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# Energy and Multipole Order of Nuclear Gamma-Rays

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The energies of some nuclear gamma-rays emitted in deuteron-induced radioactivities have been measured with a magnetic  $\beta$ -ray spectrograph with photographic film as detector. The energies measured were those of the conversion electrons, and in some cases also those of the photoelectrons ejected from a heavy element. Independent measurements of the multipole order have been obtained where possible from the internal conversion coefficient, the K/Lconversion ratio, and the lifetime of the  $\gamma$ -ray transition. In the case of eight out of the fourteen  $\gamma$ -rays measured, a fairly definite assignment of the multipole order has been made. In the nuclei Zn<sup>69</sup>, Kr<sup>83</sup>, Sr<sup>87</sup>, and Ag<sup>107 or 109</sup> the probable nuclear spins of the states involved are given.

**T** WO of the most important characteristics of a nuclear gamma-ray are its energy and its multipole order. When several gamma-rays are present or there is a branching of some sort, a knowledge of the intensity, that is the number of quanta per disintegration, is also valuable. Knowing these three quantities, one can construct a nuclear level scheme with some certainty, particularly for the lighter nuclei in which the level density is not so great as to introduce great complexity. The object of this work has been to make as definite a determination as possible of these characteristics of  $\gamma$ -rays from some artificially radioactive nuclei.

The measurements of the energy have been accomplished with a magnetic  $\beta$ -ray spectrograph according to the well-established methods. There are three independent experimental results which provide information on the multipole order of  $\gamma$ -rays, and where possible all three have been used to check one another. First, the internal conversion coefficient, that is, the ratio of the probability of the emission of a conversion electron to the probability of the emission of a

quantum, is for low energy  $\gamma$ -rays very sensitive to the multipole order. For example, Dancoff and Morrison<sup>1</sup> derived the formula:

$$\alpha_k = Z^3 \left(\frac{e^2}{hc}\right)^4 \frac{l}{l+1} \left(\frac{2}{E}\right)^{l+5/2}$$
(1)

for the conversion coefficient in the K shell, where E is the gamma-ray energy in units of  $mc^2$  and lis the multipole order. It is apparent that for high energy  $\gamma$ -rays, 300 kev or more, the change of  $\alpha$ with l is not rapid; and since the experimental measurement of  $\alpha$  is difficult, the method ceases to be of much value. If the transition takes place between two states of angular momentum **J** and **J**', the value of l will be the lowest value of  $\mathbf{J} - \mathbf{J}'$  allowed by the parity and arguments of symmetry.\* The transition may be either electric or magnetic. If the transition with l = |J - J'|, is allowed, electric  $2^i$ -pole radiation only is important. On the other hand, if this transition is

<sup>&</sup>lt;sup>1</sup>S. M. Dancoff and P. Morrison, Phys. Rev. **55**, 122 (1939). \* For a full discussion of this question see reference 1.

forbidden by the parity, either magnetic  $2^{i}$ -pole radiation, electric  $2^{l+1}$ -pole radiation, or both may be important. As an example, consider the case of a nucleus with J=9/2, J'=1/2, values which might be determined spectroscopically from hyperfine structure data. The most probable transition will be by electric  $2^{9/2-1/2}$  or  $2^4$ -pole radiation. However, if the parity of the two states is opposite, this type of radiation is forbidden, and magnetic 24-pole radiation might occur. Alternatively, since the vector difference of % and  $\frac{1}{2}$  could be 5, electric 2<sup>5</sup>-pole radiation could occur. Formula (1) is for electric radiation. Nelson and Hebb<sup>2</sup> have calculated the conversion coefficients for magnetic radiation for both the Kand the L shell. To decide, for example, between magnetic 2<sup>4</sup>-pole and electric 2<sup>5</sup>-pole radiation in the case mentioned above, is, however, very difficult on the basis of only values of  $\alpha$  (see case of Zn<sup>69</sup> below). Therefore, it is very valuable to have a second method of getting the multipole order which depends on the ratio of the Kconversion to the L conversion. Hebb and Nelson<sup>2</sup> showed that this ratio is sensitive to the multipole order, and also that magnetic and electric radiations can be distinguished in this way. Since the ratio is easier to measure than the conversion coefficient, this method is especially noteworthy. Thirdly, if the difference in angular momentum of two states is large enough (4 units for energy differences of about 150 kev and allowed electric multipole radiations), the transition will be sufficiently forbidden so as to proceed with a measurable lifetime. This phenomenon, known as isomerism, was explained in detail by Weizsäcker.<sup>3</sup> Pontecorvo, and Hebb and Uhlenbeck<sup>4</sup> showed that the process of internal conversion tends to speed up the transition, so that the lifetime of the transition calculated from the purely electromagnetic considerations must be divided by  $1+\alpha$ , where  $\alpha$  is the total conversion coefficient. The theoretical expression for the decay constant of an electric transition of energy  $\hbar\omega$  in ergs and change in angular mo-



FIG. 1. The magnetic spectrograph.

mentum  $l = \mathbf{J} - \mathbf{J}'$  is:

$$\lambda = \left(\frac{\omega}{c}\right)^{2l+1} \frac{e^2}{\hbar} \eta^2 \frac{x^{2l}}{\left[1 \cdot 3 \cdot 5 \cdot \cdots (2l-1)\right]^2} \sec^{-1}.$$
 (2)

 $\eta$  is a factor of the order of unity and x is the dimension of the radiating multipole, for which the best approximation is the nuclear radius or  $A^{\frac{1}{3}} \times 1.4 \times 10^{-13}$  cm. For simplicity in calculation this can be put in the form :

$$\log_{10}\lambda = 20.3 - 2 \log_{10} [1 \cdot 3 \cdot 5 \cdot \cdots (2l-1)] - [2l+1] [1.30 - \log_{10}E] - 2l [0.84 - \frac{1}{3} \log A], \quad (3)$$

where now E is the energy of the  $\gamma$ -ray in Mev, Ais the atomic number, and l is the change in angular momentum. If, as discussed above, the transition with the lowest possible value of  $\mathbf{J} - \mathbf{J}'$ is allowed, the radiation will be electric  $2^{|J-J'|}$ pole, and the theoretical half-life will be given by  $0.693/\lambda(l)$ . On the other hand, if the transition proceeds by magnetic  $2^{l}$ -pole or electric  $2^{l+1}$ -pole radiation, the value of l to be used in the formula is l+1. Consequently, including the effect of internal conversion, we may calculate the expected half-lives  $\tau$  from the following formulae:

$$\tau = .693(\alpha + 1)^{-1}\lambda(l)^{-1}$$

for electric  $2^{l}$ -pole radiation.

