

The Radioactivity of Se^{73}

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The disintegration of Se^{73} results in four positron groups with end-point energies and relative intensities of 1.68 Mev, 1.2 percent; 1.318 Mev, 87.4 percent; 0.750 Mev, 10.3 percent; and 0.250 Mev, 1.1 percent. Four nuclear gamma-rays are observed having energies of 67.1 kev, 361 kev, 860 kev, and 1.31 Mev. The K/L ratio of the internally converted electrons from the 67.1-kev and 361-kev gamma-ray transitions are 7.6 and 8.6, respectively. K -capture—positron branching ratios are measured as 0.45 for the 1.318-Mev positron transitions, 1.6 for the 0.750-Mev positron transition and estimated as 6 for the 0.250-Mev positron transition. Measurement of the K -Auger electrons indicates a gross K -capture—positron ratio of 0.59.

INTRODUCTION

THE radioactivity of the 7.1-hr transition from Se^{73} to As^{73} has been investigated by means of a medium sized, high resolution, magnetic spectrometer. The original interest in the problem arose from the fact that, on the basis of previously reported work,¹⁻³ the decay was simple with no gamma-rays. It was felt that this simple decay would provide a good test of the feasibility of measuring K -capture—positron branching ratios by means of relative Auger electron and positron intensities.

The results of the spectrometer measurements soon showed that the disintegration is complicated. The positron spectrum is found to be complex and is resolved into four groups with end-point energies of 0.250 Mev, 0.750 Mev, 1.318 Mev, and 1.68 Mev. Their relative intensities are 1.1, 10.3, 87.4, and 1.2 percent, respectively. From a consideration of an end-point energy of 1.68 Mev, a half-life of 425 minutes, and a relative intensity of 1.1 percent, the high energy transition appears to be once-forbidden with a $\ln ft$ of 7.1. The $\ln ft$ for the 1.318-Mev positron transition is 5.6, placing it in the allowed group. Little can be said about the forbiddenness of the two low energy positron groups because the half-life of their initial state is unknown. This half-life must certainly be short compared to 425 minutes because throughout the investigation only one period was observed.

Contrary to the previously reported work, measurements of the gamma-spectra indicate four gamma-rays. Their energies are 67.1 kev, 361 kev, 860 kev, and 1.31 Mev. Investigation of the internal conversion electrons suggests that the 67.1-kev gamma-ray is a magnetic 2^3 -pole transition in the parent Se^{73} and the 361-kev gamma-ray is a magnetic 2^2 -pole transition which follows the 1.318-Mev positron transition. The energies

of the other two gamma-rays correspond to transitions to the ground state following the two low energy positron groups.

Estimates of K -capture—positron branching ratios to several of the levels in As^{73} are made by means of gamma-ray intensities. These ratios are 1.6 for the 0.750-Mev positron transition and 0.45 for the 1.318-Mev positron decay. An estimate of 6 is given for the K -capture—positron branching ratio of the 0.250 positron transition. From the intensity of the K -Auger electrons a gross ratio of 0.59 is obtained for the total number of K -captures to total number of positrons.

EXPERIMENTAL METHOD

The energy measurements were obtained with the aid of a high resolution, 180 degree focusing, 15-cm radius of curvature, shaped magnetic field spectrometer.⁴

For most measurements a 0.6-mg/cm² mica end-window counter was used as a detector. For studying the photoelectrons and the internal conversion electrons of the 67.1-kev gamma-ray and the Auger electrons, a side-window counter was employed. The window of this counter was one double layer of zapon supported by a grid of Lektromesh.⁵ Because of slow diffusion of the counter gas through such a thin window (Approx. 150A thick), the 3-cm total pressure of two parts ethylene to one part argon was automatically maintained by means of a Cartesian Manostat.⁶

The Se^{73} activity was obtained by bombarding powdered and pressed germanium metal with alpha-particles in the cyclotron. The target material was dissolved in aqua regia, boiled to remove the excess nitric acid and diluted. Then selenium metal was precipitated with hydroxyamine hydrochloride similar to the method reported by the California group.⁷ Three milligrams of arsenic carrier and from 60 micrograms to 400 micrograms of selenium carrier were used depending on the type of spectrometer source desired. The decay of the separated selenium activity was monitored and the

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¹ Coward, Pool, McCown, and Woodward, *Phys. Rev.* **73**, 1454 and **73**, 1223(A) (1948).

² H. H. Hopkins, Jr., *Phys. Rev.* **77**, 717 (1950).

