THE BULLETIN OF OKAYAMA UNIVERSITY OF SCIENCE No. 56 A pp. 21-27 (2020)

Production and destruction of MeV-per-atom C_3 cluster ions penetrating rare gas region

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(Received November 2, 2020; accepted December 11, 2020)

The production and destruction cross sections for C_3 cluster ions penetrating rare gas (He, Ne) region in tandem accelerator facility, are calculated on the basis of impact-parameter method, where the transition of an active electron is treated quantum-mechanically, and the motion of a projectile is assumed to move along a straight-line trajectory with impact-parameter. In production process, three types of the electron-loss cross sections are incorporated, while in destruction process three destruction cross sections, depending on cluster charge state, play a role. The spatial structure of a C_3 cluster is assumed to form a linear chain and a triangle chain of equal spacing d=1.27 Å. The calculated cross sections for the 3.75 MeV C_3 clusters show a weak-dependence on the spatial structure, though strongly depending on species of gas. The maximum value of the singly charged fraction of C_3^+ clusters is obtained about 0.2 for He gas and about 0.16 for Ne gas. The gas-pressure dependences of the singly charged fraction present different features.

Keywords; electron-loss cross section, cluster ion, impact-parameter method, independent-electron model

1. Introduction

Recently, MeV/atom cluster ion beams have been applied to transmission SIMS analysis since they have an advantage of obtaining highly sensitive signals of intact large molecular ions from biological materials [1]. Under irradiation of MeV/atom cluster ions, the cluster effect or vicinage effect was reported on several topics, i.e., reduction of average charge of fragment ions [2,3], non-linear dependence of the deposited energy on the number of atoms [2,4,5], orientation dependence of the energy loss [5], strong suppression of low-energy secondary electrons [6-8], and strong enhancement of convoy electron yields [7] per projectile atom. These phenomena, which have been never seen in the single-atom ion irradiation, are closely related to space-time correlation in simultaneous electron excitation by multi-atom ions. As a typical fullerene cluster, it was reported that the electronic stopping power of carbon for a C60 cluster presents a giant resonance effect [9].

From a viewpoint of application, intense cluster-ion beams are recently requested. In order to obtain them, appropriate choice of a charge-changing gas and the optimization of the gas pressure is needed. In this sense, the collision of cluster-ion with a gas presents a basic and important information on the electron-loss cross sections in this energy region

Quite recently, we presented a theoretical research on the production and destruction of a singly charged C_n^+ cluster ions in a linear chain structure. In this work, the calculated electron-loss cross sections and the destruction cross sections presented a sub-linear

dependence on the number of constituent atoms. The resultant gas-pressure dependences of the singly charged C_2^+ and C_4^+ ions at 2.5 MeV penetrating Ne gas were in good agreement with the experimental data [10].

As for C_3 cluster, two spatial structures are proposed, i.e., linear structure and ring (triangle) structure. It is not so easy to distinguish experimentally with each other. Chiba et al. [3] reported the relation between the average charge of fragmented ions and the spatial structure of 1 MeV/atom C_3^+ ions traversing a thin carbon foil. The experimental data is quite consistent with the theoretical results [2]. From this point, it is of theoretical interest to provide information on how different the magnitude of electron-loss cross sections for a C_3 cluster in a linear structure from that in a ring structure, and the resultant singly charged fraction derived from them. In this paper, the Bohr radius and the Bohr speed are denoted by $a_0 = 0.529\times10^{-8} cm$ and $v_0 = 2.19\times10^{8} cm/s$. In addition, $m, e, \hbar$ denote the rest electron mass, the elementary charge, and the Planck constant divided by 2π, respectively.

