Nonadditivity of convoy- and secondary-electron yields in the forward-electron emission from thin carbon foils under irradiation of fast carbon-cluster ions

doi: 10.1103/PhysRevA.73.060901

PHYSICAL REVIEW A **73**, 060901(R) (2006)

Nonadditivity of convoy- and secondary-electron yields in the forward-electron emission from thin carbon foils under irradiation of fast carbon-cluster ions

S. Tomita,[1,*] S. Yoda,[1] R. Uchiyama,[1] S. Ishii,[2] K. Sasa,[2] T. Kaneko,[3] and H. Kudo[1]
[1]*Institute of Applied Physics, University of Tsukuba, Tsukuba, Ibaraki 305-0006, Japan*
[2]*Tandem Accelerator Complex, University of Tsukuba, Tsukuba, Ibaraki 305-8577, Japan*
[3]*Department of Applied Physics, Okayama University of Science, 1-1 Ridai-cho, Okayama 700-0005, Japan*
(Received 13 April 2006; published 26 June 2006)

We have measured energy spectra of secondary electrons produced by fast-carbon-cluster C_n^+ $(n=1-4)$ bombardment of thin carbon foils (3.2, 7.3, 11.9, and 20.3 $\mu g/cm^2$). For clusters of identical velocity, the convoy-electron yield is enhanced with increasing cluster size n, while the yield of secondary electrons is reduced. The yield of convoy electrons normalized to the number of injected atoms increases proportionally with cluster size n. This proportionality suggests that there is only a weak vicinage effect on the number of primary electrons scattered by the projectile. The vicinage effect observed in low-energy secondary electrons must therefore arise from either transport or transmission through the surface.

DOI: 10.1103/PhysRevA.73.060901 PACS number(s): 34.50.Dy, 36.40.−c, 78.70.−g

When closely spaced atoms, e.g., clusters, are incident on a surface, the interference in the collision-induced processes by the neighbor atoms can affect the emission of secondary particles. This is called the "vicinage effect," which has been observed for fast molecular- or cluster-ion beams. The vicinage effects were first found for small molecules not only in sputtering yield [1–4] but also in the stopping power [5] and secondary-electron emission from thin foils [6]. In the 1990s, the studies were extended to larger molecules, such as hydrogen and carbon clusters. The vicinage effect is generally stronger for larger molecules. The strong deviation of secondary-electron yields is observed for cluster injection [7–9], while enhancement is observed in sputtering yields [10,11]. Vicinage effects are also observed in stopping powers [12–15], average charge states [16], pulse height defects in solid-state detectors [12,17], etc. Recent studies for fast large molecules have been reviewed by Jacquet and Le Beyec [18].

In this paper, we investigate secondary-electron emission under fast-cluster irradiation. In general, the secondary electrons are produced in three steps: (i) production of scattered electrons by the projectile ion, (ii) transport of the scattered electrons through the foils, and (iii) transmission through the surface. Therefore, if the vicinage effect stems from the electronic stopping [step (i)], a simple relation between secondary-electron yield γ and electronic stopping power $\frac{dE}{dx}$,

$$\gamma \propto \frac{dE}{dx}, \tag{1}$$

is expected. Billebaud *et al.* [8] bombarded carbon foils with hydrogen clusters, and found that the vicinage effect observed in secondary-electron yield resulted neither from energy-loss process [15] nor from the charge state of the projectile ion [16]. For carbon clusters also, we have observed a reduction of secondary-electron yields at a backward angle [9] which is much stronger than that observed in the energy loss of carbon-cluster ions [12,13]. Therefore, the simple relation Eq. (1) does not hold for cluster bombardment. The observed deviation should contain further detailed information about cluster-solid interactions.

Here arises a question of whether the deviation stems from a difference in the primary ionization or elsewhere. The deviation from Eq. (1) is also observed when the number of primary scattered electrons is not proportional to the electronic stopping power [19]. The weak vicinage effect observed in electronic stopping [12–15] implies that the effect on the amount of primary ionization should be small, but no direct evidence has been reported so far. In this Rapid Communication, we report nonadditivity of convoy- and secondary-electron yields, which provides information concerning the number of primary scattered electrons under cluster irradiation.

