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Z oscillation of mean charges of energetic ions emerging from a carbon foil: Correlation with the shell structure of ions

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Equilibrium mean charges $\bar{q}$ of ions passing through a carbon foil were observed to oscillate versus projectile atomic number Z at ion energies 0.55, 1, and 2 MeV/u. The maxima of the oscillation take place for closed-shell ions having the electrons in mean number $Z-\bar{q}=10$ and 28.

Several empirical or semiempirical formulas have been proposed on the equilibrium mean charges $\bar{q}$ of energetic ions emerging from a carbon foil.[1-4] They are the function of a variable composed of ion velocity v and projectile atomic number Z. In these formulas, once the ion velocity is given, the values $\bar{q}$ always increase monotonically with the increase of Z. Contrary to this, recent measurements by Lennard and Phillips[5] at around 0.02 MeV/u for ions $5 \leqq Z \leqq 26$, and by Shima *et al.*[6,7] at around 1 MeV/u for ions $Z \leqq 53$, indicate that the $\bar{q}$ values are not always a monotonic function of Z. In spite of the possible interpretation for this phenomenon given in Refs. 5-7 in their specific region of v and Z, the comprehensive understanding for the variation of $\bar{q}$ over the wide range of v and Z has not been clarified yet.

In this work, the Z dependence of $\bar{q}$ has been investigated systematically by extending the ion energies up to 0.55, 1, and 2 MeV/u and ion species up to $Z=77$. As the result, the Z oscillation of $\bar{q}$ has clearly been observed at each energy. In addition, we found that this oscillation is closely correlated in common with the shell structure of ions covering from the low velocity data of 0.02 MeV/u to the present high-velocity data.

Experiment was performed at the University of Tsukuba using the 12 MV tandem accelerator. Experimental procedure for the measurement of charge distribution is given in detail elsewhere,[8] and so, it is described briefly. Ion beams emerging from a foil were charge analyzed using a split-pole-type magnetic spectrograph, and the ions with different charge states were collected with a Faraday cup. The beam integration for each charge state q was normalized by the elastically scattered beam counts to obtain the charge distribution $F(q)$. Several carbon foils with different thicknesses were prepared to confirm the charge equilibration condition and one of the foils between 30 and 120 μg/cm^2 thick was adopted depending on the combination of the ion species of $5 \leqq Z \leqq 77$ and ion energies of 0.55, 1, or 2 MeV/u. The thickness of the carbon foil was measured preliminarily using the method of α-particle energy loss.

Since the mean charges are intended to be compared for different ion species at the common emergent velocity v from the carbon foil, the incident energy was so adjusted that the actual emergent velocity v_{ob} may be almost $\sqrt{2}\times\sqrt{0.55}$, $\sqrt{2}\times\sqrt{1}$, or $\sqrt{2}\times\sqrt{2}\sqrt{\mathrm{MeV}/u}$. The deduction of $\bar{q}$ at v from the observed mean charge $\bar{q}_{\mathrm{ob}}$ at v_{ob} was done by using the empirical relation of $\bar{q}$ and v, $\bar{q}/Z=1-\exp[-v/(v'Z^{0.45})]$, given by To and Drouin[3] as

$$\bar{q}=\bar{q}_{\mathrm{ob}}\frac{1-\exp[-v/(v'Z^{0.45})]}{1-\exp[-v_{\mathrm{ob}}/(v'Z^{0.45})]}\,, \tag{1}$$

where $v'=3.6\times10^{8}$ cm/sec.

Obtained results for $\bar{q}$ divided by Z are shown in Fig. 1(a). Experimental errors are less than the size of the plotted marks. The data already reported by others[9] and by our group,[9] whose ion energy is less than 20% of the quoted energy, are also plotted after correcting the original data by using Eq. (1). Thick solid lines connecting the data points are drawn simply to guide the eye. For comparison, the $\bar{q}/Z$ values based on the empirical formula by Shima *et al.*[2] are drawn with dotted curves.