 $\tau = .693(\alpha + 1)^{-1}\lambda(l+1)^{-1}$ 

for magnetic  $2^{l}$ -pole radiation.

As can be seen from Eq. (3), for  $\gamma$ -rays of the

<sup>&</sup>lt;sup>2</sup> M. H. Hebb and E. Nelson, Phys. Rev. **58**, 486 (1940). <sup>8</sup> C. F. v. Weizsäcker, Naturwiss. **24**, 813 (1936).

<sup>&</sup>lt;sup>4</sup> B. Pontecorvo, *Travaux du Congrès du Palais de la Découverte* (Paris, 1937); M. H. Hebb and G. E. Uhlenbeck,

*Découverte* (Paris, 1937); M. H. Hebb and G. E. Uhlenbeck, Physica 5, 605 (1938).

 $<sup>\</sup>dagger$  I am indebted to Dr. E. Segrè for the correct form of this equation.

order of 100 kev in energy,  $\lambda$  may change by as much as a factor of 10<sup>5</sup> due to a change of 1 in *l*. Hence, disagreement with an experimental result by as much as a factor of 50 does not obviate an assignment to a certain value of *l*.

Although none of the above-mentioned methods of determining the multipole order is definitive, an agreement of all three can be considered convincing evidence of the correctness of the interpretation.

#### EXPERIMENTAL TECHNIQUE

Figure 1 is a diagram of the  $\beta$ -ray spectrograph<sup>†</sup> used in these experiments. Except where marked, the spectrograph was made of brass. The interior was lined with white Bakelite to reduce scattering. The source was usually placed in a small Al boat 1.5 cm long and of the order of 1 mm wide and held in position by the ends of a wire U, the base of which was clamped in the tapered brass plug, A. This allowed a variation of the perpendicular distance from the source to photographic film. The distance was usually 1.5-2 cm and was measured with calipers to about 0.2 mm. In case a foil was used as source it was mounted on a thin piece of brass which, in turn, was clamped in the plug A, the arrangement being similar to that of Valley and McCreary.<sup>5</sup> The photographic film, introduced at C, fitted into a grooved track attached to the Pb block. The brass plug C was fitted with a gasket and held in place by two screws. Removing the plug B allowed light to produce a fiducial mark on the film 12 cm from the center of the slit. Thus radii of curvature up to about 6 cm could be used. The film used was Agfa x-ray film. It was developed in standard x-ray developer for 8 minutes. The distance from the fiducial mark to the lines or edges on the film was usually measured with calipers, although several checks were made by measurement with a microphotometer. The lid to the spectrograph was fitted with a gasket and screwed down to be vacuum tight. A small Hg diffusion pump backed by a Cenco Hyvac pumped out the spectrograph to a vacuum of the order of  $10^{-4}$  mm of Hg. In many instances, the use of the Hg pump was unnecessary.

The magnetic field was supplied by an electromagnet run on storage batteries. By checking the strength of the field about twice a day, it was kept constant within 1 percent. The measurement of the fields was performed with a flip coil and ballistic galvanometer. The sensitivity of the galvanometer was determined both with a 50-mh mutual inductance in the usual way, and with a Hibbert magnetic standard which reproduced a point on a sensitivity curve previously taken with a standard solenoid. The two methods of calibration agreed within 1 percent. No natural radioactive standard was available to check the values of the magnetic field, but the energies of the electrons from the 6.7-hr. Cd and Ga<sup>67</sup> agree well with those of Valley and McCreary.<sup>5</sup> For high energies, the value of the energy of Pb photoelectrons from annihilation radiation is a good check.

In most cases the element of which the source consisted was separated chemically from the target bombarded in the cyclotron. When the element bombarded is different from the element used as a source, this method provides a means of obtaining a concentrated activity, which is essential for this type of work. The active material was then placed in the Al boat. In the case of Kr and Se formed by Se+D, Se was distilled in vacuum onto a 1-mil Al foil, which was then bombarded in the cyclotron and placed in the spectrograph. When the energy of the gamma-ray quanta, rather than that of the electrons, was measured, the source was placed in an inverted boat made of Pb or whatever material was desired as the source of photoelectrons. Alternatively, the source would be made up as above, and the Pb placed directly on top of the boat.

Absorption and half-life measurements were taken with a Lauritsen electroscope fitted with a 0.2-mil Al window, or with an ionization chamber filled with Freon or air and attached to an FP-54 d.c. amplifier. For half-life measurements extending over long periods of time, the sensitivity of the detectors were determined with a uranium standard each time. The measurement of the internal conversion coefficient is a measurement of the ratio of the number of electrons to the number of quanta. What one measures with electroscope or ionization chamber is the ioniza-

<sup>&</sup>lt;sup>‡</sup> Except for a few small changes it was designed by Professor S. W. Barnes. <sup>5</sup> G. E. Valley and R. L. McCreary, Phys. Rev. 56, 863 (1939).

tion due to electrons or quanta. The ionization is proportional to the energy lost in chamber, or to the number multiplied by the energy and the percentage absorbed. The ionization of electrons is much greater than that of quanta, and the fraction of the energy lost in an ionization chamber by either electrons or  $\gamma$ -rays is not an easily measurable quantity, so the comparison is a very uncertain one. Nevertheless, in certain cases in which the effect of the conversion electron and of the  $\gamma$ -ray can be separated by absorption measurements, the method is feasible. If the electrons have a range r, an energy  $E_e$ , the  $\gamma$ -rays an energy  $E_{\gamma}$ , the chamber a depth d (assumed less than r), filled with a gas of absorption coefficient  $\mu$  and density  $\rho$ , the ratio of the ionization due to electrons to that due to quanta is:

$$R = \frac{N_e E_e d/r^*}{N_{\gamma} E_{\gamma} (1 - e^{-(\mu/\rho) d\rho})}.$$
 (4)