2. Theoretical model.

2-1. Rate equation and fraction of cluster ions

In this section, we treat the fraction of a singly charged positive cluster ion produced in tandem accelerator, in which firstly a singly charged negative cluster ion originated at ion source is accelerated with high (MV) voltage to the entrance of charge-changing region, and secondly converted partly to a singly charged positive cluster ion with the same cluster size, and thirdly accelerated again with the same amount of high voltage. Then the intensity of the singly charged cluster ion beam is dominantly determined by charge-changing and destruction (cluster size becomes smaller) processes in the charge-changing region. Then, the fraction of a cluster in charge state i at penetrating depth x of charge-changing gas layer, $\phi_i(x)$, is governed by the following rate equation:

$$\frac{d\phi_i(x)}{N\,dx} = \sum_{j(\neq i)} \sigma_{ji}\,\phi_j(x) - \left(\sum_{j(\neq i)} \sigma_{ij} + \sigma_{id} \right)\phi_i(x) \quad .$$

Here, σ_{ij} denotes the cross section for changing from charge-state i to charge-state j of a cluster in collision with a rare gas atom, σ_{id} is the destruction cross section of a cluster in charge state i, and N denotes the number of charge-changing gas atoms per unit volume. Under the initial condition of $\phi_{-1}(0)=1$, $\phi_j(0)=0$ $(j=0,+1)$, we solved the above rate equation. Here, by considering the kinetic energy of a cluster, the electron-capture process is neglected so that the electron-loss cross sections and the destruction cross sections are taken into account. Then we obtained the analytical expressions of $\phi_i(x)$ $(i=-1,0,+1)$ as follows:

$$\phi_{-1}(x) = e^{-N\sigma_{-1}x}$$

$$\phi_0(x) = \sigma_{-10}\frac{e^{-N\sigma_{-1}x} - e^{-N\sigma_0 x}}{\sigma_0 - \sigma_{-1}}$$

$$\phi_1(x) = A\frac{e^{-N\sigma_{-1}x} - e^{-N\sigma_{1d}x}}{\sigma_{1d} - \sigma_{-1}} + B\frac{e^{-N\sigma_{1d}x} - e^{-N\sigma_0 x}}{\sigma_{1d} - \sigma_0}$$

Here we have

$A = \sigma_{-11} + B$, $B = \dfrac{\sigma_{-10}\sigma_{01}}{\sigma_0 - \sigma_{-1}}$, $\sigma_0 = \sigma_{01} + \sigma_{0d}$,

$\sigma_{-1} = \sigma_{-10} + \sigma_{-11} + \sigma_{-1d}$.

Thus, we found that $\phi_1(x)$ is described by six cross sections, i.e., three charge-state dependent destruction cross sections σ_{-1d}, σ_{0d}, σ_{1d} and three electron-loss cross sections $\sigma_{-11,}\sigma_{-10}, \sigma_{01}$.

2-2. One-electron loss Probability

We describe the one-electron loss probability for a cluster colliding with a charge-changing gas atom. In general we have to treat multi-electron and

multi-center system, but to do it rigorously is almost impossible. Therefore we restrict our attention to one excited electron and assume to neglect transition of other electrons. We call an excited electron as the *active electron* and others as the *frozen* electrons. A carbon cluster ion is assumed to be composed of the isolated ion or atoms, by which the initial state wave function of the active electron is expressed in terms of analytical functions. The background of this assumption is based on the fact that the interaction range between the active electron and a charge-changing gas atom is rather short so that the collision between a cluster ion and a gas atom can be considered as a binary collision of one constituent ion/atom in a cluster with the gas atom.

The transition of the active electron in a constituent atom/ion in a cluster is formulated by means of the time-dependent Schrodinger equation with the perturbation theory. Considering the collision system in the rest frame of the cluster, the target gas-atom moves along a straight-line trajectory with impact parameter vector $\vec{b}$ at velocity $\vec{v}$. The transition amplitude from the initial state $\varphi_j(\vec{r})$ with energy E_j to the ionized one $\varphi_k(\vec{r})$ with kinetic energy $E_k = (\hbar k)^2/(2m)$ is given [11] with $q^2 = q_\perp^2 + q_z^2$ and $q_z = (E_k - E_j)/(\hbar v)$ by

$$C_{k,j}(\vec{b}) = \frac{-v_0}{i\pi v}\int d^2q_\perp \frac{\exp(i\vec{q}_\perp\bullet\vec{b})}{q_\perp^2 + q_z^2}\rho_{ext}(\vec{q})\langle\varphi_k|\exp(-i\vec{q}\bullet\vec{r})|\varphi_j\rangle$$

