Correlated Electron Emission from Thin Carbon Foils Bombarded by 1.8 MeV/u Ar Ions

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A correlation of secondary electrons emitted in the forward and in the backward directions has been studied for 1.8 MeV/u Ar ions bombarding carbon foils. Appreciable correlation has been observed not only for thin targets but also for thick targets where no correlation was expected. It is shown that a classical trajectory Monte Carlo simulation including conversion of bulk plasmons into electron-hole pairs which extends over the target can reproduce the observation qualitatively.

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lon induced secondary electron emission has been investigated for a long time, mainly dealing with a static nature of secondary electron production such as secondary electron yield [1], angular and energy distribution for various ions and targets [2]. In addition to these, a dynamic nature has recently become an object of study, where correlated evolution of secondary electrons under the influence of projectile potential has been studied for convoy and Rydberg electron production [3], a wakeriding electron production [4], etc. In the present study, we discuss another dynamic nature observed in a correlation between forward-emitted and backward-emitted electrons (referred to as "FB correlation" hereafter) [5,6], which reveals a new "cross section" of the secondary electron production relating with the transport property of electrons in solid.

When an energetic charged particle traverses through a foil, a number of target electrons are ionized along the ion path. Electrons directly produced by the incident ion (referred to as "daughter" electrons hereafter) travel in the foil suffering elastic and inelastic collisions with target atoms. When a daughter electron is energetic enough, it produces "granddaughter" electrons and their descendants through cascading ionization [7]. These electrons are emitted from the foil if their energies exceed the surface barrier when they reach the surface. A naive consideration indicates that the FB correlation is appreciable if the target is thick enough to allow cascading collision but at the same time thin enough to allow these cascading electrons to escape from both sides of the foil. A typical thickness satisfying the above two conditions will be around the inelastic mean free path (IMFP) of binary encounter electrons. The present experimental study has revealed, however, that the FB correlation is appreciable not only for thin foils but also for foils more than 10 times thicker than the IMFP. We point out the possibility that such an unusual long correlation distance results from the conversion of bulk plasmons into electron-hole pairs. The key idea is that a bulk plasmon extends spatially in the foil, and so can be converted into the pair at a point away from the volume where ordinary cascading electrons are scattered.

To study the FB correlation, multiplicity distribution of secondary electrons (MUSE) [5,6,8] was measured in the forward and in the backward directions simultaneously. The target foil tilted 45° with respect to the incident beam was biased to about -15 kV. A pair of SSDs were installed at the ground potential facing to the front and back surfaces of the target foil separated ~ 3 cm apart. Secondary electrons emitted from the foil are accelerated toward the SSD, and deposit an energy of a multiple of 15 keV. Accordingly the pulse height of the SSD output is proportional to the number of electrons emitted from the target foil. The charge states of exiting ions were measured in coincidence with the electron signal. Projectiles used were 1.8 MeV/u Ar ions with incident charge state $q_i = 7$ and 10–16. The targets were amorphous carbon foils of $\sim 2 \ \mu g/cm^2$ and $\sim 10 \ \mu g/cm^2$. It is found that the exiting charge state distributions $F(q_e)$ for these targets are in equilibrium for all incident charge states studied. The experiments were performed at the SF cyclotron of the Institute for Nuclear Study, University of Tokyo. Further details of the experimental setup and data handling have been given elsewhere [6,8].

The primary quantity we get from the MUSE experiment is the number distribution, $f(n_f, n_b; q_i, q_e)$, where n_f and n_b are the number of electrons emitted in the forward and in the backward directions, respectively. The number distribution is normalized with respect to n_f and n_b , i.e.,

$$\sum_{n_f,n_b} f(n_f,n_b;q_i,q_e) = 1 \; .$$

Figure 1 shows an example of $f(n_f, n_b; q_i)$ for 1.8 MeV/u Ar⁷⁺+C_{foil} (2 μ g/cm²), where $f(n_f, n_b; q_i) \equiv \sum_{q_e} F(q_e)$ $\times f(n_f, n_b; q_i, q_e)$. A single peak is seen at $n_f \sim 70$ and $n_b \sim 20$. The distribution with respect to n_f is almost symmetric, but a long tail follows in the direction of n_b .

