Dependence of secondary electron emission on the emergent angle of 2.5-MeV protons penetrating a thin carbon foil

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With 2.5-MeV proton beams incident on a carbon foil of 1.8 μ g/cm² in thickness, the statistical distributions of the number of simultaneously emitted secondary electrons (SEs) have been measured as a function of the emergent angle of ions penetrating the foil in the range from 0.0 mrad to 2.0 mrad for every 0.5-mrad step. The measurement of SEs was carried out at the forward and backward directions of the incident beam separately. For all of the measured angles, the probability of simultaneous *n* electron emission per projectile, W_n , exhibits roughly an exponential decrease with increasing n . Up to \sim 1 mrad, however, the decreasing rate becomes smaller with an increase in the emergent angle. On the other hand, W_n reaches saturation at larger angles. This behavior is common to the forward and backward SE emission. In terms of this angular dependence, the average SE yields per projectile at the forward direction, γ_F , and at the backward direction, γ_B , increase as high as about 50% at 1.0 mrad compared with corresponding ones at 0.0 mrad and saturate at larger angles. As a result of a Monte Carlo simulation taking account of the impact parameter dependent energy loss in a single collision of a proton with a carbon atom, it is found that the calculated energy losses exhibit a quite similar emergent angle dependence to that of the measured SE yields.

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I. INTRODUCTION

Kinetic emission of secondary electrons (SEs) from solid surface under fast ion bombardments is one of the very important phenomena in ion-solid interactions and has been studied intensively for a long time $[1,2]$. Sterngrass has proposed that the SE emission is described by the three-step processes. First, the creation of excited electrons via collisions of projectiles with target atoms in the solid. Then the transport of liberated electrons through the bulk to the surface including higher-order ionizations by high energy internal SEs. Finally, there is the transmission through the surface-potential barrier [3]. Since the total energy per unit length deposited to the excited electrons produced in the first step is proportional to the electronic stopping power of the target material, S_e , a linear relation between the electronic stopping and the SE yields γ , the average number of the emitted electrons per projectile, is predicted theoretically $\lceil 3-5 \rceil$ as

$$
\gamma_i = \Lambda_i S_e \,, \tag{1}
$$

where Λ_i is the so-called material parameter depending only on the target material $\left[3-5\right]$. The index *i* can stand for *B* for the backward (from the beam-entrance side), F for the forward (from the beam-exit side), or *T* for the total SE yields $(\gamma_{\rm T} = \gamma_{\rm F} + \gamma_{\rm B})$. For proton impact on several kinds of metal, approximately linear relations between γ_{FB} and S_e have been observed experimentally over a wide energy range of a few keV to tens of MeV [2].

In previous experiments with energetic protons (several MeV) $[6–11]$, as well as protons and helium ions of intermediate energies (lower than 1 MeV) $[12–17]$, it has been reported that the energy loss of foil-transmitted ions increases at larger emergent angles due to the impactparameter dependence of the electronic energy loss in a single ion-atom collision. Similar behavior is observed also for heavier ions $[18]$. Since relatively large number of internal SEs or those with higher energies are produced at small impact-parameter collisions, the angular dependence is expected to be observed also in the SE yields. In order to obtain a clear angular dependence of the energy loss with energetic light ions, a finite energy resolution of particle detectors imposes the measurement with rather thick target foils on us. This weakens the constraint on the impact-parameters in the individual collisions. On the other hand, there is less restriction for the measurement of number distributions of simultaneously emitted SEs. In energy-loss measurements with intermediate- or low-energy light ions or with heavy ions penetrating a thin foil, the thickness nonuniformity of target foils hinders us from investigating the details of the impactparameter dependent inelastic energy loss $[19–22]$. As is discussed later, however, this effect is diminished in the SE emission.

In the present work, the statistical distributions of the number of simultaneously emitted SEs from a carbon foil of 1.8 μ g/cm² by 2.5-MeV proton impact have been measured as a function of the emergent angle of foil-transmitted ions in the range from 0.0 mrad to 2.0 mrad. The measured SE yields are compared with energy losses calculated by a Monte Carlo simulation taking account of the impactparameter dependent electronic stoppings in a single collision of a proton with a carbon atom.