Here the ionized state is described by a distorted plane wave with wave number $\vec{k}$ as

$$\varphi_k(\vec{r}) = \frac{e^{i\vec{k}\bullet\vec{r}}}{(2\pi)^{3/2}}e^{\pi\gamma/2}\Gamma(1+i\gamma)F(-i\gamma,1,-i(kr+\vec{k}\bullet\vec{r})),$$

with $\gamma = Z_e/(a_0k)$ and Z_e is the effective charge of the residual ion. Here, $\Gamma(1+i\gamma)$ is the gamma function and $F(-i\gamma,1,-i(kr+\vec{k}\bullet\vec{r}))$ is the hyper-geometric function. The initial state is described by the Hartree-Fock wave function, tabulated in the reference [12]. $\rho_{ext}(\vec{q})$ is the electron density of a neutral gas atom in Fourier space, expressed in Thomas-Fermi Moliere statistical model with the Thomas-Fermi screening length $a_{TF} = 0.8853Z^{-1/3}a_0$ as

$$\rho_{ext}(q) = Z\sum_{i=1}^{3}\frac{\alpha_i\, q^2}{q^2 + (\beta_i/a_{TF})^2}.$$

In the above, the values of constants are $(\alpha_1,\alpha_2,\alpha_3) = (0.10,\ 0.55,\ 0.35)$ and $(\beta_1,\beta_2,\beta_3) = (6.0,\ 1.20,\ 0.30)$. The initial state wave-function is given by a Slater-type function of the form $\varphi_j(\vec{r}) = \sum_i C_i N_i r^{n_i-1}e^{-\varsigma_i r}Y_0^0(\theta,\phi)$ with $N_i = (2\varsigma_i)^{n_i+1/2}/\sqrt{(2n_i)!}$, where $Y_0^0(\theta,\phi)$ denotes the spherical harmonics and the angular part is assumed to be spherically symmetric. The expansion coefficients C_i and the orbital parameters n_i, ς_i are given in reference [12]. The electron-loss probability $P_j(b)$ for the initial j-state is obtained [10] as

$$P_j(b) = \sum_{\vec{k}}\left|C_{\vec{k},j}(\vec{b})\right|^2$$

by integrating over the wave vector of the ionizing electron.

2.3 Independent-atom and -electron model (IAEM)

On the basis of the one-electron loss probability in two-body collision, we are going on the electron-loss cross section for a C_3 cluster colliding with a neutral rare-gas atom. Figure 1 shows the schematic view where a gas atom as a projectile moves parallel to z axis, colliding with a C_3 cluster ions in a ring structure as a target. The positions of three atoms/ions in the C_3 cluster projected to XY-plane are indicated together with the impact

parameters. The cluster is assumed to be composed of independent isolated atoms/ions (Independent-atom model, IAM) and the multiple ionization of a cluster is treated in the independent-electron model (IEM), where, the 2s and 2p electrons in an isolated C^+, C^0, C^- are taken into account. In the energy region considered (~1MeV/atom), the 1s electrons are hard to be ionized so that their excitation is now neglected. Let us take the isolated-atom picture, where ,i.e., the clusters C_3^+, C_3^- and C_3^0 are assumed to be composed of $C^+ + C^0 + C^0$, $C^- + C^0 + C^0$, and $C^0 + C^0 + C^0$, respectively, with spacing of $R_0 = 2.4a_0$ on a plane described by the azimuth angle φ and the polar angle θ. The impact parameters with respect to three carbon atom/ion are denoted by

$$b_1 = \sqrt{b^2 + (R_a \sin\theta)^2 - 2R_a \sin\theta\, b\cos(\varphi - \alpha)} \quad ,$$

