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The Alpha-Ray Spectrum of Polonium*

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Measurements of the alpha-ray spectra of polonium mounted on nickel, silver, and cadmium by means of a 180-degree-focusing magnetic spectrograph are reported and discussed. The weak line series reported by Chang was not apparent. The hypothesis is proposed that the alpha-ray spectra seen to date have not been caused by the element polonium, but by the diffusion of the polonium into the mounting metal.

I. INTRODUCTION

THE nucleus of the element polonium has been shown to emit gamma-radiation of 0.80-Mev energy and also softer gamma-radiation whose energy has not been confirmed.[1–4] The existence of multiple gamma-rays from polonium has been confirmed by the detection of gamma-gamma coincidences in this laboratory by M. L. Wiedenbeck. An alpha-ray line at 5.300 Mev has been accurately measured.[5,6] Its intensity has been determined to be about 100,000 times that of the gamma-radiation.[2,3]

In 1946, W. Y. Chang[7] reported the discovery of a long series (12 actually measured) of very weak alpha-ray lines spaced at about 0.1-Mev intervals below the 5.300-Mev line. He found their intensities to be about 10,000 times weaker than that of the 5.300-Mev line, inconsistent with the gamma-ray data, as pointed out by Feather.[8] Chang recognized that the existence of the weak line series was in severe conflict with the present theory of alpha-ray emission in that if both are taken at their face value, they imply large spin differences (of the order of 10) between the alpha-ray transitions, and spin differences of this order have never been observed elsewhere in alpha-ray spectra. He also plotted log λ *versus* energy for the line series and showed that the sequence diverged sharply from the Geiger-Nuttall relation of the uranium family.

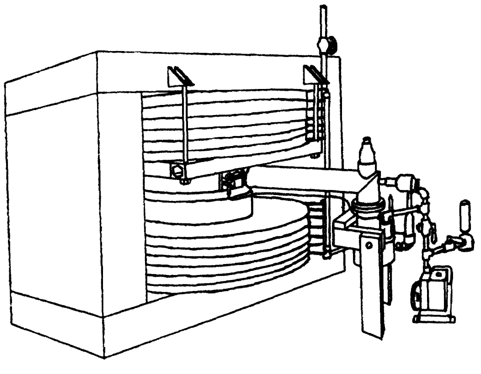

FIG. 1. View of spectrograph.

II. SPECTROGRAPH

In order to investigate the alpha-ray spectrum of polonium further, a 180-degree-focusing magnetic spectrograph of 24-inch pole diameter was designed and constructed. A general view is shown in Fig. 1 and a scale drawing of the vacuum chamber in Fig. 2.

The design was based on Kai Siegbahn's theory of beta-ray spectrographs.[9] For the resolving power R, defined as

$$R=\delta(Hr)/Hr,$$

he derives the formula

$$R=Q/r+2(1-\cos\alpha) \qquad (1)$$

and the relation for greatest intensity at a given resolution:

$$Q\cong 2r(1-\cos\alpha), \qquad (2)$$

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** Now at Yale University, New Haven, Connecticut.

[1] I. Curie and F. Joliot, J. de phys. et rad. **2**, 20 (1931).

[2] H. C. Webster, Proc. Roy. Soc. **A136**, 428 (1932).

[3] W. Bothe, Zeits. f. Physik **96**, 607 (1935).

[4] S. DeBenedetti and F. H. Kerner, Phys. Rev. **71**, 122 (1947).

[5] S. Rosenblum and G. Dupouy, J. de phys. et rad. **4**, 262 (1933).

[6] W. B. Lewis and B. V. Bowden, Proc. Roy. Soc. **A145**, 235 (1934).

[7] W. Y. Chang, Phys. Rev. **69**, 60 (1946).

[8] N. Feather, Phys. Rev. **70**, 88 (1946).