³ H. H. Hopkins, Jr., and B. B. Cunningham, *Phys. Rev.* **73**, 1406 (1948).

⁴ J. A. Bruner and F. R. Scott, *Rev. Sci. Instr.* **21**, 545 (1950).

⁵ Langer, Motz, and Price, Jr., *Phys. Rev.* **77**, 798 (1950).

⁶ L. M. Langer and R. D. Moffat, *Phys. Rev.* **80**, 651 (1950).

⁷ W. Wayne Meinke, *Chemical Procedures used in Bombardment Work at Berkeley*, AEC-2738 (1949), p. 93.

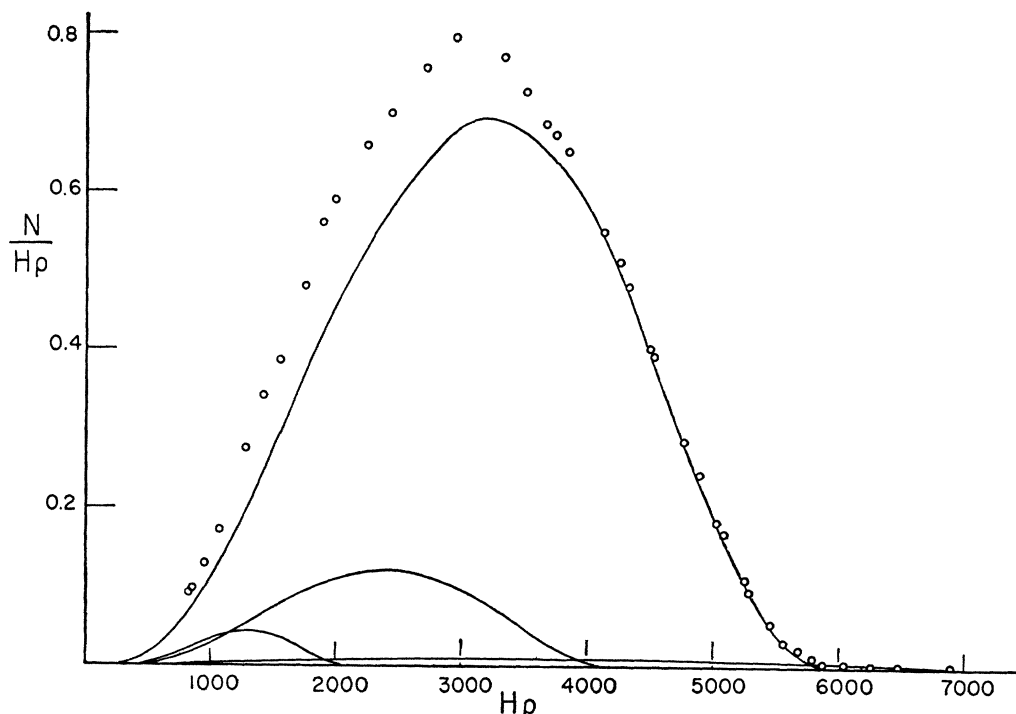


FIG. 1. Momentum distribution of the positrons of Se^{73} .

half-life was found to be 425 minutes. This period was also obtained in the spectrometer by means of repeated runs over all parts of the spectrum.

Two types of positron sources were used. A thin chemically deposited source was made by dissolving the precipitated selenium in one drop of concentrated nitric acid, evaporating almost to dryness, and placing the remaining liquid on an insulin treated zapon film.⁸ The selenium was then uniformly precipitated as the metal by adding a drop of hydriodic acid. The two sources prepared in this way were 0.4 cm wide and 2 cm long with thicknesses of 0.2 mg/cm² and 0.4 mg/cm². These sources were used to investigate the positron distribution above 100 kev and the internally converted electrons from the 361-kev gamma-ray transition.

An extremely thin positron source was made by evaporation. The selenium activity obtained from the chemical separation of the target was dissolved in nitric acid, placed in a platinum boat, and treated with hydriodic acid to precipitate the selenium metal. After heating this boat under an infrared lamp only selenium metal remained. This platinum boat was then used as the filament of a high vacuum evaporator. A masked zapon film was placed three quarters of an inch from the filament. After evacuating and heating the filament, approximately one-eightieth of the activity was evaporated on to the 0.3-cm by 2.0-cm exposed zapon film area. Each of the two sources prepared in this way were estimated to be less than one microgram in weight.

These sources were used in the investigation of the Auger electrons, the internally converted electrons of the 67.1-kev gamma-ray and the low energy positron distribution.