If the  $\gamma$ -rays lose an appreciable amount of energy in the ionization chamber window by the ejection of electrons, a term  $N_{\gamma}E_{\gamma}K(1-e^{-\mu x})_w$ must be added to the denominator. Here  $KE_{\gamma}$ represents the average energy which the electrons ejected from the window lose in the chamber.  $N_e/N_{\gamma}$  is, of course,  $\alpha$  which is to be determined. As an example, experiments on  $\mathrm{Sr}^{\mathrm{sr}}$  gave the following data:

 $E_{\gamma} = 386 \text{ kev}, \ E_{e} = 370 \text{ kev}, \ d = 10 \text{ cm}, \ r = 100 \text{ cm}$  $\mu/\rho \cdot \rho d = 0.092 \cdot 10 \cdot 0.0012 = 0.0011,$  $(\mu/\rho \cdot \rho x)_{\text{window}} = 0.095 \cdot 0.007 = 0.00067,$  $R = 23.6/1.1, \quad K = 10/100,$  $\frac{23.6}{1.1} = \alpha \frac{3701 \, \text{\% o o}}{386[(1 - e^{-0.0011}) + 0.1(1 - e^{-0.00067})]} = 82\alpha,$  $\alpha = 0.26.$ 

Obviously, it is easier to compare the numbers in two groups of electrons having about the same energy. This is possible when the absorption of a  $\gamma$ -ray in a material of roughly the same atomic number as that of the source is mostly by photoelectric effect, for the photoelectrons then have the same energy as the conversion electrons. In addition, the two sources of electrons should have the same thickness, that of the "source" of photoelectrons being necessarily r, their range. Under these conditions the ratio of the ionization from conversion electrons to that from the photoelectrons of the transition effect is the ratio of the numbers in the two groups. If the same source is used, first bare, to get the ionization from the conversion electrons, then covered with absorber of photoelectric absorption coefficient  $\mu$ for the  $\gamma$ -ray, the ratio of the ionizations is:

$$R = N_e / N_{\gamma} (1 - e^{-(\mu/\rho)\rho r}).$$
 (5)

The uncertainties are the distribution in angle of the ejected photoelectrons which can be roughly estimated from the geometry, and the part played by the Compton electrons. If the  $\gamma$ -ray energy is high compared with the K binding energy, a heavy material like Pb may be used as absorber without changing the energy distribution of the electrons absorbed in the chamber too much. As an example of this type of measurement, for Ga<sup>67</sup>, Cu was used as a source of photoelectrons from the 93-kev  $\gamma$ -ray. Absorption curves showed the curves for conversion and photoelectrons to be very similar.  $r\rho$  is 10 mg/cm<sup>2</sup>;  $\mu/\rho$  is 0.47 cm<sup>2</sup>/g;

$$R = 9.5/0.064 = \alpha/(0.010 \cdot 0.47)$$
  $\alpha = 0.7.$ 

If a K-electron capture is followed by internal conversion, absorption measurements can separate the effect of the electrons from that of the x-rays, and the numbers can be compared.<sup>6</sup> In cases of low atomic number the disparity in ionization is often not so great. Likewise, the number of  $\gamma$ -rays can be compared with the number of x-rays. To take the first case, if f is the fluorescent yield, there will be f x-rays per disintegration and  $\alpha/(1+\alpha)$  conversion electrons. If the electrons all come from the K shell, there will be  $f\alpha/1+\alpha$  additional x-rays. With the same symbols as in Eq. (4):

$$R = \frac{N_e E_e d/r}{f(1+(\alpha/1+\alpha))NE_x(1-e^{-(\mu/\rho)_x\rho d})},$$

<sup>\*</sup> This assumes that the energy loss per unit length of path is constant over the whole path length. The assumption is erroneous since the range is not a linear function of the energy, but it is simple and accurate enough for this purpose.

<sup>&</sup>lt;sup>6</sup> This is the method used by L. W. Alvarez, Phys. Rev. 54, 486 (1938).

where N is the number of disintegrations.  $N_e/N = \alpha/1 + \alpha$  so that:

$$R = \frac{\alpha}{1 + 2\alpha} \frac{E_e d/r}{(1 - e^{-(\mu/p)_x p d})}.$$
 (6)

Similar considerations apply to the comparison of x-rays and  $\gamma$ -rays. The formula is

$$R = \frac{1}{(1+2\alpha)f} \frac{E_{\gamma}(1-e^{-(\mu/\rho)_{\gamma}\rho d})}{E_x(1-e^{-(\mu/\rho)_x\rho d})}$$
(7)

in the case that the absorption in the ionization chamber window is unimportant. The correction term mentioned for Eq. (4) must be added if it is not. As an example, for the case of the 6.7 hr. Cd, the data were:

 $E_{\gamma} = 92 \text{ kev}, \quad E_x = 22 \text{ kev}, \quad d = 10 \text{ cm}.$ Gas in ionization chamber CCl<sub>2</sub>F<sub>2</sub> at 1.67 atmospheres.  $\frac{1}{2}$  conversion electrons come from K shell.

 $(\mu/\rho)_x d\rho = 0.33, \quad (\mu/\rho)_y d\rho = 0.017,$ 

$$f = 0.75, \quad R = 3/1000,$$

$$\frac{3}{1000} = \frac{92(1 - e^{-0.017})}{22(1 - e^{-0.33})} \frac{1}{0.75} \frac{2}{(2 + 3\alpha)},$$

$$\alpha = 75.$$

The uncertainty in the above types of measurement may be as great as  $3 \times$  the value found, but since the values of  $\alpha$  for values of *l* differing by one may differ by a factor of 10, the method is useful. Ellis and Aston<sup>7</sup> determined  $\alpha$  by comparing, in a  $\beta$ -ray spectrograph, the intensity of conversion electrons with that of photoelectrons ejected by quanta. This method is useful if  $\alpha$  is small; but it requires a considerable amount of not too certain calculation concerning the distribution in energy and direction of the photoelectrons. An accurate method is to compare, by means of a  $\beta$ -ray spectrograph equipped with counter, the number of disintegration electrons with conversion electrons. This has been used, for example, by Flammersfeld.8