[9] Kai Siegbahn, Arkiv. f. Mat., Astr. o Fys. **30A**, No. 20 (1944).

where H is the magnetic field strength, r the radius of curvature of the particle path, Q the source width, and α the half-angle of divergence of the particle paths in the horizontal plane. $\mathrm{Sin}\alpha = s/d$, where $2s$ is the width of the slit and d is the distance from source to slit. The constants of the spectrograph were $d=25.42_5$ mm $\pm 0.02_5$ mm, $2s=1.72_7$ mm$\pm$0.05 mm, $Q=0.32_3$ mm$\pm 0.01_2$ mm, which give at the maximum radius of 260 mm

$$R=0.23 \text{ percent.}$$

The alpha-rays were detected with Eastman nuclear track plates (type NTA) inclined at a 45-degree angle to the plane of the particle paths and accurately located within the spectrograph in order that positions on the plates could be correlated with positions in the spectrograph.

The inclination of the plates necessitated corrections to the measured plate positions, as may be seen from Fig. 3, where XYZ is a space coordinate system with OZ the vertical axis and OY horizontal in the plane of the spectrograph slit midway between the top and bottom of the vacuum chamber. The monoenergetic image of a line source is a line, BO, called the "track line." This geometrical correction allows us to correct the measured value of a, the distance from the slit to the point concerned, to that value which would have been observed if φ had been zero:

$$\delta a = (y_0 - y)(\cos\theta)d/a, \qquad (3)$$

where y_0 is the value of the coordinate across the plate in the plane of the slit, y is the point coordinate, θ is the plate inclination angle (45 degrees), and a is the distance from the image to the slit.

A simple calculation shows that if the position of the plate is known to within 1 mm and the position of the line in the plate with reference to the end of the plate to within 0.25 mm, the momentum will be known to within 0.1 percent. The spectrograph construction and method of plate measurement enabled them to be measured to within 0.1 mm. Expansion of the "frame" (see Fig. 2) introduced no error because of the use of the quartz rod which determined the slit-plate distance.

The vacuum chamber had a brass wall and iron lids, top and bottom, leaving $1\frac{3}{4}$ inches clear inside. It was lined with 0.010-inch cellulose acetate. The "trunk" and "slide" permitted daylight loading of the spectrograph. All parts in the vacuum chamber were made of 24S-T aluminum alloy, except for the quartz rod and brass screws.

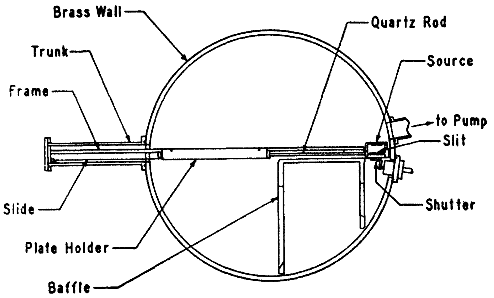

FIG. 2. Spectrograph vacuum chamber, to scale.

The electromagnet was powered by a motor-generator controlled by a current-regulating circuit (a slight modification of the circuit devised by Lawson and Tyler[10]) which maintained the magnet current within very close limits indefinitely. Simple annular shims gave the magnetic field distribution shown in Fig. 4, varying only 0.4 percent over the 260-mm maximum radius at the field strength used, 15,000 gauss.

III. EXPERIMENTAL PROCEDURE

The sources consisted of polonium metal deposited by the Canadian Radium and Uranium Corporation[11] on one $1\frac{7}{16}$-inch by 0.020-inch edge of a strip of metal $1\frac{7}{16}$ inches by 0.020 inch by $\frac{1}{8}$ inch. We were informed that "deposition was made spontaneously by immersion in a polonium solution relatively free from metallic ions. The plating time was approximately 6 hours" for a 0.1-mc source. The sources are listed in Table I.

The mountings were made at this laboratory

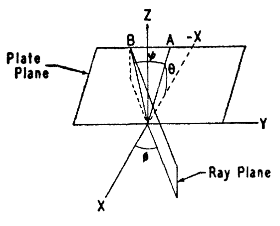

FIG. 3. Track-plate geometry.

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[10] J. L. Lawson and A. W. Tyler, Rev. Sci. Inst. **10**, 304 (1939).

[11] 630 Fifth Avenue, New York 20, New York.

TABLE I. List of sources used.