The photo- and Compton electron distributions of the gamma-rays were examined by placing a strong Se activity in a rectangular aluminum boat. A 23-mg/cm² uranium radiator, measuring 0.4 cm by 2.0 cm, covered the front face of this boat. The aluminum was not thick enough to stop all the positrons. This decreased the intensity of the annihilation photolines and Compton distribution.

RESULTS

The momentum distribution of the positrons of Se^{73} is shown in Fig. 1. The complex spectrum has been resolved into four groups on the basis of the Fermi plot shown in Fig. 2. All experimental points in Fig. 2 contain a screening correction to the coulomb wave functions,⁹ and the low energy points were obtained with the thin evaporated sources and normalized to the high energy distribution.

When the positron distribution was first analyzed using data obtained with the chemically deposited sources, a residual positron distribution remained after three Fermi subtractions. Such an excess of positrons at low energy might arise from finite source thickness. Also, since all of the high energy groups had been treated as allowed transitions, the possibility existed

⁸ L. M. Langer, Rev. Sci. Instr. **20**, 216 (1949).

⁹ John R. Reitz, Phys. Rev. **77**, 10 (1950).

that the excess at low energy was caused by the forbidden shape of the 1.68-Mev positron group. To test the first hypothesis the low energy positrons were measured with an extremely thin evaporated source. No change was observed in the low energy positron distribution. To test the second hypothesis the Fermi analysis was repeated with the assumption that the 1.68-Mev group has an "a" shape.^{10,11} This refinement made no difference in the resulting end-point energies and the relative intensity values given in Table I.

The distribution of the Compton and photoelectrons ejected from the uranium radiator is shown in Fig. 3. The experimental points below 200 keV were obtained with a zapon window counter and were normalized with the high energy distribution. *K* and *L* photolines can be seen which correspond to the 361-keV gamma-ray, the 511-keV annihilation radiation, and the 860-keV gamma-ray. In addition a weak Compton distribution can be seen corresponding to the 1.31-Mev gamma-ray. At low energies the three photopeaks correspond to the *L*_{I,II}, the *L*_{III}, and the *M* photolines of uranium resulting from the ejection of the corresponding electrons by a 67.1-keV gamma-ray.

The relative intensities of the four gamma-rays were estimated by comparing relative areas of the photolines and using Gray's formula for the variation of photoelectric cross section with energy. The intensity of the 67.1-keV gamma-ray was approximated by comparing the observed *L* lines with the *L* line of the 361-keV gamma-ray transition. The intensity of the 860-keV gamma-ray was compared to that of the 361-keV transition by means of the *K* lines and 1.31-Mev gamma-ray intensity was estimated from the Compton distribution. The energies and relative intensities of these gamma-rays are indicated in Table I.

Internal conversion electrons were found corresponding to the 67.1-keV and the 361-keV gamma-ray transitions. Figure 4 shows the *K* and *L* internally converted electrons from the 67.1-keV gamma-ray obtained from an evaporated source and using the zapon side window counter. The intensity of this source was normalized to that of the positron distribution up to 250 keV with the zapon side-window counter and then these measurements were overlapped with data using the mica end-window counter as the detector. A comparison of the two conversion lines of the 67.1-keV gamma-ray gives a *K/L* ratio of 7.6 and the area of the *K* line is 25.7 percent of the 1.318-Mev positron distribution.

Because of the low energy of the 67.1-keV gamma-ray, it is possible to measure the *K-L* energy difference between the conversion electrons accurately enough to establish the element in which the conversions occur. The measured *K-L* energy difference in selenium is 11.07 keV, in arsenic 10.48 keV, and in germanium 9.70

