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# CHARGE DISTRIBUTIONS OF ALPHA-RECOIL ATOMS FROM ELECTRODEPOSITED $^{210}$Po SOURCE

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Charge distributions have been measured for $^{206}$Pb recoil atoms emerging from a $^{210}$Po source electrodeposited on platinum. It has been found that more than 50% of the recoils are neutrals when the average energy loss of the recoils is about 18 keV. The observed large fraction for the neutrals indicates that a strong neutralization of the recoil ions occurs with increasing energy loss.

## 1. Introduction

At present numerous studies [1] on the charge distributions have been performed for high-energy heavy ions in the MeV range generated by heavy-ion accelerators. However, at very low energies below 10 keV/amu, the charge distributions have scarcely been measured. The specific nature of the heavy-ion matter interactions in this energy range is that the energy loss of the ion is essentially determined by the interaction with nuclei, not by that with atomic electrons, i.e., the nuclear stopping power dominates the electronic one [2]. This situation is in sharp contrast to the high-velocity case. Thus, for low-velocity heavy ions, it is of particular and fundamental interest to study the charge distributions in connection with energy loss.

In this respect, it should be attempted to investigate the charge distributions of $^{206}$Pb daughter atoms recoiling from $\alpha$ decay of $^{210}$Po, since the $^{206}$Pb atom has a nascent kinetic energy of 103 keV corresponding to the recoil velocity of $v_r = 0.14\ v_b$, where $v_b$ is the Bohr velocity of $e^2/\hbar$. $^{210}$Po is a unique nuclide having nearly 100% $\alpha$ transition to the ground-state of a stable daughter nucleus. (The feeding of the 803-keV level of $^{206}$Pb is $\sim 10^{-5}$.) This means that the charge state of $^{206}$Pb recoil cannot be influenced by internal-conversion and Auger-transition processes, which, in general, seriously alter the charge distributions.

There have been several charge-state measurements [3,4] for recoils from $\alpha$-decay nuclides, either in gas or solid phase. However, most of the nuclides investigated are of complex decay schemes. By using spontaneously deposited $^{210}$Po source onto platinum, Cano and Dressel [4] reported that approximately 90% of the $^{206}$Pb recoil atoms were emitted singly ionized and 10% were doubly ionized in a positive charge state. However, their measurement was made only for a thin and new source, from which the recoil ions were emitted with the original kinetic energy of 103 keV. They also observed the degradation of the mean recoil energy with time. They attributed this to the Po-atom behavior: Po atoms penetrate the platinum surface several atomic layers before disintegration as the source ages.

In this work the charge distributions of recoil ions from $^{210}$Po were investigated with the aim of studying the influence of the energy loss of recoils on the charge distribution. Accordingly, measurements were made for the energy-degraded recoil ions emitted from the electrodeposited source. Experimental details of source preparation, apparatus, and data analysis are described. Some discussion on the results is presented, including the consideration of the effect of $\alpha$ decay on the electronic structure of the recoil atoms.

## 2. Experimental

The $^{210}$Po source was prepared by electrodeposition onto platinum. The chemical form of the solution purchased from Amersham International plc, England, was $Po(NO_3)_2$ in 5 ml 3N $HNO_3$ with an activity of 3 mCi. About 0.4 ml of the solution ($\sim 240\ \mu$Ci) was transferred to a small Teflon vessel installed in a glass system and evaporated to dryness with dry air passing through the system. The residue was visible as a faint yellow color. To purify the solution, a solvent-extraction