Beams of carbon-cluster ions were delivered by the 1 MV tandem accelerator at the University of Tsukuba. Negatively charged cluster ions were produced with a Cs sputter ion source and extracted with a potential difference of 20 kV. The extracted ions were mass selected by a 30° magnet and injected into the accelerator. After acceleration, the cluster ions were mass selected by deflecting them through 11° with a magnet, and, finally, injected into a scattering chamber in which thin carbon foils and an electron spectrometer were located.

The experimental setup was almost the same as one already described in the literature [9]. The cluster ions from the accelerator impinged on an amorphous carbon foil where a voltage of $V=-50$ V was applied. About 1 cm downstream from the target, a grounded plate was placed, so that the electrons emitted in the forward direction were accelerated and detected in an electron spectrometer. The acceleration is effective enough to distinguish the secondary electrons from stray electrons and to increase transmission of the spectrometer for low-energy electrons. The effects of residual magnetic fields are also avoided by the acceleration. On the other hand, it must be noted that the spectrum shape, especially below 20 eV, is influenced by the focusing of the electrons before entering the spectrometer. However, a comparison of

*Electronic address: tomita@bk.tsukuba.ac.jp

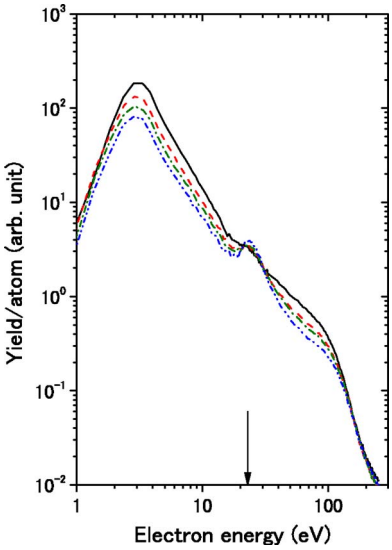

FIG. 1. (Color online) Energy spectra of secondary electrons produced by irradiation of C^+ (solid line), C_2^+ (dashed line), C_3^+ (dash-dotted line), and C_4^+ (dash-dot-dotted line) on a 7.3 $\mu g/cm^2$ carbon foil. Arrow indicates the energy of electrons that have the same velocity as the projectile ions.

electron yield at the same electron energy can be performed even with the focusing effects.

The spectrometer is a 45° parallel-plate electrostatic spectrometer of the double-deflection type. The electrons were counted by a channeltron detector whose anode potential was set to 200 V to increase the detection efficiency of low-energy electrons. Energy calibration was performed by measuring *KLL* Auger electrons from the carbon foil produced by irradiation with 1 MeV protons. The energy resolution of the spectrometer is 3%, and therefore the energy resolution ΔE_e for the electron energy E_e is given by $\Delta E_e = 0.03(E_e - eV)$.

Energy spectra were measured by varying the voltage on the spectrometer and counting the number of electrons as a function of the voltage. The beam current of cluster ions was monitored with a Faraday cup located downstream of the carbon foil during the measurements. Careful measurements of the ratio of the current with and without a carbon foil were made to normalize the electron yield to the number of incident ions. The energy of the carbon clusters C_n^+ $(n=1-4)$ was 0.5 MeV/atom, and the beam current was a few picoamperes. The foils used were purchased from Arizona Carbon Foil. The thicknesses, determined by a combination of Rutherford backscattering yield and energy-loss measurements, were 3.2, 7.3, 11.9, and 20.3 $\mu g/cm^2$. The vacuum pressure was $\sim 5 \times 10^{-6}$ Pa during the beam irradiation.

Typical energy spectra of the emitted electrons under irradiation of 0.5 MeV/atom C_n^+ are shown in Fig. 1. The spectra were normalized to the number of injected atoms instead of to the number of cluster ions. Also, no correction was made for the detection efficiency of the channeltron or for the focusing effect discussed above, while the energy dependence of the acceptance of the spectrometer has been corrected. In order to compare the relative yield at the same energy, the ratio of the yield of 0.5 MeV/atom C_n^+ to that of C^+ was obtained and shown in Fig. 2. The negative vicinage