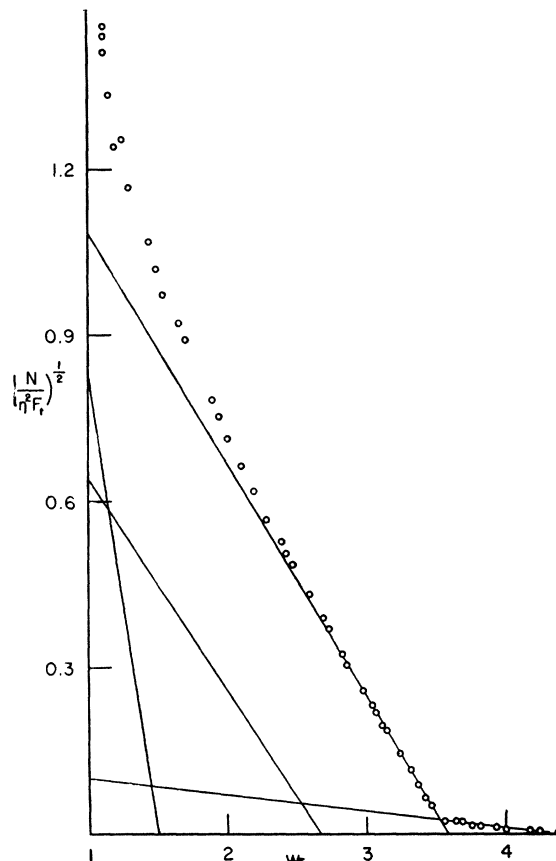


FIG. 2. Fermi plot of the positron spectrum of Se^{73} .

keV, as calculated from Compton and Allison.¹² The measured energy difference of the peaks of the two lines in Fig. 4 is 11.04 keV, indicating that the 67.1-keV gamma-ray transition occurs in Se^{73} .

TABLE I. Beta- and gamma-rays of Se^{73} .

Transition	Energy in keV	Intensity (percent)
Positron 1	250	1.3
2	750	11.8
3	1318	100
4	1680	1.4
Gamma 1	67.1	9.7
		(from photolines)
2	361	163
4	860	11.3
		(from photolines)
5	1310	8
		(from Compton electrons)
Int. conv. of gamma 1		
<i>K</i>	54.45	25.7
<i>L</i>	65.45	3.4
Int. conv. of gamma 2		
<i>K</i>	350.5	1.9
<i>L</i>	360	0.22
<i>K</i> -Auger electrons		
<i>K-2L</i>	9.07	38.5
<i>K-L-M</i>	10.2	

¹⁰ E. J. Konopinski, *Revs. Modern Phys.* **15**, 209 (1943).

¹¹ L. M. Langer and H. C. Price, *Phys. Rev.* **76**, 641 (1949).

¹² A. Compton and S. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand Company, Inc., New York, 1935), p. 784.

