Physical Properties of Polonium. II. XRay Studies and Crystal Structure

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Structures are proposed which account for the powder diffraction patterns obtained from samples of polonium containing various amounts of lead. Unusual temperature effects are recorded for α-polonium. It is suggested that the Pb and Po are present in interpenetrating primitive cubic lattices which are indistinguishable because the atomic numbers are nearly the same.

INTRODUCTION

A STUDY of polonium by x-ray diffraction powder photographs was undertaken to determine whether the abrupt change in electrical conductivity with temperature[1] is caused by a phase transformation. The existence of two forms of polonium has been confirmed and reported earlier.[2] The fact that only microgram quantities of polonium are available and the difficulties of handling a material of such great radioactive toxicity have not permitted the growth of single crystals for x-ray diffraction. Structures have been found which account for the data available from the powder diagrams.

EXPERIMENTAL

X-Ray Apparatus

Two cameras were used; one was a 7.5-cm radius Debye camera covering a range of $\sin^2\theta$-values from 0.003 to 0.97; the other was a back reflecting cylindrical type of radius 5 cm covering $\sin^2\theta$ from 0.018 to 0.985. Iron and copper radiations from General Electric CA-6 diffraction x-ray tubes were used with double-coated Blue Brand Eastman x-ray film. Direct tests on this film showed no appreciable film shrinkage under the conditions of the experiments.

The films were measured without magnification with a pointer attached to a slide and vernier. The smallest division which could be read on the vernier was 0.1 mm. The measurement of any one sharp line had a possible error of ±0.1 mm; the diameter of any sharp ring had a possible error of ±0.2 mm.

Preparation of Sample

Because of the characteristics of polonium discussed in the introduction only glass-enclosed powder samples were used. From 30 to 100 micrograms of polonium were put into the very end of a thin wall capillary with three successive vacuum distillations. The capillary was drawn from a low absorbing lithiaborosilicate glass (Corning No. 707) which can be sealed directly to Pyrex. A drawing of the apparatus used is shown in Fig. 1.

The procedure for the preparation of powder samples was as follows:

(1) The capillary and the end of the distillation apparatus up to the point (A) was thoroughly outgassed to get rid of oxygen, evacuating to a pressure of 10^{-6} mm of Hg for 15–20 hours while holding at a temperature of 400–425°C in a tube furnace.

(2) The polonium was electroplated on a 40-mil platinum wire. The wire was placed in a small closed-end platinum tube (B) which was in turn placed in a small close-fitting quartz tube (C).

(3) The distillation apparatus was filled with a positive pressure of helium and the quartz tube (C) was inserted at the end (E) while helium was flowing through the apparatus. The apparatus was again evacuated to a pressure of approximately 10^{-6} mm of Hg.

(4) The quartz tube was slid to the position (F) by tilting the apparatus on a ball joint. A high frequency induction coil was placed around the entire apparatus at (F) and the platinum tube heated rapidly to a red heat. This distilled the polonium from the platinum wire in from 5 to 10 seconds. The polonium condensed as a shiny metallic mirror on the walls of the apparatus near the capillary. The quartz tube served as insulation
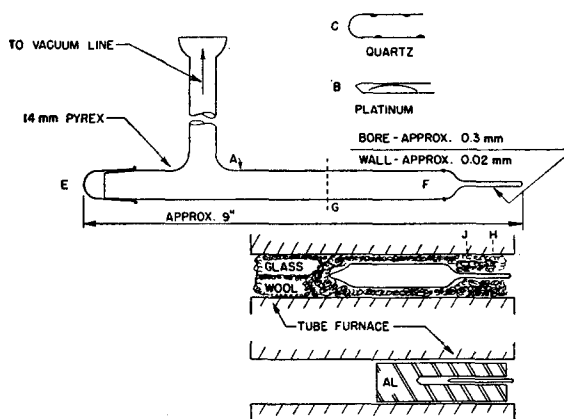

FIG. 1. Apparatus for preparation of x-ray sample.

[1] C. R. Maxwell, J. Chem. Phys. 17, 1286 (1949).
[2] W. H. Beamer and C. R. Maxwell, J. Chem. Phys. 14, 569 (1946).

TABLE I. Calculated and observed spacing and intensities for α-polonium. Simple cubic with $a = 3.345A$.

