CHARGE DISTRIBUTION OF THE DAUGHTER ATOM IN α DECAY OF ^{210}Po

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The evaporation model developed by Russek and collaborators for multiple ionization of outer-shell electrons in ion–atom collisions has been applied to the charge distribution of the daughter atom produced by α decay of ^{210}Po. The calculated result indicates that most of the daughter atoms are positively charged ions with a mean charge of +1.41. The other ionization mechanisms, the recoil effect and the shake-off effect, are also considered. In comparison with the recent experimental results, it is shown that the evaporation model predicts more adequately than the recoil effect and the shake-off effect.

In α decay, atomic electrons in the daughter atom experience recoil and a decrease in the nuclear charge. The inner-shell electrons are strongly bound to the nucleus and their orbital velocity is so large that the α-decay process may be considered to be adiabatic. On the other hand, for the outer-shell electrons the total energy transfer to them during α decay is large compared to the binding energy per electron and the change in the nuclear charge is sudden in comparison with their orbital period. These facts suggest that the inner-shell electrons remain in the daughter atom, but the residual atom is, in general, in ionized states due to outer-shell ionization. In this letter, in order to obtain physical insight into the mechanism of producing the charge-state distribution of the daughter atom, we have attempted to calculate the ionization probabilities by utilizing three different theoretical models; the recoil effect [1], the shake-off effect [2], and the evaporation model [3,4]. ^{210}Po is a unique and most suitable nuclide in studying the charge distribution due to α decay, as explained in ref. [5]. In all the three models, the six outer-shell electrons in 6s, $6p_{1/2}$ and $6p_{3/2}$ shells are taken into account for ionization.

In the calculations of the recoil effect [1] and the shake-off effect [2], all electrons are assumed to be ejected independently. When n_i is the number of electrons emitted from the ith shell, the probability for n-electron ejection is given by the summation of all possible combinations with $n=\sum_i n_i$ of the following probability,

$$\prod_i \binom{2}{n_i} p_i^{n_i}(1-p_i)^{2-n_i}\,, \tag{1}$$

where p_i is the single-electron ejection probability for the ith shell by the recoil effect or by electron shake-off.

The kinetic energy of an α particle emitted from ^{210}Po is 5.3 MeV and the corresponding recoil energy of the residual nucleus, ^{206}Pb, is estimated to be 103 keV. Using the simple theory of Migdal [1] and the mean radius of each atomic shell taken from the table of Lu et al. [6], the charge distribution of the daughter atom due to the recoil effect is calculated and shown in table 1. The degree of ionization, n, is given for the parent atom. Therefore, $n=0$ corresponds to the charge state of -2 for the daughter

Table 1
Comparison of calculated and measured charge distributions (%) and mean charges.

n	Charge state of daughter atom	Calculated probability			Measured probability [a]	
		recoil	shake-off	evaporation	17 h	23 d
0	−2	78.7	63.0			
1	−1	19.3	30.3	0.4		
2	0	1.9	6.0	10.8	60 ±6	73 ±7
3	1	0.1	0.6	43.8	36 ±3	25 ±1
4	2			37.8	4.3±1.1	2.3±0.5
5	3			7.0		
6	4			0.2		
mean charge		−1.76	−1.56	1.41	0.45	0.30

[a] Ref. [5].

atom, $n=1$ to -1, $n=2$ for the neutral atom, and so on.

On the other hand, the electron shake-off probability in α decay is given by [2]

$$P_{nlj}=1-|\langle\psi_{nlj}(Z-2)|\psi_{nlj}(Z)\rangle|^2-P_F\,. \tag{2}$$

Here $\psi_{nlj}(Z)$ is the wave function of the electron in the orbit nlj with atomic number Z, where n and l are the principal and orbital angular-momentum quantum numbers, respectively, and $j=l\pm\frac{1}{2}$ is the total angular momentum. P_F is the electron transition probability to occupied states:

$$P_F=\int_{n'}\frac{N'}{2j+1}|\langle\psi_{n'lj}(Z-2)|\psi_{nlj}(Z)\rangle|^2\,, \tag{3}$$

where the summation over n' runs over all the occupied shells and N' is the number of electrons in the $n'lj$ orbital.

The claculation of shake-off probabilities has been performed with the atomic wave functions in the Dirac–Fock–Slater method [7]. The charge distribution by the shake-off effect is obtained using these results and is listed in table 1.

It can be seen from the table that in both models most of the outer-shell electrons retain their initial quantum state after α decay, and thus the daughter atom is a negative ion. This conclusion is in disagreement with the recent experimental result [5] that more than 50% of the recoils are neutral. A possible reason for the discrepancy is attributed to the fact that the α-decay process is a so drastic event for outer-shell electrons that the models based on the single-electron transition picture such as the recoil effect and the shake-off effect are inadequate. The outer-shell ionization process due to α decay, as a whole atomic decay, is quite similar to the case of multiple ionization during ion–atom collisions, since α decay can be considered to be a part of the collision process of α + daughter atom with only an out-going part and impact parameter $b=0$. In the 1950s and 1960s, Russek and collaborators [3] developed a statistical model for multiple ionization of outer-shell electrons. In this model, the ionization mechanism is treated as a two-step process. First, a small part of the kinetic energy of translational motion of the atom is transferred to the atomic electrons during atomic collisions. Second, this excess energy is distributed among atomic electrons and there is a probability that some electrons evaporate from the heated atom. They introduced the following assumptions to simplify the problem:

(1) The energy transferred is statistically distributed among the outer-shell electrons only.

