Energy Loss and Resultant Charge of Recoil Particles from Alpha Disintegrations in Surface Deposits of ^{210}Po and ^{241}Am†

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Recoil particles from the ^{210}Po$(\alpha)^{206}$Pb and ^{241}Am$(\alpha)^{237}$Np disintegrations in surface deposits on platinum planchets have been studied experimentally using both a low-resolution magnetic spectrometer, employing an electron multiplier detector, and a hemispherical ionization chamber. The average energy of the ^{206}Pb recoil ions emitted from thin deposits of ^{210}Po showed an exponential degradation with time after preparation, starting with the nascent energy of 103 keV and decreasing to $\frac{2}{3}$ of this value in five days. Approximately 90% of the ^{206}Pb recoil atoms are emitted singly ionized and 10% are doubly ionized. The ^{237}Np recoils from thicker deposits of ^{241}Am apparently were mostly neutral but a small number were emitted with six units of charge and with an average kinetic energy of only 65 ± 15 keV compared with their nascent energy of 92.4 keV. Ejected recoils are accompanied by large numbers of secondary particles having energies less than 40 eV. With thin source deposits there are about five positively charged ions and three detected neutral secondary particles emitted per disintegration; but with thicker deposits these numbers rise to 15 positive ions and 10 neutrals, respectively. Negative ions may also be present but were excluded from observation by the experimental conditions. The average energy loss for the ^{206}Pb recoil particle in passing through argon gas was determined to be 121 ± 6 eV/ion-pair. The extrapolated range for a nascent energy ^{206}Pb recoil particle in argon at STP is 78 ± 2 μ. This value agrees with the range reported by Jessi and Sidauskis and lies within 2% of the value calculated from an energy-loss theory originally developed by Bohr.

INTRODUCTION

THE daughter particle recoiling from α decay of a parent nuclide will have a nascent kinetic energy equal to $(m_\alpha/m_R)E_\alpha$, consistent with conservation of momentum and energy. For the ^{210}Po$(\alpha)^{206}$Pb and the ^{241}Am$(\alpha)^{239}$Np disintegrations, these recoil energies are 103 and 92.4 keV, respectively. Prior to disintegration, the parent system consists of a neutral atom, presumably in its ground state but possessing an unstable nucleus. After disintegration, the system will have made a transition to its final state consisting of an energetic α particle moving in one direction and the recoil nucleus together with its excited atomic electrons moving in the opposite direction. During the process, the nuclear charge decreases by two units, the ejected α particle traverses the atomic electron field, and the residual nucleus accelerates to a recoil velocity of about 3×10^7 cm/sec. Since the characteristic periods of the inner electrons surrounding the nucleus are small compared with the transit time of the α particle, the transition is nearly adiabatic. Accordingly, the atomic electron system may be expected to remain virtually intact and move with the recoiling body. In the absence of excitation this recoil system could be a doubly charged negative ion. However, it is more likely that the recoil atom will be left in a state of positive ionization. Goldstein[1] has estimated an order of magnitude for the probability of such ionization due to the change in nuclear charge and due to the possible collision of the emitted α particle with the extranuclear electrons. A more recent estimate[2] places the average energy of excitation possessed by the residual recoil atom at about 150 eV. Thus, there is sufficient energy to leave the recoil atom multiply ionized.

Wertenstein[3] tried to determine the charge carried by RaD recoil particles from RaC' α disintegrations in a vacuum. He reached the conclusion that all recoil atoms are initially neutral and that they become singly or multiply ionized only upon interaction with other atoms in their path. Walmsky and Makower[4] examined the recoil particles from the RaA (α)RaB disintegration using a magnetic vacuum spectrometer with a photographic plate for a detector. They found that a large fraction of the recoils were singly charged and that the recoil-energy spectrum was broad, indicating a significant energy loss in escaping from the source deposit. Confirmation of these results was obtained by Wood and Makower[5] for the recoils from RaC deposited on a platinum wire. In a careful study, McGee[6] has found that the ejected recoil particles from deposits on platinum or nickel surfaces are accompanied by large numbers of both positive and negative secondary ions which probably were present in the measurements of Wertenstein and led him to erroneous conclusions. Using a sweep field to remove these secondary ions and outgassing the emitting surfaces by elevating the temperature of the source deposit in a vacuum, McGee was able to establish that the recoil atoms leave a clean platinum or nickel surface with an average of one unit of positive charge; however, he found that recoils from an oxide-coated surface are all neutral, and of those emitted from a surface covered by an adsorbed gas layer, about 50% are neutral. No circumstances under

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[1] L. Goldstein, J. Phys. Radium 8, 235 and 316 (1937).
[2] R. H. Thomas, Progr. Theoret. Phys. (Kyoto) 12, 252 (1954).
[3] L. Wertenstein, Compt. Rend. 161, 696 (1915).
[4] Walmsley and Makower, Phil. Mag. 30, 253 (1915).
[5] A. B. Wood and W. Makower, Phil. Mag. 30, 811 (1915).
[6] J. D. McGee, Phil. Mag. 13, 1 (1932).

which the recoil atom carried a negative charge were found.

The charge acquired by the recoil atoms from isolated distintegrations in a monatomic gas was determined by Mund, Capron, and Jodogne.[7] Their results indicate that recoil atoms of RaA from the α disintegration of radon under reduced pressure possess a positive charge of two units. This is consistent with the expected[2] atomic excitation energy of RaA and probably is typical of the nascent charge carried by the recoil atom from the α disintegration of any of the heavy nuclides. However, when a recoil particle is emitted from a surface deposit, its charge state is determined mainly by its interaction with that surface rather than by the disintegration itself. For clean metallic deposits on metallic surfaces the resultant charge of the escaping recoil is governed by the electron work function which is approximately equal to the first ionization potential of the atom. Therefore, most of the recoil atoms from such a surface may be expected to leave with a single positive charge. On the other hand, if the surface consists of oxide crystallites mechanically supported by the backing material, quite different charge magnitudes may be expected.