$h^2+k^2+l^2$	Calculated $1/d^2$	Observed $1/d^2$	Calculated intensity	Observed intensity
1	0.0894	0.0896	100	100 VS
2	0.1787	0.1795	91	80 VS
3	0.2681	0.2692	38	40 MS
4	0.3575	0.3595	21	25 M
5	0.4469	0.4486	62	55 S
6	0.5362	0.5371	50	45 S
8	0.7150	0.7169	18	10 W
9	0.8045	0.8065	42	30 M
10	0.8937	0.8978	32	25 M
11	0.9831	0.9866	32	15 MW
12	1.0725	—	11	0 —
13	1.1619	1.1628	35	15 MW
14	1.2512	1.2526	75	30 M
16	1.4300	1.432	12	5 F
17	1.5193	1.5193*	132	20 W
18	1.6087	1.608*	144	15 VW

* Average of resolved α_1- and α_2-measurements.

and prevented the red hot platinum from breaking the glass tube.

The apparatus was open to the vacuum line during the distillation so that any permanent gases liberated from the platinum and quartz were continually removed. During the distillation the pressure of the permanent gases in the system did not exceed 10^{-4} mm.

(5) The quartz tube and platinum were returned to position (E) and the apparatus was sealed off at point (G). This tube was wrapped in glass wool and placed in a tube furnace so that the end of the capillary was exposed to the open air. The tube furnace was held at 400°C for two or three hours. Due to the temperature gradient in the apparatus the polonium migrated into the capillary but since the temperature gradient in the capillary was not sharp, the polonium was distributed over a considerable length (H) of the capillary.

(6) The capillary was sealed off from the main apparatus at the point (J), and placed in a close-fitting hole in an aluminum block which was held at 400°C for another few hours. The temperature gradient in the capillary was now sharp and the polonium migrated to the end of the capillary.

(7) The capillary was then mounted and photographed in the usual manner. By this procedure a small amount of polonium was concentrated into one spot in a capillary without exposure to air or danger of mechanical loss.

The capillaries used were 0.3 to 0.4 mm O.D. with a wall thickness of approximately 0.02 mm or less. The alpha-particles which are released by radioactive decay and which have a range in air of 5 cm at this altitude (7300 ft.) often penetrated the capillary walls (five cm of air is equivalent to 0.02 mm of glass). The alpha-particles from sample II used in obtaining films 27 and 28 reported in Tables I and II had a range of over two cm in air after penetrating the capillary walls.

Calibration of Cameras

The technique described for the preparation of polonium samples was developed with zinc as a stand-in for polonium. Zinc was selected because it has approximately the same vapor pressure as polonium. These samples were used to calibrate the cameras for the investigation. The zinc was obtained by electrodeposition from a solution of Baker's C.P. zinc sulfate. The constants for the crystal structure of zinc were taken from Barrett[3] and were: $a = 2.6595A$; $c = 4.9368A$. The values for the camera radius which were calculated from individual zinc lines did not vary with the diffraction angle as did the values calculated from the pattern of a nickel wire of comparable size photographed in the same mounting. This indicates very small absorption in small metal samples prepared by this technique.

Estimation of Intensities

Estimations of intensities were made visually. In addition to the usual method of reporting intensities from very strong (VS) to faint (F), a numerical value was assigned to each line.

An arbitrary high value in agreement with the value of the calculated intensity was assigned to the first line. The intensity of the second line was estimated in terms of the first line and a value assigned. The intensity of the third line was estimated in terms of the assigned value for the second line and so forth. The error in any estimation was thus carried through all subsequent estimations. Any non-linearity in the exposure characteristics of the film affects these numerical values.

The intensity estimations were made difficult by the presence of an over-all background from the gamma-radiation associated with the radioactive decay. An exposure of 100 hours to the gamma-radiation from one sample completely blackened the film. The average exposure for unfiltered copper radiation was 10–12 hours, while CuKα-radiation secured by filtering with one mil nickel foil required an exposure of 30 hours. The α-phase polonium patterns were exposed to gamma-radiation for several additional hours while the sample was cooled before x-ray exposure.

Temperature Measurement and Control

Only a rough estimate can be made of the temperature of the polonium samples when the diffraction patterns were photographed. The heat evolved in stopping the alpha-particles is 27.42 calories/curie/hour, so the sample is maintained at a temperature above that of its surroundings. The actual temperature of the sample depends upon the distribution of the polonium in the capillary, thickness of the capillary walls, and thermal contact with the surrounding air.

[3] C. S. Barrett, *Structure of Metals* (McGraw-Hill Book Company, Inc., 1943), p. 554.

