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Monte Carlo Simulations of Charge States of Heavy Ions After Passing Through Solids

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Electron rearrangement of fast heavy ions passing through solid targets has been studied by the Monte Carlo simulation. Starting from the electron configuration of ions in solids, the change in electronic state due to successive radiative and Auger transitions was traced from the time when the ion emerges from the solid surface to the time when it is detected. The calculations were made for various ions with a fixed energy and the results for charge distributions are shown as a function of distance between the target and the detector.

KEY WORDS: Charge state of ions/Solid target/Monte Carlo method/

1. INTRODUCTION

The charge distribution of fast heavy ions passing through matter has been of fundamental importance from early days of atomic plysics and extensive experimental and theoretical studies have been reported in a number of review articles.[1-5] When fast heavy ions travel in the medium, their charge state fluctuates by loss and capture of electrons. The charge distribution depends on the initial charge state and the velocity of the ion as well as on the properties of the medium.

The experimental measurements show that the mean charge of ions emerging from solid targets is higher than that from gas targets. This effect is called *density effect*. In passing through matter, there are three processes which change the electronic state of projectile ions: excitation and ionization of electrons in ions and electron capture from target atoms. In gases, the time between successive collisions of ions with atoms is longer than the lifetime of excited states. The excited ion loses its energy through radiative or Auger transitions and goes to a ground-state configuration. On the other hand, the collision time is short in solids and the excited states play an important role on charge states of ions.

Several phenomenological models for charge states of ions in solids have been proposed. In the Bohr-Lindhard (BL) model,[6] the electron in ions is assumed to be excited to high-energy states by collision. The probability of loss of electrons for ions in these highly-excited states is larger than than that in the ground state because of decrease in the binding energy, while the probability of electron capture for an excited ion is small. This suggests that in solids the electron loss cross section is higher and the electron capture cross section is lower than the corresponding values in gases. In their model, the main part of the increase in the mean charge of ions takes place inside of solids.

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On the other hand, Betz and Grodzins (BG)[7] assumed that the electron is not excited in a single collision to so high-energy states as in the BL model, but a number of electrons can be excited in successive collisions. When these excited ions emerge from the solid surface into vaccuum, their mean charge increases substantially through successive emission of Auger electrons. Therefore, in the BG model the main reason for the density effect is due to the Auger transitions of ions after passing through solids.

Based on these two models, a lot of argumennts have been done on the charge state of heavy ions in solids. However, in most experiments the charge distribution of ions was measured at a certain distance from the target. This means that the experimental charge distribution corresponds to the distribution after electron rearrangement due to Auger cascade between the target and the detector. Although there has been reported an experimental attempt[8] to observe Auger electrons outside the target, no theoretical calculations have been performed for electron rearrangement of ions after passing through solid targets. This rearrangement effect is critically important in the BG model, where there are many excited electrons in the ion and the final charge state is determined through Auger transitions.

In the present work, we perform the Monte Carlo simulation for electron rearrangement of ions after passing through solid targets. We assume an initial electron configuration of the ion emerging from the solid target and the calculations are made for various ions as a function of travelling time of the ion between the target and the detector. For this purpose, we fix the energy of the ion and the charge distributions of ions are shown for various target-to-detector distances.

2. METHOD OF COMPUTATION

The method to calculate the rearrangement of electronic states is in principle same as that used for vacancy cascade following inner-shell ionization.[9-11] Only difference consists in the fact that the time for electron transition is taken into account. The flow diagram for the present work is shown in Fig. 1.

The initial configuration of ions is estimated from the experimental data for the mean number of atomic shell electrons in solids. During the electron rearrangement, two processes are considered: x-ray emission and Auger transition. It is assumed that the innermost vacancy is filled first.