To see the strength of the FB correlation explicitly, a



FIG. 1. The number distribution $f(n_f, n_b; q_i)$ for 1.8 MeV/u Ar⁷⁺ bombarding a carbon foil of $\sim 2 \mu g/\text{cm}^2$ tilted 45° with respect to the incident beam. The total number of incident ions is $\sim 7 \times 10^5$. Each data point is obtained by summing the neighboring four channels over n_f and n_b . The peak count at around $n_f \sim 70$ and $n_b \sim 20$ is $\sim 1.5 \times 10^4$. No smoothing procedures are employed.

correlation function defined by

$$\Delta f(n_f, n_b) \equiv f(n_f, n_b) - f_f(n_f) f_b(n_b) \tag{1}$$

is used, where $f_f(n_f) \equiv \sum_{n_b} f(n_f, n_b)$. A sum rule holds for the correlation function: $\sum_{n_f} \Delta f(n_f, n_b) = \sum_{n_b} \Delta f(n_f, n_b)$ n_b) = 0. If the emission in the forward direction occurs independent of the emission in the backward direction, $\Delta f(n_f, n_b) = 0$ for all n_f and n_b . Figure 2 shows a contour plot of $\Delta f(n_f, n_b)$ for 1.8 MeV/u Ar⁷⁺+C_{foil}(2 µg/cm²). It is seen that $\Delta f(n_f, n_b)$ consists of a sharp peak, a plateau, and two valleys. Typical values of $\Delta f(n_f, n_b)$ were $(1.6\pm0.1)\times10^{-3}$ at the peak around $n_f\sim60$ and $n_b \sim 20$, and $(-1.2 \pm 0.1) \times 10^{-3}$ at the valley around $n_f \sim 85$ and $n_b \sim 20$, i.e., the structures observed in Fig. 2 are statistically significant. Comparing with Fig. 1, the sharp peak of $\Delta f(n_f, n_b)$ is seen near the peak of $f(n_f, n_b)$ but slightly shifted to smaller n_f . The positive part of $\Delta f(n_f, n_b)$ appears around a line $n_f \sim n_b + 40$, i.e., a collision event to produce a certain amount of electrons more (less) than the average on one side of the foil is likely to produce the same amount of more (less) electrons also on the other side. In this case, the distribution is termed to have "positive linear correlation." In the present Letter, we will concentrate on this linear correlation and disregard quadratic and higher order correlations although they are not necessarily negligible as is



FIG. 2. The contour plot of the correlation function $\Delta f(n_f, n_b; q_i)$ for the number distribution shown in Fig. 1. The shadowed areas show that the correlation function Δf is negative. As in Fig. 1, no smoothing procedures are employed.

seen in Fig. 2.

To parametrize the strength of the linear correlation, we employ correlation coefficients defined by

$$\eta \equiv \frac{\sum_{n_f, n_b} (n_f - \langle n_f \rangle) (n_b - \langle n_b \rangle) f(n_f, n_b)}{\left[\sum_{n_f} (n_f - \langle n_f \rangle)^2 f(n_f)\right]^{1/2} \left[\sum_{n_b} (n_b - \langle n_b \rangle)^2 f(n_b)\right]^{1/2}} = \frac{\langle n_f n_b \rangle - \langle n_f \rangle \langle n_b \rangle}{\left(\langle n_f^2 \rangle - \langle n_f \rangle^2\right)^{1/2} \left(\langle n_b^2 \rangle - \langle n_b \rangle^2\right)^{1/2}},$$
(2)

where $\langle n_f \rangle$ for example is the average of n_f with respect to $f(n_f, n_b)$. η varies from -1 to 1 depending on the sign and the strength of the linear correlation. $\eta(q_i)$ is given in Table I for thin $(2 \ \mu g/cm^2)$ and thick $(10 \ \mu g/cm^2)$ targets. It is seen that $\eta(q_i)$ is about +0.1 for all q_i measured *irrespective of the foil thickness*, which is unexpected because a carbon foil of $10 \ \mu g/cm^2$ (~500 Å) is more than 10 times thicker than the IMFP of the binary encounter electrons (<3.5 keV). Typical values of the IMFP are ~60 and ~10 Å for the binary encounter electrons and for 50 eV electrons, respectively [9]. Further inspection of $\eta(q_i, q_e)$ for $2 \ \mu g/cm^2$ and 10 $\mu g/cm^2$ foils shows that the dependence on q_e is again very weak. Only a slight decrease of $\eta(q_i, q_e)$ with increasing q_e has been discernible.