$$b_2 = \sqrt{b^2 + (R_2)^2 + 2R_2\, b\cos(\varphi - \alpha)} \quad ,$$

$$b_3 = \sqrt{b^2 + (R_3)^2 - 2R_3\, b\cos(\varphi - \alpha)} \quad ,$$

with

$$R_2 = \frac{1}{2}(R_0\cos\theta + R_a\sin\theta),\ R_3 = \frac{1}{2}(R_0\cos\theta - R_a\sin\theta),$$

and $R_a = R_0/\sqrt{3}$. The angle α denotes the azimuth angle of the impact parameter.

The one electron-loss probability for the 2s or 2p electron on the j-th (j=1-3) carbon atom/ion is denoted by $P_{2s}(b_j)$ or $P_{2p}(b_j)$. The multiple electron-loss cross sections for a C_3 cluster is given by the combination like $P_{2s}(b_1)P_{2p}(b_2)[1-P_{2s}(b_3)]$, where $1-P_{2s}(b_3)$ means that the 2s electron is not lost from the atom/ion characterized by b_3. Thus, multiple electron-loss probability from a ring C_3 cluster is represented as $P(b_1,b_2,b_3) = P(b,\theta,\varphi,\alpha)$. In order to obtain the corresponding multiple electron-loss cross section σ, we should integrate the probability over the impact parameter and average it over the azimuth and polar angles.
Then we obtain

$$\sigma = \frac{1}{2}\int_0^{\pi} d\theta \sin\theta \int_0^{\infty} db\, b \int_0^{2\pi} d\alpha P(b,\theta,\varphi,\alpha).$$

The integral over the azimuth and polar angles of the cluster means the random orientation of the cluster. In this way, we calculated the six cross sections appeared in the fraction formula.

3. Numerical Results and Discussion.

Figure 2 shows the structure dependence of the six cross sections calculated for the 3.75 MeV C_3 clusters in collision with He and Ne gases. The number in the axis of abscissas indicates the type of the cross section: $1 = \sigma_{-10}, 2 = \sigma_{-11}, 3 = \sigma_{-1d}, 4 = \sigma_{0d}, 5 = \sigma_{0d}, 6 = \sigma_{1d}$. In every case, the cross sections for Ne gas is larger than that for He gas. This is due to that the magnitude of the electro-static field of a Ne atom is stronger than a He atom. As for the structure dependence, a noticeable difference is not found, though there is a slight variation. As will be seen later, this variation brings a bit change in the singly charged fraction.

Figure 3(a)-(d) show the calculated fractions, $\phi_j(x)$ $(j = -1, 0, 1)$ of the 3.75 MeV linear and ring $C_3^{\,j}$ ions in collision with He gas ((a) and (b)) and Ne gas ((c) and (d)). In every figures, the blue lines, the red lines, and the green lines indicate respectively $\phi_{-1}(x)$, $\phi_0(x)$ and $\phi_1(x)$ as a function of gas pressure Nx in units of 10^{14} atoms/cm^2. In tandem accelerator, negative cluster ions are

accelerated initially so that they are partly changed to neutral and positive cluster ions as passing deeply through the gas region. In He gas, $\phi_1(x)$ overcomes other fractions beyond 20×10^{14} atoms/cm^2, while in Ne gas it does beyond 13×10^{14} atoms/cm^2. As for $\phi_0(x)$, it reaches the maximum value of 0.26-0.27 in He gas at $Nx=10\times10^{14}$atoms/cm^2, and 0.23-0.24 in Ne gas at $Nx=6\times10^{14}$atoms/cm^2. The total particle fraction $\phi_{total}(x)=\phi_{-1}(x)+\phi_0(x)+\phi_1(x)$ is decreasing with increasing the gas pressure. This is because the decay process is incorporated.

