour product decays to Ga^{69} passing through the isomeric state of 57 minute half life as in the following nuclear level scheme:—

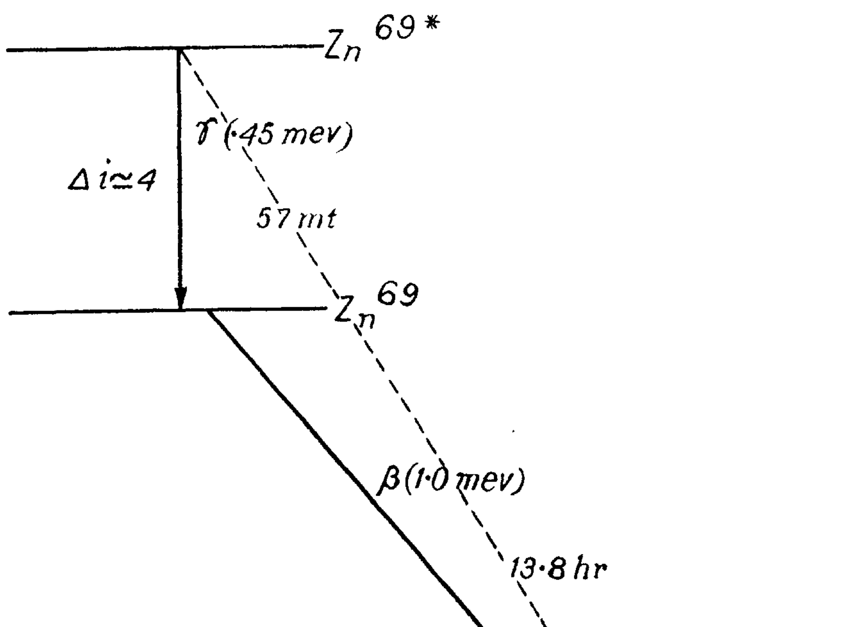

FIG. 1.

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Kennedy, Seaborg and Segre (1939) and Dubridge (1939) tried to detect electrons that might be expected from the internal conversion of such a high energy γ-ray due to a forbidden transition. The spin change due to the γ-ray transition was deduced to be 5 by Kennedy *et al.* (1939), from an internal conversion formula due to Hebb and Uhlenbeck (1938). The conversion coefficient for such a γ-ray is expected to be about 13% from the formulae worked out by Dancoff and Morrison (1939). The figure is naturally rough owing to uncertainties in the theory. The experiments of Kennedy *et al.* (1939) and Dubridge (1939), for finding out the conversion coefficient did not yield any positive result, though their experimental set up was such that they expected to detect a conversion coefficient as low as 10%. The former author also attempted to detect β-γ coincidences which were expected if the β and the γ-ray be genetically related but did not obtain any positive result. In view of the incomplete nature of the above investigations, a further attempt to study Zn^{69} isotope was thought desirable. The following experiments were made:—

(1) Accurate measurement of the energy value of the γ-ray from Zn^{69} and to find out if there were other intermediate nuclear levels associated with Zn^{69*} (13·8 hr.) and Zn^{69} (57 min.).

(2) Measurement of the internal conversion coefficient of the γ-ray by the ionisation chamber method.

(3) Determination of the ratio σ_1/σ_2, the ratio of the capture cross-sections for production of the two isomers and to test its value in the light of Weizsacker's theory.

Experimental Procedure and Results.

The radioactive Zn^{69} isotope was produced by neutrons and deuterons on Zn. For neutron bombardment chemically pure zinc globules were rolled into thin sheets of approximately 1·5 mil. thick. Commercial zinc sheets of 1 mm. thickness were used to absorb any resonance neutrons. Equal weights of zinc sheets were taken in three samples. First sample intended to be bombarded by slow neutrons only was put in a box $3 \times 3 \times \frac{1}{2}$ cm. made up of 1 mm. zinc sheets to absorb resonance neutrons. The zinc box was placed at a distance of 12″ from the beryllium target of the cyclotron with 10″ of paraffin all around the sample to slow down the neutrons coming from the beryllium target. The second sample intended to discover resonance effects was wrapped in Cd shields 1 mm. thick and was kept at the same distance from the target with the paraffin all around as before. The third was put in a zinc sheet box as in the first sample and wrapped in Cd