Source	Strength	Mounting	Prepared
B	0.25 mc	nickel	Dec. 8, 1947
E	0.1 mc	silver	Jan. 19, 1948
F	0.1 mc	cadmium	Jan. 26, 1948

as follows: For source *B*, machined from nickel sheet which presumably had been rolled into sheet form; for source *E*, rolled down to 0.020-inch from 0.065-inch sheet and machined; and for source *F*, rolled down to 0.020 inch from a piece about 0.065 inch thick cut from a cast rod. Each was carefully cleaned of burrs and the edge to be plated polished on a buffing wheel, and each appeared smooth and clean under a 15× glass both before and after coating. In the spectrograph the source was masked to 0.012-inch width by two aluminum jaws.

Exposures were made at pressures of less than 0.10 micron. Immediately after the completion of exposure the plates were developed on the time-temperature curve passing through 5 minutes at 68 degrees Fahrenheit in D-11 or 2 minutes at 68 degrees Fahrenheit in D-8. All plate handling and processing was done in total darkness.

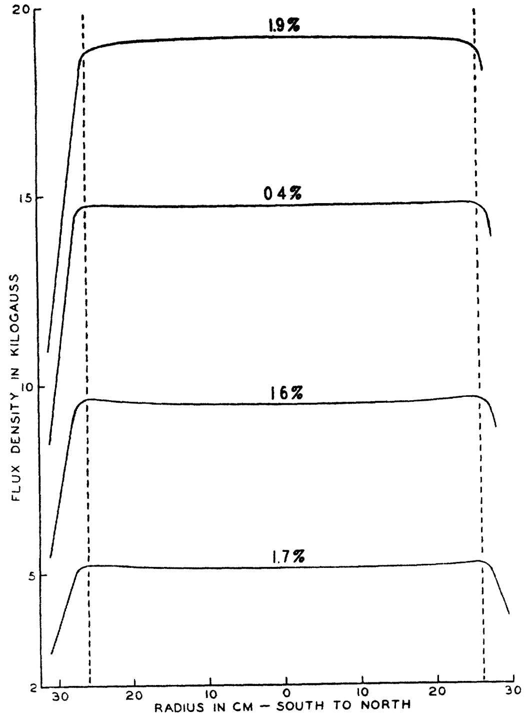

FIG. 4. Magnetic field distribution.

The data were obtained by measuring the alpha-ray track density as a function of position on the plate. This was done by sampling the tracks by dark-field microscopic examination at systematic intervals down the plate. Areas were marked by an ocular grating of 49 squares calibrated by a stage micrometer. Plate coordinates were taken from a special calibrated mechanical stage with verniers on both motions reading to 0.1 mm. The appearance of the microscope field may be seen in Fig. 5, taken from plate No. 39 at 5.15 Mev. The magnification used throughout was about 150×, and counting was done over the central vertical strip of seven squares (each about 0.1 mm by 0.1 mm).

Most of the tracks were parallel and of the same length. These were the tracks counted. The others, caused by contamination of the vacuum chamber or scattering, were readily disregarded because of their different range and direction. The large blobs were dirt on the emulsion. For the spectra studied, the track density varied over a range of 7000 to 1—too great a range to be counted on one plate, so three accurately measured exposure times were used for each spectrum and the data combined. It is conservatively estimated that the error in counting tracks was about 2 percent.

IV. DATA

(a) Reduction

The original data were numbers of tracks per square as a function of the plate coordinates x and y. It was necessary to reduce them to track densities per hour of exposure as a function of alpha-particle energy. From the geometry and constants of the spectrograph:

$$r=(1/2)(521.3-\delta a)-x/2, \tag{4}$$

where δa is given by formula (3). From these values of r, the values of the momentum Hr were obtained using:

$$Hr=r\cdot(Hr)_0/r_0,$$

where $(Hr)_0$ was the known value for the 5.300-Mev line, $3.316_5\times10^5$ gauss cm. The quantity r^0 was the measured position of the 5.300-Mev line.