In order to see structure dependence of $\phi_1(x)$ in detail, we present figure 4, where fig.4(a) and 4(b) display the He gas case and Ne gas case, respectively. In both figures, the red line and the blue line indicate the ring structure and linear structure, respectively. In case of He in fig. 4(a), the ring structure yields a bit larger fraction in the region of $Nx>20\times10^{14}$atoms/cm^2, and the peak shifts by $\Delta(Nx)=2\times10^{14}$atoms/cm^2 without changing the peak value. On the other hand, in the case of Ne gas, the shape of $\phi_1(x)$ is almost the same, irrespective of the structures, but the peak value in the ring structure is suppresses by 0.01 at $Nx=11\times10^{14}$atoms/cm^2.

In summary, we presented a theoretical model for the charged and neutral fractions of the 3.75 MeV C3 cluster ions in the ring structure passing through He and Ne gas region as well as in the linear structure. The electron-loss and decay cross sections estimated display a small variation as well as the fractions do. We found a contrastive feature in the singly charged fraction.

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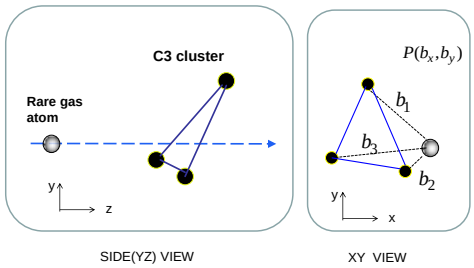

Fig.1: Schematic view of collision system (see text).

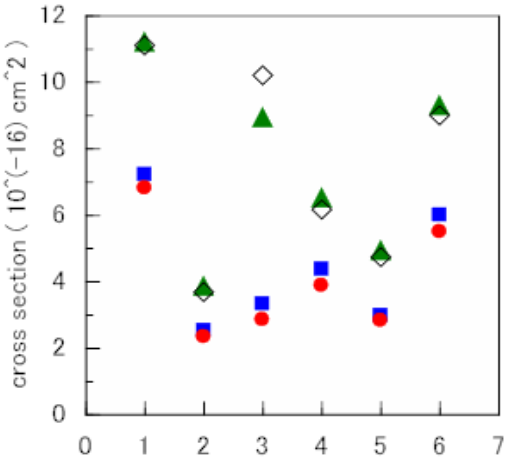

Fig.2: The structure dependence of the cross sections for 3.75 MeV C_3 cluster ions colliding with He and Ne atoms: red circles (C_3 ring with He), blue squares (C_3 linear with He), open diamonds (C_3 ring with Ne), green triangles (C_3 linear with Ne). The numbers in the axis of abscissas indicate $1=\sigma_{-10}$, $2=\sigma_{-11}$, $3=\sigma_{-1d}$, $4=\sigma_{01}$, $5=\sigma_{0d}$, $6=\sigma_{1d}$.

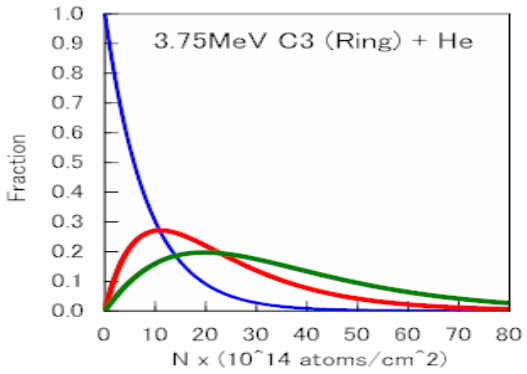

Fig.3(a): Fraction of the ring C_3 cluster in He: blue (ϕ_{-1}), red (ϕ_0), green (ϕ_1).