method was applied. The residue was dissolved in 0.5 ml of 3N HCl + 0.25M KI. The solution, transferred to a small test tube, was added with about 2 ml of isopropyl ether, then $^{210}$Po was extracted to the isopropyl ether by stirring with $N_2$ gas bubbles introduced into the solution. The isopropyl ether was added with 0.5 ml of 3N HCl, to which $^{210}$Po was extracted again. The 3N HCl solution was evaporated and the residue was dissolved in 1 ml of 0.5N HCl. With this solution, the electrodeposition was made onto a 0.02-mm-thick Pt plate with an active area of 2.4-mm diameter. The invisible source of an activity of ~ 30 $\mu$Ci was obtained by applying the current of about 200 $\mu$A for 20 min. The other source of ~ 100 $\mu$Ci with an active area of about 6-mm diameter was prepared in similar fashion, but the current used for the deposition varied from 800 $\mu$A to 5 mA for 10 min. Inspection of the source showed a slightly dark surface. The amount of $^{210}$Po indicated by the source intensity corresponds to several monolayers, but the microscopic structure and chemical composition of the source remained unknown. The sources were exposed to air for ~ 6 h before insertion into the vacuum system of the apparatus.

A schematic diagram of the apparatus is shown in fig. 1. The source was installed in a source holder with a double slit-collimator. To avoid undesired Po contamination, the source holder, effectively confining the knock-out Po [5], was separately evacuated from the main chamber. The typical pressure of the chamber was kept at $2 \times 10^{-6}$ Torr by using diffusion pumps and cold traps. A surface barrier detector (SBD) was mounted on the wall of the chamber and served to monitor the Po contamination. The detector is sensitive to the contamination because the gold layer of its entrance window tends to favourably attach Po. During the whole course of the measurements, it was found that the contamination detected had a negligible influence on the present experiment.

Collimated particles emerging from the source were analyzed with an electrostatic deflector by supplying ± 2 kV between a 25-mm gap distance corresponding to the electric field of 1.6 kV/cm. The charge-analyzed ions were detected by a channeltron (Murata, EMS-6081B) mounted in a copper box with 1.6-mm high and 6.0-mm wide entrance slit. The channeltron was biased with the cone at ground potential and the anode at + 3.0 kV. The signals were fed to a conventional amplifying circuits, and the pulse heights were analyzed and stored in PDP 11/04 computer. To obtain charge spectrum, the channeltron was moved along the vertical axis of $Y$ shown in fig. 1, with a step of $\Delta Y = 0.63$ mm by using a gear device driven by a small motor. The motor was controlled by the computer to enable us automatic sequential measurements.

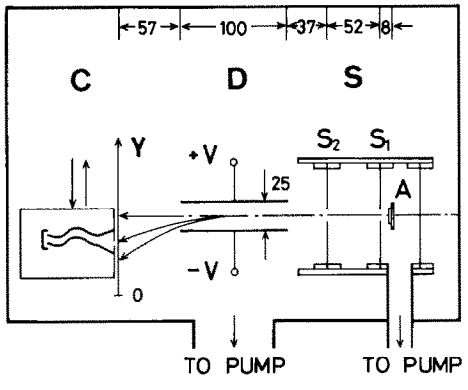


Fig. 1. Schematic diagram of the apparatus: S, source holder; A, $^{210}$Po source; S1, S2, aluminum collimating slits with 1.0-mm-diameter openings; D, electrostatic deflector with a gap distance of 25 mm; C, channeltron mounted in a copper box with 1.6-mm high and 6.0-mm wide entrance slit. The channeltron was moved along the axis of $Y$ with a step of $\Delta Y = 0.63$ mm. Dimensions are given in mm.

## 3. Results and discussion

The charge spectra obtained with the 30 $\mu$Ci source are shown in fig. 2. The peaks for singly- and