Figure 1(a) shows that the mean charges $\bar{q}$ oscillate versus Z at ion energies 0.55, 1, and 2 MeV/u. The presence of the maxima of $\bar{q}$ at around $Z=26$ for 1 MeV/u and $Z=15$ for 0.02 MeV/u had been known.[5,6] At present, by extending the Z region, another maxima of $\bar{q}$ have been observed. Figure 1(a) further indicates that the phase of the oscillation differs according to the ion velocity, which appears to vary in accord with some systematics.

In considering this problem, the ionization potential (IP) for multiply charged ions calculated by Carlson *et al.*[10] is plotted versus Z in Fig. 1(b) where IP is strongly dependent on the shell structure of ions. On this map, we plot the hypothetic IP of multiply charged ions attached with the mean number of electrons, $Z-\bar{q}$, for each combination of Z and ion energy. First, we notice that the mean IP of ions having the mean charges $\bar{q}$ oscillate as a function of Z for given ion velocity. Second, from Figs. 1(a) and 1(b), we notice that the Z position exhibiting the maxima of the oscillation of $\bar{q}$ corresponds to that of ions having the Ne- or Ni-like structure. In fact, if the relation $Z-\bar{q}=10$, and 28 is drawn with thin solid curves in Fig. 1(a), the first maxima of $\bar{q}$ for each set of data of 0.02 to 2 MeV/u fall on the curve $\bar{q}/Z=1-10/Z$ and the second maxima of $\bar{q}$ for 0.55 and 1 MeV/u fall on the curve $\bar{q}/Z=1-28/Z$. In Fig. 1(a), the curve corresponding to the He-like ions, $\bar{q}/Z=1-2/Z$, is also drawn, but the oscillatory behavior is rather weak in comparison with that of Ne- or Ni-like ions.

Since the mean charges $\bar{q}$ vs Z are correlated with the shell structure of ions, the similar correlation should be present for charge distribution $F(q)$ vs Z. In Fig. 2, the