No quantitative information concerning the residual energy of the recoil particle after escaping from a surface has been reported previously. Also, in most of the earlier measurements, only the average of the charge distribution for recoil atoms has been determined. Although Kolhörster[8] has established that the number of recoils is equal to the number of α particles emitted from surface deposits of ThC and ThC', Haissinsky and others[9] have obtained evidence from α-particle ranges that Po atoms penetrate into the surface of gold and platinum before decay. Accordingly this information injects an uncertainty as to how many of the ^{206}Pb recoils may successfully exit from such a deposit on a platinum surface. Lack of definite answers to these questions places results of earlier measurements[10–13] on gas ionization and range of recoil particles from α decay in doubt since it has been assumed, generally, that for one half of the disintegrations the recoil atom leaves the surface with its full nascent energy and for the other half of the disintegrations the recoil atom remains within the source.

This paper describes an experimental investigation of the charge and energy carried by recoil particles from α decay in surface deposits of ^{210}Po and ^{241}Am on platinum using a specially designed magnetic spectrometer and a 16-stage electron multiplier for the detector whose pulse output was fed to a conventional scaler through a low-noise linear amplifier equipped with a variable discriminator bias. These particular nuclides were selected because they have convenient half-lives of 138 days and 462 years, respectively, and because their atoms had distinctly different physical attachment to the backing material. For example, the ^{210}Po was plated by spontaneous deposition[14] on polished platinum surfaces while the ^{241}Am was deposited as microcrystals, probably of the oxide and of the nitrate. Following these measurements, the range of the recoil particles and the energy loss due to ionization in argon gas were determined for the ^{206}Pb recoils from the ^{210}Po disintegration under conditions for which the initial charge and energy were known. For this, a hemispherical ionization chamber operated within a pressure range from a millitorr to one atmosphere was used. With a vibrating-reed electrometer, ionization currents were measured as a function of chamber pressure and the resulting curves were analyzed to obtain the component due to the recoil particles.

EXPERIMENTAL CONDITIONS

Since the deflection of a particle in a magnetic field is proportional to the ratio of its charge to its momentum, magnetic deflection alone cannot reveal the charge or the energy of the particle independently. This ambiguity is particularly significant for the recoil particles from a surface deposit because the residual charge and the residual momentum may be interrelated due to surface interactions. To resolve the charge and the

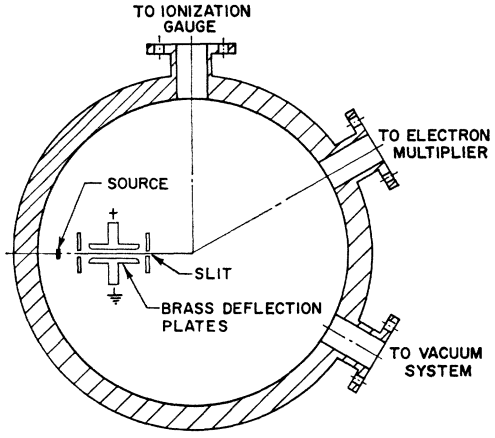

FIG. 1. Cross-sectional view of the vacuum spectrograph with velocity selector used for measuring the charge and energy of recoil particles from α disintegrations occurring in surface deposits. A magnetic field, uniform in the region of the electric deflection plates, was applied normal to the plane of the diagram. Deflection of charged particles continued in the fringing magnetic field which dropped to a very low intensity at the magnetically shielded electron multiplier. The entire system was evacuated to less than 10^{-5} Torr.

[7] Mund, Capron, and Jodogne, Bull. Soc. Chim. Belges, No. 1, Jan. (1931).
[8] W. Kolhörster, Z. Physik **2**, 257 (1920).
[9] M. Haissinsky, H. Faraggi, A. Coche, and C. Avignon, Phys. Rev. **75**, 1963 (1949).
[10] L. Wertenstein, Compt. Rend. **155**, 450 (1912).
[11] B. Bianu and L. Wertenstein, Compt. Rend. **155**, 475 (1912).
[12] A. B. Wood, Phil. Mag. **26**, 586 (1913).
[13] W. P. Jessi and J. Sadauskis, Phys. Rev. **102**, 389 (1956).
[14] M. Haissinsky, *Nuclear Chemistry and Its Applications* (Addison-Wesley, Reading, Massachusetts, 1964), pp. 578 and 580.

energy independently, a vacuum spectrometer was constructed as illustrated in Fig. 1. The source was mounted on a narrow platinum strip placed perpendicular to the plane of the diagram. Disintegration particles passed through an entrance slit 1 mm wide into a conventional velocity selector consisting of crossed electric and magnetic fields which were, respectively, parallel and perpendicular to the plane of the diagram. The exit slit of the velocity selector was also 1 mm wide but the separation between deflecting plates within the velocity selector was set at $\frac{2}{3}$ cm so that particles striking these plates would not be scattered out through the exit. Owing to spatial restrictions, the length of the deflecting plates could be no longer than 4 cm. These dimensions limited the resolution of the velocity selector to about 30% but this was adequate to identify the components of the recoil beam. Since the transmission of a particle through the velocity selector is independent of the charge, the output formed a collimated beam of particles nearly homogeneous in velocity but possibly differing in charge. This beam was then deflected by the magnetic field to accomplish an analysis of the charge spectrum. The 16-stage electron multiplier used for the detector had Cu-Be dynodes. It was operated with an accelerating potential for positive ions of about 3 kV and so was insensitive to electrons and negative ions.

To obtain recoil particles from these sources it was necessary to use very thin deposits. This consequently restricted the permissible source activity to about 6000 disintegrations/sec and similarly limited the counting rate at the detector. Accordingly, resolution was sacrificed to gain transmission through the spectrometer but the resolution remained adequate to permit discrimination between charge groups differing by one electronic unit. Also, because of the low resolution of the velocity selector, some α particles in a wing of the velocity-resolution profile successfully passed through the slits even when the fields were set for the recoil velocities. These, however, could be distinguished suitably from the recoil particles and therefore caused no error in interpretation. The spectrometer together with its electron multiplier detector was operated at a pressure of about 10^{-5} Torr. At this low pressure, collisions between the recoil particles and residual gas atoms in the system that might induce a charge exchange were extremely unlikely.