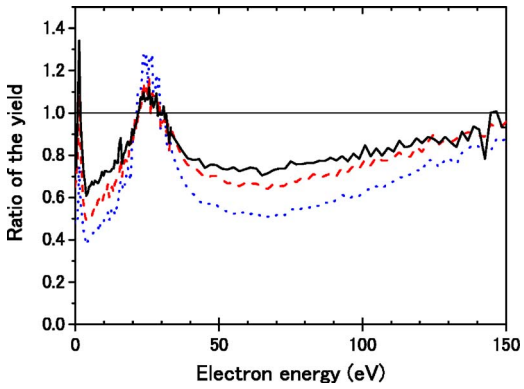

FIG. 2. (Color online) Ratios of secondary-electron yield produced by irradiation of C_2^+ (solid line), C_3^+ (dashed line), and C_4^+ (dotted line) to that of C^+. The electrons are emitted from 7.3 $\mu g/cm^2$ carbon foil bombarded by 0.50 MeV/atom C_n^+.

effect, i.e., the reduced electron yield for cluster injection, is clearly recognized at energies $E_e < 150$ eV while the yield for cluster injection is larger around the peak located at $E_e \sim 23$ eV. The difference can be seen in the yield of low-energy electrons, which is similar to the case of the screening effect observed for atomic ions [20]. However, a comparison of the electron-energy spectra for C_3^+ and C_3^{2+} at the same velocity shows no discernible difference between the two cases, which indicates that the vicinage effect observed here is not due to the preequibrium effect [21], which depends on the initial charge state of injected ions.

To investigate the vicinage effect, the yields of low-energy electrons at $E_e \sim 3$ eV as a function of foil thickness are shown in Fig. 3 for 0.5 MeV/atom C_n^+ ions. The electron yield is almost the same for all cases except for the 3.2 $\mu g/cm^2$ thickness. This means that the observed electron yields mainly originate from electron production within a depth of ~ 5 $\mu g/cm^2$ from the surface. This depth is comparable to that observed for the backward electron emission from carbon films by fast gold clusters [22]. The electron yield is reduced with increasing cluster size. The yield for

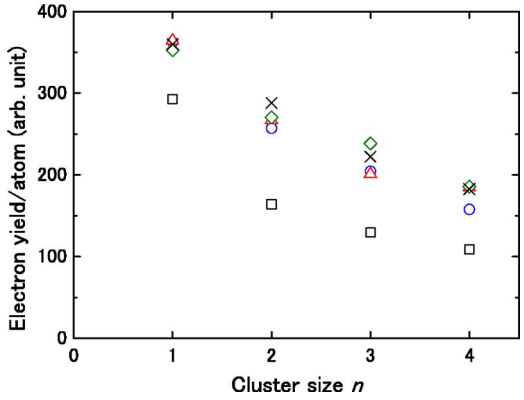

FIG. 3. (Color online) Size dependence of the yield of low-energy electrons produced by 0.5 MeV/atom C_n^+ ions for 3.2 (open squares), 7.3 (open circles), 11.9 (open triangles), and 20.3 $\mu g/cm^2$ carbon foils (open diamonds). Crosses show the relative intensity of the backward electrons emitted from highly oriented pyrolytic graphite [9], which are normalized to the yield of C_1^+.

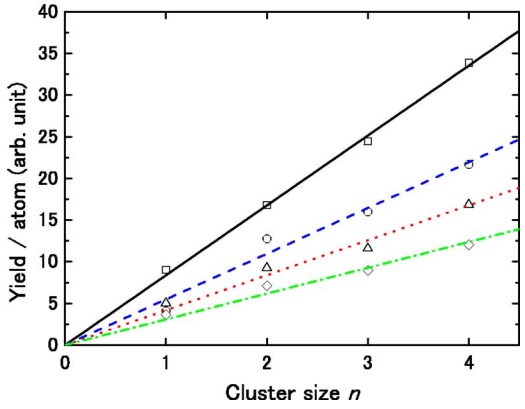

FIG. 4. (Color online) Yield of convoy electrons produced by 0.5 MeV/atom C_n^+-ion bombardment of 3.2 (open squares), 7.3 (open circles), 11.9 (open triangles), and 20.3 $\mu g/cm^2$ carbon foils (open diamonds). Lines are from least-squares fits.

C_4^+ is reduced to 0.5 relative to that for C^+ and the reduction is observed even for the thick 20.3 $\mu g/cm^2$ foil. According to the model proposed by Kaneko [23], the interatomic distance after penetration of 20.3 $\mu g/cm^2$ is so large (~4 Å for C_4^+) that no vicinage effect on the electronic stopping power is expected. The yield of low-energy electrons measured in the backward direction [9] is shown as crosses in Fig. 3, indicating that the vicinage effect is essentially the same for the backward electrons, which is consistent with the results of hydrogen-cluster irradiation [8].