For a given electron configuration, the computer program calculates x-ray emission rates and Auger rates for the innermost vacancy from all possible atomic shells. This sum of both transition rates gives the total probability for filling the vacancy per unit time, P. When we consider a very short time interval, we can assume that the transition occurs randomly. In this case, the distribution of the time interval is expressed by the exponential distribution with the mean value of $1/P$. The random sampling of the time interval, DT, can be made by

$$DT = -\ln R/P, \tag{1}$$

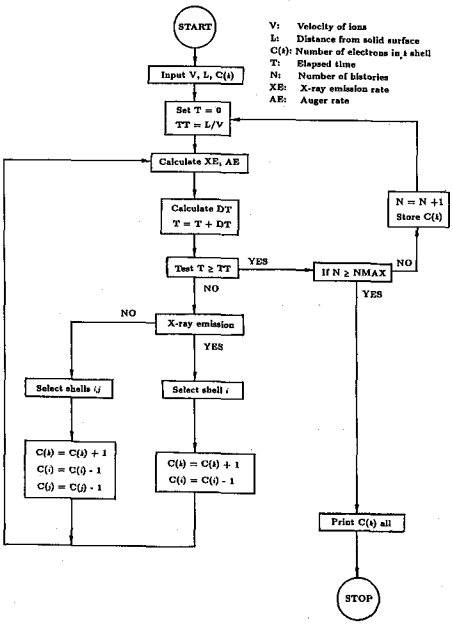

Fig. 1. Flow diagram of the Monte Carlo method. The symbol V indicates the velocity of the ion, L the distance between the target and the detector, $C(i)$ the number of electrons in i shell, DT the time interval of electron transition, T the elapsed time, N the current number of histories, NMAX the maximum number of histories, XE the total x-ray emission rate, and AE the total Auger transition rate.

where R is a uniform random number in the interval [0,1] . Using the time interval DT, the time elapsed after the ion emerges from the target, T, is increased by DT.

If the time T is shorter than the time necessary for ions to travel from the target to the detector, TT, the transition is determined whether radiative or not. For radiative transition, a random number decides a new position of the vacancy, which is produced by electron transfer accompanying x-ray emission, using relative x-ray transition rates obtained above. In the case of Auger transition, two new vacancy states are selected from partial Auger rates and the number of ionic charge is increased by one.

This procedure is repeated until no electronic transition takes place because all inner-shell vacancies are filled or until the time T is larger than TT. Then the final charge state of the ion is recorded and the computer program generates a new history. After 10000 histories, the charge distribution of ions is printed out.

3. RESULTS AND DISCUSSION

The calculations of charge state distributions have been carried out for Ar, Cu, and Br ions after passing through C targets. The energy of ions is fixed and the charge distributions are expressed as a function of the distance between the target and the detector. All the computations were made on FACOM M-760/10 computer at Institute for Chemical Research of Kyoto University.

We take the radiative transition rates between various atomic shells from the table of Manson and Kennedy.[12] The Auger rates for K shell are taken from the tabulated values calculated by Kostroun *et al.*,[13] those for L subshells from the

theoretical results of McGuire,[14] and M-subshell values are obtained from the values of McGuire.[15] The outermost shell, i. e. M shell in Ar and N shell in Cu and Br, is treated as one shell and transitions between subshells are not considered. When there are no tabulated data, the atomic data are obtained by interpolation from the values for the nearest elements.

The atomic transition rates obtained above correspond to the values for singly ionized atom. For multiply-charged ions, these values change because the number of electrons available in the transition is smaller and their binding energies are larger. We use the simple method proposed by Larkins[16] to estimate the transition rates in multiply-charged ions. His method is based on the assumption that the radiative and Auger transition rates in ions are proportional to the number of electrons available to a particular transition. The radiative transition rate in ions is obtained from that for the neutral atom by multiplying a factor n/n_0, where n is the number of electrons in a particular shell of ions and n_0 is that of the neutral atom in the ground state. The Auger rates are calculated with the scaling factor $nn'/n_0n'_0$ when each electron in two differnt shells is concerned in the transition, where n' and n'_0 correspond to n and n_0 in another shell, or $(n-1)n/\{n_0(n_0-1)\}$ when two electrons in the same shell are involved.

Mizogawa *et al.*[17] measured the K-x-ray intensity ratios for 1.25-MeV/u Ar ions in the carbon foil and estimated the mean charge of the projectile in the target. From their experimental results, we chose the electron configuration of 50-MeV Ar ions emerging from the carbon foil as $(1s)^2(2s)^2(2p)^1(3s)^2$. Figure 2 shows the charge state fractions $F(q)$ of Ar ions with charge q after passing through the carbon target for the target-to-detector distances between 0 and 2 m. It can be seen that the atomic transitions take place within very short period and final charge distribution remains unchanged for distances larger than 0.01 mm.