The hemispherical ionization chamber used to measure the range and energy loss for the recoil particles is shown schematically in Fig. 2. The system was opened to install new ^{210}Po sources prepared in the same way as the sources used for measurement of recoil energy and charge. It was then evacuated to about 10^{-3} Torr, flushed with argon gas, evacuated again, and after two such operations was finally filled with argon gas of 99.9% purity to the desired pressure as indicated by a mercury manometer isolated from the system by a cold trap. Pressure reductions from the initial value were

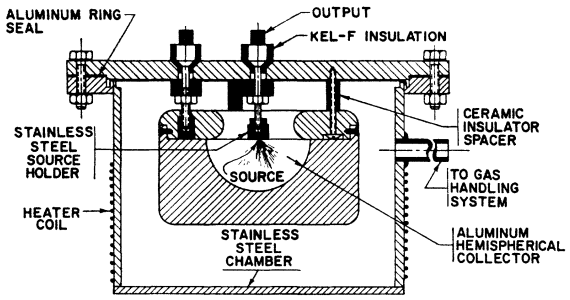

FIG. 2. Cross-sectional view of the ionization chamber for measuring energy loss and range of recoil particles. A positive collector potential of 20 to 750 V was used and ionization currents were read from a vibrating-reed electrometer connected to the central terminal holding the source. When the chamber was used as a proportional counter the central terminal was connected to the amplifier input.

accomplished by means of a calibrated metering system which employed an auxiliary expansion chamber coupled to the ionization chamber through a manually operated valve. After each expansion, the auxiliary chamber was closed off from the main system and evacuated. Through repeated applications of this process, the system pressure could be reduced in steps by a factor of 0.855 per step. This procedure introduces a cumulative error, but even for the lowest pressures employed, this error was less than 3%.

Ionization-current measurements contained a background of about 10% due to cosmic rays, radioactive contaminants within the chamber, and insulator strain currents. These were separately evaluated and introduced as corrections to the data. Collector currents were measured by a charge accumulation method within an accuracy of about 2%. Collector voltages employed were sufficient to ensure ion current saturation but not so large as to cause gas multiplication of ions.

It was convenient to use the ionization chamber as a proportional counter for determination of the absolute value of source activity. This has the advantage of offering exactly the same geometrical and physical conditions as those occurring for the ionization current measurements. The recoil particles themselves could not be detected in this way since they generated only about 900 ion pairs before reaching the end of their range; whereas, the equivalent input noise level of the amplifier-ion chamber system was about 2000 ion pairs. However, the α particles could easily be detected and counted with 100% efficiency. Independent measurements with the vacuum spectrometer were used to determine the ratio of recoil particles to α particles emitted by the source. Consequently, the recoil-particle emission rate could be fixed rather accurately from the α-particle count.

RESULTS

A preliminary survey quickly showed, as might be expected, that the energy and the charge of the recoil particles emitted from a surface source deposit depends

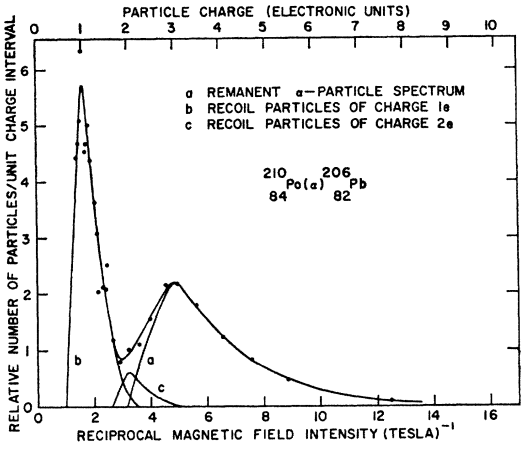

FIG. 3. Charge spectrum of particles emitted from the ^{210}Po-(α)^{206}Pb disintegration in a thin surface deposit on polished platinum.

upon the conditions of source preparation and upon its age after preparation. It was found that recoil particles escaped with nearly their full nascent energy provided that the source deposit was thin and freshly prepared. As either the age or the thickness of the source increased, the recoil energy was degraded. Thin, new sources, for purposes of this paper, are defined as deposits of sufficiently small thickness that the recoil particle escapes with at least 90% of its nascent energy. For ^{210}Po deposited on platinum, this requirement indicates a surface activity of less than 0.6 μC/cm^2 and an age less than 24 h. Such sources emitted essentially monoenergetic recoil particles bearing one or two units of positive charge. No negatively charged recoil particles were found even though they could have been detected if emitted by the source.

The charge spectrum for recoil particles from new, thin ^{210}Po sources deposited on platinum is shown in Fig. 3. For these data, the electric field in the velocity selector was adjusted at each value of the magnetic field to maintain a constant ratio of E/B appropriate to the recoil velocity. Three distinct components can be identified. One is an α-particle component which is a remanent from the fringe in the velocity-selector profile of the α-particle line. This was identified through the introduction of a Mylar film, 6 μ thick, into the particle beam just ahead of the detector. While attenuation of the α particles by the film was negligible, it stopped the recoil particles completely. Thus, the α-particle component was isolated and measured independently. It is displaced slightly with respect to the $2e$ recoil spectrum line, with which it would be expected to coincide, because these fringe α particles do not follow a normal trajectory through the system. From the resultant analysis of the spectral components as displayed in Fig. 3 it is evident that about 90% of the recoil atoms leaving a platinum surface are singly ionized and about 10% are doubly ionized. This observation agrees with the measurements of the average charge reported by McGee[6] as well as the photographic records obtained by Walmsley and Makower[4] and by Wood and Makower.[5] It is also consistent with the state of ionization expected for ^{206}Pb atoms escaping from a metal surface whose electronic work function is about 5 eV since the first ionization potential for Pb is 7.4 eV and the second is 15.0 eV.

Because these sources were necessarily weak, counting rates were correspondingly low. Typical values were 60 counts/min with a background of 41 counts/min. The background was nearly constant and was checked frequently to establish its value within a statistical uncertainty of $\pm 5\%$. In general, the measured points plotted in Fig. 3 have a statistical uncertainty of $\pm 10\%$ which is reflected in the scatter about the smooth curves displaying the estimated best fit to the data. It was not possible to decrease the statistical uncertainty by using longer counting intervals because the source characteristics changed with age. For this reason, it was desirable to complete a spectrum scan within an 18-h period and, consequently, the counting intervals per data point were limited to 30 min or less. These limitations on counting rate and counting time are inherent in the physical phenomena investigated and therefore could not be improved significantly.