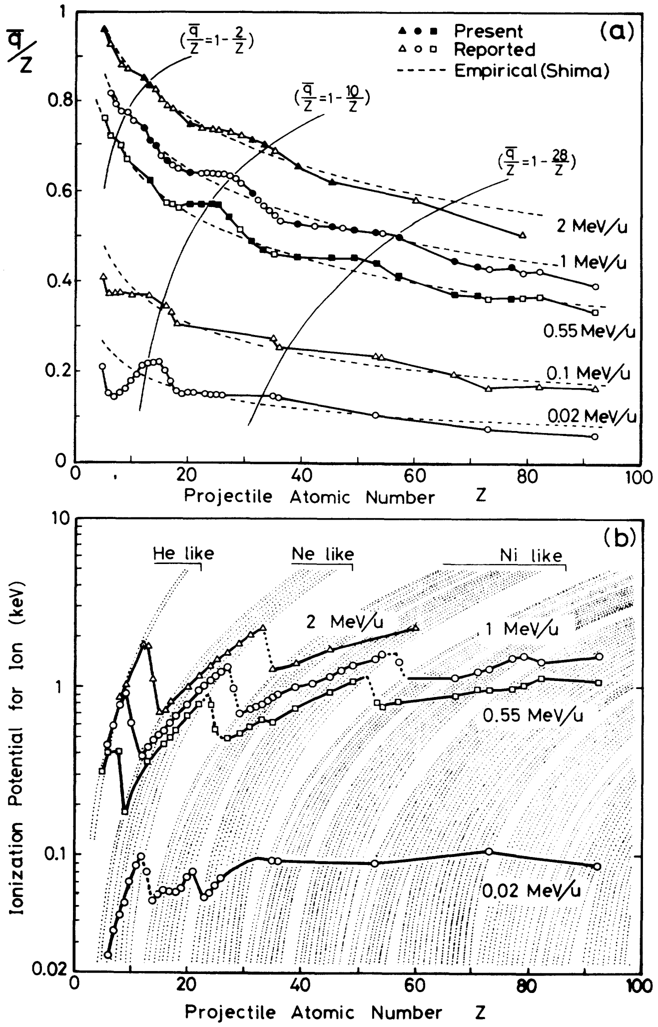

FIG. 1. (a) Equilibrium mean charges $\bar{q}$ divided by the atomic number of ions, Z, are plotted against Z for ions after the passage through a carbon foil. Present data are shown with solid marks. Thick solid lines connecting the data points are drawn only to guide the eye. Dotted lines indicate the $\bar{q}$ values evaluated from the empirical formula by Shima *et al.* Explanation for thin solid curves denoted with $\bar{q}/Z = 1 - 2/Z$, $1 - 10/Z$, or $1 - 28/Z$ is given in the text. (b) Calculated ionization potential for multiply charged ions by Carlson *et al.* plotted as a function of atomic number of ions Z. Data points indicate the hypothetic ionization potentials for multiply charged ions attached with mean number of electrons, $Z - \bar{q}$, where $\bar{q}$ means the equilibrium mean charges of ions observed after the passage through a carbon foil.

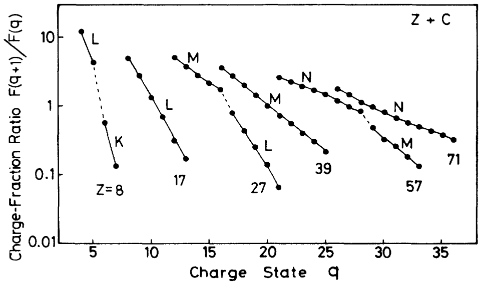

FIG. 2. Charge-fraction ratio $F(q+1)/F(q)$ of ions, O($Z = 8$), Cl($Z = 17$), Co($Z = 27$), Y($Z = 39$), La($Z = 57$), and Lu($Z = 71$) after the passage through a carbon foil at around 1 MeV/u. Dotted lines indicate the boundary of charge states where the change of slopes takes place. K, L, M, or N, means the shell of ions in which the outermost electron is included.

ratios of charge fractions between adjacent charges, $F(q+1)/F(q)$, are shown as a function of charge state q for data at around 1 MeV/u. Data displayed in the figure are those of typical ion species whose $\bar{q}$ values fall on the vicinity of the maxima ($Z = 8$, 27, 57) or minima ($Z = 17$, 39, 71) of Z oscillation of $\bar{q}$ in Fig. 1(a). We notice that the curve approximated by a single straight line (minima of $\bar{q}$) and the curve composed of two straight lines having different slopes (maxima of $\bar{q}$) appear alternately with increasing Z. The former corresponds to ions whose outermost electrons are distributed in a single shell (the shells in which the outermost electrons are included are denoted in Fig. 2). The latter corresponds to ions whose outermost electrons are distributed among two shells because the boundary charge state where the slopes of two straight lines change corresponds to $q = Z - 2$, $Z - 10$, or $Z - 28$. The presence of such a change of slopes in $\mathrm{Zn}[F(q+1)/F(q)]$ vs q at the boundary charge state of adjacent shells is known as the shell effect of charge distribution first reported by Moak *et al.*[11] At present, it should be emphasized that the occurrence of the shell effect becomes the criterion to cause the Z oscillation of $\bar{q}$.

From Figs. 1 and 2, it is clear that the shell structure of ions is closely correlated with the Z oscillation of $\bar{q}$ of ions emerging from a carbon foil. Based on this correlation, we discuss the origin of the Z oscillation of $\bar{q}$ in the following by dividing the charge distribution formation process into two: (a) the charge-exchange process during the collision inside the foil and (b) the rearrangement process at the emergence from the foil.