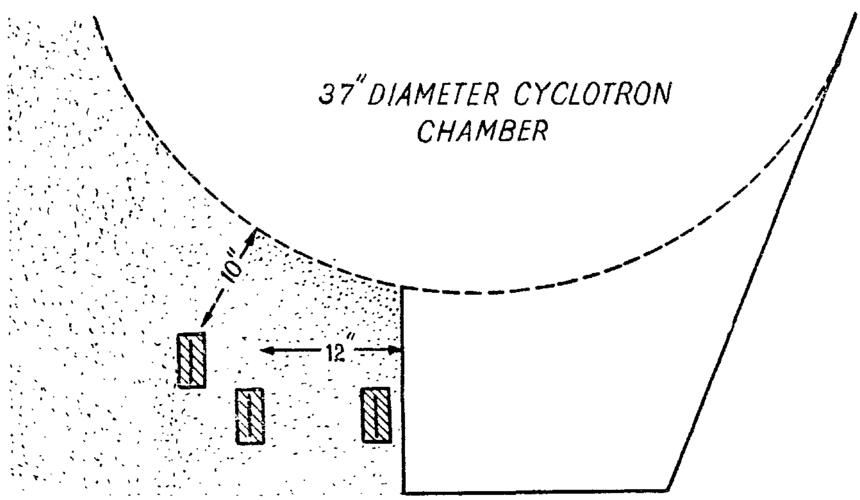

Fig. 2.

sheets and placed as close to the beryllium target as possible without any paraffin intervening to absorb the fast neutrons only from the beryllium-deuteron reaction. The slow and resonance neutrons would be effectively absorbed by the shields of cadmium and zinc, so that only fast neutron effects would be discernible. The same experiments were repeated with boron shields (made of borax) instead of cadmium in all the experiments without any detectable difference. The geometry of the samples with respect to the target is shown in diagram (fig. 2). The zinc sample after irradiation was dissolved in hydrochloric acid and a little copper and nickel added as tracers. The copper and nickel are then separated by precipitation with hydrogen sulphide and dimethylglyoxime