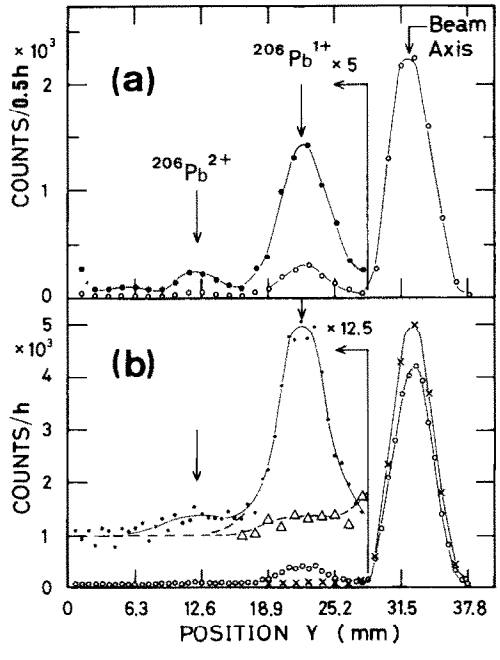


Fig. 2. Charge spectra of particles emerging from the 30-$\mu$Ci $^{210}$Po source electrodeposited on a platinum plate. The electric field of 1.6 kV/cm was applied to the deflector. (a) Spectrum observed 17 h after source preparation. (b) Spectrum for 23 d after the preparation (○, ●), together with the spectrum obtained without the electric field (×, △), and the broken lines indicating the background as explained in the text.

doubly-ionized $^{206}$Pb recoil ions are clearly seen in fig. 2(a), which was observed 17 h after the source preparation. However, the data obtained 23 d after the preparation, fig. 2(b), showed that the $Pb^{2+}$ peak appeared as a smooth bump, reflecting the increasing energy dispersion as time passed. Also shown in the figure is a spectrum observed without the electric field of the deflector. This spectrum showed no structure in the vicinity of $Pb^{+}$ position. Furthermore, when the electric field was reduced from 1.6 kV/cm to 1.4 kV/cm, the both peaks were found at the expected positions, so that it was confirmed that the observed peaks resulted from the recoils. With the measured distance of 10.1 mm between the $Pb^{+}$ peak position and the beam axis, and the dimensions of the apparatus, fig. 1, the average energy of the recoil ions was estimated to be 85 keV, which corresponds to the mean energy loss of 18 keV. In contrast to the observation of Cano and Dressel [4], the average energy of the recoils was found to already be degraded by 18 keV when only 17 h elapsed after the source preparation. Moreover, more gradual degradation of recoil energy was observed as time passed, although the energy (position) resolution of the apparatus was limited, being only 40 keV (fwhm). This may be due to the different surface conditions resulting from the spontaneous deposition and the electrodeposition of $^{210}$Po onto platinum.

Emission of secondary particles such as oxygen atoms from $\alpha$ active sources has been known [4], but neither precise knowledge of the particles nor mechanism for the emission has been clear. When the electric field of the deflector was removed, the peak observed at beam axis contained the total of these secondaries, recoils, and $\alpha$ particles. Since the electric field of 1.6 kV/cm was strong enough to deflect the particles except $\alpha$'s, the total of neutral secondaries, neutral recoils, and $\alpha$ particles formed the peak at beam axis when the electric field was applied. Thus, the spectral component of $\alpha$ particles was measured in the vicinity of beam axis by using SBD being mounted instead of the channeltron, because SBD is insensitive to the other particles than $\alpha$'s. The result is given in fig. 3, together with the results observed by the channeltron with and without the electric field. The latter results are a part of those shown in fig. 2(b). The sorting of the each component was attempted in the following way. Fricke et al. [6] made the elaborate measurements for the channeltron detection-efficiency for $H^{+}$, $He^{+}$, $Ne^{+}$, $Ar^{+}$, $Kr^{+}$ and $Xe^{+}$ ions with energies of 4–15 keV. The efficiency was found to range between 80 and 100% and, for Xe ions, to be very close to 100%. It can be said from their observations that the recoil ions having the energy of around 85 keV are energetic enough to produce secondary electrons from the entrance surface of the channeltron and thus yield detectable pulses. Furthermore, the pulse-height distributions were recorded for every step of the measurements to monitor the gain variations of the pulses, which were found to be negligible during the experiment. The discrimination level used for integration of the counts was set as low as possible but comfortably above the system noises. From these considerations, the counting efficiency for the recoil particles as well as $\alpha$'s can be reasonably assumed to be 100%. However, the efficiency is unknown for the secondaries because of the lack of knowledge about them. Accordingly, the detected number of the secondaries must be regarded as a lower limit. The background subtraction was made by using, for $Pb^{+}$, the data of the spectrum observed without the electric field, and, for $Pb^{2+}$, the data at the region below $Y = 6.3$ mm. In addition, an overlap of $Pb^{+}$ and $Pb^{2+}$ peaks was taken into account. This caused a relatively large error of 26% for the determination of $Pb^{2+}$ counts. An example of the background thus determined was depicted with the broken lines in fig. 2(b).