From the experimental charge distribution of 42-MeV Br ions from the carbon target, Betz[18] estimated the electron configuration of Br ion in the target based on the BL and BG models. In both cases, K and L shells are fully occupied. The M-and N-shell configuration is $(3s)^2(3p)^6(3d)^3(4p)^1$ in the BL model and $(3s)^2(3p)^4(3d)^2(4s)^2(4p)^3(4d)^3$

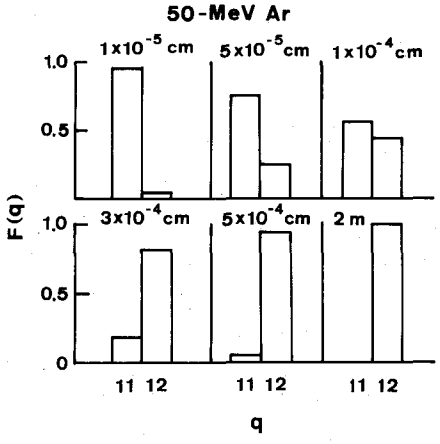

Fig. 2. Charge distribution of 50-MeV Ar ions passing through C target as a function of the target-to-detector distance.

$(4f)^1$ in the BG model. In the case of the BL model, only single radiative ($4p \rightarrow 3d$) transition is possible. For the electron configuration of the BG model, the charge state distributions of Br ions are demonstrated in Fig. 3 as a function of the distance between the target and the detector

Shima *et al.* [19] measured the projectile K-x-ray energy shifts and $K\beta/K\alpha$ x-ray intensity ratios for 63-MeV Cu ions on the carbon target and roughly estimated the number of K-, L-, and M-shell electrons in the target. The mean number of electrons is 2 for K shell, (5.6 ± 0.4) for L shell, and 3.3~7.4 for M shell. For simplicity, we use an electron configuration $(1s)^2(2s)^2(2p)^4(3s)^2(3p)^3$ plus two N-shell electrons. The calculated charge distributions of Cu ions at the detector are shown in Fig. 4 against the distance from the target. In this case, electron rearrangement occurs within distances smaller than 0.01 cm and the charge distribution does not change for larger distances.

In the present model, we have made Monte Carlo simulation on the electron

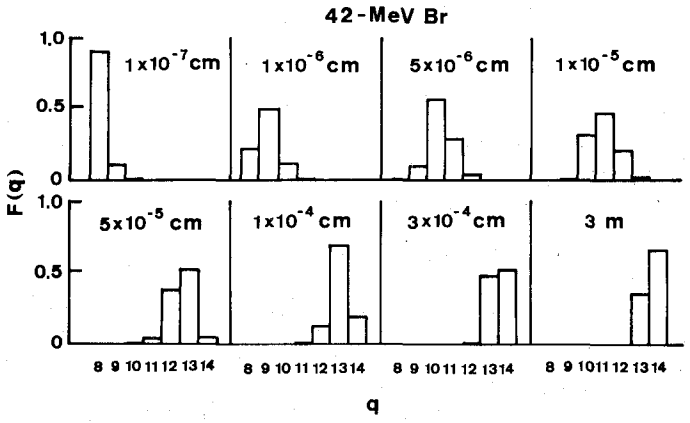

Fig. 3. Charge distribution of 42-MeV Br ions passing through C target as a function of the target-to-detector distance.