As for (a), charge-exchange process of ions repeating the residual excitation collision in solid targets is complicated. In order to simplify the consideration on the Z dependence of mean charge of ions inside the foil, we focus upon the representative charge state $q = \bar{q}$, where $\bar{q}$ value is not always an integer. Furthermore, only one-electron transfer for the ground-state ions is dealt with. Then, the determination of mean charge may be attributed to the balance of the electron-loss cross sections $\sigma_l(\bar{q},\bar{q}+1)$, from charge state $\bar{q}$ to $\bar{q}+1$, and the electron-capture cross sections $\sigma_c(\bar{q}+1,\bar{q})$, from charge state $\bar{q}+1$ to $\bar{q}$. As for the trial function for the charge state $q = \bar{q}$, we must start from the expression having no oscillatory trend versus Z. At present, we adopt the empirical relation of $\bar{q}$ vs Z by Shima, Ishihara, and Mikumo[2] which is shown in Fig. 1(a).

Since the mean IP of ions with charge state $\bar{q}$ oscillates against Z [see Fig. 1(b)], it is expected that both electron-loss and -capture cross sections would oscillate against Z. In Fig. 3, calculated cross sections $\sigma_l(\bar{q},\bar{q}+1)$ derived from binary encounter approximation[12] (BEA) and $\sigma_c(\bar{q}+1,\bar{q})$ by Oppenheimer, Brinkman, and Kra-

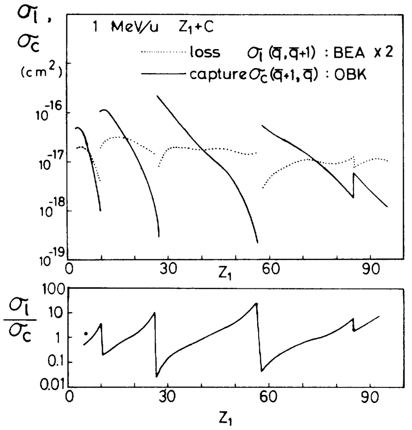

FIG. 3. Calculated electron-loss cross sections $\sigma_l(\bar{q},\bar{q}+1)=2\sigma_{\mathrm{BEA}}$ and electron-capture cross sections $\sigma_c(\bar{q}+1,\bar{q})=\sigma_{\mathrm{OBK}}$ for 1 MeV/u ions with charge state $q=\bar{q}$ colliding with carbon atoms. The calculation was done for the ground-state ions by taking into account of the number of electrons or vacancies occupied in the outermost shell of ions (upper). Electron-loss-to-capture cross-section ratios $\sigma_l(\bar{q},\bar{q}+1)/\sigma_c(\bar{q}+1,\bar{q})$. Notice that the phase of the Z oscillation of cross-section ratios agrees with that of observed $\bar{q}$ values shown in Fig. 1(a) (lower).

mers approximation[13] are shown against Z for ions at 1 MeV/u. Plotted values for σ_l are the BEA cross sections multiplied by a factor of 2 considering the electron loss of ions in solids being composed of direct ionization plus excitation followed by ionization in the successive collision. Since the number of electrons or vacancies contained in the outermost shell of ions is taken into account for the calculation, the resultant relation for σ_l or σ_c vs Z shows the discontinuity of cross sections at the boundary of the shell in which the outermost-shell electron is included. Electron-loss-to-capture cross-section ratios $\sigma_l(\bar{q},\bar{q}+1)/\sigma_c(\bar{q}+1,\bar{q})$ are also shown in Fig. 3. We notice that the phase of the Z oscillation of $\bar{q}$ at 1 MeV/u shown in Fig. 1(a) corresponds to that of the cross-section ratios for the representative charge state of ions with $q=\bar{q}$. This fact suggests that the Z oscillation of electron loss as well as the capture cross sections plays an essential role for the occurrence of the Z oscillation of $\bar{q}$.

As for the process (b), there is the possibility that the mean charges of ions inside the foil increase by $\Delta\bar{q}$ due to the Auger electron emission during the rearrangement process outside the foil.[14] In their explanation of Z-dependent $\bar{q}$ at around 0.02 MeV/u, Lennard and Phillips[5] pointed out that the Z position exhibiting the enhancement of $\bar{q}$ corresponds to that of the Auger electron yield measured by Schneider *et al.*[15] If we introduce the concept of the correlation between the ionic shell structure and the Z oscillation of $\bar{q}$, it is easily estimated that such correspondence is not specific to the ions of 0.02 MeV/u.