The ratio of the K to the L conversion has, in most cases, been measured photometrically on films from the spectrograph, using as a standard a film exposed to Ga<sup>67</sup> electrons for various lengths of time. If the ratio is large or the background large, the difficulty is obvious. Another method attempted in some cases is to obtain equal blackenings at the same point on two films by exposing the L line at the required higher field for longer times. This has the disadvantage that the background will be higher for the longer exposure. In the case of Ga<sup>67</sup>, both methods have been used to give a K/L ratio of 8, which is in agreement with that determined by Valley and McCreary.<sup>5</sup>

As has been noted before, a transition may consist of both electric and magnetic radiation. From the formulae of Hebb and Nelson<sup>2</sup> one can calculate the K/L ratio for both kinds of radiation. Then if the observed ratio  $N_K/N_L$  lies between the two, the percentage of the transitions occurring by electric radiation is:\*

$$\frac{E}{M+E} = \frac{1 + \alpha_K + \alpha_L}{1 + \alpha_K + \alpha_L + \left[ (\alpha_K - N_K \alpha_L / N_L) / (\beta_L - N_K \beta_K / N_L) \right] (1 + \beta_K + \beta_L)},$$
(8)

where  $\alpha_K$  and  $\alpha_L$  are the electric conversion coefficients,  $\beta_K$  and  $\beta_L$  the magnetic, and E and M the number of the transitions per sec. proceeding by electric and magnetic radiation, respectively.

#### EXPERIMENTAL RESULTS

The experimental results are summarized in Table I. These take precedence over previously

reported results.9-13 For the cases in which the data are insufficient to decide the type of

- <sup>8</sup> A. Flammersfeld, Zeits. f. Physik 114, 227 (1939).
   <sup>\*</sup> I wish to thank Mr. E. Nelson for information on this point.
  - <sup>9</sup> A. C. Helmholz, Phys. Rev. 57, 248 (1940).

<sup>&</sup>lt;sup>7</sup> C. D. Ellis and G. H. Aston, Proc. Roy. Soc. A129, 180 (1930).

 <sup>&</sup>lt;sup>10</sup> H. Walke, Phys. Rev. 57, 163 (1940).
 <sup>11</sup> A. Langsdorf, Jr., and E. Segrè, Phys. Rev. 57, 105 (1940).

<sup>&</sup>lt;sup>12</sup> L. W. Alvarez, A. C. Helmholz, and E. Nelson, Phys. Rev. 57, 660 (1940).

<sup>&</sup>lt;sup>13</sup> G. T. Seaborg, J. J. Livingood, and J. W. Kennedy, Phys. Rev. **57**, 363 (1940).

Nucleus*	Gamma-Ray Energy in kev	Half-Life of Transition	Multipole Order and Type of Radiation†
Sc44	$268 \pm 5$	52 hours	$E 2^4$ or $E 2^5$ (?)
Zn <sup>69</sup>	$439 \pm 5$	13.8 hours	$E 2^5$ or $M 2^4$
Zn <sup>67</sup>	$92.5 \pm 1$		$E 2^2$
	$180 \pm 2$		$E 2^{1} (?)$
	$297 \pm 3$		$E 2^{1} (?)$
Sc79 or 81	$99 \pm 1$	1 hour	$E 2^4$ and $M 2^3$
Kr <sup>83</sup>	$46 \pm 1$	110 min.	$E 2^4$ and $M 2^3$
	$29 \pm 1$		$E 2^{1}$ (?)
Sr <sup>87</sup>	$386 \pm 5$	2.8 hours	E 25
43	$97 \pm 1$	90 davs	$\overline{E}$ 2 <sup>5</sup> and M 2 <sup>4</sup> or
			$E 2^4$ and $M 2^3$
Ag107 or 109	$92.5 \pm 1$	40 sec.	$E 2^4$
Te <sup>127</sup>	86 + 1.5	90 days	$\overline{E}$ 2 <sup>5</sup> and M 2 <sup>4</sup>
Te <sup>129</sup>	102 + 1.5	32 days	E 2 <sup>5</sup> and $M$ 2 <sup>4</sup>
Te <sup>131</sup>	177 + 3	1.2  days	E 2 <sup>5</sup> and $M$ 2 <sup>4</sup> or
		duj b	$\overline{E}$ 2 <sup>4</sup> and $\overline{M}$ 2 <sup>3</sup>

TABLE I. Gamma-ray energies and multipole orders.

\* Nucleus in which  $\gamma$ -ray transition takes place. † E denotes electric, M magnetic radiation.

radiation, the most likely possibilities are given. For the sake of convenience in the discussion of individual cases below, the values of l usually refer to those appropriate in the half-life calculation. Hence, for example, if l=5, it must be remembered that the radiation may be magnetic 2<sup>4</sup>-pole.

#### Sc44

The activity of Sc<sup>44</sup> was discovered by Walke.<sup>10</sup> It is produced by the  $K^{41}(\alpha, n)Sc^{44}$  and  $Ca^{43}(d, n)Sc^{44}$  reactions. Walke found both a 52-hour period and a 4.1-hour period belonging to this isotope. The 52-hour period showed, besides the positrons characteristic of the 4.1-hour activity, a group of electrons of energy about 250 kev. To see whether these were really conversion electrons from the  $\gamma$ -ray in an isomeric transition, a sample of Sc prepared by deuteron bombardment of Ca was placed in the spectrograph and showed a line of electrons at 260 kev. Recently Smith<sup>14</sup> has measured more accurately the energy of this  $\gamma$ -ray and found 268 kev. The good agreement of this result with Walke's absorption data seems proof that these are the conversion electrons from the  $\gamma$ -ray responsible for the isomerism. The conversion coefficient was estimated by comparing the number of conversion electrons with the number of photoelectrons from Pb. The x-rays from Sc are not energetic enough to be measured. The result for  $\alpha$  was 0.5.

The conversion coefficient calculated from the formula of Morrison and Dancoff is 0.60 for l=5, 0.15 for l=4, both within the error of the measured value. The calculated half-life for l=5is  $2.5 \times 10^5$  hours, for l=4, 13 min. On this basis the value of l=4 seems preferable. The L conversion could not be observed, nor is much variation of the K/L ratio with l expected at this energy and value of Z. An accurate value of  $\alpha$ could probably decide the correct value of *l*.