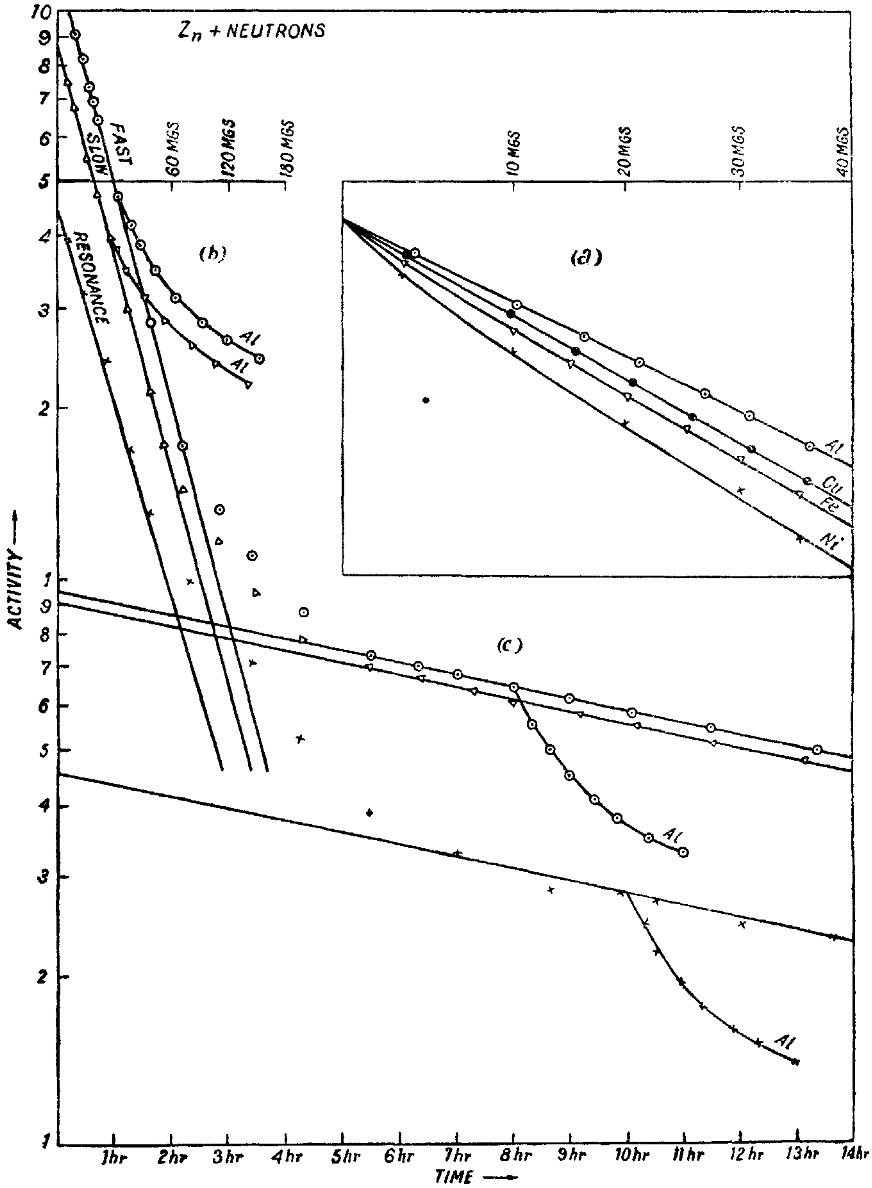

FIG. 3.

respectively. The solution is next evaporated to dryness and then zinc is recovered and mounted on cardboard discs with cellophane covers glued on to protect the samples. The activities of the samples were measured with an FP 54 electrometer with a modified Dubridge and Brown valve circuit and appropriate ionisation chambers. The absorption curves with aluminium as absorber were obtained and the corresponding decay periods were also measured.

Attempts were made to detect the possibility of a γ-ray transition in Ga^{69} in the following way. The zinc after bombardment was separated from the nickel and gallium fractions. The separation from gallium was carried out by ether extraction several times so that the final ether extract contained no detectable activity. The activated zinc was then allowed to decay for various periods of time from half an hour to about ten hours and at each stage a little gallium was added and the gallium fraction was separated by ether extraction, evaporated and the activity of the gallium was measured with a freon chamber. No activity could be detected. This eliminated the possibility of any transition within the limit of 1% which is claimed for these experiments. Various thicknesses of aluminium absorbers were employed and the decay periods measured. No change in the decay periods were observable.

The absorption curve of β-rays from the 13·8 hour period was measured with aluminium absorbers and a slight kink was observed (*vide* fig. 3) at the range of 0·11 gm./cm.^{2} of absorber. This indicated the existence of a homogeneous group of electrons at 470 ± 50 kev. which was interpreted as being due to the internal conversion of the γ-ray 13·8 hour product. A magnetic β-ray spectrograph was used to measure the energy of these homogeneous electrons from which the energy of the γ-ray could be next calculated. The energy of the γ-ray was also obtained by allowing it to impinge against lead and measuring the energy of the photo-electrons. To confirm if the homogeneous electrons are due to internal conversion, a lead absorber (thickness 0·6 gm./cm.^{2}) was put in and the absorption curve was again taken with aluminium foils and the kink showed up more distinctly as was expected. A strong bombardment was made for about 30 minutes on chemically pure Zn with deuteron and the zinc then chemically treated to remove Ga, Cu and Ni. The zinc precipitate was dried and distributed in a small Al boat $1 \times 0{\cdot}1 \times 0{\cdot}1$ cm., and then it was inserted within the magnetic spectrograph.[1] The conversion line was obtained and measured $H\rho = 2640$. The K and L photo-electrons from the annihilation radiation of positrons were obtained with a lead absorber on the same plate which enabled accurate measurement of the energy of the conversion line by comparison. The results obtained have been quoted in part by Helmholz.

K—photoelectrons from conversion of annihilation radiation in lead $H\rho = 2315$.
L— do. do. $H\rho = 2605$.
Energy of zinc conversion electrons $= 436 \pm 5$ kev.
Energy of γ-ray $= 436 \pm 5 + K$ binding energy of zinc $= 447 \pm 5$ kev.