In order to convert from momentum to energy, a table was prepared, accurate to four digits, over the range 3.513 Mev to 5.343 Mev, based on the general relativistic relation:

$$E_t = c(p^2 + m^2c^2)^{\frac{1}{2}} \text{ ergs.}$$

This relation, the equation between the magnetic and centrifugal forces on the particle, and the values of the physical constants[12] give the generating formula:

$$E = 4.822_2 \cdot 10^{-11} \cdot (Hr)^2 - 3.12 \cdot 10^{-25} \cdot (Hr)^4 \text{ Mev.}$$

The track density data were converted from tracks per square to tracks per square millimeter per hour of exposure, corrected for source decay, and for the change from momentum space to energy space.

(b) Polonium on Nickel

To correlate this data with that of Chang,[7] the first spectrum studied was that of 0.25 millicurie of polonium mounted on nickel. The data were taken from plates No. 34, 35, and 36, listed in Table II. (The data from plate No. 28 confirmed the absence of alpha-particles of energy higher than 5.300 Mev. At the field strength used, alpha-particles up to 11.4 Mev would have been seen.)

The reduced data are plotted in Fig. 6. Each point represents a counted field. The data from plate No. 34 are those below 5.2 Mev and to the right of the drawn-in curve at 5.3 Mev. The data from plates No. 35 and 36 are plotted to an intensity scale reduced 500 times and connected by a solid line. Plate No. 39 was exposed as a check. Data at 4.0 Mev and 5.0 Mev fell within the data plotted in Fig. 6.

(c) Polonium on Silver

In order to investigate the possibility that the spectrum might be influenced by the mounting, the next source studied was 0.1 millicurie of polonium mounted on silver. Data were obtained from plates No. 44, 46, and 47, with No. 45 providing a satisfactory check. The exhaustive counting done for the nickel-mounted source was not done for these plates, and the data are presented in Fig. 7 with the expected mean deviations indicated as calculated from the formula

$$\delta N = N/u^{\frac{1}{2}},$$

where u is the total number of tracks counted to determine N.

TABLE II. List of plates exposed.

Plate No.	Date	Source	Exposure	Current	Developer
28	12/12/47	*B*	1:55 hr.	148*A*	D-11
34	12/16/47	*B*	25:00 hr.	50*A*	D-11
35	12/17/47	*B*	16 min.	50*A*	D-11
36	12/18/47	*B*	101 sec.	50*A*	D-11
39	12/19/47	*B*	25:00 hr.	50*A*	D-8
44	1/21/48	*E*	25:00 hr.	50*A*	D-8
45	1/23/48	*E*	25:00 hr.	50*A*	D-8
46	1/24/48	*E*	3600 sec.	50*A*	D-8
47	1/25/48	*E*	300 sec.	50*A*	D-8
48	1/28/48	*F*	25:00 hr.	50*A*	D-8
49	1/30/48	*B*	25:00 hr.	50*A*	D-8

(d) Polonium on Cadmium

Data from 0.1 millicurie of polonium mounted on cadmium were obtained from plate No. 48. The results are plotted in Fig. 8.

Finally plate No. 49 was exposed to the nickel-mounted source, and its data showed that there had been no change in the spectrograph characteristics during the course of the experiments.

V. CONCLUSIONS

(a) Discussion of Data

The spectrum on nickel, of which the most extensive study was made and for which the

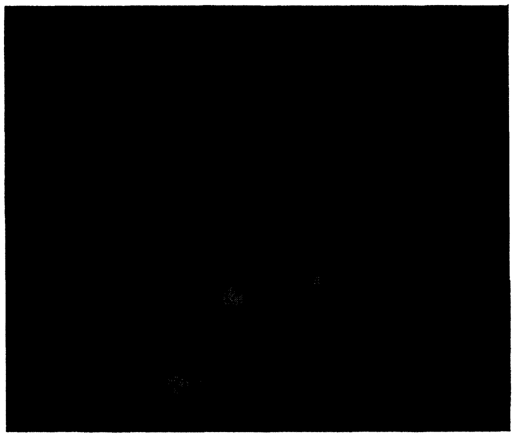

FIG. 5. Microscope field showing tracks from polonium on nickel at 5.15 Mev.

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[12] R. T. Birge, Rev. Mod. Phys. **13**, 233 (1941).