The peak at $E_e \sim 23$ eV corresponds to electrons that have the same velocity as the projectile ions (0.5 MeV/atom). The electrons are called convoy electrons. The position of the peak does not depend on the cluster size, but the area of the peak is enhanced for C_4^+ relative to C_1^+, in contrast to the negative vicinage effect observed in the continuum yield (Fig. 1). There are no peaks observed in the backward direction [9], and the yield does not depend on the initial charge state of projectile ions. Convoy electrons have been studied intensively for swift atomic ions [24]. They arise either from capture of target electrons into the continuum state of the projectile or from loss of projectile electrons into the continuum [24]. The measured electron yield also stems from field-ionized Rydberg electrons (ionization occurs either during acceleration or in the spectrometer) [25]. Thus the measured yield is generated from a highly excited state or continuum state of the projectile ions.

In Fig. 4, the size dependence of the convoy-electron yield is shown for the four different carbon-foil thicknesses. It is interesting that the yield per injected atom shows a clear proportionality with n. The yield for C^+ decreases for thick foils but the proportionality still remains even for the thickest foil of 20.3 $\mu g/cm^2$. This observed proportionality can be understood if the convoy electrons stem from electron capture of primary scattered electrons to the continuum state of the projectile ion. Considering the weak vicinage effect observed for energy loss by cluster ions [12,13], the number of scattered electrons, and accordingly, the number of captured electrons, should increase proportionally with the cluster size n. This provides evidence for the final-state interaction between projectiles and free secondary electrons traveling with velocities around the projectile velocities near the exit surface of the target foils as shown in Ref. [26].

The results obtained for the yield of the convoy electrons suggest that the number of scattered electrons is proportional to the number of projectile atoms. This leads us to conclude that the strong vicinage effect in the continuum spectra stems from either electron transport inside the foil or transmission through the surface. The energy of the convoy electrons (~23 eV) is not essentially different from the energy of the electrons where strong reduction of the yield is observed. Therefore, the effect takes place only for free electrons inside the material, not for the electrons moving together with projectile ions.

One of the proposed mechanisms for the vicinage effect is a sweeping-out-electron effect in which the number of target electrons is reduced locally due to sweeping by preceding projectile atoms [27]. In this model, not only the yield of secondary electrons but also the number of primary scattered electrons is reduced, which does not agree with the present results. Therefore, some other effects should be taken into account. There is a possibility that the local reduction of target electrons creates a positively charged region behind the ion track, which induces an electric potential Φ. Then the electrons that have lower energy than the induced potential Φ are trapped and the yield of low-energy electrons is reduced. A similar concept has been proposed for atomic ions and called the track potential, to explain the deviation from Eq. (1) [28,29]. However, we do not observe that the position of the convoy peak changes with the size of the projectile cluster, which would be expected for some additional potential Φ. The application of the model for conducting targets is also questionable because of the fast relaxation time of the electronic system (10^{-16}–10^{-17} s) and it is shown that the concept is valid only for insulators [19,30].

In summary, we have measured the energy spectra of the secondary electrons produced by fast-carbon-cluster irradiation of thin carbon foils. The yield of convoy electrons is higher than that for atomic ions with the same velocity and increases proportionally with the size of the injected cluster. This implies that the yield of the primary scattered electrons is roughly proportional to the cluster size. The interpretation leads to the idea that the vicinage effect on secondary-electron yield must arise from either electron transport inside the foil or transmission through the surface.

The authors wish to thank our colleagues in the accelerator laboratory for their technical assistance. S.T. is indebted to Professor J. S. Forster and Professor P. Hvelplund for critically reading the manuscript.