II. EXPERIMENT

The experiment was performed with 2.5-MeV proton beams obtained with a 1.7-MV tandem Van de Graaff accel-

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erator at Nara Women's University. The proton beam was transported to a target carbon foil using the same method described in Refs. $[23,24]$. The incident beam is collimated with two diaphragms of 0.2 mm in diameter and 224 cm apart. A baffle of 1.0 mm in diameter was placed 5 cm behind the second diaphragm to prevent edge-scattered particles from hitting the target. The target foil was placed 7 cm behind the baffle and tilted by 45° relative to the normal angle of incidence. The foil was floated at a potential of -30 kV. The emitted electrons are accelerated to a grounded electrode that is parallel to the foil and 40 mm away. At the grounded electrode a solid-state Si detector (SSD) of 100 mm² sensitive area faces the target foil. The thickness of the carbon target foil was determined to be 1.8 μ g/cm² by measuring the transmitted fraction of 2.5- $MeV H⁰$, while accounting for the electron-loss and capture cross sections involved [25]. The independently movable vertical and horizontal slits were located about 70 cm behind the target and used to define the emergent angle of transmitted protons. Their widths were determined to be 0.265 mm (vertical) and 0.300 mm (horizontal) with a Tiyoda LTG bi-AII microscope of ± 1 μ m position resolution. The angleresolved protons were detected by a Si photodiode of 800 mm2 sensitive area. The geometrical parameters determining the angular spread of the incident and detected protons have been taken into account properly in the Monte Carlo simulation to evaluate energy losses depending on the emergent angle. The energy spectrum of simultaneously emitted electrons was measured in coincidence with these protons. The measured emergent angle was in the range from 0.0 mrad to 2.0 mrad for every 0.5-mrad step. The forward and backward measurements were carried out separately.

It should be mentioned that our measurements were carried out with a typical pressure of $\sim 5 \times 10^{-7}$ Torr, and no prior treatment to the target foil had been applied. As a consequence, the present data seem to include some effects by adsorbed contaminants. It is well known that the SE emission is very sensitive to the surface conditions of the foil $[2]$. On the other hand the SE yields may also be affected by surface modifications induced by the incident beam; however, the coincidence measurement requires very low beam intensity, and the obtained results seem to be free from such modifications. As a consequence, there might be some discrepancies between the present absolute values and those obtained with sputter-cleaned foils in an ultrahigh vacuum. But the relative change of the SE emission depending on the emergent angle of transmitted ions is expected to be independent of the surface condition of the foil.

III. EVALUATION OF EMISSION STATISTICS

The analysis of electron energy spectra detected by the SSD was carried out by a similar method to that presented in Ref. [26]. The energy spectrum $S(E)$ was fitted to the following equation,

$$
S(E) = \sum_{n=1}^{n_{\text{max}}} Y_n F_n(E),
$$
 (2)

FIG. 1. The measured emission probability as a function of *n* emitted SEs for a given event in which a 2.5-MeV proton emerges from a carbon foil of $1.8 \pm 0.1 \mu$ g/cm² at any of the angles listed.

where $F_n(E)$ and Y_n denote the normalized energy distribution and the number of total events having *n* emitted electrons, respectively. $F_n(E)$ is expressed by the superposition of $n+1$ Gaussian functions that correspond to the number of electrons backscattered through the detector surface. All of the parameters such as the electron backscattering probability at the detector surface, its *K* factor, the energy resolution of the SSD, and so forth were also determined simultaneously. In the present analysis, n_{max} , the maximum number of simultaneously emitted SEs observed in the spectra ranged from 11 to 15.