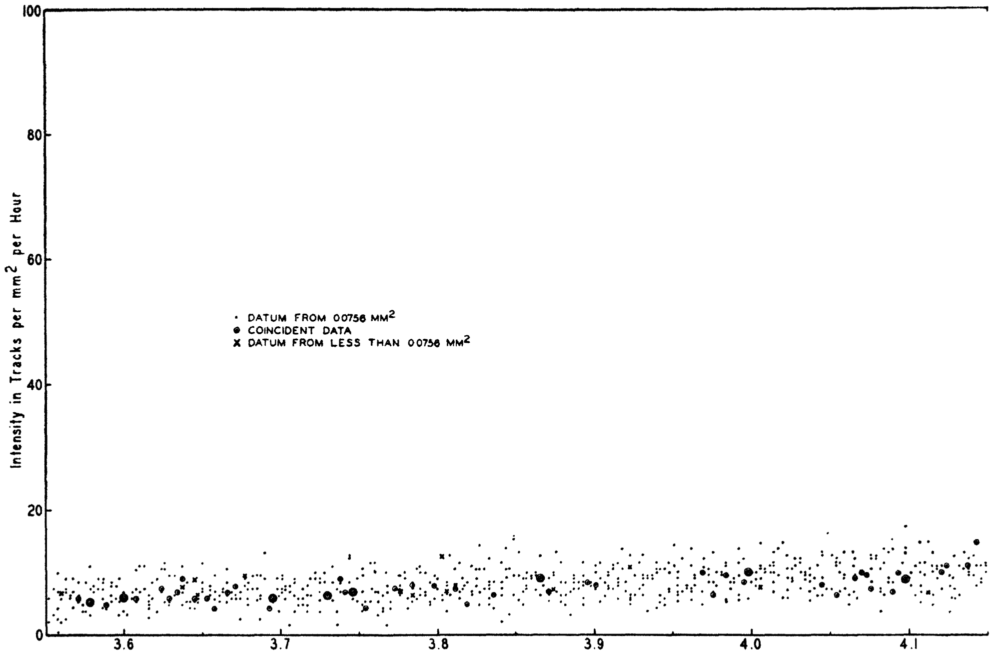

FIG. 6(a). Alpha-ray spectrum of polonium mounted on nickel.

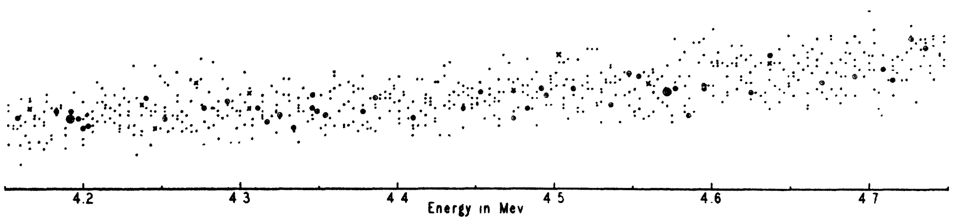

FIG. 6(b).

statistics are very good, shows no evidence of the weak line series discovered by Chang. The other spectra are similar in this regard, but differ considerably in detail. On nickel, the spectrum has a line at 5.300 Mev and falls gradually to low values on the low energy side.

On silver, the 5.300-Mev line has the same half-width, but shows a striking plateau from 5.1 Mev to 5.25 Mev. On cadmium, the plateau begins at a higher energy and intensity, has greater slope, but ends at about the same energy as on silver. (The ratios of maximum intensities are not believed significant since the source strengths were not known accurately.)

Thus, it can readily be seen that the mounting metal had a strong influence on the spectrum obtained. The structural data[13] on the metals concerned are listed in Table III.

The question arises, why did the weak line series which was so clearly apparent in the data of Chang not appear here? The resolution of the instrument was adequate since the half-width of the 5.300-Mev line as measured by this instrument was 0.012 Mev and the half-widths of the lines seen by Chang were about 0.03 Mev. The examination intervals were close enough to resolve these lines also. Straggling in the source or scattering in the vacuum chamber might have raised the intensity at the lower energies so as

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[13] *Handbook of Chemistry and Physics* (Chemical Rubber, Cleveland, 1939), twenty-third edition, p. 1590.