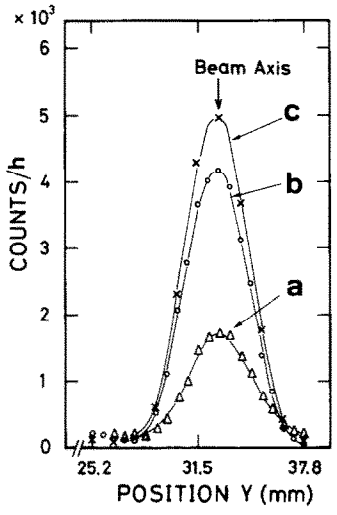


Fig. 3. Comparison of the spectra observed in the vicinity of beam axis. (a) $\alpha$-particle component observed by a surface barrier detector. (b) and (c) a part of the spectra shown in fig. 2(b), obtained by a channeltron with and without the electric field applied to the deflector, respectively.

The results of the sorting revealed that the amount of neutral secondaries was more than that of $\alpha$'s but less than twice it, and that of charged secondaries was more than 25% of that of $\alpha$'s. The measured charge-state fractions of recoils *per* $\alpha$ *disintegration* ($f_i$, $i = 0, 1, 2$) were $f_1 = 0.36 \pm 0.03$ and $f_2 = 0.043 \pm 0.011$ for 17-h data, and $f_1 = 0.25 \pm 0.01$ and $f_2 = 0.023 \pm 0.005$ for 23-d data, respectively. The fraction ratio of $f_1/f_2$ was found to be $0.12 \pm 0.03$ for 17-h data and $0.092 \pm 0.019$ for 23-d data, respectively. Provided that the number of recoils emitted from the source is equal to that of $\alpha$'s, the fraction for the neutral recoils can be deduced to be $f_0 = 0.60$ for 17-h data, and 0.73 for 23-d data, respec-

tively. However, Cano and Dressel observed the decrease of the number of recoils with the decrease of average recoil energy. They found the ratio of the number of recoils to that of $\alpha$'s to be 0.9 when the recoil energy decreased to about 65 keV. Thus, the deduced fractions for the neutrals ($f_0$) were associated with approximately 10% uncertainties. These large fractions for the neutrals can be ascribed to the associated energy loss of the recoils, since the previous experiment [4] in which all recoils were found to be charged was made only for the recoils with the original kinetic energy of 103 keV, namely, for those being not subject to energy loss. Therefore, it is evident that the neutralization of recoil ions proceeds with the increasing energy loss. Indeed, the observed increase of the fraction for neutrals as time elapsed indicates the proceeding neutralization.

Fig. 4 presents the charge spectra obtained with the 100 $\mu$Ci source. The spectrum, fig. 4(a), similar to that in fig. 2 was observed 15 h after the source preparation. The energy dispersion observed was caused by the rather rough preparation of the 100 $\mu$Ci source, as described in the former section. After 84 d, as shown in fig. 4(b), the spectrum was found to be deformed to a great extent due to large energy dispersion associated with the mean energy loss of about 33 keV. Nevertheless, it is clear that $Pb^+$ ions were dominant when compared with $Pb^{2+}$ ions. Since $\alpha$ measurements were not performed, the charge fraction could not be determined. However, it seems that the disappearance of $Pb^{2+}$ peak supports the statement that the recoils are neutralized with increasing energy loss.

