Surface-plasmon-assisted secondary-electron emission from an atomically flat LiF(001) surface

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Abstract

The production process of secondary electrons at a LiF(001) surface is investigated utilizing specular reflection of 0.5 MeV protons. From the observed secondary-electron yield, the position-dependent secondary-electron production rate is derived. Comparing the obtained production rate with the observed surface stopping power, the conversion efficiency of the excited surface plasmons into electron–hole pairs is estimated to be almost 100% similar to KCl(001) [6], while the efficiency for SnTe(001) was about 30% [5]. Observation of the surfaces by atomic force microscopy reveals that the LiF(001) is atomically flat while the SnTe(001) has many surface steps. The large conversion efficiency can be ascribed to the flatness of the LiF(001), which prohibits decay of the surface plasmons into photons. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

Secondary-electron emission (SEE) induced by ion impact on solids has been extensively studied (see for example [1]). There are two different mechanisms of the SEE: When the potential energy of the incident ion is larger than twice the work function of the target, potential electron emission (PEE) can occur. The other mechanism, called kinetic electron emission (KEE), is a process of direct transfer of kinetic energy from the incident ion to the target electrons. The mechanism of KEE is explained by a so-called three step model [2]. The three steps are production of excited electrons over the vacuum level inside solids, transportation to the surface, and transmission through the surface barrier. Although these three processes are analyzed separately in theoretical studies, the separation is usually difficult in experimental studies. If the excitation of electrons takes place outside solids, the electron is ejected directly to the vacuum without other processes and the production process can be studied separately from other processes. This can be done by utilizing specular reflection of fast ions at single crystal surfaces.
When a fast ion is incident on a single crystal surface with a grazing angle smaller than a critical angle (≈6 mrad for 0.5 MeV H$^+$ on LiF(0 0 1)), the ion is specularly reflected from the surface without penetration inside the crystal [3,4]. Secondary electrons induced by the ion are produced outside the crystal and not subject to the transportation and transmission processes. We have demonstrated that the position-dependent secondary-electron production rate can be derived from the observed secondary-electron yield induced by the specularly reflected ions [5,6]. The obtained production rate was explained in terms of the direct excitation of surface electrons by ion impact as well as the decay of bulk and surface plasmons into electron–hole pairs. The conversion efficiency of the excited surface plasmons into electron–hole pairs was estimated to be about 30% for SnTe(0 0 1) and almost 100% for KCl(0 0 1). This large difference was suggested to be due to the difference in the surface conditions. The surface of SnTe, which was prepared by vacuum evaporation, had many surface steps, while the KCl(0 0 1), which was prepared by cleavage, was atomically flat [6]. The surface roughness allows decay of surface plasmons into photons, which is prohibited at a perfect flat surface [7]. This results in a large conversion efficiency of surface plasmons into electron–hole pairs for flat surfaces. In the present paper, we extend the study to another surface, namely LiF(0 0 1) to confirm our explanation. The surface of LiF(0 0 1) prepared by cleavage is atomically flat as is KCl(0 0 1) and so a large conversion efficiency is expected if our explanation is correct.

2. Experimental

Details of the experimental procedure are described elsewhere [5]. Briefly, a single crystal of LiF cleaved along (001) in air was mounted on a goniometer in a UHV scattering chamber. The surface was heated at 250°C to prepare a clean surface and also to avoid surface charging. A beam of 0.5 MeV protons from the 1.7 MV Tandetron accelerator of Kyoto University was collimated by a series of apertures to less than 0.1 × 0.1 mm$^2$ and to a divergence angle less than 0.3 mrad. The beam was incident on the LiF(0 0 1) at glancing angles $\theta_i$ of 2–6 mrad. A movable aperture of $\phi = 1$ mm, placed 425 mm down stream from the target, was used to select specularly reflected protons. The energy spectrum of specularly reflected protons were measured by a silicon surface barrier detector (SSBD). The mean energy loss of the reflected protons was obtained from the observed energy spectrum. The secondary electrons induced by the protons were detected by a microchannel plate (MCP, effective diameter $\phi = 20$ mm and placed at ~10 mm in front of the target surface) in coincidence with the specularly reflected protons. The MCP was biased at +700 V to collect all secondary electrons emitted from the target. The pulse height distribution of the MCP signals was registered by a pulse height analyzer.