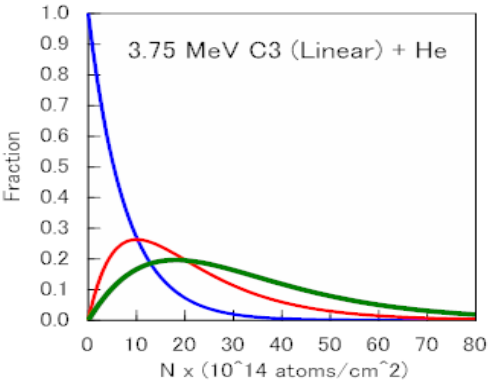

Fig.3(b): Fraction of the linear C_3 cluster in He: blue (ϕ_{-1}), red (ϕ_0), green (ϕ_1).

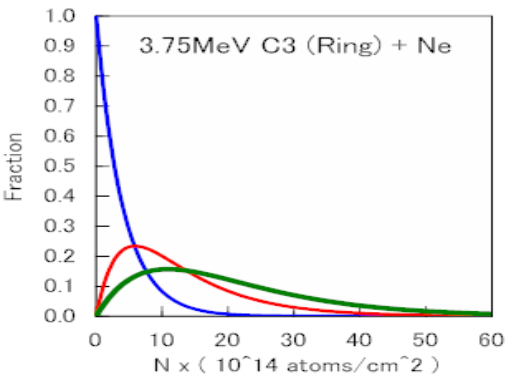

Fig.3(c): Fraction of the ring C_3 cluster in Ne: blue (ϕ_{-1}), red (ϕ_0), green (ϕ_1).

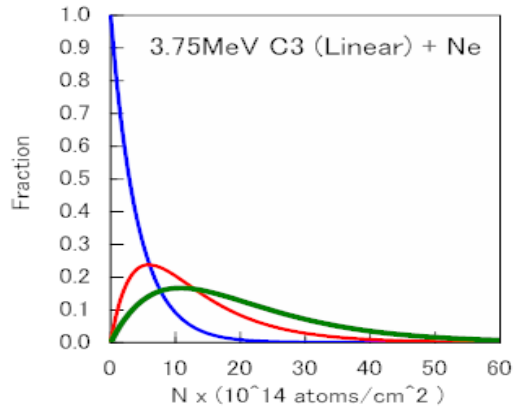

Fig.3(d): Fraction of the linear C3 cluster in Ne: blue (ϕ_{-1}), red (ϕ_0), green (ϕ_1).

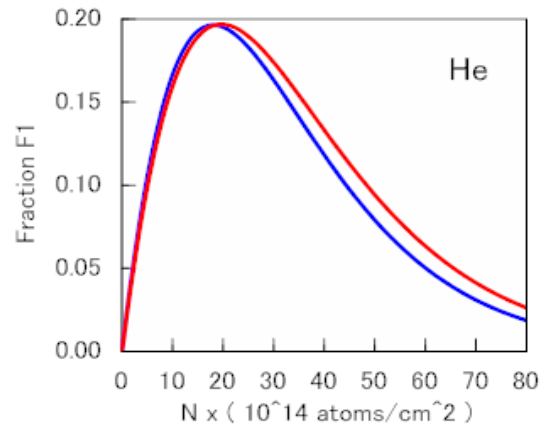

Fig.4(a): The structure dependence of ϕ_1 for the 3.75 MeV C_3 cluster ions in He gas: red (ring), blue (linear).

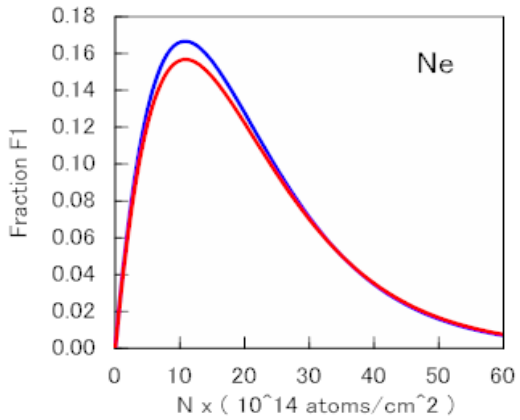

Fig.4(b): The structure dependence of ϕ_1 for the 3.75 MeV C_3 cluster ions in Ne gas: red (ring), blue (linear).