Consider the ions observed as the Ne-like ions (maximum of $\bar{q}$) in $\bar{q}$. Inside the foil, such ions are expected to have, on an average, a few L vacancies, some M electrons and only a few N electrons,[16] and the charge increase during the rearrangement is caused by LMM and LMN Auger process. Consider, on the other hand, the off Ne-like ions (minimum of $\bar{q}$) whose outermost electrons are distributed simply in L shell outside the foil. In this case, they would have several L electrons and only a few M electrons, on an average, inside the foil. Because of the rapid decrease of nonradiative transition probability with decreasing M electrons, the charge increase of the off Ne-like ions caused by the LMM Auger process would be lower than that by LMM or LMN Auger process of Ne-like ions. The similar argument is possible to the comparison of $\Delta\bar{q}$ between the Ni-like ions ($\bar{q}=Z-28$) and the off Ni-like ions. Consequently, because of the difference of the post-foil Auger electron emission yield, the charge increase of ions whose outermost electrons are distributed among two shells would be higher than that of ions whose outermost electrons are in a single shell.

In summary, the Z oscillation of $\bar{q}$ observed at present is strongly correlated with the shell structure of ions. This phenomenon is attributed to the charge-exchange process inside the foil and the rearrangement process outside the foil. Quantitative investigation on the relative importance of these processes is the future problem.

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[1] V. S. Nikolaev and I. S. Dmitriev, Phys. Lett. **28A**, 277 (1968).

[2] K. Shima, T. Ishihara, and T. Mikumo, Nucl. Instrum. Methods **200**, 605 (1982).

[3] K. X. To and R. Drouin, Phys. Scr. **14**, 277 (1976).

[4] O. Sayer, Rev. Phys. Appl. **12**, 1543 (1977).

[5] W. N. Lennard and D. Phillips, Phys. Rev. Lett. **45**, 176 (1980).

[6] K. Shima, M. Kakuta, S. Fujioka, and T. Ishihara, Nucl. Instrum. Methods B **14**, 275 (1986).

[7] K. Shima, E. Nakagawa, T. Kakita, M. Yamanouchi, Y. Awaya, T. Kambara, T. Mizogawa, and Y. Kanai, Nucl. Instrum. Methods B **33**, 212 (1988).

[8] K. Shima, T. Ishihara, T. Miyoshi, and T. Mikumo, Phys. Rev. A **28**, 2162 (1983).

[9] Data were adopted from two sets of data compilation, and the recent publication. A. B. Wittkower and H. D. Betz, At. Data **5**, 113 (1973); K. Shima, T. Mikumo, and H. Tawara, At. Data Nucl. Data Tables **34**, 357 (1986); J. A. Martin, Oak Ridge National Laboratory, Physics Division Progress Report No. ORNL-6420, 1988 (unpublished), p. 10.

[10] T. A. Carlson, C. W. Nestor, Jr., N. Wasserman, and J. D. McDowell, At. Data **2**, 63 (1970).

[11] C. D. Moak, H. O. Lutz, L. B. Bridwell, L. C. Nothcliffe, and S. Datz, Phys. Rev. Lett. **18**, 41 (1967).

[12] J. D. Garcia, R. J. Fortner, and T. M. Kavanagh, Rev. Mod. Phys. **45**, 111 (1973).

[13] H. D. Betz, in *Methods of Experimental Physics,* edited by P. Richard (Academic, New York/London, 1980), Vol. 17, p. 73.

[14] H. D. Betz, Rev. Mod. Phys. **44**, 465 (1972).

[15] D. Schnieder, N. Stolterfoht, D. Ridder, H. C. Werner, R. J. Fortner, and D. L. Matthews, IEEE Trans. Nucl. Sci. **NS-26**, 1136 (1979).

[16] Electron populations of excited ions inside a foil is estimated by many authors. See for instance, K. Shima, S. Fujioka, Y. Tajima, T. Ishihara, and M. Yamanouchi, Nucl. Instrum. Methods A **262**, 132 (1987).