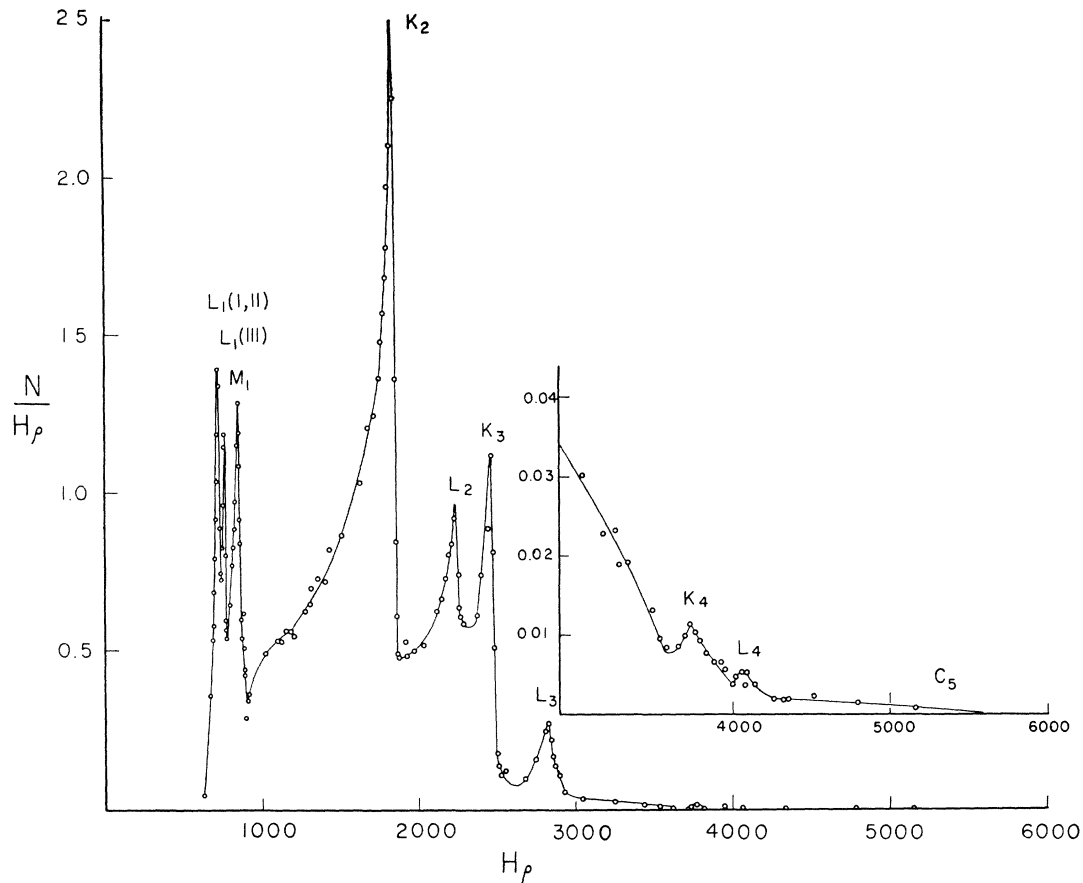


FIG. 3. Compton and photoelectrons ejected from an uranium radiator by the gamma-rays of Se^{73} .

The internally converted electrons from the 361-keV gamma-ray transition are shown in Fig. 5. These lines were obtained with the chemically deposited sources and have been normalized with the positron distribution. The K/L ratio of the areas of the two partially separated lines is 8.6, and the total area of the two lines is 2.14 percent of the 1.318-MeV positron distribution.

Figure 6 shows the K -Auger electron distribution as obtained from an evaporated source and using a zapon side-window counter. The peaks at 9.07 keV and 10.2 keV correspond to the $K-2L$ and $K-L-M$ energies in arsenic. This is in good agreement with the values of 9.09 keV and 10.34 keV as calculated from Compton and Allison.¹² The intensity of the K -Auger electrons is 38.5 percent of the 1.318-MeV positron intensity.

The measured energies and relative intensities of the internal conversion and K -Auger electrons are given in Table I.

DISCUSSION OF RESULTS

The decay scheme of Fig. 7 is proposed for Se^{73} after the following considerations.

All the gamma-rays and all parts of the positron spectrum decay with a half-life of 425 minutes. This indicates that all of these transitions belong to the

same decay. (The half-life of the ground state of Se^{73} is presumed to be very short.)

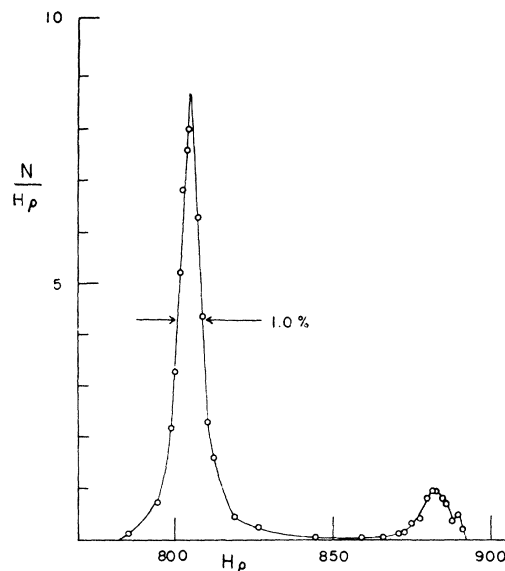


FIG. 4. Internal conversion electrons of the 67.1-keV gamma-ray of Se^{73} .

The end-point energies of 1.68 Mev and 1.318 Mev, and the relative intensities of 1.4 percent and 100 percent give $\ln ft$ values of 7.1 for the 1.68-Mev positron group and 5.6 for the 1.318-Mev positron group. This indicates that the high energy positron transition is once-forbidden while the intense 1.318-Mev positron transition is allowed.

The energy of the 361-keV gamma-ray equals the difference between the end-point energies of the two high energy positron groups. The K/L ratio of 8.6 for the internal conversion electrons of this gamma-ray suggests that the 361-keV gamma-ray is magnetic quadrupole radiation.¹³ This would conform to a spin change of two and no parity change. This type of radiation is compatible with the forbiddenness of the two positron transitions.

The $K+L$ internal conversion coefficient for a 361-

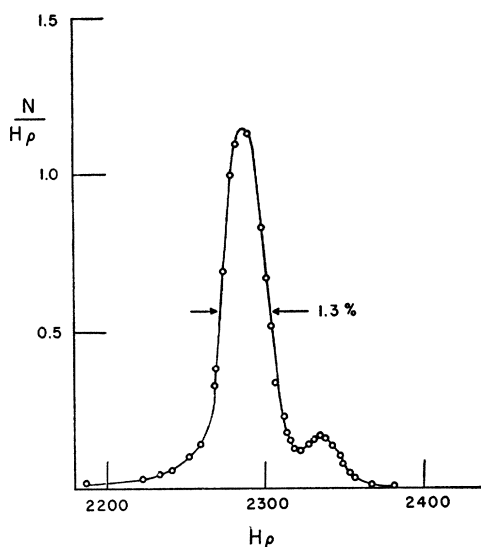


FIG. 5. Internal conversion electrons of the 361-keV gamma-ray of Se^{73} .