#### Zn69†

The isomerism in Zn was investigated by Kennedy, Seaborg, and Segrè,15 who found that the 13.8-hour period consisted of a  $\gamma$ -ray transition to the ground state of Zn, which then decayed to Ga<sup>69</sup> with a half-life of 57 min. They observed no conversion electrons in absorption measurements, but estimated the  $\gamma$ -ray energy as 470 kev and set an upper limit of 0.1 for the value of  $\alpha$ . Valley and McCreary<sup>5</sup> also failed to find evidence of conversion. The conversion electrons would be few in number for such a high energy  $\gamma$ -ray and might be masked by the disintegration electrons. However, Zn was bombarded with 8-Mev deuterons and placed in the spectrograph. A 24-hour exposure showed a line at  $H\rho$  2640. Another spectrogram with the Zn source covered with Pb showed K and L edges due to photoelectrons ejected by the same  $\gamma$ -ray. The energy deduced from the single line which must be the K conversion and from the Pb photoelectrons is 439 kev. The value of the magnetic field was checked in these cases by obtaining the K and L photoelectron edges from Pb for the 511-kev annihilation radiation.

It was estimated from the strengths of the samples used that the conversion coefficient was

TABLE II. Comparison of observed and theoretical values  $\alpha$  and  $\alpha_K/\alpha_L$  for Ga<sup>67</sup>.

Calcu	ALCULATED FOR ELECTRIC RADIATION				
	l = 1	l=2	l=3	Observed	
$\alpha_K/\alpha_L$ Internal con- version coeff.	10.5 0.07	8 0.83	4.5 7.9	8 0.75	

† The writer was aided in the work on this element by Dr. B. D. Nag. <sup>15</sup> J. W. Kennedy, G. T. Seaborg, and E. Segrè, Phys. Rev. **56**, 1095 (1939).

<sup>&</sup>lt;sup>14</sup> G. P. Smith, Phys. Rev. 59, 937A (1941).

of the order of 0.1–0.01. From Dancoff and Morrison's formula and l=5, we obtain  $\alpha = 0.036$ . The calculated half-life with l=5 is 272 hours, with l=4, 2 sec. Therefore the value of 5 seems quite definite. No *L* conversion could be observed so the K/L ratio could not be determined. Surprisingly enough, the calculated magnetic conversion coefficient is 0.039, so it is not possible to tell whether this is electric 2<sup>5</sup>-pole or magnetic 2<sup>4</sup>-pole radiation.

Zn<sup>69</sup> can be made by neutron bombardment of Zn<sup>68</sup> which has a spin 0. Ga<sup>69</sup> has a spin of  $\frac{3}{2}$ ,<sup>16</sup> so it is natural to assume that the ground state of Zn<sup>69</sup> has a spin of  $\frac{1}{2}$ . Then the isomeric state would have a spin of  $\frac{1}{2}$ , or at least  $\frac{9}{2}$  if the electric transition with l=4 was forbidden.

#### Ga<sup>67</sup>

Alvarez<sup>6</sup> first found an internally converted  $\gamma$ -ray in Ga<sup>67</sup> formed in the reaction Zn<sup>66</sup>(d, n)Ga<sup>67</sup>, which decays by K-electron capture to  $Zn^{67}$ . Alvarez estimated the conversion coefficient as 2.\* Beta-ray spectrograms showed two lines at  $H\rho$  1007 and 1065, the K and L conversion lines in Zn of a 92.5-kev gamma-ray. This result is in agreement with that of Valley and McCreary.<sup>5</sup> The source was obtained by dissolving Zn in 6NHCl, shaking with ether to extract GaCl<sub>3</sub>, evaporating the ether to a small volume, and then putting it in the source boat drop by drop. To verify Alvarez' conclusion that the  $\gamma$ -ray is only partially converted a source was covered with Sn. The resulting film showed an edge at 64 kev due to the K photoelectrons ejected from Sn (K binding energy 29 kev) by the 92.5-kev gammaray. This picture also showed an edge at 151 kev. To prove that this came from a gamma-ray of 180 kev, another source was covered with a thin sheet of gold about 0.5 mg/cm<sup>2</sup> thick. The resulting picture then showed lines at 100 and 167 key, the K and L photoelectrons from the 180-key gamma-ray, and another at 216 key resulting from a 297-kev gamma-ray. Although these gamma-rays are probably dipole radiations and the internal conversion coefficients only of the order of 0.01, sufficiently strong samples were obtainable so that exposures of several days



FIG. 2. Examples of  $\beta$ -ray spectrograph films.  $H\rho$  increases to the right. (a) Lines from Se and Kr. (b) K and L lines from Sr<sup>87</sup>, differing by only 2 percent in  $H\rho$ . (c) K and L lines from element 43. (d) K, L, and M lines from 6.7-hr. Cd. (e) Film from radioactive Te isotopes. K, L, and M lines of 86-kev  $\gamma$ -ray, K and L lines from 102 and 107-kev  $\gamma$ -rays.

showed conversion lines at 170 kev and 288 kev. It might be thought that these were photoelectrons from the source, but the photoelectric absorption coefficient is very low in Ga for these  $\gamma$ -rays, and, since the source is separated without carrier, there would not be enough material to produce the effect. Since the source boat is made of Al, photoelectrons from it would differ in energy. The *L* conversion of these two gammarays is probably very small and was not observed. The K/L ratio of the 92.5-kev gamma-ray is 8 to 1, as measured from single films and by exposing one film to eight times as much radiation as another.

Several different measurements of the internal conversion coefficient were made. The experiment of Alvarez comparing the numbers of electrons and x-rays was repeated. The ionization of the photoelectrons ejected from a sheet of copper was compared with that of the conversion electrons as mentioned above. In addition, the ionization of the x-rays was compared with that of the unconverted gamma-ray. The gamma-ray absorption curve is difficult to resolve so that this method is not so satisfactory. Besides the three

<sup>&</sup>lt;sup>16</sup> N. A. Renzetti, Phys. Rev. 57, 753 (1940).

<sup>\*</sup> His value of  $\frac{2}{3}$  refers to the ratio of conversion electrons to conversion electrons plus gamma-rays.

gamma-rays mentioned, there may be even a fourth of higher energy. The results of all these determinations were quite consistent, and the value of  $\alpha$  ranged for the most part from 0.5 to 1. From Table II one can see that this definitely establishes the gamma-ray as electric quadrupole. The conversion electrons of the 180-kev gammaray were not observable on an absorption curve, so that no estimate of the coefficient could be made. For the 297-kev gamma-ray, only a slight indication of the electrons was discernible, and from this and the gamma-ray intensity one deduces a coefficient appropriate to l=1.