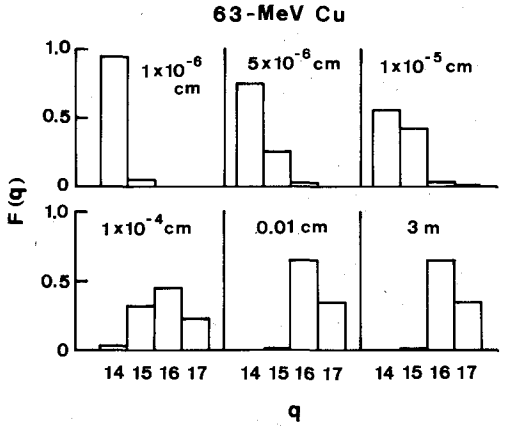

Fig. 4. Charge distribution of 63-MeV Cu ions passing through C target as a function of the target-to-detector distance.

rearrangement of heavy ions after passing through solid targets. For fixed projectile energy, the charge distributions are shown as a function of the distance from the target to the detector. It is seen that for ions with ~MeV/u the electron rearrangement takes place shortly after the ion emerges from the target. The present method has been applied to study the origin of the shell effect in the equilibrium charge state of ions[20] and found to be very useful to study charge states of ions in solids.

In the present model, we considered only x-ray and Auger-electron emission processes in vacancy cascade and neglected the effect of shakeoff process When the vacancy is created, there is a probability that another atomic electron is emitted due to sudden change in the atomic potential.[21] When we take this process into consideration, the fraction of highly-charged ions increases.[9] In addition, we neglected the change in the binding energies of electrons in highly-charged ions produced during vacancy cascade. For multiply-charged ions, some Auger channels may close due to increase in binding energies of electrons concerned. This leads to smaller fraction of highly-charged ions in charge distribution.

REFERENCES

(1) S.A. Allison, *Rev. Mod. Phys.*, **30**, 1137 (1958).
(2) H.D. Betz, *Rev. Mod. Phys.*, **44**, 465 (1972).
(3) H.D. Betz, in "*Methods of Experimental Physics*," Vol. 17, ed. by P. Richard, Academic, New York, (1980), pp. 73-148.
(4) M.A. Kumakhov and F.F. Komarov, "*Energy Loss and Ion Ranges in Solids*," Gordon and Breach, New York, (1981).
(5) H.D. Betz, in "*Applied Atomic Collision Physics*," Vol. 4, ed. by S. Datz, Academic, New York, (1983), pp. 1-42.
(6) N. Bohr and J. Lindhard, *K. Danske Vidensk. Selsk., Mat.-Fys. Meddr.*, **28**, No. 7 (1954).
(7) H. Betz and L. Grodzin, *Rhys. Rev. Lett.*, **25**, 211 (1970).
(8) R.A. Baragiola, P. Ziem, and N. Stolterfoht, *J. Phys. B*, **9**, L144 (1973).
(9) T. Mukoyama, *Bull. Inst. Chem. Res., Kyoto Univ.*, **63**, 373 (1985).
(10) T. Mukoyama, *J. Phys. Soc. Jpn*, **55**, 3054 (1986).
(11) T. Mukoyama, T. Tonuma, A. Yagishita, H. Shibata, T. Koizumi, T. Matsuo, K. Shima, and H. Tawara, *J. Phys. B*, **20**, 4453 (1987).
(12) S.T. Manson and D.J. Kennedy, *At. Data Nucl. Data Tables*, **14**, 111 (1974).
(13) V.O. Kostroun, M.H. Chen, and B. Crasemann, *Phys. Rev. A*, **3**, 533 (1971).
(14) E.J. McGuire, *Phys. Rev. A*, **3**, 1801 (1971).
(15) E.J. McGuire, *Phys. Rev. A*, **5**, 1043 (1972).
(16) F.P. Larkins, *J. Phys.B*, **4**, L29 (1971).
(17) T. Mizogawa, Y. Awaya, T. Kanbara, Y. Kanai, M. Kase, H. Kumagai, P.H. Mokler, and K. Shima, *Phys. Rev. A.* **42**, 1275 (1990).
(18) H.D. Betz, *Nucl. Instr. and Meth.*, **132**, 19 (1976).
(19) K. Shima, S. Fujioka, Y. Tajima, T. Ishihara, and M. Yamanouchi, *Nucl. Instr. and Meth.*, **A262**, 132 (1987).
(20) K. Shima, T. Mukoyama, T. Mizogawa, Y. Kanai, T. Kanbara, and Y. Awaya, *Nucl. Instr. and Meth.*, in press.
(21) T. Mukoyama and T. Taniguchi, *Phys. Rev. A*, **36**, 693 (1987).