Secondary-electron emission from nonmetallic (perovskite) surfaces under slow proton impact: Absence of apparent threshold

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The secondary-electron emission (SEE) yield has been measured at low-energy proton impact for clean Cu metal and nonmetallic targets. The observed SEE for Cu below 3 keV shows the apparent thresholdlike behavior and decreases roughly as v_p^4 when the proton velocity (v_p) decreases; meanwhile, there seems to be no such behavior in nonmetallic targets whose SEE decreases as v_p^1. The SEE for these two different types of targets is also compared with the electronic stopping powers. [S1050-2947(99)03904-9]

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As the slow (a few keV) proton velocity v_p decreases, its electronic stopping power S_e in rare gas as well as in metal solid targets is known to deviate from the expected v_p^1 dependence [1] and decreases very rapidly ($\approx v_p^4$) at the velocity of roughly 10^7 cm/s, showing a thresholdlike behavior. This sudden decrease of the S_e at low energies is theoretically understood to be due to the non-negligible size of the excitation/ionization energy of the target elements, which is the lower limit of integration of the energy transferred to the target, over the projectile kinetic energy, as discussed by Semrad [2], Golser and Semrad [3], and Schiefermüller *et al.* [4].

Recently, Eder *et al.* [5] have reported the absence of such an ''apparent'' threshold effect in the S_e of slow protons in insulators such as Al_2O_3 and SiO_2 as well as LiF. Compared with those in rare gas and metal targets where the direct binary collision interactions are mostly responsible to the S_e, the physical mechanisms for the absence of the apparent threshold in insulating nonmetallic targets can be due to the electron promotion in a quasimolecule formed during collisions, through which one of the innershell electrons promotes to higher excited states, followed by the autoionization.

On the other hand, in the secondary-electron emission (SEE) yield under slow proton impact, the apparent threshold behavior from clean metal targets by Lakits *et al.* [6,7] and Spierings *et al.* [8] and the absence of such threshold in LiF by Vana, Aumayr, and Winter [9] and Stracke *et al.* [10] have also been reported recently.

It has been known that the secondary-electron emission phenomena from solids are closely related with the electronic stopping power [11]. Various approaches are being studied for establishing their relationship in a wide range of the collision parameters such as the collision energy, particle and their charges and targets.

In this paper we would like to show significant difference in the impact energy dependence of the SEE in clean metal Cu targets and in nonmetallic perovskite-structured $SrCeO_3$ (5% Yb) solid targets under slow proton impact. The present experiment has been performed under an ultrahigh vacuum ($\approx 4\times 10^{-11}$ Torr) chamber equipped with a target sputtering system by argon ion beam and an Auger electron spectrometer to check the surface cleanness.

The present observed results of the SEE from clean Cu surfaces under proton impact, combined with data by Baragiola, Alonso, and Oliva-Florio [12] and Hasselkamp *et al.* [13] at higher energies, are shown in the lower part of Fig. 1 where, as the proton energy decreases, the SEE starts to show sharp reduction below 3 keV and levels off to a roughly constant value at further lower energies.

This behavior can be understood as follows [14]. The total SEE is given as a sum of the contribution of two processes: (1) The kinetic emission process (KE) due to the kinetic energy of the incident particles, which increase with their kinetic energy and (2) the potential emission process (PE) due to their potential energy, which is independent of their kinetic energy. The apparent constant SEE at the lowest energies (below 0.5 keV) observed in the present paper is consistent with the total SEE, which is dominated by the PE. Using the empirical formula proposed by Baragiola, Alonso, and Oliva-Florio [12], the contribution of the PE (in this case, by protons) to the SEE is predicted to be 0.055 that is consistent with the observed value of 0.07 at the lowest incident energies. The rising SEE at energies above 0.6 keV is related entirely to the KE. It is easily noted that the *partial* SEE due to the KE up to 3 keV, which has been obtained after subtraction of the contribution of the PE from the total SEE and shown with a dotted curve in Fig. 1, increases rapidly as $v_p^{3.5}$ and then as v_p^1 above 3 keV. It is now worth noting that this $v_p^{3.5}$ dependence of the partial SEE between 0.6 and 3 keV is in rough agreement with the v_p^4 dependence of the S_e, as mentioned previously [1].

Though the SEE from solids depends on a number of processes such as (a) electron production, (b) electron transport mechanisms to surface, and (c) release mechanisms from surfaces and are much more complicated than that in gas targets, the SEE is known to be parametrized nicely with the electronic stopping power S_e as follows [11]:

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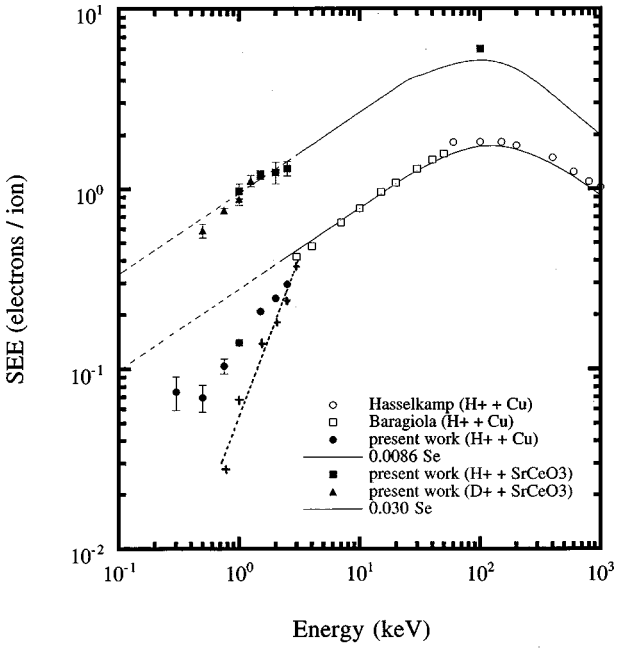