The diffraction patterns photographed with freshly prepared samples at thermal equilibrium with static air always showed the high temperature, or β, phase only. The low temperature, or α, phase pattern was only obtained after prolonged cooling in a stream of refrigerated air. On returning the sample to thermal equilibrium with static air at room temperature, the diffraction pattern was predominantly that of β-polonium although the stronger α-polonium lines were always present. Thus it appears that the temperature of the polonium samples when in equilibrium with static air at room temperature was near the α- to β-transformation temperature. In view of the electrical resistivity measurements the temperature of the polonium sample under these conditions is estimated as $75\pm15°C$.

To obtain the α-phase alone the polonium samples were cooled by a stream of dry air which had been refrigerated by passing through a copper coil immersed in dry ice and trichloroethylene. A constant, brisk rate of flow was used to obtain as good heat transfer as possible and thus decrease the temperature differential between the metal and the air stream.

The temperature of the air stream was determined by exploring the air stream in the vicinity of the sample with a thermocouple made of 3-mil wire. This temperature was found to be approximately $-10°C$. The temperature of the polonium sample under this condition is estimated as $10\pm10°C$.

RESULTS

Crystal Structure of α-Polonium

The diffraction pattern of α-polonium can be interpreted as that arising from a simple cubic crystal with a parameter of $3.345\pm0.002A$.

Columns (3) and (6) of Table I list the spacings and intensities observed on the best α-Po film. Columns (2) and (5) list the corresponding values calculated for a simple cubic structure with $a=3.345A$.

The film from which the values in Table I were taken was photographed with Ni filtered $CuK\alpha$-radiation. The film had a very high background since it was exposed to gamma-radiation for 55 hours while the sample was cooled and photographed. The spacing corresponding to $(h^2+k^2+l^2)$ equal to 12 was not observed on this film but has been observed on other films with less background. A freshly prepared sample was used so that the lead content (approx. 1.5 percent) would be as low as possible.

The agreement between observed and calculated values of $1/d^2$ is quite satisfactory. A variation of 0.002A in the parameter would cause the observed and calculated patterns to disagree by an amount larger than the experimental error in measuring this particular film. Hence, the precision with which the parameter may be determined is $\pm0.002A$ although the parameter

TABLE II. Calculated and observed spacings and intensities for β-polonium. Simple rhombohedron $a=3.359A$, $\alpha=98°\ 13'$.

Pseudo-cubic $h^2+k^2+l^2$	Rhombo-hedral indices	Calculated $1/d^2$	Observed $1/d^2$	Calculated intensity	Observed intensity
6	100	0.0931	0.0931	200	200 VS
10	1$\bar{1}$0	0.1552	0.1550	108	100 S
14	110	0.2173	0.2169	78	60 MS
16	11$\bar{1}$	0.2483	0.2486	65	50 M
24	111 200	0.3725	0.3733	13 40	50 M
26	2$\bar{1}$0	0.4035	0.4045	73	60 MS
30	$\bar{2}$11	0.4656	0.4669	30	15 MW
34	210 21$\bar{1}$	0.5277	0.5279	53 53	60 MS
40	2$\bar{2}$0	0.6203	0.6231	22	8 W
46	211 2$\bar{2}$1	0.7139	0.7153	19 38	15 W
54	22$\bar{1}$ 300 3$\bar{1}$0	0.8380	0.8393	17 17 34	30 MW
56	220 $\bar{3}$11	0.8691	0.8708	17 17	5 VW
64	22$\bar{2}$ 31$\bar{1}$	0.9932	0.9941	16 32	10 W
66	310 3$\bar{2}$0	1.0243	1.0255*	32 32	10 W
70	221 $\bar{3}$21	1.0863	1.0868*	17 34	5 VW
74	3$\bar{2}$1	1.1484	1.151	35	3 F
80	311	1.2415	—	18	not obs.
86	32$\bar{1}$ $\bar{3}$22	1.3346	1.3344*	44 22	2 F
90	320 3$\bar{3}$0	1.3967	1.3947*	48 24	1 F
94	32$\bar{2}$ 4$\bar{1}$0 $\bar{4}$11	1.4588	1.4586*	58 58 27	15 MW
96	222 3$\bar{3}$1 400	1.4898	1.4902*	10 62 31	10 W
104	4$\bar{2}$0	1.6140	1.6126*	110	3 VW
106	321 $\bar{4}$21	1.6450	1.641	144 144	6 W

* Value is average of resolved α_1- and α_2-measurements.

change associated with the large uncertainty in temperature is much larger than this.