keV magnetic quadrupole gamma-ray is 0.0132 as calculated from an extrapolation¹⁴ to $Z=33$ in Rose's tables.¹⁵ This predicts a total relative population of 163 percent for the final state of the 1.318-Mev positron transition. This is compared with 100 percent for the positron intensity to this same level. The excess of 63 percent arises from K -capture to this level and from feeding by a 0.500 Mev cross-over gamma-ray (indicated by dotted lines in Fig. 7). The photolines of this gamma-ray are masked by that of the annihilation radiation.

The intensity of the 67.1-keV gamma-ray can be estimated in the following way. The calculated ratio of the relative intensity of the 361-keV gamma-ray to that

¹³ N. Tralli and I. S. Lowen, Phys. Rev. **76**, 1951 (1949).
¹⁴ E. Segre and A. C. Helmholz, Revs. Modern Phys. **21**, 271 (1949).
¹⁵ Rose *et al.*, unpublished data.

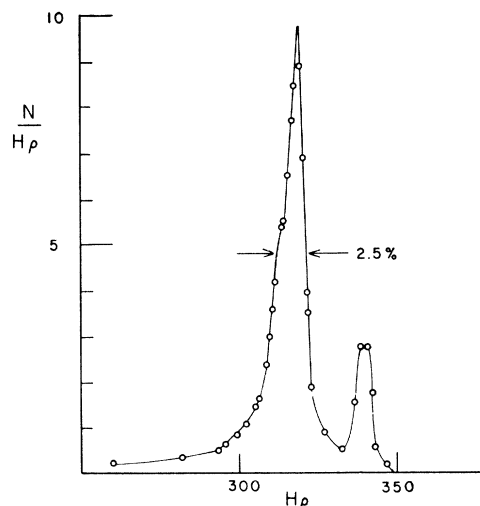


FIG. 6. K -Auger electron distribution of Se^{73} .

of the 67.1-keV gamma-ray, as obtained from the L photoelectron areas, is

$$N_{\gamma}(361)/N_{\gamma}(67.1) = 100/6.$$

The observed ratio of the K internally converted electrons arising from these two gamma-rays is,

$$N_K(67.1)/N_K(361) = 13/1.$$

Since $\beta_K = N_K/N_{\gamma}$, then

$$\beta_K(67.1) = \beta_K(361) \frac{N_{\gamma}(361)N_K(67.1)}{N_{\gamma}(67.1)N_K(361)},$$

and $\beta_K(67.1)$ is approximately three. Using this measured K -internal conversion coefficient, the population of the final state of the 67.1-keV gamma-ray is 37.6 percent of the 1.318-Mev positron transition. This

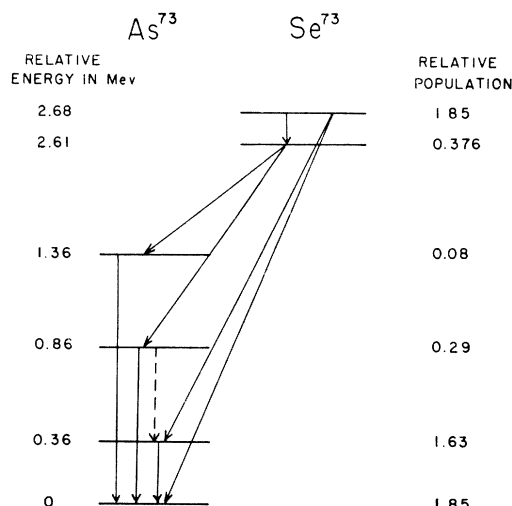


FIG. 7. Disintegration scheme for the Se^{73} - As^{73} decay.

intensity is compatible with the other gamma-ray intensities (as given in Table I), if the intensity of the 500-keV cross-over gamma-ray is 18 percent.