We obtain a result of the expected γ as

$$
\gamma = \sum_{n=1}^{n_{\text{max}}} nW_n \quad \text{with} \quad W_n = Y_n/N,
$$
 (3)

where N and W_n denote the number of detected protons and the probability of the simultaneous emission of *n* electrons, respectively. In the coincidence measurement, the probability of no SE emission, W_0 , can also be determined by

$$
W_0 = 1 - \sum_{n=1}^{n_{\text{max}}} W_n.
$$
 (4)

IV. RESULTS AND DISCUSSION

Figure 1 represents the distributions of emission probabilities, W_n , for $n \le 10$. Due to the poor counting statistics, data points for $n > 10$ deviate upward or downward from smoothly decreasing curves as *n* increases. For all of the measured spectra, however, the SE yields obtained from the summation up to $n=10$ in Eq. (3) did not differ by more than 2% compared with those from $n = n_{\text{max}}$. For all of the measured angles, W_n exhibits roughly an exponential decrease with *n*, but up to \sim 1 mrad the decreasing rate becomes smaller with the increase of the emergent angle. On the other hand, *Wn* appears to reach a saturation value for each *n* at larger angles. This behavior is common to the forward and backward SE emission. The plots in Fig. 2 shown with squares represent the emergent angle dependence of γ_F and

FIG. 2. The emergent-angle dependence of the measured SE yields (squares) and corresponding energy losses (circles) calculated by a Monte Carlo simulation. The simulation takes into account the impact-parameter dependence of the energy loss in a single collision of a proton with a carbon atom $[30]$. Energy losses are normalized to the SE yields at 0.0 mrad.

 γ_B . The 5% errors shown in the plots are associated with the uncertainty of the 45° tilt angle of the target. In terms of this emergent angular dependence of W_n 's, γ_F and γ_B increase up to 1.0 mrad and approach a constant value at larger angles. At each angle, γ_F is slightly larger than the corresponding γ_B due to the preferential forward emission of high-energy internal SEs. Both for the forward and backward directions, an enhancement of as high as \sim 50% is observed in the SE yields at the large angles.

Before the interpretation of the present result, we compare the present W_n 's with distributions theoretically proposed to describe the SE emissions. At first, a Poisson distribution is expected not to reproduce well the experimental data due to the contribution from high-energy internal SEs leading to the production of other SEs by the cascade multiplication. The comparison with a Poisson distribution is given in our previous paper [24]. Furthermore, Benka *et al.* have also pointed out the inadequacy of the Poisson distribution [28]. On the other hand, some authors have indicated that a Pólya distribution can reproduce very well the *W_n*'s obtained from their experiments $[26-30]$. In order to obtain the best fit to our data, we have varied the parameter b given in Eq. (2) of Ref. [29]. In this fitting, we have kept the parameter μ for the mean SE yields a constant, since it is well determined by our measurements. However, we have been unsuccessful at reproducing our W_n values. In contrast to the measurements by other authors $[26-30]$, the present data were obtained by a transmission experiment. This may affect the applicability of the Pólya distribution. At the present stage, however, we cannot discuss any more about the poor agreement between our data and the Pólya distribution.

Here we discuss the origin of the increase followed by the saturation of γ values at larger angles. Figure 3 represents the angular distribution due to multiple scattering of 2.5- MeV protons transmitted through a carbon foil of 2.55 $(51.8\times\sqrt{2})$ μ g/cm² evaluated with the theory of Sigmund and Winterbon $[31]$. The dashed curve corresponds to single scattering by an unscreened Coulomb potential. As is clear

FIG. 3. The angular distribution due to multiple scattering of 2.5-MeV protons emerging from a carbon foil of $2.55 (=1.8)$ $\sqrt{2}$) μ g/cm² evaluated with the theory of Sigmund and Winterbon $\lceil 31 \rceil$ (solid curve). The dashed curve corresponds to single scattering by an unscreened Coulomb potential.

from this figure, for angles larger than ~ 0.5 mrad the emergent angle is mainly determined by single close collisions. An evaluation using the Molière potential $\left[32\right]$ predicts that a collision with an impact parameter of 0.06 a.u. should give a scattering angle of 1.0 mrad. Applying a Dirac-Fock method calculation $\begin{bmatrix} 33 \end{bmatrix}$ shows that the *K*-shell electron density of a carbon atom reaches its maximum value at the radius of about 0.174 a.u., and a proton incident on a carbon atom with a impact parameter equal to this radius is scattered at ~ 0.3 mrad. Therefore, while protons emerging at angles around 0.3 mrad seem to lose their energies depending on the impact parameter, those emerging at angles larger than \sim 1 mrad penetrate deeply inside the *K*-shell electron cloud of a carbon atom and lose approximately constant energies, irrespective of the impact parameter. Furthermore, at the latter region a small decrease of the impact parameter gives rise to a large increase of the scattering angle. Hence, the observed behavior is quite reasonable.