The same experiments were repeated with a thin lead sheet over the aluminium boat carrying the zinc and the K and L photo-electrons in lead were obtained in another plate as a check on the previous measurements. The values of the γ-ray obtained from these are 450 ± 6 kev.

Determination of Internal Conversion Coefficient.

The determination of internal conversion coefficient was next attempted with a freon filled ionisation chamber at 1 atmos. pressure and using a very thin aluminium window ·005 mm. thick. The internal conversion of the γ-ray in the K shell was expected to give rise to K, X-rays characteristic of zinc. Paraffin absorbers of ·8 cm. thickness were used to cut off β-rays without completely cutting off the X-rays, critical absorbers were used to detect and estimate the relative intensity of X-ray to the γ-ray. The

[1] The author is thankful to Dr. A. C. Helmholz for the loan of his spectrograph and his collaboration in the magnetic spectrographic part of the work.

samples were made very thin and as strong as possible. The electrons were all absorbed by 0·48 gms. of paraffin per cm.^{2} Ni, Cu, Fe and Al foils of ½ mil thickness and upwards were used as absorbers and Ni foils absorbed the X-rays critically as expected. The absorption of the X-rays in the paraffin absorber and the window of the ionisation chamber and the absorption of the γ-rays in the paraffin absorber are taken into account in the following way:

$$R = \frac{N_X E_X\left(1-e^{-(\mu/\rho)_X \rho d}\right) - N_X E_X\left(1-e^{-\mu' x}\right) + K' N_X E_X\left(1-e^{-\mu''\gamma}\right)}{N_\gamma E_\gamma\left(1-e^{-(\mu/\rho)_\gamma \rho d}\right) + K'' N_\gamma E_\gamma\left(1-e^{-\mu'_a x}\right)}.$$

where in the numerator of the r.h.s. the second term represents the X-ray energy lost in the absorber and in the third term $K'E_X$ represents the average energy which the electrons ejected from the window lose in the chamber. Similarly $K''E_\gamma$ due to the γ-ray energy which the electrons ejected from the absorber lose in the chamber. R is the ratio Ne to Nr, where i_X is the ionisation current due to X-rays absorbed in the ionisation chamber and $i_X + i_\gamma$ is the total ionisation current in the chamber due to γ-rays and X-rays. Here

E_X = energy of the K X-rays from zinc = 11 kev.
E_γ = energy of the γ-ray = 450 kev.

$(\mu/\rho)_\gamma \rho d = {\cdot}001 \qquad (\mu/\rho)_X \rho d = 0{\cdot}28$

$K' = 1/100. \qquad K'' = 2/100.$

$\mu' x = {\cdot}02. \qquad \mu'_a x = {\cdot}001, \quad \mu''\gamma = {\cdot}002.$

$R \simeq {\cdot}06.$

Discussion.

In the first part of the experiment, while the activities of the samples were being measured with the electrometer circuit and the absorption curves with Al absorbers were being plotted, a search was made to detect the 38 minute due to Zn^{63} and 250 day periods due to Zn^{65}, as well as the possibility of the existence of other half lives of β- and γ-rays associated with Zn^{69}. Such investigation was thought necessary since complicated cases may occur as in the case of uranium X_2 or Z unravelled by Feather and Bretscher (1938). No activities apart from a very weak background of 250 day activity due to the positron emitting isotope Zn^{65} were found. The γ-ray absorption curve showed only one homogeneous γ-ray above 100 kev. within the limits of experimental error, which was the well-known γ-ray at 480 kev. Below 100 kev., the β-ray background made it difficult to estimate whether any weak γ-rays were present. There still remains the possibility of the presence of a γ-rays of less than 100 kev. energy. The occurrence of such a γ-ray would necessitate the presence of at least four intermediate levels, with less than 100 kev. difference between each other.

Attempts to detect the possibility of a γ-ray transition in Ga^{69} gave negative results with 1% accuracy. The possibilities of other transitions were eliminated by observing the decay periods with various thicknesses of Al absorbers. No change in the decay periods was observable and the possibilities of an alternative β-transition ruled out within the limits of accuracy of the experiment, i.e. 2% of the total. This negative result strengthens the argument for a genetically related isomer. The positive evidence from chemical separation of isomers and from β-γ coincidence experiments had been attempted but failed to give any positive results.