By setting the magnetic field of the spectrometer at a fixed value corresponding to the peak of the $1e$ recoil line and scanning with the electric field of the velocity selector, it was possible to measure the velocity of these recoil particles. The resulting data are shown in Fig. 4. In this, the curve shape is consistent with the resolution profile expected for a monoenergetic beam of particles entering the velocity selector. The peak falls, within experimental error, at the nascent velocity for the recoil

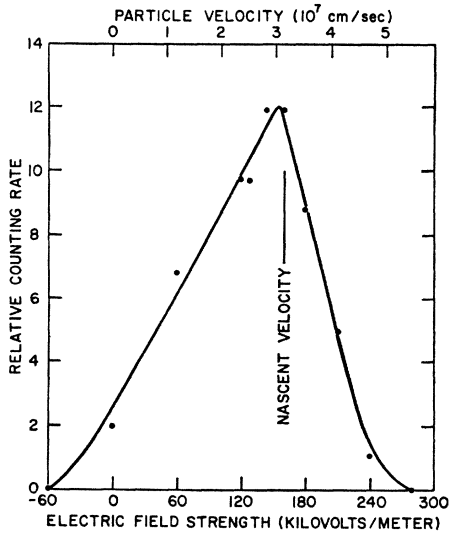

FIG. 4. Velocity spectrum of Pb^+ recoil particles from the ^{210}Po(α)^{206}Pb disintegration in a thin source deposit on polished platinum.

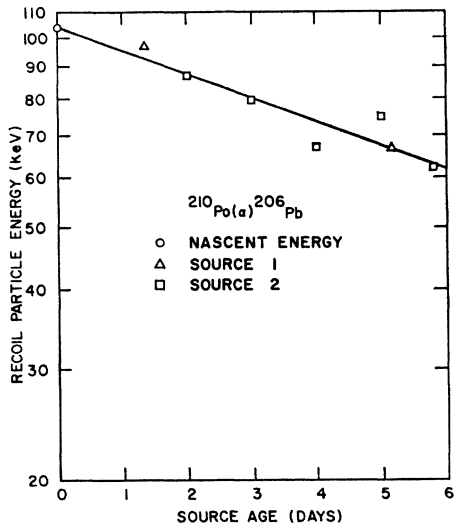

FIG. 5. Average energy of the recoil particles from the $^{210}Po(\alpha)^{206}Pb$ disintegration as a function of source age.

particles and therefore one may conclude that the ^{206}Pb atoms recoiling from the α decay of ^{210}Po on a platinum surface leave with negligible energy loss provided the source deposit is less than 24 h old. As the age of the source deposit advances, however, there is a progressive degradation in the residual energy of the escaping recoil particle. This effect is demonstrated in Fig. 5. Two sources, prepared independently but by the same technique and approximately the same activity show approximately the same exponential degradation of the mean recoil energy with time. These mean energies were determined from successive measurements of the recoil-particle residual velocity, using the velocity selector as described above. The positions of the peaks in a sequence of velocity distribution curves similar to Fig. 4 were taken as the mean velocities and from these the corresponding energies were computed. Evidently, a half-value period of 8.2 days for degradation of the recoil-particle energy is characteristic of thin ^{210}Po sources plated on a smooth platinum surface. These results, apparently, are independent of minor differences in surface conditions, since source 1 in Fig. 5 was inserted into the vacuum spectrometer on the first and sixth day after preparation. During the interval between, it was stored in dry air while source 2 was kept continuously in the vacuum and measured at the times indicated. To lose half their energy, the recoil particles would have to make from 200 to 300 collisions with gas atoms along their path. This far exceeds any conceivable thickness of adsorbed gas on the surface and therefore it is not surprising that storage in air or in vacuum makes little difference. Instead, these results suggest that, as the source ages, ^{210}Po atoms may penetrate the platinum surface several atomic layers before disintegration. Such a suggestion is consistent with observations by Haissinsky[9] on α-particle emission from ^{210}Po on platinum. However, this migration is not a normal thermal diffusion which would be negligibly slow at the ambient temperature of 25°C encountered in these measurements. Absence of significant diffusion has been confirmed by the measurements of Rona and Schmidt.[15] It appears, therefore, that the polonium migrates into interstitial microcracks and other surface imperfections before decay.[16] Since the sources employed in these measurements had an average surface deposit of less than 5×10^{11} atoms/cm^2, the platinum surface would be very sparsely populated if the polonium were distributed uniformly. However, Chamie,[17] Harrington,[18] and Haissinsky[9] have obtained evidence that thin deposits of ^{210}Po are not uniform but, instead, tend to clump at discrete site locations. From the present data it seems plausible that a new deposit may consist of individual ^{210}Po atoms spread more or less randomly over the surface but as the source ages these migrate to deeper locations prior to decay.

Quite different results were obtained with the ^{241}Am sources. Because of the longer half-life of ^{241}Am it was necessary to use thicker source deposits to attain a significant counting rate at the detector. The sources, in this case, were microcrystals of the nitrate deposited from solution on polished platinum surfaces. A preliminary survey established that emitted recoil particles indeed were degraded in energy. Accordingly, the velocity selector in the spectrometer was set for $v=2.2\times10^7$ cm/sec corresponding to a 35% loss in energy for the recoil particles. The resulting charge spectrum for particles of this velocity is shown in Fig. 6. Again, the remanent α-particle component is prominent, but after its subtraction there is no evidence of significant numbers of charged recoil particles bearing a charge between 0 and $6e$. At $6e$ a small peak occurs followed by

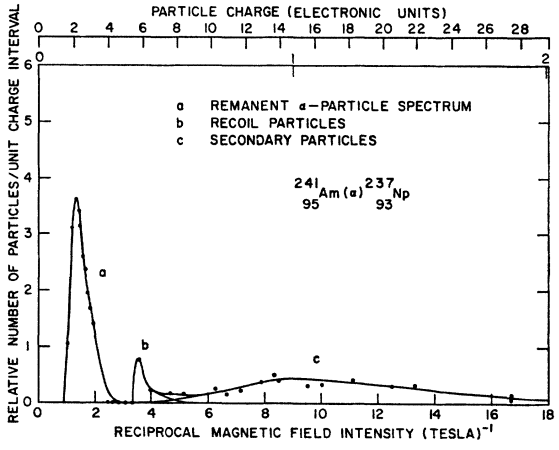

FIG. 6. Charge spectrum of particles emitted from the $^{241}Am(\alpha)^{237}Np$ disintegration in a thick surface deposit on polished platinum.