The agreement between the calculated and observed intensities is excellent for the first six lines. The observed intensity values for the high angle lines were

TABLE III.

	Ratio	α-Po	β-Po
Goldschmidt	1.04	3.42	3.49
Westgren and Almin	1.123	3.69	3.77
Pauling		3.47	3.54
This work	1.07	3.52	3.60

considerably less than the calculated intensity values. There is a general weakening of the entire observed pattern with increasing diffraction angle and not an isolated weakening of a particular line. This trend in the intensities of the higher angle lines was observed with both α-Po and β-Po and in the calibration photographs taken with a zinc sample prepared in the same manner. Comparison should be made between adjacent or near adjacent lines. When the ratio between the observed intensity values of adjacent lines is compared with the ratio between the calculated values for the same lines, satisfactory agreement is obtained for the entire pattern.

The calculated density for this structure is 9.32 g/cc which is in the satisfactory agreement with the measured value of 9.4±0.5 g/cc.

The structure belongs to the space group O_h^1.

Crystal Structure of β-Polonium

The diffraction pattern of β-polonium can be interpreted as that arising from a simple rhombohedral crystal with a parameter of 3.359±0.002A and an angle of 98° 13'±3'.

Columns (4) and (6) of Table II list the spacings and intensities observed on the best β-Po film. Columns (3) and (5) list the corresponding spacings and intensities calculated for a rhombohedral cell with $a=3.359$ and $\alpha=98°$ 13'. In Column (2) are listed the corresponding rhombohedral indices. The spacings of this pattern occur in the ratio of simple whole numbers, and cubic indices may be assigned to each line. Column (1) lists the $(h^2+k^2+l^2)$ values corresponding to the pseudo-cubic indices.

The agreement between observed and calculated spacings is completely satisfactory. When the ratio between the observed intensity values of adjacent lines is compared with the ratio between the calculated values for the same lines, satisfactory agreement is obtained for every line except line "94." The presence of a high background makes it impossible to say whether this apparent discrepancy is real or not. The line is very sharp, and for this reason may appear more intense.

Two or more indices are assigned to thirteen of the twenty-two spacings observed in β-Po. This accidental agreement in the spacings of different families of planes must be very close since the lines observed on the photographic film are quite sharp. Exact agreement for all such accidental superpositions is obtained when

$\alpha=98°$ 12' 40". The exactness of this superposition is a function of α alone and does not depend upon the parameter a. The exactness of the superposition of planes 400 and 331 observed together as line "96" is the most susceptible to slight changes in α. A change of ±3' would cause the spacing of these planes to be different by an observable amount. Similar consideration of planes 410 and 411 observed as line "94" sets a limit of ±5'. The distance between lines "94" and "96" is also very dependent upon α. A change of more than ±3' would be easily detected.

Because of the pseudo-cubic nature of the β-Po pattern, many crystal lattices will give completely satisfactory agreement between the calculated and observed values of the spacings. A number of possible structures in the cubic, orthorhombic and hexagonal systems have been considered and ruled out on intensity calculations.

The patterns reported in Table I and II were obtained from the same sample. The β-polonium pattern was photographed immediately after the preparation of the sample. The lead content at this time was 0.5 percent.

The calculated density for this structure is 9.51 g/cc which is in satisfactory agreement with the measured value of 9.4±0.5 g/cc.

This structure belongs to space group D_{3d}^5.

The Po-Pb Alloy System

The effect of the growth of lead in the polonium on the x-ray pattern was studied by following a sample for one half-life, 138 days. No new lines were observed on any of the films taken over this period indicating that lead forms a solid solution with α-polonium to the extent of at least 50 atomic percent. The photographs taken the first few days showed only the pattern of β-polonium. The sample was cooled and the pattern of the α-phase was obtained when the sample contained 4 percent lead. All subsequent photographs taken with the sample in thermal equilibrium with static air showed some lines of α-Po. At first the pattern was predominantly that of β-Po with only the stronger α-Po lines appearing. With time the α-Po lines gained in intensity while the β-Po lines lost. The β-Po lines were not observed when the sample contained more than 25 atomic percent lead. This change in the relative amounts of the
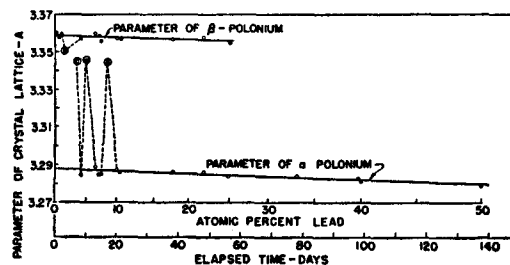

FIG. 2. Parameter of crystal lattice of Pb-Po alloys.
o = uncooled sample, ⊙ = cooled sample.

two phases present is probably a function of the lead content and of the temperature which is decreasing with the decay of the sample.