The decay taking into consideration the genetic relationship is

$$P = P_0 e^{-\lambda_1 t} \quad \ldots\ldots \quad (1) \text{ for the } \gamma\text{-ray activity}$$

$$Q = \left(Q_0 - \frac{\lambda_1 P_0}{\lambda_2 - \lambda_1}\right) e^{-\lambda_2 t} + \frac{\lambda_1 P_0}{\lambda_2 - \gamma_1} e^{-\lambda_1 t} \quad \ldots\ldots \quad (2) \text{ for the } \beta\text{-ray activity}$$

where P_0 = the number of radioactive nuclei in excited state (A) at time $t = 0$,
P = the number of radioactive nuclei in excited state (A) at time t,
Q_0 = the number of radioactive nuclei in the normal state (B) at time $t = 0$,
Q = the number of radioactive nuclei in the normal state (B) at time t,
λ_1 = decay constant of the excited state (A), i.e. the γ-ray activity,
λ_2 = decay constant for the normal state (B), i.e. the β-ray activity,
σ_1, σ_2 = cross-sections for the production of isomers of decay constants λ_1 and λ_2 respectively.

In equation (2), the underlined terms come as a result of genetic relationship, and would not hold if isomers were unrelated.

The ratio $\frac{P_0}{Q_0} = \frac{\sigma_1}{\sigma_2}$. Since the periods of bombardment were short compared to the half life no corrections were made for the growth which were assumed linear with time, in most cases examined. The experiments were all repeated with fast and slow neutrons and deuteron bombardment on zinc. The initial activities P_0 and Q_0 were calculated from (1) by extrapolating to initial intensities. The ratio of the cross-sections σ_1/σ_2 for the production of the two activities are shown below for slow, resonance and fast neutron and deuterons in the deuteron reaction which may be said to be equivalent to a neutron capture. The gradual decrease of σ_1/σ_2 with velocity

Nature of the bombarding particle.	Value of σ_1/σ_2
Neutrons slow	4·95
,, fast	2·2
,, resonance	5·1
Deuterons	2·9

of neutron indicates evidence in favour of Weizsacker model of formation of these nuclei, i.e. the capture of the neutrons at a small number of high levels. The spacing of these levels must be fairly wide for if the levels were close together there will be a large number of possible transitions to both levels and the ratio of σ_1/σ_2 would approximate to unity and would not change sufficiently with the energy of the bombarding particles. The absence of any resonance levels seem definitely established as well as the fact that with increasing energy of the bombarding neutrons σ_1/σ_2 tends to unity. Fast neutrons used in these experiments had a maximum energy of 9 mev., and if the experiment could have been done with lithium neutrons further evidence could be obtained.

The value of the energy of the γ-ray obtained with the help of the magnetic spectrograph is 450 ± 6 kev. when the measurements were carried out on the K and L photo-electrons in lead. This value is in very good agreement with the measurement from the internal conversion line in zinc. The value obtained is somewhat less than the previous values given by Seaborg *et al.* of 47 mev. from absorption measurements.

The value ·06 of the conversion coefficient bears out the expected value from the large energy and long life of the γ-ray. The value of ·06 is, however, subject to large uncertainties and can be regarded as giving the order of magnitude of the conversion coefficient of the 450 kev. γ-ray of Zn.

The angular moment change in the γ-ray transition is expected to be 5 from Dancoff and Morrison's formula. Ga^{69} has spin of $\frac{3}{2}$ and the lower normal state of Zn^{69} can be expected to have spin of $\frac{1}{2}$. The spin of the isomeric state has, therefore, probably a spin of $\frac{11}{2}$.

The energy of 450 kev. for the γ-ray is, perhaps, a better value than the previous one of Kennedy *et al.* of 480 kev. The absence of any resonance capture of neutrons seems fairly well demonstrated. Indications have been obtained of the increase of the number of levels in which neutron capture takes place with the increasing energy of the incident neutrons.

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