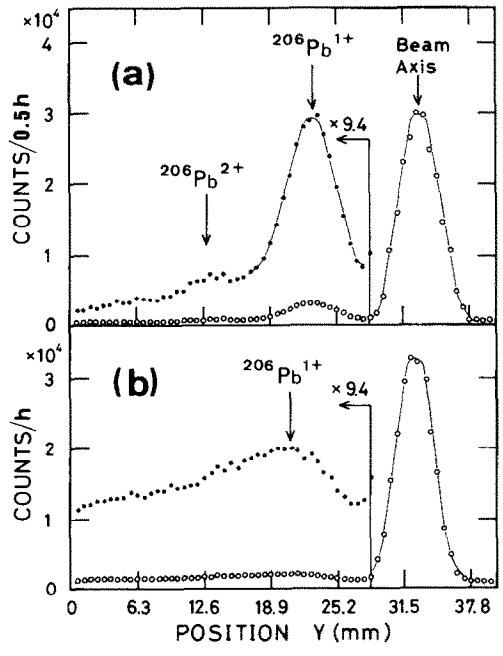


Fig. 4. The same as fig. 2, but for the 100 $\mu$Ci source. (a) Spectrum obtained 15 h after source preparation. (b) Spectrum for 84 d after the preparation.

In order to discuss further the charge changing of recoil ions in matter, the primary charge distribution initiated by $\alpha$-decay perturbation on the recoil atoms, namely, the one before being influenced by the ion–matter interactions, should be known. This primary charge distribution, however, is not well understood from either the experimental or theoretical side. Nevertheless, it can be considered in a qualitative way to give insight into the process involved and the mechanism responsible for yielding the charge distribution discussed here. Serber and Snyder [7] derived a simple equation to give an estimate of the average energy transfer, $\Delta E$, to the electronic system in nuclear transitions in which nuclear charge changes, but in the case that the transition is completely nonadiabatic. The estimate gives $\Delta E = 540$ eV for Po. In contrast to the case of $\beta$ decay, however, the estimate is not accurate for $\alpha$ decay, since the velocity of 5.3-MeV $\alpha$ particle is $7.3v_b$, and thus it is much slower than the orbital-electron velocities of the three innermost shells, but much faster than those of the two outermost shells, where the binding energies are less than 150 eV. This fact indicates that the $\alpha$-decay influence on the electronic structure can be divided into two parts; the adiabatic part for the inner-shell electrons and the sudden part for the outer-shell electrons. The adiabatic part manifests itself as a small part of the kinetic energy of $\alpha$ particle and hence appears as a part of the so-called screening correction in $Q$-value measurement [8]. Therefore, the excitation energy responsible for ionizing the recoil atom is considered to originate from the rearrangement energy deposited on the outer-shell electrons through the sudden change, experienced by these loosely bound electrons, of the effective charge. We have followed Thomas [9] to extract the average excitation energy from $\Delta E$ and have found it to be about 60 eV for $^{210}$Po. A rough estimation can be made for the charge state of the recoil ions by simply taking account of the atomic binding energies of $5d_{5/2}$, $6s_{1/2}$, and $6p_{1/2}$ for Pb atom. The estimate leads one to a charge state as high as 7, provided that the excitation energy deduced is equally distributed among the relevant electrons. Because of the lack of consideration of the interactions responsible, this may be an overestimate, but it suggests that a rather high charge state can be reached. In order to clarify this qualitative discussion, a more elaborate theoretical model should be developed for understanding the charge distribution induced by $\alpha$-decay perturbation.

In summary, by extending the measurement of Cano and Dressel [4] to energy-degraded recoils, we have

found that more than 50% of the recoils are neutrals when the average energy loss is about 18 keV, while the charged recoils are in singly- and doubly-ionized states with the fraction ratio of $f_1/f_2 \approx 0.1$ within an experimental uncertainty of 25%. From the results of the present work, it is established that the neutralization of recoil ions strongly proceeds with the increasing energy loss, indicating a decisive role of the electron capture process for the low-velocity heavy ions such as $\alpha$ recoil atoms. In addition, the observed strong neutralization of recoil ions allows us to say that the charge-states higher than the singly- and doubly-ionized states, observed in the present measurement, can be expected for the charge distributions initiated by $\alpha$-decay perturbation on the recoil atoms.

Further experimental study is needed, especially to study the correlation between charge state and energy loss. It is hoped that theoretical descriptions of the charge-changing process for low-velocity heavy ions in matter will also advance.

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