[15] E. Rona and E. A. W. Schmidt, Wien. Ber. **136**, 2a 65 (1927).
[16] E. Montel, J. Phys. Radium **10**, 78 (1929).
[17] C. Chamie, J. Phys. Radium **10**, 44 (1929).
[18] Harrington, Phil. Mag. **6**, 685 (1928).

a broad distribution whose maximum falls at about $15e$. The appearance of charge $6e$ can be understood as corresponding to the hexavalent state Am^{6+} prior to disintegration. There is no plausible explanation, however, for the appearance of higher charge states of the recoil atom. Its recoil velocity is too low to produce such an ionization state by collisions with other atoms of the source. Moreover, an estimate of the ionization energy based on an approximation developed by Slater[19] indicates about 1 keV for 12 electrons removed from the ^{237}Np atom. This far exceeds the excitation energy of 150 eV expected as a result of the α decay of ^{241}Am. However, in a nitrate crystallite the ^{241}Am is surrounded by oxygen and nitrogen atoms so that when disintegration occurs the chemical bonds will be ruptured and kinetic energy transferred to these atoms. The energy spectrum for such secondary particles is likely to be very broad with low energies of only a few electron volts predominating but with a long, thin tail extending to the maximum energy that may be communicated in a direct, head-on collision. If one assumes that the broad distribution appearing in Fig. 6 is due to oxygen atoms, then the corresponding charge scale should be expanded in the ratio of the mass of the recoil to the mass of the oxygen atom. This establishes the second scale on Fig. 6 which indicates, quite plausibly, that such oxygen atoms would carry a charge of $+1e$. The relative abundances for the various charge components in Fig. 6 are appropriate only to a velocity of 2.2×10^7 cm/sec. These change if a different velocity is selected and also change with source age so the results given here are merely typical under the conditions specified.

Appearance of secondary particles from α-active sources has been observed previously by Lawson[20] who introduced the term "aggregate recoil" to describe the spontaneous ejection of active atoms from surface deposits of ^{210}Po. He found that "sputtering" of the adjacent metal due to α-particle bombardment contributed, in general, less than 1% of the total particle flux ejected from the surface. Instead, his results indicated that the particle transport was due to eruption of particle aggregates consisting, on the average, of at least three atoms. These observations are consistent with those of McGee[6] and with the present results obtained with an electron multiplier detector. To measure total particle emission, the source holder and velocity selector in the spectrometer, previously illustrated in Fig. 1, were aligned with the aperture of the electron multiplier so that, in the absence of electric and magnetic fields, particles from the source would reach the detector by following straight-line trajectories. Individual pulses from the detector observed on an oscilloscope showed an initiating pulse followed by two or three subsequent pulses raggedly spaced along a ten-microsecond sweep. These completely disappeared when a small electric or magnetic deflecting field was applied. From this behavior it was evident that secondary particles were being ejected from the source at the instant of disintegration, thus supporting in detail the aggregate recoil hypothesis of Lawson. The frequency of occurrence of these grouped pulses greatly exceeded that expected for chance coincidences at the counting rate observed. Also, the time of flight for the assumed secondary particles was consistent with measurements of their energy using retarding electric fields. The results indicated that at least 75% of the secondary particles had energies less than 40 eV. By applying a magnetic field sufficiently great to deflect all the charged particles, including secondaries, recoils, and α particles, away from the detector it was determined by n_N, the remaining count above background, that some of the particles were neutral. The possibility that these neutrals might be photons from the source was eliminated from consideration since a transparent Mylar film 6μ thick placed in the beam reduced the intensity to 2% of its original value, a result which was not statistically different from zero. Additional sorting of the components was accomplished, first, by measuring the α-particle count n_α through Mylar film with zero deflecting fields; then, after removing the film, the count with zero deflection gave the total $n_\alpha+n_R+n_S+n_N$ of α particles, recoils, charged secondaries, and neutrals, respectively. A small electric field in the velocity selector of about 100 V/cm was sufficient to sweep away the low-energy secondaries giving the resultant count $n_\alpha+n_R+n_N$. Thus, by appropriate subtraction each component was determined. The counting efficiency for α particles, recoil particles, and charged secondaries is close to 100%, but the efficiency is unknown for the neutral particles. Accordingly, n_N must be regarded as a lower limit. Based on the assumption that secondary particles for each disintegration are ejected from the surface with an angular distribution similar to that of the α particle, but that the probability of α emission in the direction of the detector is 50%, the following results have been obtained. For new, thin ^{210}Po deposits on platinum there were an average of 5 positive ions and 3 detected neutral particles ejected per disintegration, but from the thicker deposits of ^{241}Am the numbers increased to 15 positive ions and 10 detected neutral particles per disintegration. Quite likely, negative ions and electrons were ejected as well; however, the accelerating potential of 3000 V at the input to the electron multiplier discriminated against these.

The emission rate of secondary particles also changes with time but apparently is not significantly affected by adsorption of gas or oxidation of the surface since it seems to make little difference whether the source is stored in a vacuum or in air. Typical behavior for ^{241}Am deposited on platinum is illustrated in Fig. 7. Initially, there is a slight but statistically significant increase in the particle emission rate, perhaps owing to crystal growth or some other surface readjustment processes.

[19] J. C. Slater, Phys. Rev. **36**, 57 (1930).
[20] Lawson, Wien. Ber. **127**, 1 (1918); **128** 795 (1919); Nature **102**, 465 (1919).

It is then followed by a steady decline which probably is exponential with time. This latter decrease is more rapid than can be accounted for by natural radioactive decay or the physical loss of active material through ejection of secondary particles. Since a layer of condensed material about 300 Å thick is sufficient to stop the recoils completely, then much less than this will inhibit the emission of secondaries. Accordingly, the growth and decline of secondary particle emission may be associated with ever increasing layers of inert material originating from the source eruptions being readsorbed on the surface.