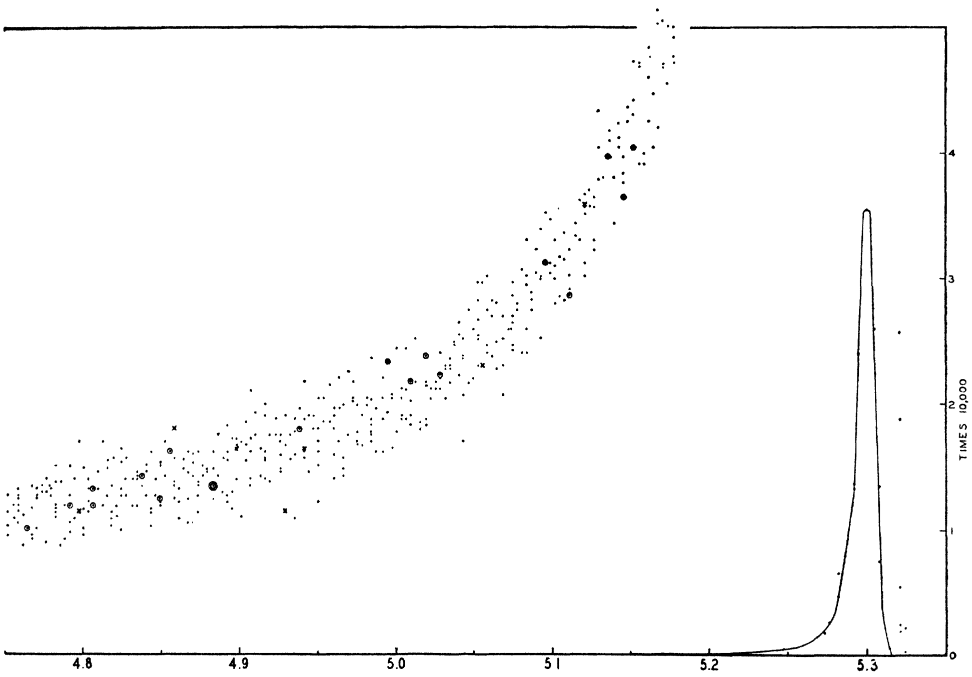

FIG. 6(c).

to cover up the weak line series. However, Chang states[7] that "the intensities at 0.25 Mev or more below the energy of the main group are less than 0.1 percent of the maximum intensity," and in Fig. 6, at 5.05 Mev, the intensity is 0.15 percent of maximum. Thus we see that the data are strictly comparable, and we are driven to the conclusion that there was no instrumental error, but that the weak line series seen by Chang did not come out of the sources used for these experiments.

(b) Suggested Explanation

The following hypothesis is proposed: None of the alpha-ray spectra seen here nor that seen by Chang is the true alpha-ray spectrum of polonium, but, with the exception of the 5.300-Mev line, all the observed spectra are due to the diffusion of the polonium into the metallic mounting.

This hypothesis would eliminate the theoretical difficulty regarding the spins and Feather's[8] objection to the intensities since it would indicate that the true spectral line intensities are of a lower order of magnitude than the intensities obtained to date and so have never been seen. It is substantiated by a paper of A. B. Focke[14] in which he reported a study of the alpha-radiation from polonium introduced into bismuth as an impurity in single crystals. He concluded that "polonium is segregated into small regions which have nearly regular spacings when viewed in several different directions."

It seems probable that when polonium is introduced into a crystal by diffusion it should also be segregated. This would cause just such regularly spaced weak alpha-ray lines as were seen by Chang. A further fact tending to confirm this explanation is the shape of the lines seen by Chang. This type of spectrograph inherently gives a line shape which is very sharp on the high energy side. The lines in Chang's paper are symmetrical and not at all sharp. This agrees with the work of Focke, since there would surely be a symmetrical distribution of the polonium atoms within the crystal about each point of segregation.