TABLE I. Correlation coefficients η as a function of incident charge state q_i for Ar^{q_i +} bombarding $\sim 2 \,\mu g/cm^2$ and $10 \,\mu g/cm^2$ carbon foil. Statistical errors are indicated together.

q_i	7	10	11	12	13	14	15	16
$2 \mu g/cm^2$	0.10	0.11	0.10	0.09	0.10	0.09	0.09	0.08
	± 0.01	± 0.04	± 0.02	± 0.02	± 0.02	± 0.02	± 0.01	± 0.02
10 µg/cm ²	0.12			0.13	0.14		0.13	
	± 0.03			± 0.03	± 0.03		± 0.03	

Secondary electron production is supposed to consist of three steps, i.e., (1) production of daughter electrons by the projectile, (2) transport of them inside the foil including cascading collision with atoms and electrons (production of granddaughter electrons and their descendants), and (3) emission from the front and/or back surfaces of the foil [7]. It is noted that the ion energy is high enough and the target foil is thin enough so that the energy decrease of the ion in the foil is negligible in the present experimental condition.

In the first step, if the stopping power is not too large, i.e., if successive ionization events are well separated in space or time and then if target electrons are always available for further ionization, we may expect the successive ionizations to be independent of each other. Actually, a carbon atom has four valence electrons, and the mean free path of ionizing collisions for 1.8 MeV/u Ar in carbon is comparable to the interatomic distance, i.e., the assumption of independence is acceptable at least qualitatively. In other words, secondary electrons belonging to different collision events of the first step do not contribute to the FB correlation. The observation that $\eta(q_i, q_e)$ shows very weak dependence on q_i and q_e is consistent with the above discussion. In the second step, on the other hand, cascading electrons belonging to the same daughter electron do correlate with one another because the total number of cascading electrons depends on the energy of the daughter electron and the target thickness. The third step concerns each electron at the moment of escape, and contributes to the correlation indirectly through the height of the surface barrier.

The above scenario of secondary electron production indicates that the spatial extension of correlating electrons is confined to a size comparable to the order of the IMFP of daughter electrons. Accordingly, no correlation is expected for foils much thicker than the IMFP [5]. However, as is shown, we have observed appreciable correlation even for a very thick foil. To understand this interesting variance, we would propose as a possible candidate to take into account a process of plasmon conversion into electron-hole pair [10,11], which is analogous to the photon conversion into electron-hole pair. As bulk plasmon extends over the media [12], the conversion can occur away from the point of its production (referred as "remote conversion" hereafter). It is noted, however, that the plasmon extension for carbon is typically limited within a few cycles of the wake wavelength (≤ 100 \sim 150 Å). In other words, a small fraction of plasmons contributes to the FB correlation for targets much thicker than the IMFP. To excite a plasmon, a certain energymomentum relation should be fulfilled [13]. The threshold energy of the ion is estimated to be around several tens keV/u, i.e., our beam energy is high enough to produce plasmons in the first as well as in the second step.

To handle the phenomena quantitatively, a classical trajectory Monte Carlo (CTMC) simulation has been

employed assuming stochastic ionization of target electrons by the projectile ion followed by cascading ionizations. As the energy and angular distribution of the daughter electron depends not so much on the projectile charge but on the projectile velocity, we used, in the simulation, (1) proton-gas data [14] for the ionization of carbon foil by the ion, (2) optical data [9] for inelastic collision of electrons by amorphous carbon, and (3) theoretical evaluation for bulk and surface plasmon excitations [10]. Auger electron emission [15], surface barrier height, and the conversion of surface plasmons have also been taken into account. As is discussed, a uniform remote conversion of bulk plasmons is assumed. Considering that (1) the band structure and the density of states (DOS) of amorphous carbon are similar to those of graphite [16], and (2) the conversion takes place between occupied and unoccupied states of high DOS with energy difference of the plasmon energy (~ 25 eV for carbon), the energy of conversion electron in vacuum is estimated to be ~ 15 eV. This estimation is consistent with the relation of the energy loss to the energy of the corresponding secondary electron [17], and the peak energy of "shock" electrons which may be another example of the conversion of plasmons but with rather high momentum [18]. It is confirmed that the CTMC simulation described above reasonably reproduces several basic quantities such as the stopping power, the energy straggling, secondary electron spectra, secondary electron yield, etc., for proton impact. The simulation predicts positive η for ions bombarding thin targets, because an energetic ion produces daughter electrons in a wide energy region up to the binary electron energy, which results in a wide number distribution of descendant electrons which are eventually divided into the forward and backward directions. Including the remote conversion of bulk plasmons, we get "positive correlation" even for foils much thicker than the IMFP, which fits our observation reasonably.