Figure 2 shows the parameters of the two phases as a function of time. The parameters plotted here are averages calculated from low angle, strong lines. These lines were selected because they appeared on all films and had no interferences. Since systematic errors in the camera affect the value of the parameter calculated from low angle lines the same lines were used to compare parameters from film to film. The values of the parameter of α-polonium designated by encircled points on Fig. 2 were calculated from films taken while the sample was cooled. The average parameter for the α-Po lines recorded on these films is 3.345A. The other points were taken while the sample was in equilibrium with static air at room temperature. The average parameter for α-Po recorded on these films is 3.286A. The difference is many times larger than the differences between the individual measurements which were averaged. We are not able to offer a suitable explanation for this temperature effect. If the structure of these alloys is in reality more complex than our data indicates, the parameter plotted in Fig. 2 may have no physical significance in the true structure. If, on the other hand, the plotted parameter represents a distance between primary lattice sites, a negative coefficient of thermal expansion is indicated. Based on the simple cubic structure and using the temperature estimates made earlier the value of this coefficient for alloys containing 4 percent to 9 percent lead is $-300\pm100\times10^{-6}$ cm/cm/°C. From diffraction patterns at 41 percent lead the coefficient is about -30×10^{-6} cm/cm/°C.

Only one film was made recording the β-phase at a temperature lower than normal. The parameter value from this film is indicated also by an encircled point on the β-polonium parameter plot of Fig. 2. The sample contained 1.5 percent lead when the film was made. The coefficient of expansion from this single parameter value is $+50\pm25\times10^{-6}$ cm/cm/°C.

Atomic Diameter of Polonium

Figure 2 shows that the parameter of α-polonium phase decreases slightly with time. Neglecting the temperature change of the sample and taking the extrapolated value of the atomic diameter of α-polonium at zero lead content as 3.288A the atomic diameter for lead was calculated to be 3.27A in a simple cube with a coordination number of six.

The atomic diameter of lead in a f.c.c. crystal with coordination number of twelve is observed to be 3.49A. The ratio between this value and the value calculated for a coordination number of six is 1.07. According to Goldschmidt[4] the expected ratio is 1.04. On the other hand Westgren and Almin[5] have, on the assumption

of constant atomic volume, calculated the ratio to be 1.123. More recently Pauling[6] has developed an equation for the change of atomic diameter with coordination number based on the idea that metallic bonds are resonating covalent bonds. His equation predicts a value of 3.31A for lead with a coordination of six.

Possible values for the atomic diameter of polonium with a coordination number of twelve are listed below using the ratios of Goldschmidt, Westgren, and Almin, the Pauling formula, and the ratio obtained for lead in the above. These values are for polonium at a temperature where the two phases co-exist, probably near 75°C (Table III).

Hume-Rothery[7] has divided the periodic table into three classes according to the crystal structures of the elements.

Class I includes those elements which have the typical metallic structures, body-centered cube, face-centered cube and close-packed hexagonal.

Class III contains the elements of Groups IV, V, VI, and VII which crystallize so that each atom has (8-N) close neighbors, where N is the number of the group to which the element belongs. The structure of sulfur and selenium (Group VI) are such that the atoms are arranged in spiral chains so that each has two close neighbors in accordance with the "(8-N) rule."

The elements of Class II form a transition group in which the crystal structure is a distorted form of the typical metallic structures. Examples of this class are zinc, cadmium, and mercury. In considering the structures of other members of this class Hume-Rothery has pointed out that the interatomic distances of thallium and lead are much greater than that of the preceding univalent element gold and that the same is true for indium and white tin in relation to silver. In the first long period the interatomic distances do not change greatly in the whole series of elements. This can be explained by assuming that these four metals of Class III are incompletely ionized.