Of the three gamma-rays, the 93 kev is the most intense, the 180 kev is least intense. This suggests that most of the processes proceed from  $Ga^{67}$  through the state in  $Zn^{67}$  93 kev above ground. However, sometimes the disintegration goes through a state 93+297 kev above ground, and least often through a state 93+297+180 kev above ground, each then cascading down. The fact that the most energy is available in the first process would explain the preponderance of that type of disintegration.

# Se<sup>79</sup> or Se<sup>81</sup>

Langsdorf and Segrè<sup>11</sup> found a case of isomerism in Se with a half-life of 1 hour and electrons of about 100-kev energy. To measure the energy of this converted  $\gamma$ -ray, a Se source was prepared as described above, and placed in the spectrograph. Two lines at  $H\rho$  1035 and 1095 were found. (See Fig. 2a). To be sure that these were due to the isomerism in Se, successive exposures were made and showed that the intensity of the lines decreased appropriately for a substance with a 1-hr. half-life. The energies of the lines are 86.8 and 96.4 kev. On addition of the K and L binding energies of Se, the  $\gamma$ -ray energy turns out to be 99 kev as has been reported. The K/L conversion ratio is 4. Because of the many activities produced by Se+d, it was not possible to separate out this activity for a test of the conversion coefficient. The fact that the daughter activity is  $\beta$ -active, going to Br, would also hinder the determination. Assuming l=4 for the transition, we calculate a half-life of 9 hours. The observed K/L ratio points then to a mixture of 50 percent electric and 50 percent magnetic radiations. Assuming l=3 or l=5 would make the calculated

half-lives in error by large factors. In addition, an electric  $2^3$ -pole  $\gamma$ -ray would not have a K/L ratio as low as 4. Therefore, the assignment to part electric  $2^4$ -pole, part magnetic  $2^3$ -pole radiation seems quite conclusive.

## Kr<sup>83</sup>

Langsdorf and Segrè<sup>u</sup> found that the decay of Br<sup>83</sup> produces a 110-min. activity in Kr<sup>83</sup>. Since Kr<sup>83</sup> is stable, this activity must be due to isomerism. They found very soft electrons indicating a  $\gamma$ -ray of about 30 kev. Br<sup>83</sup> can be produced by the Se(d, n)Br reaction, so the same pictures obtained for Se were examined for the Kr lines. In the low energy region three lines at  $H\rho$  725, 620, and 572 were found. (See Fig. 2a.) Successive exposures seemed to indicate that all three came from the same activity and that this activity was probably Kr. The Kr activity should at first grow and then decay. Actually, the first two exposures of  $1\frac{1}{2}$  hr. each showed equal intensities. Thereafter the intensities fell off corresponding to a 2-hr. half-life. This evidence in conjunction with Langsdorf and Segrè's absorption measurements indicates that these lines come from Kr<sup>83</sup>. One exposure showed evidence of a fourth line at about 15 kev. Therefore, the three lines seem to be the L conversion of a 29-kev  $\gamma$ -ray, and the K and L of a 46-kev  $\gamma$ -ray. The question then arises as to which transition has the life of 110 min. A similar case of two low energy  $\gamma$ -rays, one probably with l=4, the other l = 1 has been investigated in the 4.4-hr. isomerism of Br<sup>80.17</sup> The K/L ratio of the 46-kev  $\gamma$ -ray is 1. If l=4 for this transition, the calculated half-life is 50 hr., in good agreement with the observed value, and the radiation would be 60 percent electric, 40 percent magnetic.\* The observed K/Lratio cannot be fitted by l=1, 2, or 3, so it seems probable that this transition has l=4. The failure to observe the K line of the 29-key  $\gamma$ -ray would then be due not to the low K/L ratio but to the rapid decrease with energy of the sensitivity

<sup>&</sup>lt;sup>17</sup> A. P. Grinberg and L. J. Roussinow, Phys. Rev. 58, 181 (1940).
\* It must be remembered that the calculation of the

<sup>\*</sup> It must be remembered that the calculation of the magnetic conversion coefficient is uncertain for  $\gamma$ -ray energies not much greater than the K binding energy, as in this case. Also, in the case of high atomic numbers such as Te, the electric conversion coefficient calculations are uncertain.

of the film.<sup>18</sup> If this  $\gamma$ -ray is a dipole ray, the conversion coefficient would be 3.5, which might be observable and would provide a check of this scheme. Kr83 has a spin of %.19 If there were a level 75 kev above ground with spin  $\frac{1}{2}$ , another 29 kev above ground with spin  $\frac{7}{2}$ , and the transition  $\frac{1}{2}$  to  $\frac{7}{2}$  with electric 2<sup>3</sup>-pole radiation was forbidden, one could explain the observations. However, the explanation seems somewhat artificial.

## Sr<sup>87</sup>

The isomerism in Sr<sup>87</sup> was discovered by DuBridge and Marshall,<sup>20</sup> who found a converted  $\gamma$ -ray associated with a 2.75-hr. period. This Sr activity grows from the 80-hr. Y<sup>87</sup>. This latter activity was produced here in great quantities in the production of Sr<sup>89</sup> for biological use. Several strong samples were separated from Y and the half-life measured as 2.8 hours. The  $\beta$ -ray spectrograph showed the line found by DuBridge and Marshall. The K and L lines differ in  $H\rho$  by only 2 percent; but by using a radius of curvature of 6 cm it was possible to separate the two lines. (See Fig. 2b.) The  $H\rho$  values were 2395 and 2447, the energies 370 and 383 kev. Adding the K and L binding energies of Sr gives 386 key as the  $\gamma$ -ray energy. The K/L ratio as determined from microphotometer measurements and different length exposures is 6-7. Finally, the conversion coefficient was determined as mentioned above. The average of the measured values was 0.15, in agreement with the measurements of DuBridge and Marshall.<sup>20</sup> For l=5, the calculated half-life is 520 hours, for l=4, 4 sec. The K/L ratio for electric 2<sup>5</sup>-pole radiation is 6, and the conversion coefficient is 0.06, which is within the experimental error. From the half-life, l=5 seems preferable, and from the other data quite certain. Magnetic 2<sup>4</sup>-pole radiation would give a K/Lratio of 10. Consequently, this is probably electric radiation.