3. Results and discussion

An example of the observed pulse height distribution of MCP signals measured in coincidence with the reflected proton is displayed (solid circle) together with the distribution measured without the proton beam (open circle) in Fig. 1. The pulse height $I$ of the MCP signal is proportional to the number of secondary electrons detected [8]. The

![Fig. 1. Pulse height distribution of MCP signals detected in coincidence with the reflected protons when 0.5 MeV protons are incident on LiF(0 0 1) at $\theta_i = 4.9$ mrad. The distribution measured without the proton beam is also shown.](image)
signals measured without the proton beam correspond to the detection of single electrons or pairs of electrons which are created by an ion pump evacuating the chamber. The secondary-electron yield \( g \) (the mean number of secondary electrons produced by a single proton) can be derived from the observed distributions using the equation, 
\[
g = \langle I \rangle / (e I),
\]
where \( \langle I \rangle \) is the mean value of the pulse height distribution, \( I \) the pulse height for single electron detection and \( e \) the efficiency of the MCP (\( \approx 0.6 \) [9]).

Fig. 2 depicts the temperature dependence of the observed secondary-electron yield. While the observed yield decreases rapidly below 50°C due to the macroscopic charging of the surface, it is almost constant above 100°C showing that the macroscopic charging can be avoided by the ionic conduction at these temperatures. The following measurements were performed at 250°C to avoid the macroscopic charging.

Fig. 3 shows the observed secondary-electron yield as a function of \( \theta_i \) together with the observed energy loss. The results for SnTe(001) from our previous work [5] are also shown for comparison. Although the crystal structures of LiF and SnTe are the same (NaCl-type), electronic structures are different. SnTe is a narrow gap semiconductor with a band gap 0.19 eV, while LiF is a typical insulator. The observed secondary-electron yield for LiF is about five times larger than that for SnTe. At normal incidence case, it is known that secondary-electron yield for insulator is enhanced, sometimes by one order of magnitude, compared to metals or semiconductors [10–14]. This is attributed to a large mean free path due to a large band gap and also a large escape probability due to a lower surface barrier. The present enhancement for LiF, however, cannot be ascribed to these effects, because the electrons observed here were produced outside the crystal. The observed enhancement indicates that the electron excitation process itself is enhanced in front of the LiF(001) surface compared to SnTe(001), although the electron excitation process in insulators has usually been assumed to be suppressed due to a large band gap [11,14]. In order to see the origin of the enhancement of the secondary-electron-production process, more detailed information is extracted from the observed results.

Introducing a position-dependent secondary-electron production rate \( P(x) \), i.e., the number of secondary electrons produced by a proton per unit path length at a distance \( x \) from the surface, the secondary-electron yield is given by integrating \( P(x) \) along the proton trajectory
\[
\gamma(\theta_i) = \int_{\text{traj}} P(x) \, dz,
\]  
(1)

Fig. 2. Temperature dependence of the secondary-electron yield from LiF(001) induced by specularly reflected 0.5 MeV proton. Different experimental symbols represent results for different crystals. A typical experimental error is shown.