By extending the detection technique described above, it was possible to determine the ratio of the number of recoil particles to the number of alpha particles emitted from a source. These results are illustrated in Fig. 8 for ^{210}Po sources and in Fig. 9 for ^{241}Am sources. Since the pulse-height spectrum of the electron-multiplier-voltage output for α particles was different from that for recoil particles, the relative count at any one level of discriminator bias was not a true indication of the relative

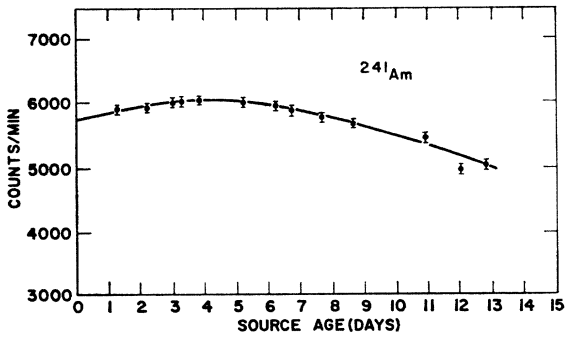

FIG. 7. Total particle emission rate in vacuum from a thick source deposit of ^{241}Am of polished platinum.

number of particles. But, by taking a sequence of readings at decreasing discriminator voltages and then extrapolating the trend to zero voltage, a determination of the ratio n_R/n_α independent of the pulse-height spectrum was obtained. Extrapolation was accomplished by taking the least-squares linear fit to the data and solving for the zero-bias intercept. It will be noticed that thin, new sources of ^{210}Po deposited on platinum all have an intercept near $n_R/n_\alpha=1$. More precisely, the average value for this intercept is 0.99 with a standard deviation of 3%. There is clear evidence of the effects of aging, however, so that at $7\frac{1}{2}$ days after preparation the ratio of n_R/n_α has dropped to 0.9 and the average residual energy of the recoil particles correspondingly has dropped from 103 to 65 keV. Effects of the shifting energy spectrum are visible in the changing slope of the extrapolation lines. Thick source deposits, as expected, show a significant reduction in the relative number of recoil particles emitted. For ^{241}Am, a source deposit sufficiently thin to permit full emission of recoil

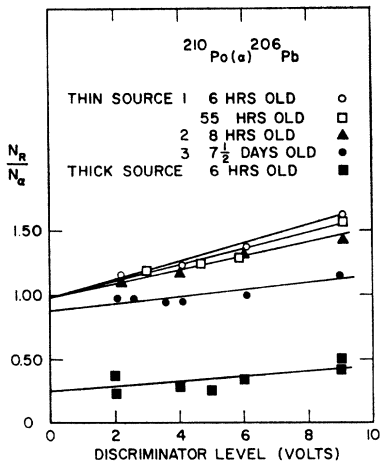

FIG. 8. Ratio of the number of recoil particles counted N_R to the number of α particles counted N_α as a function of the pulse-height discriminator level.

particles was necessarily very weak and therefore the data points have a larger scatter.

As a result of these investigations it is clear that earlier measurements of the range of recoil particles in gases and the ionization produced by them is subject to question since the condition of the source deposit usually was not well defined. For this reason, a redetermination of the range and ionization in argon gas produced by ^{206}Pb recoils from ^{210}Po α disintegration was carried out in the ionization chamber described earlier. A typical curve showing the relationship between ionization current and chamber pressure is shown in Fig. 10. At zero pressure, there was a small residual current of about 4 fA (10^{-15} A) that remained after subtracting the background and insulator strain currents. This current is generated through the transport of charge by the α particles, recoils, ions, and electrons emitted from the source. With increasing argon gas pressure, the ionization current rises approximately linearly while the range of the emitted particles decreases until, at the point A, the recoils have a range equal to the chamber radius. The rounding of the knee in the curve is related, in part, to the finite extent of the source planchet but this is a small contribution and most of the rounding is due to range straggling. Further increase in gas pressure is attended by a continuing increase in ionization current engendered by the α particles, which, at these pressures have ranges longer than the chamber radius. If this

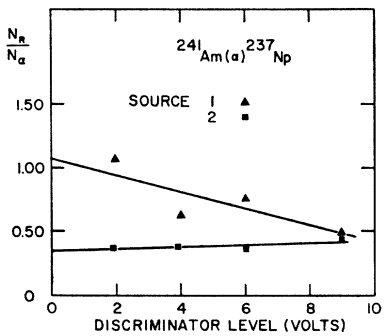

FIG. 9. Ratio of the number of recoil particles counted N_R to the number of α particles counted N_α as a function of the pulse-height discriminator level. In this case, some of the recoil particles are probably O^+ ions in addition to Np^{6+} ions.

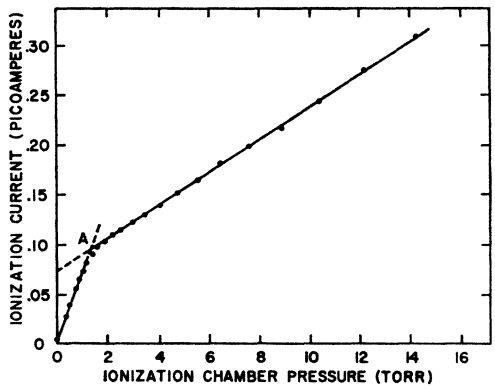

FIG. 10. Ionization current in argon gas produced by particles from the $^{210}Po(\alpha)^{206}Pb$ disintegration as a function of chamber pressure.

linear portion of the curve is extrapolated back to zero pressure, the intercept will be the ionization current assignable to the recoil particles. Although secondary particles are also emitted by the source, their contribution to the ionization current is less than 1% and was considered negligible. To confirm that the knee in the ionization curve was actually associated with the recoil particles, an aluminum foil, 6μ thick, was placed over the source. This was sufficiently thick to stop the recoils but not the α particles. Accordingly, the knee in the curve disappeared and only the linear increase in ionization current due to the α particles was then observed.

The extensions of the two linear portions of the ionization-current curve intersect at the point marked A in Fig. 10. This intersection may be used to evaluate the extrapolated range. For nascent energy recoil particles from the $^{210}Po(\alpha)^{206}Pb$ disintegration, the intersection falls at an argon pressure of 1.32±0.03 Torr. Since the hemispherical-ion-chamber radius was 4.50 ±0.01 cm, the extrapolated range in argon is 5.9±0.2 cm Torr; or alternatively, the equivalent extrapolated range in argon at STP would be 78±2 μ. Using a similar technique, Jessi and Sadauskis[13] have reported an extrapolated range of 77±8 μ for the ^{206}Pb recoils in argon at STP while a much earlier measurement by Bianu and Wertenstein[11] gave an equivalent extrapolated range of 92 μ in air at STP. Range determinations for the recoils from α disintegrations of Ac*C*, Th*C*, and Ra*A* have also been made[10,21] yielding results consistent with the present measurements.