Polonium most closely resembles this sub-group of elements. The simple cubic structure has not been observed for any other element. It has the symmetrical properties associated with true metallic bonding but it represents a rather loosely packed structure. If polonium were a member of Class III and conformed to the (8-N) rule, one would expect its interatomic distance to be of the order of magnitude of 3.10A observed for bismuth. Instead the interatomic distance is observed to be around 3.60A when corrected for change in coordination number. With the same coordination number the interatomic distance of polonium is slightly larger than that of lead. Further the melting point, electrical resistance, density, and hardness are similar to those of lead and are characteristic of metallic bonding.

[4] V. M. Goldschmidt, Zeits. f. physik. Chemie 133, 397 (1928).

[5] A. Westgren and A. Almin, Zeits. f. physik. Chemie, Abstract B 5, 14 (1929).

[6] L. Pauling, J. Am. Chem. Soc. 69, 542 (1947).

[7] W. Hume-Rothery, The Structure of Metals and Alloys (Institute of Metals, London, 1936).

It has been suggested that the lead always present in polonium samples may exert a great effect on the structure. These samples contain from 1.0 percent to 50 percent lead. At the higher concentrations of lead the NaCl type structure is likely. The atomic number of lead and polonium are 82 and 84 respectively and so intensities of lines from Pb-containing planes and from Po-containing planes would be very nearly equal. At low lead concentrations the interpenetrating lattices of Pb and Po might be maintained with Po atoms occupying the sites not filled by Pb atoms. This explanation of the simple cubic and simple rhombohedral patterns would fit well with the observed solubility of lead in the polonium and with the fact that PbS, PbSe and PbTe all have the NaCl structure.

ACKNOWLEDGMENT

The Physical Properties of Polonium. III. The Half-Life of Polonium

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The half-life of polonium has been measured by a calorimetric method. The reproducibility and precision of the data is much greater than has been previously reported. The value is found to be 138.3±0.1 percent days.

INTRODUCTION

THE half-life of polonium has been measured many times and values in the literature vary from 126 to 148 days.[1] Counting, total ionization, and calorimetric methods have been used. Schweidler[2] measured the decrease of ionizing power of a sample of polonium mounted on a copper foil over 2200 days and found the half-life to be 136.5 days. This value was accepted for some time. M. Curie[3] found the half-life to be 140 days by electrical counting methods extending over 184 days. The polonium was deposited on quartz and covered with an aluminum foil 0.01 mm thick.

There are a number of errors inherent in the counting methods, such as coincidence corrections, change of counter characteristics, etc. Several of these tend to increase the apparent half-life, for example, incomplete separation from other radioactive substances such as radium D. However, most of the errors tend to decrease the half-life by effective decrease of the amount of polonium with time. (1) Atomic aggregates of polonium are known to be knocked off the sample by alphas leaving the material; (2) diffusion of polonium into the

support material; (3) covering of the surface of the polonium by a layer of oxide or foreign material. These effects are all known to occur.

The calorimetric method, which measures the decrease of heat evolution with time of a sealed polonium sample, is affected only by these errors: (1) radioactive impurities, (2) heat evolved by secondary chemical or nuclear reactions produced by the alphas. Microcalorimetric determinations of half-life have been made by Sanielevici[4] (138.7±0.4 percent and 139.6±1 percent) and A. Dorablaska[5] (137.6±0.4 percent) but the measurements of the heat evolved were not consistent. The value of the heat evolved by a curie of polonium can be calculated from the energy of the alphas and of the recoil lead atoms. The accepted value for the energy of the alpha-particles is 5.303 Mev as measured by Rutherford, Wynn-Williams, Lewis, and Boden.[6] The alpha-spectrum for polonium has been investigated by Chang[7] and found to contain less than 0.1 percent of alphas different in energy from the 5.303-Mev value. Polonium gives off about seven quanta of gamma-rays per million alphas;[8] this can be neglected. The energy of the recoil atoms, disregarding relativity effects, is

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[1] St. Meyer and E. Von Schweidler, *Radioaktivität* (B. G. Teubner, Leipzig, 1927), second edition.
[2] E. V. Schweidler, Verh. d. D. Phys. Ges. (2), 14, 539 (1912).
[3] M. Curie, J. de phys. et rad. (6) 1, 12 (1920).

[4] A. S. Sanielevici, J. de chimie phys. 33, 779 (1936).
[5] A. Dorablaska, Roczniki Chem. (Poland) 11, 475 (1931).
[6] Rutherford, Wynn-Williams, Lewis, and Boden, Proc. Roy. Soc. A139, 617 (1933).
[7] W. Y. Chang, Phys. Rev. 69, 60 (1946).
[8] W. Bothe, Zeits. f. Physik 96, 607 (1935).