Arguments for the magnetic octipole character of the 67.1-keV gamma-ray are as follows: (1) the $K-L$ energy difference between the internally converted electrons from this gamma-ray is 11.04 keV which is within experimental error of the 11.07 keV $K-L$ difference measured¹² for selenium. According to the shell model¹⁶ a metastable state may be expected in this isotope, since many have been observed¹⁴ in the region of odd number of neutrons or protons between 38 and 50. (2) The measured K/L ratio of 7.6 is compatible with magnetic octipole radiation. (3) If, as indicated in Fig. 7, the excited state of Se^{73} decays by four modes, one would expect the decay constant to be

$$\lambda = \lambda(\text{positrons}) + \lambda(K\text{-capture}) + \lambda(\text{gamma-ray}) + \lambda(\text{int. conv.}),$$

and if the ratio

$$\frac{N(\text{beta-decay})}{N(\text{gamma-decay})} = \frac{\lambda(\text{positrons}) + \lambda(K\text{-capture})}{\lambda(\text{gamma-ray}) + \lambda(\text{int. conv.})}$$

can be measured then an estimate of the mean life of the gamma-transition can be made because λ is experimentally measured. This ratio of beta-decay to gamma-decay can be obtained from the relative intensities of the 361-keV gamma-ray (excluding the 18 percent arising from the 500-keV cross-over gamma-ray) and the 67.1-keV gamma-ray. This ratio of $1.47/0.376 = 3.9$ closely approximates the ratio of beta-to gamma-ray intensities decaying from the excited state of selenium. Putting the decay constant λ in terms of the half-life and including the experimental ratio of beta- to gamma-decay, the half-life of the excited state of Se^{73} becomes:

$$t_{1/2} = \frac{1}{4.9} \frac{\ln 2}{\lambda(\text{int. conv.}) + \lambda(\text{gamma-ray})} = \frac{1}{4.9} \cdot t_{1/2}(\text{gamma-decay}).$$

Since $t_{1/2} = 25,500$ sec this gives measured gamma-ray half-life of 125,000 seconds for the 67.1-keV gamma-ray transition. Segre and Helmholtz¹⁴ give a half-life of 50,000 seconds for a magnetic octipole transition of this energy. ‡

An estimate of the K -capture—positron ratios to several of the levels in arsenic can be made by means

¹⁶ M. G. Mayer, Phys. Rev. **78**, 16 (1950).

‡ *Note added in proof:*—Since this paper was written, an article by M. Goldhaber and A. W. Sunyar (Phys. Rev. **83**, 906 (1951)) has been brought to the attention of the author. According to their article, the measured gamma-ray half-life for the 67.1-keV transition is in good agreement with what one would expect for a transition involving a spin change of 3. The measured K/L ratio for the 67.1-keV transition is not in good agreement with their values for either magnetic or electric octipole radiation.

of the relative intensities of the positron groups and the gamma-rays as given in Table I. The intensity of the 361-keV gamma-ray fed by the 1.318-MeV beta-decay is 145 percent while the intensity of the associated positron group is 100 percent. This indicated an excess of 45 percent to be accounted for by K -capture and thus gives a K -capture—positron ratio of 0.45 for the 1.318-MeV transition. This ratio compares favorably with 0.42 as predicted by the Fermi theory.¹⁷ The sum of the intensities of the 0.860-MeV gamma-ray and the 0.500-MeV cross-over gamma-ray is in excess of the 0.750-MeV positron intensity by 18 percent of the 1.318-MeV positron intensity. Since the 0.750-MeV positron intensity is 11.8 percent of the 1.318-MeV positron intensity, this gives a K -capture—positron ratio of 1.6 for the 0.750-MeV positron transition. Theoretically, this ratio should be 2.5. In the case of the 250-keV positron transition, the estimated K -capture to positron ratio is 6, but the Fermi theory predicts 100. Because of the uncertainty in the measurement arising from the very low intensity of this transition, this discrepancy does not appear serious.

An estimate of the gross K -capture to positron decay can be obtained from the intensity of the Auger electrons. In order to do this the fluorescent yields for selenium and arsenic must be known. The values of 0.591 for arsenic and 0.620 for selenium were obtained by a Z extrapolation¹⁸ of the fluorescent yield of krypton recently obtained by West and Rothwell.¹⁹ Before calculating the relative gross K -captures from the total Auger intensity, the Auger electrons arising from K -internally-converted electrons of the gamma-rays must be obtained. These are

$$N_A(67.1) = W_A(\text{Se})N_K(67.1) = 0.380 \times 25.7 = 9.8$$

plus

$$N_A(361) = W_A(\text{As})N_K(361) = 0.409 \times 1.92 = 0.8,$$

where N_A is the relative number of Auger electrons arising from the K -conversion of the indicated gamma-ray, W_A is the K -Auger electron yield (one minus the K -fluorescent yield), and N_K is the relative intensity of the K -internal-conversion electrons.