An accurate determination of the recoil-particle emission rate from the source was accomplished by counting the emission of α particles as described earlier and using the knowledge that new, thin sources of ^{210}Po on platinum emitted recoil particles in the ratio $n_R/n\alpha=0.99\pm0.03$. Sources less than 10 h old were employed. With these data, the recoil-particle ionization current obtained from the zero-pressure intercept as in Fig. 10 was reduced to an average ionization charge per recoil particle. Then, with the further knowledge that these particles were emitted with an energy of 100±3 keV it was possible to express an average energy loss per ion-pair created. In argon gas at sufficiently low pressure that recombination of ions is not significant, the average energy loss per ion pair W_R by the ^{206}Pb recoil particles is 121±6 eV/ion pair. A similar determination for the α particles emitted by the source gave an average energy loss of $W_\alpha=26.8\pm1.1$ eV/ion pair which compares very favorably with the value of 24.3 obtained by Schmieder[22] and 27.7 eV/ion pair determined by Stetter.[23] Except for the work of Stone and Cochran[24] only relative values of energy loss per ion pair for the recoil particles compared with that for the α particles from the same disintegration has been reported previously. Jessi and Sadauskis[13] gave $W_R/W_\alpha=4.5\pm0.2$ in argon for the particles from the $^{210}Po(\alpha)^{206}Pb$ disintegration and Madsen[25] cites 4.4±0.3 while the present results yield a value of 4.4±0.2 for this ratio. This is remarkably good agreement in view of the difficulties associated with source thickness and aging.

EVALUATION

In discussing the energy loss of heavy particles it is useful to adopt a terminology introduced by Bohr (Ref. 35). Particles colliding with relative velocities less than the characteristic Bohr velocity $v_0=e^2/\hbar$ for atomic electrons will experience a quasiadiabatic interpenetration of the respective electron fields and the encounters will be nearly elastic. Ionization does occur, but the efficiency is low and most of the energy loss experienced by the incident particle can be accounted for by the energy transfer to the recoiling resident atoms of the stopping medium. For the measurements reported here, the initial particle velocity was $v=0.142\,v_0$. However, the relative kinetic energy between the ^{206}Pb particle and the argon gas atoms initially was 16.8 keV. This is well above the threshold for electronic excitation and, therefore, ionization is energetically possible although the detailed process by which it is produced is obscure. Because of the reduced ionization efficiency from that associated with fast particles, such as the α particles, and because of the energy transfer that occurs in numerous quasiadiabatic ionic collisions, the energy loss per ion pair is much larger for the slow, massive ^{206}Pb particles than for α particles, as the measurements disclose. This realization, however, brings up a question regarding the measurement of range by means of the ionization produced. Under the conditions experienced in these investigations, 90% of the ^{206}Pb recoil atoms initially were singly ionized and 10% were doubly ionized. But measurements by Phipps, Lowry, and Boring[26] show that the energy loss per ion pair for

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low-energy ions is quite insensitive to their ionic charge. Thus, the two charge states present should have little effect on the results. On the other hand, there is a danger that near the end of its trajectory, a particle will produce no more ionization and the range obtained from ionization measurements will be less than the physical range. Knipp and Teller,[27] and later Knipp and Ling,[28] have discussed the factors affecting ionization yields for heavy, slow ions while Schmitt and Leachman[29] have obtained evidence that an ionizing defect exists for fission fragments. But more recently, Sevier,[30] using Ilford $K5$ emulsions, has found that the final rest positions of the fragments from the fission of ^{252}Cf were located $0.131\pm0.053\,\mu$ before the ends of the fragment tracks with no clear indication of extended range owing to an ionizing defect. The distance between the final position and end of the track is approximately the radius of the last exposed silver halide grain. Using different methods, Wellish[31] and Dee[32] independently have determined that about 85% of the recoil particles reach the end of their range in air with one unit of positive charge and the rest are neutral. Only in gases having complex organic molecules, such as ether, do all of the recoil particles reach the end of their range as neutral atoms. With cloud chambers operating at reduced pressure, Akiyama[33] and Joliot[34] have been able to obtain photographs of recoil tracks showing that there is no significant range defect for these particles but that the ionization continues to the end of the particle trajectory. The photographs clearly indicate that the α-particle track which signals the decay of AcA occurs precisely at the end of the track left by this particle as it recoiled from the previous decay of An in the cloud chamber. Thus, it appears that ionization measurements indeed yield a valid determination of recoil-particle range.

In the photographs published by Joliot[34] are many examples of collisions between recoil particles and gas atoms, indicating that forward projection of oxygen or nitrogen atoms is quite probable. This evidence offers further support for the earlier interpretation of the broad spectrum curve in Fig. 6 as due to oxygen ions projected from the source by the recoil particles.

Bohr,[35] in an extended treatment of the penetration of atomic particles through matter, has examined the problem of energy loss by slow moving ions in a gas. Further developments based on this fundamental exposition by Bohr have been introduced by Knipp and Ling[28] and also by Lindhard, Scharff, and Schiott.[36] These discussions lead to the same theoretical formulas and differ basically only in the collision cross section assumed by each. In the center-of-mass system the scattering due to collisions between shielded nuclei is assumed to be spherically symmetric at low relative velocities. The collision total cross section established by Bohr as an approximation for shielded Coulomb fields is

$$\sigma=(\pi a^2/e)\zeta\,, \tag{1}$$

where e is the base of the natural logarithms and $a=a_0(Z_1^{2/3}+Z_2^{2/3})^{-1/2}$ is an effective mutual screening radius between the incident particle of atomic number Z_1 and a gas atom of atomic number Z_2, while $a_0=\hbar^2/m_e e^2$ is the characteristic Bohr radius for the hydrogen atom. The screening parameter ζ is defined

$$\zeta=\frac{M_1+M_2}{M_2}\frac{Z_1Z_2e^2}{aE_1}\,, \tag{2}$$

with M_1 and M_2 representing the masses of the incident and gas atoms, respectively, e the electronic charge, and E_1 the energy of the incident particle in the laboratory coordinate system. The conditions under which the approximate cross section in Eq. 1 may be applied require that $\zeta>1$. This means that the distance of closest approach between scattering centers is greater than the screening radius, a condition labeled "excessive screening" by Bohr. For the 103-keV ^{206}Pb recoil particles, $\zeta=12.0$ which falls well within the limit prescribed for excessive screening.