Another check is a calculation of the range of 5.300-Mev alpha-particles in nickel correspond-

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[14] A. B. Focke, Phys. Rev. **46**, 623 (1934).

ing to the 0.1-Mev spacing of the line series. It is 10^{-5} cm which is the order of magnitude of the spacing of the "slip-bands" of metals[15] and agrees with the spacings found by Focke.[14]

This argument would make it appear that the line series should have been found in the data of *this* paper. However, all the mountings used in these experiments were rolled during preparation so that each was thoroughly cold-worked, and the crystals were oriented at random so that any such effect would be expected to average out to nothing.

It may be possible to use polonium salts mounted on plastic films to obtain the true alpha-ray spectrum, and such work is being undertaken at this laboratory. If the above explanation proves correct, the technique of alpha-ray spectrography may be a useful method for the investigation of the structure of metals since it would enable the structure to be studied

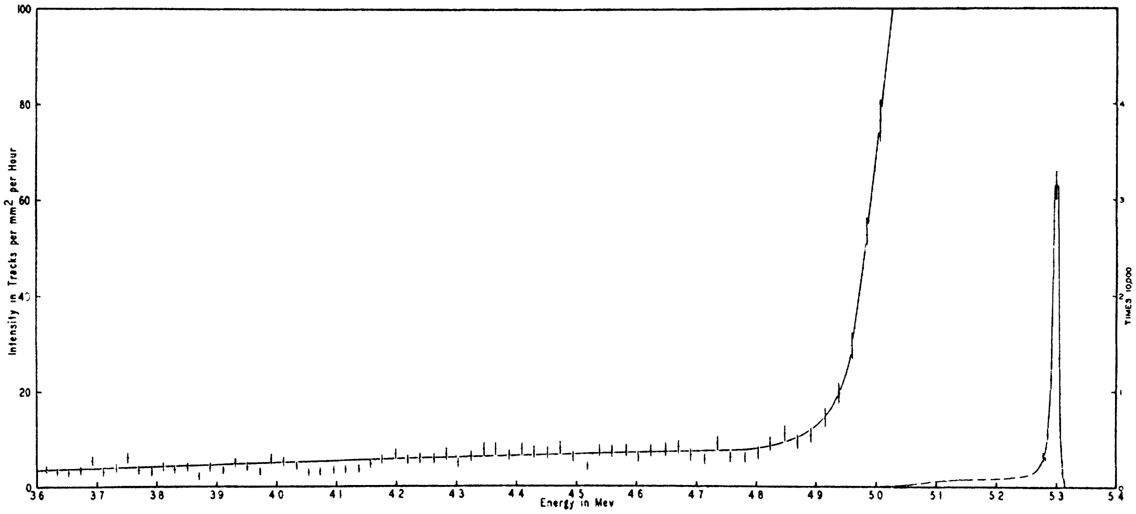

FIG. 7. Alpha-ray spectrum of polonium mounted on silver.