In the angular range from 0° to 4° , a similar emergent angle dependence has been observed in the energy loss of 5-MeV protons penetrating through uniform Au and Cu foils of a few mg/cm² in thickness [9–11]. In contrast to the present SE yields, however, the enhancement of the energy loss at larger angles was only 1–2% of that observed at 0.0 mrad. Although the measured angular range of energy losses was about 40 times larger, the Au and Cu foils used were three orders of magnitude thicker than the present carbon foil; therefore, the present experiment for the SE emisssion is more sensitive to the impact parameter.

The Monte Carlo simulations carried out under our experimental conditions here give us a quantitative comparison to the results of the SE yield measurement. In this simulation, we have accounted for the impact-parameter dependent energy loss of a proton colliding with a carbon atom according to the theoretical treatment by Kaneko $[34]$. This theory is based on the dielectric-function method and on localdensity-electron models for carbon atoms in a solid. The Molière potential [32] was employed to determine the relation between the impact parameter and the scatting angle. The procedures of the simulation were quite similar to those given in our previous paper $[23]$.

Target atoms were assumed to be spheres of radius *R* and to be distributed randomly. *R* is taken to be $5a_B$. In this connection a preliminary simulation with $R=2a_B$ gives very similar results as that for which $R=5a_B$. If a projectile traverses these spheres it loses a part of the kinetic energy and is scattered by the target atoms. Initially, the depth at which the first collision occurs is determined by a pseudorandom number generated by the code. Then the impact parameter of the collision and the direction after scattering is determined with two other pseudorandom numbers. Besides recording the energy loss at the impact parameter of the collision, we also follow the polar and azimuthal angles of the scattering with respect to the incident direction. Subsequent collisions are treated in the same manner until the projectile exits the foil. The total deflection angle can be calculated from the polar and azimuthal angles according to the approximation described in Ref. [7]. This angle as well as the corresponding total energy loss is registered for each projectile. As is described in Ref. $[23]$, it is quite important to take into account the geometrical parameters, such as the angular divergence of the incident beam and the finite acceptance of the emergent angle defining slits in the present SE measurement. The circles in Fig. 2 represent the result of the simulation. In both the forward and backward directions the energy loss is normalized to the SE yields at 0.0 mrad. As is clear from the figure, the measured SE yields and the calculated energy losses exhibit quite a similar emergent angular dependence. This similarity quantitatively confirms a simple prediction that the SE yields depend almost linearly on the energy deposited to excited electrons in the individual collisions of a projectile with target atoms in a solid.

Finally, we discuss the effect of the thickness nonuniformity of the target foil on the present data. It is well known that the thickness nonuniformity gives rise to a spurious enhancement of the energy losses at large emergent angles $(19-22)$. In some cases this effect prevents us from evaluating the contribution of electronic stopping power due to solely the impact parameter dependence. For the SE yields, however, the situation is quite different. From our previous measurement the escape depth of internal SEs in a carbon foil by 2.5-MeV proton impact was estimated to be 0.85 $\pm 0.10 \ \mu$ g/cm² [24]. With this value the thickness increase of, for example, 20% in the present target foil raises the SE yields at most by 5%, which is a sufficiently small effect compared to the measured enhancement of SE yields at larger angles. So, the thickness inhomogeneity does not play a significant role in our experiments.

V. SUMMARY

The emission statistics of the SEs from a thin carbon foil by 2.5-MeV proton impact have been measured as a function of the emergent angle of the transmitted protons. Up to \sim 1 mrad the measured SE yields increase with the emergent angle. At larger angles the SE yields tend to approach a constant value. Monte Carlo calculations used to simulate the energy loss under the present experimental conditions show good agreement with the measured SE yields. This agreement suggests that the SE emission measurement is a very effective method to investigate the details of ion-solid interactions.

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