Fig. 3. Secondary-electron yield and energy loss at specular reflection of 0.5 MeV protons from LiF(001). Results for SnTe(001) are also shown for comparison [5].
where the trajectory lies in the $x$–$z$ plane. Using the surface continuum potential $V(x)$ the equation of the trajectory is written as

$$\left( \frac{dx}{dz} \right)^2 = \frac{V(x_m(\theta_i)) - V(x)}{E}, \quad (2)$$

where $x_m(\theta_i)$ is the closest approach distance to the surface and $E$ the proton energy. Substituting Eq. (2), Eq. (1) can be written as

$$\gamma(\theta_i) = 2\sqrt{E} \int_{x_m(\theta_i)}^{\infty} \frac{P(x)}{\sqrt{V(x_m(\theta_i)) - V(x)}} \, dx. \quad (3)$$

This is an integral equation of the Abel type and the solution is given by

$$P(x) = -\frac{1}{2\pi E} \frac{dV(x)}{dx} \left\{ \gamma(0) \sqrt{\frac{E}{V(x)}} \right. \right.$$ 

$$+ \int_0^{\pi/2} \frac{d\gamma(\theta_i)}{d\theta_i} \bigg|_{\theta_i = \sqrt{\frac{E}{V(x)} \sin (\alpha)}} \, d\alpha \bigg\}. \quad (4)$$

Using Eq. (4), the production rate can be derived from the observed secondary-electron yield. The position-dependent stopping power $S(x)$ can be derived from the observed energy loss $\Delta E(\theta_i)$ using an equation similar to Eq. (4).

Fig. 4 shows the obtained position-dependent secondary-electron production rates for 0.5 MeV protons on LiF(001) and SnTe(001) together with the position-dependent stopping powers. Both the production rate and the stopping power decreases with increasing distance from the surface. Although the secondary-electron production rate is usually assumed to be proportional to the stopping power in phenomenological theories [2], the observed production rate is not proportional to the stopping power for both LiF and SnTe. This is because the dominant process of the secondary-electron production depends on the distance $x$ from the surface. While the surface-plasmon-assisted process is dominant at larger $x$, bulk plasmon process and single electron excitation process are dominant at small $x$. The corresponding energy losses for single electron emission are different for these processes.

The ratio of the stopping power to the secondary-electron production rate $S(x)/P(x)$, i.e., the corresponding energy loss for single electron emission, is also shown in Fig. 4. The ratio decreases with increasing $x$ and approaches to constant values of 36 and 70 eV/electron at $x > 2$ Å for LiF and SnTe, respectively. In this large $x$ region, surface-plasmon-assisted process is exclusively dominant for the secondary-electron production. The surface plasmon can decay either via electron-hole pair production or via photon emission. At an ideal smooth surface, decay of the surface plasmon into a photon is prohibited because the laws of energy and momentum conservation are not satisfied at the same time. Real surfaces, however, are not perfect and the conservation law of the transverse momentum is violated, which allows the decay of surface plasmons to photons [7]. Thus the conversion probability of surface plasmons to secondary electrons depends on the surface conditions.

Using surface plasmon energies, 18 and 11 eV for LiF [15,16] and SnTe, respectively, and as-
assuming that half of the electrons produced by surface plasmon decay are ejected into the vacuum with the other half impinging into the solid, the conversion probabilities of the surface plasmons into electron–hole pairs are estimated to be ~100% and ~30% for LiF and SnTe, respectively, indicating that the LiF surface is much smoother than SnTe. This is consistent with observations performed using atomic force microscopy. While a number of pyramidal hillocks were observed on the SnTe(001), wide terraces (several hundred nm wide) separated by atomic-height steps were observed on the LiF(001). The large enhancement of the secondary-electron production process for LiF(001) at large ℓ is thus ascribed to the large conversion probability of the surface plasmon into an electron-hole pair. It should be noted that even at smaller ℓ the secondary-electron production process is enhanced for LiF compared to SnTe. This might be explained by the negative electron affinity of LiF which makes single electron excitation very efficient for the production of excited electrons over the vacuum level.

4. Conclusions

We have measured secondary-electron yield induced by 0.5 MeV protons specularly reflected from LiF(001). The position-dependent secondary-electron production rate is derived from the observed yield as a function of the distance ℓ from the surface. The probability of surface plasmon decay to an electron-hole pair is estimated to be ~100%. The obtained complete conversion efficiency is ascribed to the flatness of the LiF(001) in atomic level.

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References