The spin of the ground state of Sr is %.<sup>21</sup> If we omit from consideration, as unlikely, spins of  $1\frac{7}{2}$ or 1%, the spin of this excited state must then be  $\frac{1}{2}$ , and the transition with l = 4 must be forbidden.

#### **ELEMENT 43**

Kalbfell<sup>22</sup> reported two internally converted  $\gamma$ -rays from long-lived element 43. He was at the time unable to assign them to the 90- or 62-day periods measured by Cacciapuoti.23 The same sample used by him, and another kindly supplied by Dr. E. Segrè both gave a pair of lines at  $H_{\rho}$  960 and 1080. (See Fig. 2c). These indicate a  $\gamma$ -ray energy of 97 kev. A part of one of these samples was followed for 200 days with an electroscope, and decayed with a half-life of  $91\pm 2$  days. Absorption curves showed that almost all the ionization was due to electrons of energy about 90 kev. Therefore, the assignment of this  $\gamma$ -ray to the 90-day period is certain. Seaborg and Segrè<sup>24</sup> reported that the x-rays from a sample of the two long-lived activities were Mo K x-rays. However, several absorption measurements on the same sample using their technique of critical absorption, showed that most of the x-rays belonged to element 43. Since the sample had aged considerably since their experiments, the conclusion is that the x-rays due to the 62-day period are Mo x-rays arising from K capture, those from the 90-day period are element 43 x-rays, arising from the internally converted  $\gamma$ -ray of the isomeric transition. This interpretation encounters the objection that no isotopes of 43 are supposed to be stable,<sup>25</sup> and no activity of half-life less than 15 years growing from the 90-day period has been observed.

On the assumption that this is an isomeric transition, l must be 4 or 5. The observed K/Lratio is 2. With l=5, this would mean 70 percent magnetic 2<sup>4</sup>-pole, 30 percent electric 2<sup>5</sup>-pole radiation, a conversion coefficient of 470, and a half-life of  $10^5$  days. On the other hand, l=4gives 50 percent electric, 50 percent magnetic radiation,  $\alpha = 61$ , and a half-life of 0.1 day. No unconverted  $\gamma$ -rays were observed, but the samples used were not strong enough even were  $\alpha$ equal to 61. Since the calculated half-lives are both in error by a factor of 10<sup>3</sup>, there seems no adequate way of determining the multipole order unless by determining  $\alpha$ .

- <sup>22</sup> D. C. Kalbfell, Phys. Rev. 55, 422 (1939).
   <sup>23</sup> B. N. Cacciapuoti, Phys. Rev. 55, 110 (1939).
   <sup>24</sup> G. T. Seaborg and E. Segrè, Phys. Rev. 55, 808 (1939).
   <sup>25</sup> H. Jensen, Naturwiss. 26, 381 (1938).

<sup>&</sup>lt;sup>18</sup> For example, B. v. Borries and M. Knoll, Physik. Zeits. **35**, 279 (1934). <sup>19</sup> H. Korsching, Zeits, f. Physik **109**, 349 (1938).

<sup>&</sup>lt;sup>20</sup> L. A. DuBridge and J. Marshall, Phys. Rev. **56**, 706 (1939); **58**, 1 (1940).

<sup>&</sup>lt;sup>21</sup> M. Heyden and H. Kopferman, Zeits. f. Physik 108, 232 (1938).

ENERGY SPIN KEV  $\sim 800 \frac{7}{2}$  499% FIG. 3. Possible level CAPTURE 92.5  $\frac{9}{2}$   $0 \frac{1}{2}$   $A_6$  Co

Ag<sup>107</sup> or 109

The observation of the 40-sec. Ag activity growing from the 6.7-hr. Cd has been described previously.<sup>12</sup> The  $\beta$ -ray spectrograph showed the same lines observed by Valley and McCreary.8 (See Fig. 2d.) The source in this case was prepared by dissolving the bombarded Ag in HNO<sub>3</sub>, precipitating AgCl, adding 1 mg of Cd as carrier, and precipitating CdS. An absorption curve of the x-rays and  $\gamma$ -rays showed, besides the Ag K x-rays, the unconverted  $\gamma$ -rays of about 90 kev and a hard  $\gamma$ -ray of about 700 kev. The conversion coefficient calculation was given as an example above. The average observed value of 100 agrees well with 110 calculated for electric 2<sup>4</sup>-pole radiation; and the observed K/L ratio of 1 also agrees well with the calculated value of 1. The calculation of the lifetime given in the original note<sup>12</sup> was done with the formula given by Bethe.<sup>26</sup> Using the correct formula (2), the calculated half-life is 5000 sec., which is poor agreement but somewhat better than for l=3, which gives 0.01 sec.

Over 5 half-lives, the 700-kev  $\gamma$ -ray decayed with a half-life of 6.7 hours, so it certainly belongs to this activity. It occurs in only about 1/100 of the disintegrations and must come from an excited state in Ag, through which the disintegration proceeds less often because of the reduced energy available.

The spin of the ground state of Ag is  $\frac{1}{2}$ ,<sup>27</sup> so that the spin of the excited state must be at least  $\frac{1}{2}$  or perhaps  $\frac{9}{2}$ . A possible level scheme showing the mode of disintegration is given in Fig. 3.

# Te127, Te129, Te131

Seaborg, Livingood and Kennedy<sup>13</sup> found three cases of isomerism in Te from the reaction Te(d, p)Te. The half-lives of the upper isomeric states and the isotopes responsible are 1.2-day Te<sup>131</sup>, 32-day Te<sup>129</sup>, and 90-day Te<sup>127</sup>. In each case they have been able to make fairly complete isomer separations by the chemical method, suggesting that the  $\gamma$ -rays are fairly highly converted. The sources observed in the spectrograph at the time of the report by Seaborg, Livingood and Kennedy were weak, and since then much stronger sources have been available. Te was scraped from the target bombarded with 16-Mev deuterons and separated chemically.\* It was then placed in the spectrograph and an exposure of two days made. Two lines at  $H_{\rho}$  1375 and 1515 appeared. In longer exposures at later times these lines were absent; hence we may ascribe a  $\gamma$ -ray of energy 177 kev of which these are the K and L conversion lines, to the 1.2-day period. Kalbfell has observed these lines also but with weaker sources. The K/L ratio is 2. The half-life calculated with l=5 is  $3.4 \times 10^4$  hr., with l=4, 4.5 min. The K/L ratio could be explained on either basis by assuming part electric and part magnetic radiation. The conversion coefficient could not be measured because of the other Te activities. It would be 20 for l=5, 4 for l=4. A good determination of the conversion coefficient would seem, at present, to be the only method of deciding the *l* value.