When these Auger electrons are subtracted from the total Auger intensity, the remainder $N_A = 0.279$ is due only to K -capture.

If this is divided by the K -Auger electron yield in arsenic the result of 63.2 percent is equal to the total relative number of K -captures as compared with the 1.318-MeV positron intensity. In terms of the total positron intensity the gross K -capture—positron ratio is 0.59.

¹⁷ E. Feenberg and G. Trigg, Revs. Modern. Phys. **22**, 399 (1950).

¹⁸ H. S. W. Massey and E. H. S. Burhop, Proc. Roy. Soc. (London) **A153**, 661 (1936).

¹⁹ D. West and P. Rothwell, Phil. Mag. **41**, 873 (1950).

CONCLUSIONS

The decay of Se^{73} is complex. With the help of the Fermi theory the positron distribution has been resolved into four groups with end-point energies and relative intensities as given in Table I. Four gamma-rays have been observed. Their energies and relative intensities are given in Table I. The 67.1-keV gamma-ray has been found to be a magnetic octipole transition in the parent selenium. The 361-keV gamma-ray follows the 1.318-MeV positron group and is magnetic quadrupole radiation. The other gamma-rays and positron groups have energies and intensities compatible with the decay scheme proposed in Fig. 7.

Estimates of K -capture—positron branching ratios have been made for several of the positron transitions.

For the intense 1.318-MeV positron group this ratio is 0.45 as compared to the theoretical value of 0.42. The measured ratio for the weak 0.750-MeV positron group is 1.6 and the theoretical value is 2.5, and for the very weak 0.250-MeV positron transition the measured K -capture—positron ratio is estimated as 6 and the corresponding theoretical value is 100. From the relative intensity of the K -auger electrons, a value of 0.59 was obtained from the gross ratio of K -capture to positrons.

The author wishes to express his gratitude to Professor Lawrence M. Langer for his valuable advice and encouragement. He would also like to thank Professor M. B. Sampson and the cyclotron crew for the numerous bombardments necessary for this work and Mr. E. Plassmann for his assistance in making some of the measurements.

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A Canonical Field Theory with Spinors*

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In this paper, we have constructed a general field theory in covariant form which incorporates the usual covariant generalization of Dirac matrices. The field equations are derived from a lagrangian that is a second-order differential covariant (a scalar density of weight 1) constructed in the covariant spinor formalism by the same method that in riemannian geometry leads to the curvature tensor. It is possible to show that, in spite of the apparently greater wealth of geometrical elements, this theory is completely equivalent to the general theory of relativity. The field equations satisfy the usual differential identities and, in addition, "spin" identities; there are four "strong" conservation laws which can be used to obtain equations of motion for singularities. Since we do not know at present whether the equivalence with the theory of relativity may not be lost in the process of quantization, we consider eventual quantization desirable and have, in this paper, converted the theory into the canonical form.

INTRODUCTION

IN a series of papers, Bergmann and co-workers¹⁻³ developed the theory of canonization of covariant field theories, in the hope that some of the difficulties in quantum field physics might be overcome by the adoption of the "best" classical (nonquantized) field theory and its subsequent quantization.

The question now arises which theory is to be considered the "best" one. We could consider Einstein's theory of gravitation (with the electromagnetic field included). In that theory the laws of physics are

generally covariant, i.e., unchanged by all types of coordinate transformations for which the jacobian of the transformation is non-zero. There are other possibilities, for example, the recent theory of Einstein,⁴ in which he attempts to unify the gravitational and electromagnetic fields. This theory also assumes the basic laws of physics to be generally covariant.

There are, however, indications that the basic laws of physics contain spinors as well as tensors. In the Dirac theory of the electron, anticommuting quantities arise with transformation laws different from those of tensors. The success of Dirac's theory of the electron and, among others, the ample evidence of atomic spectra, furnish strong indications that the basic laws of physics contain spinors.

In this paper we shall develop a classical field theory which is generally covariant and contains spinors. We

* This paper incorporates the results of the Ph.D. dissertation of the first author, accepted by the Graduate School, Polytechnic Institute of Brooklyn.

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¹ P. G. Bergmann, *Phys. Rev.* **75**, 680 (1949).

² P. G. Bergmann and J. H. M. Brunings, *Revs. Modern Phys.* **21**, 480 (1949).

³ Bergmann, Penfield, Schiller, and Zatzkis, *Phys. Rev.* **80**, 81 (1950).

⁴ A. Einstein, *The Meaning of Relativity* (Princeton University Press, Princeton, New Jersey, 1950), third edition.