(2) The energy scale is divided into cells of equal width and the statistical weight of the cells is taken to be the same for bound and continuum states.

(3) The ionization energy is assumed to be uniform; i.e. the same for all outer-shell electrons and does not depend on the number of electrons evaporated.

This model, called the evaporation model, has been successfully used to estimate the multiple ionization probabilities for low-energy ion–atom collisions.

Russek and Meli (RM) [4] improved the evaporation model as follows. The excitation of the residual ion is allowed and a factor corresponding to the average matrix element is introduced. Furthermore, the ionization probability is defined as the unit volume of the phase space instead of the unit energy cell.

In the present work, we have applied the evaporation model of RM to calculate the charge distribution of the daughter atom in α decay of ^{210}Po. According to the RM model, the probability that n electrons among N outer-shell electrons are ejected is given by [4]

$$P_n^{(N)}(E_\mathrm{T})=\frac{\binom{N}{n}S_n(E_\mathrm{k}/\epsilon_1)}{\sum_{i=1}^{N}\binom{N}{i}S_i(E_\mathrm{k}/\epsilon_1)}, \qquad (4)$$

with

$$E_\mathrm{k}=E_\mathrm{T}-\sum_i \epsilon_i - E_\mathrm{R}, \qquad (5)$$

where E_T is the total energy transferred to the electronic system, E_k is the kinetic energy available to the electrons, ϵ_i is the ith ionization energy, and E_R is the excited energy of the residual ion. We assume that the average matrix element, g, in the original RM model is equal to unity. The integrated volume of phase space for the nth ionization state is expressed as [4]

$$S_n(E)=\frac{2^{\{(n-1)/2\}}\pi^{\{n/2\}}E^{(3n-2)/2}}{(3n-2)!!}, \qquad (6)$$

where $\{a\}$ represents the integral part of a.

In the case of α decay in ^{210}Po, the total energy transferred to the electrons can be estimated to be about 60 eV, in a manner similar to the method of Thomas for Th [8]. We consider that this energy is distributed to 6s and 6p electrons only, i.e. the number of outer-shell electrons is taken to be $N=6$. The mean ionization energy ϵ of these electrons is estimated as the average value of the binding energies tabulated by Sevier [9] for the Po atom to be 5.2 eV. We assume that the mean excitation energy of the residual ion E_R is zero. The result calculated in this fashion within the framework of the evaporation model is shown in table 1. It is clear from the table that the most probable charge state of the daughter atom is +1 and the mean charge is equal to +1.41.

In order to study the influence of the energy loss of recoils on the charge distribution, Ito and Maeda [5] made a measurement for the ^{210}Po source prepared by electrodeposition onto platinum. Thus, their measurement was made for the energy-degraded recoil ions. Two series of measurements were made: one was made 17 h after the source preparation and the other was done 23 d after. As shown in table 1, they observed a large fraction of neutral atoms with a mean energy loss of about 18 keV and, furthermore, an increasing fraction of neutral atoms as time elapsed, being associated with the increasing dispersion of the energy. The change of the fraction and the energy dispersion as time passed was due to the fact that Po atoms penetrate the platinum surface several atomic layers before disintegration as the source ages. Their result suggests that the neutralization of recoil ions strongly proceeds with increasing energy loss, indicating an important role of the electron capture for low-velocity heavy ions such as α recoil atoms. It can be said, therefore, that charge states higher than neutral, observed in their experiment, can be expected for the primary charge distribution initiated by α-decay perturbation on the recoil atoms.

As shown in table 1, the calculated mean charge by the evaporation model is about one higher than the measured ones. When compared with the calculated results of the recoil effect and the shake-off effect, the result by the evaporation model is in a more favorable direction; charge states higher than neutral are predicted from the neutralization effect as discussed above. The complex nature of charge-changing process in matter prevents us studying further. However, comparison of the calculated and the measured results suggests that about one electron is captured by the recoil ion before emerging from the source. Indeed, a rough estimation based on this hypothesis leads one to the corrected fractions of 54.6% (10.8%+43.8%) for neutral, 37.8% for +1 and 7% for +2. This result is in reasonably good agreement with the measured data. At the present stage, however, both theoretical and experimental information on charge-changing processes for low-energy heavy ions in solids are very scarce. In order to compare the present theoretical estimation with the experimental results in greater detail, it is hoped that further theoretical and experimental studies on the charge state of heavy ions in solids will be performed.

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