Because the source element is the same as the bombarded one, it is difficult to get high specific activity. However, exposures of 4–6 days after the 1.2-day period had died away showed five lines at  $H_{\rho}$  807, 997, 1020, 925, and 1105. These correspond to the K, L, and M conversion in Te of an 86-kev  $\gamma$ -ray, the K and L of a 102-kev  $\gamma$ -ray (see Fig. 2e. The other pair of lines observed seem to come from a 107-kev  $\gamma$ -ray, whose assignment is uncertain). The L line of the former seems more intense than the K, while for the latter they seem about equal. The lines are faint and the background large, so that an accurate determination of the intensity ratio has not been possible. To determine which  $\gamma$ -ray

<sup>&</sup>lt;sup>26</sup> H. A. Bethe, Rev. Mod. Phys. 9, 226 (1937).

<sup>&</sup>lt;sup>27</sup> D. A. Jackson and H. Kuhn, Proc. Roy. Soc. A158, 372 (1937).

 $<sup>^{\</sup>ast}\,I$  wish to thank Mr. G. Friedlander for doing the chemical work.

belongs to which period, pictures were taken after 30 and 60 days. After 60 days the 86-kev  $\gamma$ -ray was relatively more intense than at the beginning so that it must be the  $\gamma$ -ray of the 90-day isomeric transition, the 102 kev that of the 32-day transition.

A  $\gamma$ -ray in Te with l=5 and an energy of 102 kev corresponds to a calculated half-life of 2400 days, too large by a factor of 75. On the other hand, the assumption of l = 4 gives a half-life only 1/5000 the observed. The value of 5, therefore, seems quite probable. Using this value, one then calculates that a K/L ratio of 1 is consistently explained by 45 percent electric 25-pole radiation, 55 percent magnetic 2<sup>4</sup>-pole radiation. Similarly, a  $\gamma$ -ray with l=5 and an energy of 86 kev in Te, gives a calculated half-life of 40,000 days,  $400 \times \text{longer}$  than the observed 90 days. However, l=4 gives only 10 minutes as the calculated halflife, and since this is only 1/13,000 the observed value, l=5 seems much to be preferred. The estimated K/L ratio of 0.75, which is probably not wrong by more than 30 percent, then gives for this case also a mixture of 50 percent electric, 50 percent magnetic radiation. The calculated conversion coefficients are large, 2000 and 10,000, respectively; and no unconverted  $\gamma$ -rays could be observed.

#### CONCLUSION

The results discussed above indicate that in many cases of nuclear  $\gamma$ -rays, it is feasible to determine the type of the radiation, electric, magnetic, or both, and the multipole order. Although present formulae for the lifetimes of  $\gamma$ -ray transitions usually indicate a preferable value of the multipole order, in only two or three out of ten cases is the agreement of theory and experiment satisfactory. Therefore, an improvement of the theory on this point would be valuable.

In conclusion, I wish to thank Professor E. O. Lawrence, Professor L. W. Alvarez, and Dr. E. Segrè for their interest and assistance in this work; and Mr. E. Nelson for helpful discussions of the theory. This work has been aided by the financial assistance of the Research Corporation.

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#### PHYSICAL REVIEW

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# Artificial Radioactivity of Ti<sup>45</sup>

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A radioactive isotope possessing a half-life of  $3.08\pm0.06$  hours has been produced by four different types of bombardment. Evidence is presented which indicates that the activity should be assigned to Ti<sup>45</sup>. The four nuclear reactions are:

$Ca^{42} + He^4 \rightarrow Ti^{45} + n^1$	$Sc^{45}+H^2 \rightarrow Ti^{45}+2n^1$
$Sc^{45}+H^1 \rightarrow Ti^{45}+n^1$	$Ti^{46} + n^1 \rightarrow Ti^{45} + 2n^1.$

Nuclear spin considerations indicate that a fifth reaction,  $Ti^{46}(\gamma,n)Ti^{45}$ , is improbable, as was verified experimentally. Analysis of cloud-chamber pictures reveals the maximum positron energy to be 1.2 Mev. On a Sargent diagram, the reaction  $Ti^{45} \rightarrow Sc^{46} + e^+$ , is a permitted one. The half-life of  $3.08 \pm .06$  hours for  $Ti^{45}$  is the weighted average of 3.17, 3.10, and 3.02 hours from Sc(p,n); 3.04 hours from Sc(d,2n); 3.17 hours from  $Ca(\alpha,n)$ ; 3.0 hours from Ti(n,2n).

#### INTRODUCTION

THE elements in the titanium region, shown in Fig. 1, have been quite thoroughly examined for radioactive isotopes by Walke and others. Walke<sup>1</sup> has reported many new radioactive isotopes in potassium, calcium, scandium, titanium, and vanadium, of which the two in scandium,  $Sc^{43}$  (4 hours) and  $Sc^{44}$  (4.1 hours) have

<sup>1</sup>H. Walke and others, Phys. Rev. **51**, 143A, 439, 1011A, 1033 (1937); **52**, 669, 777 (1937); **57**, 163, 171, 177 (1940); Proc. Roy. Soc. **A171**, 360 (1939).

<sup>\*</sup> On sabbatical leave from Bethany College, Bethany, West Virginia.



FIG. 2. Examples of  $\beta$ -ray spectrograph films.  $H_{\rho}$  increases to the right. (a) Lines from Se and Kr. (b) K and L lines from Sr<sup>37</sup>, differing by only 2 percent in  $H_{\rho}$ . (c) K and L lines from element 43. (d) K, L, and M lines from 6.7-hr. Cd. (e) Film from radioactive Te isotopes. K, L, and M lines of 86-kev  $\gamma$ -ray, K and L lines from 102 and 107-kev  $\gamma$ -rays.