FIG. 1. Energy dependence of the SEE from cleaned Cu metal target (lower part) and from nonmetallic, cleaned $SrCeO_3$ target (upper part) as a function of proton impact energy. Data at high energies for Cu target are taken from Baragiola, Alonso, and Oliva-Florio [12] and Hasselkamp *et al.* [13]. The solid lines represent the S_e calculated by TRIM code with different constants $\alpha = 0.0086$ and $\alpha = 0.030$ for a clean Cu and $SrCeO_3$ targets, respectively [see Eq. (1)]. The dashed lines show the extrapolation of the results by TRIM toward lower energies. On the other hand, a dotted line shows the best fitting to the *partial* SEE due to the KE for clean Cu targets that has been obtained after substraction of the contribution of the PE from the total SEE and is found to change as $v_p^{3.5}$.

$$\mathrm{SEE} = \alpha S_e , \tag{1}$$

where α is constant.

Experiments performed so far indicate that α is roughly constant over a wide range of the proton impact energy range from 3 keV up to 10 MeV. Indeed, in our recent analysis including the experimental data available, the constant α above 3 keV has also been found to be practically independent of the clean metal target materials. The lower solid curve in Fig. 1 calculated with TRIM code indicates that the best fit to the observed SEE above the apparent threshold (>3 keV) is found to be obtained with $\alpha = 0.0086$ nm/eV for the present clean Cu target [15] (note that, as the original TRIM code is intended to be used above 10 keV [1], the S_e calculated by TRIM may not be accurate below 3 keV anymore).

The situation is quite different below 3 keV where, as mentioned already [2,3], the S_e is expected to significantly deviate from the *extrapolation* of TRIM calculated above 3 keV (the dashed line in Fig. 1). Though it is presently difficult to accurately calculate the electronic stopping powers S_e near and below the apparent threshold as no relevant mechanism has been understood well, it has been experimentally established that both of the electronic stopping power S_e [2–4] and the SEE yields including the present and the previous results [6,7] in clean metal targets decrease sharply below the apparent threshold as the proton collision energy decreases, showing the thresholdlike behavior. Thus, the constant α in Eq. (1) is expected to be practically unchanged over the whole proton impact energy range, except for the lowest impact energy region (<0.6 keV in proton+Cu collisions) where the PE plays a role.

On the other hand, the observed SEE for nonmetallic, clean $SrCeO_3$ surfaces, as shown in the upper part of Fig. 1, has been found to indicate two distinct features. Firstly, the SEE from the nonmetallic surfaces is quite large, compared with that for clean metal surfaces and this enhancement can be largely due to the increased electron transport to surfaces [16]. Secondly, without showing any apparent thresholdlike behavior, the observed SEE does decrease roughly as v_p^1 as the proton velocity decreases below 3 keV down to the lowest energies.

Thus, above 3 keV, where TRIM code works nicely, the SEE observed in the present paper has been found to be proportional to the electronic stopping cross section S_e, as suggested in Eq. (1), where the constant α is found to be 0.030 nm/eV for this particular nonmetallic solid target, which indeed is by a factor of 4 larger than that for clean metallic (Cu) targets.

Presently, there still is no clear understanding why there is the absence of the apparent threshold in the SEE from such nonmetallic surfaces at lower energies. One of the most likely electron emission mechanisms, which has been discarded so far, can be the electron promotion mechanism, similar to that in the S_e already discussed [5]. In such an electron promotion process within a quasimolecule formed during slow heavy particle collisions, an electron promoted into one of the excited states is emitted into vacuum. Though similar to that in LiF surfaces shown by Stracke *et al.* [10], the present case is found to have a slightly different feature in the energy correlation diagram involving H (neutralized atomic hydrogen projectile) and O atom, one of the constituents of this nonmetallic target: the diabatic energy levels of H ($1s$:13.598 eV) and O ($2p$:13.618 eV) are almost degenerated at the isolated-atomic state limit. Thus, in contrast to LiF [10], an electron, either in the $2p$ state of O atom or in the $1s$ state of H atom, can promote via $3d\sigma$ orbit to highly excited states. Here we should note that the probabilities of the electron promotion during the quasimolecular formation involving H and O atoms in slow ion-atom collisions are generally expected to decrease slowly as the collision energy decreases [17–20], which is similar to the observed SEE behavior ($\propto v_p^1$).

On the other hand, the probabilities of the electron promotion are very low in the metallic targets under proton impact as practically no energy-matching levels exist between the projectile and targets. That is why the apparent threshold is observed in clean metallic targets.

Similarly, other constituents (Sr and Ce) in the present nonmetallic $SrCeO_3$ targets are expected to play only a minor role in such an electron promotion mechanism involving hydrogen atom.

It is concluded that the present results in nonmetallic perovskite $SrCeO_3$ targets as well as those in LiF previously observed by Stracke *et al.* [10] suggest that, over a wide range of the proton impact energy, the SEE from the nonme-

tallic targets decrease as v_p^1 down to the lowest impact energies investigated when the proton velocity decreases, without showing any apparent threshold. This is in sharp contrast to the SEE observed in gas [3] and clean metal [8] targets at low energies (<3 keV), which deviates significantly from the v_p^1 dependence expected from the simple binary collision model and shows the apparent thresholdlike behavior, where the SEE has roughly the $v_p^{3.5}$ dependence.

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