By considering the shielding approximation differently, Knipp and Ling propose a collision cross section

$$\sigma=\pi a^2\zeta\,\ln 2\,, \tag{3}$$

while Lindhard, Scharff, and Schiott use

$$\sigma=(\pi^2a^2/2e)\zeta\,. \tag{4}$$

These latter also have carried out numerical calculations using the Thomas-Fermi statistical model,[36] but the corresponding cross section cannot be expressed in closed form. All of the cross sections listed in Eqs. (1), (3), and (4) can be expressed by the prototype formula

$$\sigma=\eta\pi a^2\zeta\,, \tag{5}$$

since they differ only in the numerical factor η. From σ, the stopping cross section S_n due to shielded nuclear collisions may be obtained by integrating the energy transfer per collision

$$E_2=[2M_1M_2/(M_1+M_2)^2](1-\cos\theta)E_1 \tag{6}$$

over all scattering angles θ.

$$S_n(E_1)=(1/4\pi)\int_{4\pi}\sigma(E_1)E_2(\theta,E_1)d\Omega\,, \tag{7}$$

$$=\tfrac{1}{2}\sigma E_m\,,$$

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where

$$E_m=[4M_1M_2/(M_1+M_2)^2]E_1 \tag{8}$$

is the maximum energy transferred in a direct collision. In addition to the energy loss through shielded nuclear collisions there is a small contribution from electronic excitation. According to Lindhard, Scharff, and Schiott[36] a satisfactory expression for the electronic stopping cross section S_e is

$$S_e=\xi_e 8\pi a(Z_1Z_2e^2/(Z_1^{2/3}+Z_2^{2/3}))(v/v_0). \tag{9}$$

This formula was derived from the Thomas-Fermi statistical model and applies for $Z_1>10$ and $v<v_0Z_1^{2/3}$ down to very low velocities. The coefficient ξ_e is closely approximated by $Z_1^{1/6}$.

The range R of the incident particle in the gas, computed from the familiar integral

$$R=(1/N)\int_0^{E_1}(S_n+S_e)^{-1}dE_1, \tag{10}$$

is then

$$R=\frac{2E_0}{kN}\left[\frac{v}{v_0}-\frac{\eta K}{2k}\ln\left(1+\frac{2k}{\eta K}\frac{v}{v_0}\right)\right], \tag{11}$$

where N is the number of atoms per cm^3 in the stopping medium and

$$E_0=\tfrac{1}{2}M_1v_0^2,$$
$$k=8\pi aZ_1^{1/6}(Z_1^{2/3}+Z_2^{2/3})^{1/2}Z_1Z_2e^2,$$
$$K=4\pi aZ_1Z_2e^2M_1(M_1+M_2)^{-1}.$$

Since the coefficient k which is associated with the electronic stopping cross section is smaller than K the coefficient associated with the nuclear stopping cross section, the range formula of Eq. (11) may be approximated by the following expansion.

$$R=\frac{2E_0}{N\eta K}\left(\frac{v}{v_0}\right)^2\left[1-\frac{4}{3}\frac{k}{\eta K}\frac{v}{v_0}+\cdots\right]. \tag{12}$$

Here, the leading term expresses the range expected when considering only energy losses due to shielded nuclear collisions while the second term is a correction due to losses from electronic excitations. The effect of choosing the various collision cross sections is contained in the coefficient η which may assume the values e^{-1} for the cross section obtained by Bohr, $\pi(2e)^{-1}$ for that of Lindhard, Scharff, and Schiott, and ln 2 for that of Knipp and Ling.

A comparison of these theoretical predictions with existing measurements of the range of heavy particles in argon[37,38] is given in Fig. 11. For this purpose it is useful to employ dimensionless range and energy parameters

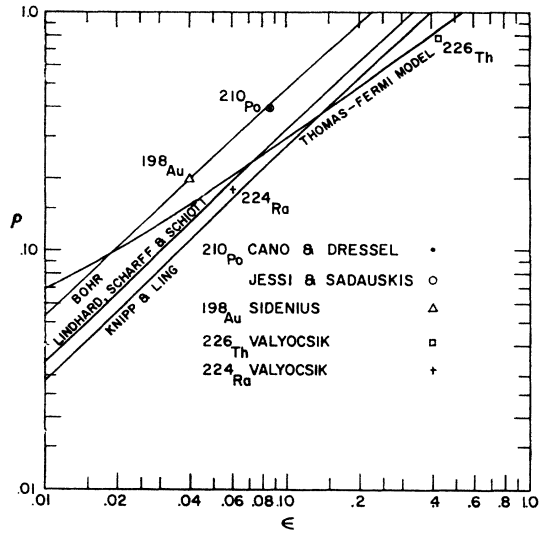

FIG. 11. Comparison of theoretical predictions with observed ranges of recoil particles in argon in terms of the dimensionless range and energy parameters ρ and ϵ.

introduced by Lindhard, Scharff, and Schiott.

$$\rho=4\pi a^2N(M_1M_2/(M_1+M_2)^2)R,$$
$$\epsilon=(a/Z_1Z_2e^2)(M_2/M_1+M_2)E,$$

which permit a universal display independent of the incident particle or the stopping medium. The curves are calculated from Eq. (12) except for the Thomas-Fermi curve which was obtained from Ref. 36. While the data and the theoretical predictions broadly overlap there is insufficient evidence to decide in favor of one or another calculation. This same ambiguity persists over a wide range of energy and for a large sampling of data as demonstrated in a compilation assembled by Linhard, Scharff, and Schiott.[36] In general, measurements agree with the Thomas-Fermi calculations within $\pm 20\%$ except at the lower energies, $\epsilon<0.2$, where the theory is expected to be inaccurate. Here the data seem to indicate a range greater than predicted. The present measurement which gives $78\pm 2\,\mu$ for the range of ^{206}Pb in argon agrees very well with the value of $79.7\,\mu$ calculated from the Bohr formula and also agrees with a range of $77\pm 8\,\mu$ reported by Jessi and Sadauskis.[13] The Bohr formula, however, is derived from an approximate theory and so the close agreement must be interpreted as an evaluation of the approximations employed.

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