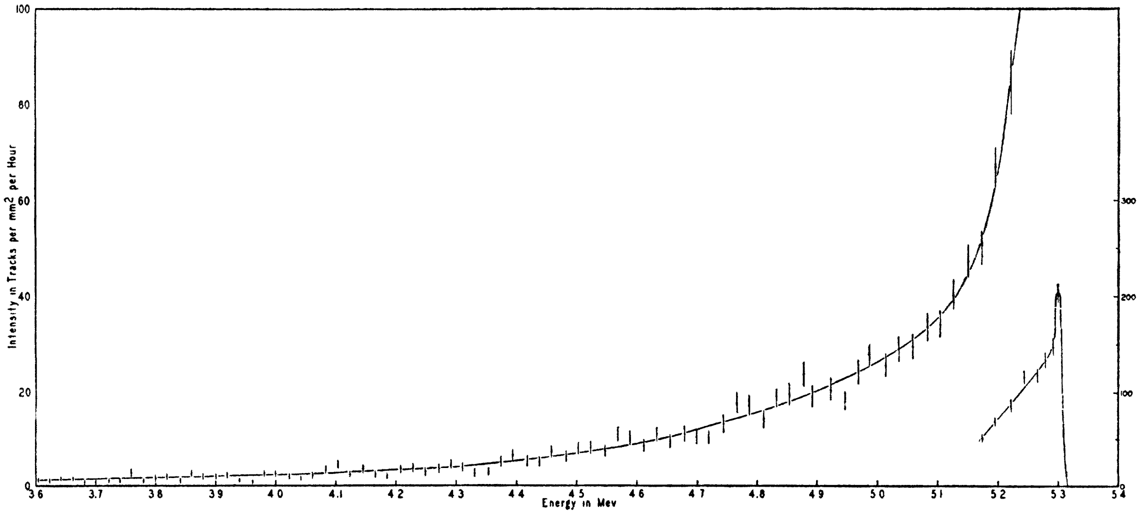

FIG. 8. Alpha-ray spectrum of polonium mounted on cadmium.

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[15] F. Seitz, *The Physics of Metals* (McGraw-Hill Book Company, Inc., New York, 1943), p. 74. Also R. D. Heidenreich and W. Shockley, J. App. Phys. **18**, 1029 (1947).

without the application of stress or the introduction of a significant quantity of the radioactive impurity.

VI. ACKNOWLEDGMENTS

The author wishes to express his appreciation for the great interest and many helpful suggestions of Professor M. L. Wiedenbeck and for the benefit of discussions with Professors G. E. Uhlenbeck, H. R. Crane, D. M. Dennison, and G. Y. Rainich. It is also a pleasure to thank the University of Michigan for granting him a Horace H. Rackham Special Fellowship during the work on this problem.

TABLE III. Structural data of metals concerned.

Metal	System	Lattice constants
Po	monoclinic	7.42, 4.29, 14.10
Ni(β)	cubic, face-centered	3.517
Ag	cubic, face-centered	4.0776
Cd	hexagonal, close-packed	2.97, —, 5.61

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Statistical Error in Absorption Experiments

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In certain exponential absorption experiments, notably measurements of cross sections by transmission, it is important to achieve minimum statistical error in a limited time or to minimize the counting time required to measure the absorption coefficient with a preassigned accuracy. The conditions required to attain these ends, i.e., the geometry for optimum transmission, and the best apportionment of counting times among the incident and transmitted beams and background, have been investigated for a wide range of relative backgrounds (10^{-3} to 10^2), and for two geometries: I. Beam area fixed, absorber thickness alone is varied. II. Beam area and absorber thickness are both disposable parameters, while the total amount of absorber intercepting the beam remains fixed. In both cases the incident flux density and the background rate are assumed constant. The optimum transmissions are shown to be, in general, considerably smaller than those commonly used in absorption experiments. Thus, in Case I, a useful rule is to employ a transmission of about 0.1 for low backgrounds, 0.2 for moderate backgrounds, and 0.3 for high backgrounds. The following have also been determined: (a) minimum statistical error for a given total counting time, (b) statistical error and the best distribution of counting times for non-optimum geometry, and (c) sensitivity of the accuracy or total counting time to deviations from optimum transmission.

INTRODUCTION

THERE are many physical measurements of absorption or transmission of radiation in which the transmission is an exponential function of the absorber thickness. Measurements of this kind occur, for example, in the fields of optics, x-rays, and nuclear physics. In the latter field the purpose is usually to determine a total cross section.

In most measurements of this type, the thickness of absorber can be chosen at will within reasonable limits, i.e., the transmission is a disposable parameter. Suppose for the sake of definiteness that the detector is a counter. If a thick absorber is interposed in the beam, the transmitted intensity is low and a relatively long time is required to collect an adequate count. In fact, for sufficiently low transmissions, the transmitted beam may become comparable in magnitude to the background. This obviously results in inefficient counting. On the other hand, if a very thin absorber is used, the intensity of the transmitted beam approaches that of the incident beam, and again the geometry is statistically unfavorable. Therefore, some intermediate value of the transmission should be employed, and of course this is commonly done.

It may sometimes be desirable to know what value of the transmission is *best* if the greatest accuracy is to be obtained in a given time or if a preassigned accuracy is to be attained in the least time. Moreover, the dependence of this optimum