Finally, it is noted that the CTMC simulation predicts negative correlation if a monoenergetic electron bombards a foil with a thickness of the order of the IMFP, i.e., η changes its sign from positive to negative and then is close to 0 as the foil thickness increases. This negative η appears because the total number of descendant electrons are more or less definite for a foil with the thickness of the order of IMFP. The descendant electrons are shared in the forward and in the backward directions, i.e., $\langle n_f \rangle + \langle n_b \rangle$ is roughly constant. For heavy ions with many loosely bound electrons, the production of semimonoenergetic electron is realized by the electron loss to the continuum (ELC) process. Actually, the slight decrease of $\eta(q_i, q_e)$ with increasing q_e indicates the participation of the ELC electrons.

In conclusion, we have found appreciable forwardbackward (FB) correlation in secondary electron emission not only for a 2 μ g/cm² carbon foil but also for a 10 μ g/cm² foil. A CTMC simulation shows that the origin of the FB correlation is attributable to the cascading ionization of target electrons. It is proposed as a possible candidate that the conversion of bulk plasmons into electron-hole pairs taking place all over the target is responsible to explain the discernible correlation for the thick foil ($\sim 10 \ \mu g/cm^2$).

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- M. Roesler et al., in Particle Induced Electron Emission I, Springer Tracts in Modern Physics Vol. 122 (Springer, Berlin, 1991); in Particle Induced Electron Emission II, Springer Tracts in Modern Physics Vol. 123 (Springer, Berlin, 1991), and references therein.
- [2] E.g., N. Oda et al., Nucl. Instrum. Methods 170, 571 (1980); and Y. Yamazaki, in Proceedings of the Third Workshop on High Energy Ion-Atom Collisions, edited by D. Berényi and G. Hock, Lecture Notes in Physics Vol. 294 (Springer-Verlag, Berlin, 1988), p. 322.
- [3] E.g., J. Burgdoerfer, in Interaction of Charged Particles with Solids and Surfaces, edited by A. Gras-Marti et al. (Plenum, New York, 1991), p. 459; Y. Yamazaki et al.,

Phys. Rev. Lett. **61**, 2913 (1988); and J. Burgdoerfer and C. Bottcher, Phys. Rev. Lett. **61**, 2917 (1988).

- [4] J. Burgdoerfer, J. Wang, and J. Mueller, Phys. Rev. Lett.
 62, 1599 (1989); Y. Yamazaki *et al.*, J. Phys. Soc. Jpn.
 59, 2643 (1990).
- [5] A. A. Kozochkina, V. B. Leonas, and M. Witte, Nucl. Instrum. Methods Phys. Res., Sect. B 62, 51 (1991); V. B. Leonas, Usp. Fiz. Nauk 161, 73 (1991) [Sov. Phys. Usp. 34, 317 (1991)].
- [6] T. Azuma *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. B 67, 636 (1992).
- [7] E.g., E. J. Sternglass, Phys. Rev. 108, 1 (1957).
- [8] Y. Yamazaki and K. Kuroki, Nucl. Instrum. Methods Phys. Res., Sect. A 262, 118 (1987); H. G. Clerc *et al.*, Nucl. Instrum. Methods 113, 325 (1973).
- [9] J. C. Ashley, J. Electron Spectrosc. Relat. Phenom. (to be published); J. Ashley (private communication).
- [10] P. Nozières and D. Pines, Phys. Rev. 109, 741 (1958).
- [11] T. E. Everhart, N. Saeki, R. Shimizu, and T. Koshikawa, J. Appl. Phys. 47, 2941 (1976); L. S. Caputi and L. Papagno, Phys. Lett. 93A, 417 (1983).
- [12] R. A. Ferrel, Phys. Rev. 111, 1214 (1958); W. Steinmann and M. Skibowski, Phys. Rev. Lett. 16, 989 (1966).
- [13] P. E. Batson and J. Silcox, Phys. Rev. B 27, 5224 (1983).
- [14] W. E. Wilson and L. H. Toburen, Phys. Rev. A 11, 1303 (1975).
- [15] L. H. Toburen, W. E. Wilson, and H. G. Paretzke, Phys. Rev. A 25, 713 (1982).
- [16] R. C. Tatar and S. Rabii, Phys. Rev. B 25, 4126 (1982);
 R. F. Willis and B. Fitton, Phys. Rev. B 9, 1926 (1974).
- [17] F. J. Pijper and P. Kruit, Phys. Rev. B 44, 9192 (1991).
- [18] M. Burkhard et al., Phys. Rev. Lett. 58, 1773 (1987).