[1] H. H. Andersen and H. L. Bay, J. Appl. Phys. **45**, 953 (1973).
[2] D. Thompson and S. Johar, Appl. Phys. Lett. **34**, 342 (1979).
[3] A. R. Oliva-Florio, E. Alonso, R. Baragiola, J. Ferron, and M. Jakas, Radiat. Eff. Lett. Sect. **50**, 3 (1979).
[4] H. H. Andersen, in *Fundamental Processes in Sputtering of Atoms and Molecules*, edited by P. Sigmund, special issue of K. Dan. Vidensk. Selsk. Mat. Fys. Medd. **43**, 127 (1993).
[5] W. Brandt, A. Ratkowski, and R. H. Ritchie, Phys. Rev. Lett. **33**, 1325 (1974).
[6] D. Hasselkamp and A. Scharmann, Phys. Status Solidi A **79**, K197 (1983).
[7] N. V. de Castro Faria, B. Farizon Mazuy, M. Farizon, M. J. Gaillard, G. Jalbert, S. Ouaskit, A. Clouvas, and A. Katsanos, Phys. Rev. A **46**, R3594 (1992).
[8] A. Billebaud, D. Dauvergne, M. Fallavier, R. Kirsch, J. C. Poizat, J. Remillieux, H. Rothard, and J. P. Thomas, Nucl. Instrum. Methods Phys. Res. B **112**, 79 (1996).
[9] H. Kudo *et al.*, Jpn. J. Appl. Phys., Part 2 **45**, L565 (2006).
[10] H. H. Andersen, A. Brunelle, S. Della-Negra, J. Depauw, D. Jacquet, Y. Le Beyec, J. Chaumont, and H. Bernas, Phys. Rev. Lett. **80**, 5433 (1998).
[11] M. Fallavier, R. Kirsch, S. N. Morozov, J. C. Poizat, J. P. Thomas, and N. Wehbe, Phys. Rev. B **68**, 140102(R) (2003).
[12] K. Baudin *et al.*, Nucl. Instrum. Methods Phys. Res. B **94**, 341 (1994).
[13] C. Tomaschko, D. Brandl, R. Kügler, M. Schurr, and H. Voit, Nucl. Instrum. Methods Phys. Res. B **103**, 407 (1995).
[14] K. Narumi, K. Nakajima, K. Kimura, M. Mannami, Y. Saitoh, S. Yamamoto, Y. Aoki, and H. Naramoto, Nucl. Instrum. Methods Phys. Res. B **135**, 77 (1998).
[15] E. Ray, R. Kirsch, H. Mikkelsen, J. C. Poizat, and J. Remillieux, Nucl. Instrum. Methods Phys. Res. B **69**, 133 (1992).
[16] B. Mazuy, A. Belkacem, M. Chevallier, M. J. Gaillard, J. C. Poizat, and J. Remillieux, Nucl. Instrum. Methods Phys. Res. B **28**, 497 (1987).
[17] M. Seidl, H. Voit, S. Bouneau, A. Brunelle, S. Della-Negra, J. Depauw, D. Jacquet, Y. Le Beyec, and M. Pautrat, Nucl. Instrum. Methods Phys. Res. B **183**, 502 (2001).
[18] D. Jacquet and Y. Le Beyec, Nucl. Instrum. Methods Phys. Res. B **193**, 227 (2002).
[19] H. Rothard, M. Jung, J.-P. Grandin, B. Gervais, M. Caron, A. Billebaud, A. Clouvas, R. Wünsch, C. Thierfelder, and K.-O. Groeneveld, Nucl. Instrum. Methods Phys. Res. B **125**, 35 (1997).
[20] A. Koyama, H. Ishikawa, Y. Sasa, O. Benka, and M. Uda, Nucl. Instrum. Methods Phys. Res. B **33**, 338 (1988).
[21] H. Rothard, J. Schou, and K. O. Groeneveld, Phys. Rev. A **45**, 1701 (1992).
[22] M. Fallavier, R. Kirsch, J. C. Poizat, J. Remillieux, and J. P. Thomas, Nucl. Instrum. Methods Phys. Res. B **164–165**, 920 (2000).
[23] T. Kaneko, Phys. Rev. A **66**, 052901 (2002).
[24] M. Breinig *et al.*, Phys. Rev. A **25**, 3015 (1982).
[25] Z. Vager, B. J. Zabransky, D. Schneider, E. P. Kanter, Gu Yuan Zhuang, and D. S. Gemmell, Phys. Rev. Lett. **48**, 592 (1982).
[26] Y. Yamazaki and N. Oda, Phys. Rev. Lett. **52**, 29 (1984).
[27] E. Parilis, Nucl. Instrum. Methods Phys. Res. B **193**, 240 (2002).
[28] J. E. Borovsky and D. M. Suszcynsky, Phys. Rev. A **43**, 1433 (1991).
[29] O. Benka, A. Schinner, T. Fink, and M. Pfaffenlehner, Phys. Rev. A **52**, 3959 (1995).
[30] G. Schiwietz and G. Xiao, Nucl. Instrum. Methods Phys. Res. B **107**, 113 (1996).