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Surface Plasmon Excitation at Topmost Surface in Reflection High-Energy Positron Diffraction

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To investigate the excitation process of surface plasmon by fast positrons, we measured the positron energy loss spectra from Si(111)-7 × 7, Al(111)-1 × 1, and Bi(001)-1 × 1 surfaces under the total reflection condition using an energy-filtered reflection high-energy positron diffraction. We observed the multiple surface plasmon excitations, which can be represented by the Poisson distribution with the mean excitation number of 2.4-2.8. The energy loss spectrum is nearly independent of the glancing angle of the incident positron beam. We also measured the electron energy loss spectra at similar glancing angles and determined the mean excitation number to be 1.4-1.8. We found that due to the total reflection positrons are able to excite more surface plasmons as compared to electrons.
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Keywords: Surface plasmon; Reflection high-energy positron diffraction (RHEPD); Total reflection

I. INTRODUCTION

When fast electrons pass through a thin metal film, the energy is lost efficiently by the plasmon excitations [1]. In the case of grazing incidences at surfaces, the surface plasmon excitation is also an important energy loss process. Lucas and Šunjić showed theoretically the mean excitation number (n_s) of surface plasmon by electrons as a function of the glancing angle as

$$n_s = \frac{e^2}{8\epsilon_0 h v \sin\theta}, \quad (1)$$

where v and θ are the velocity and the glancing angle of electrons, respectively, and e, ϵ_0, and h are the charge of electrons, the static dielectric constant, and the Planck constant, respectively [2]. In 1995, Horio et al. measured the electron energy loss spectra from a Si(111)-7×7 surface using an energy-filtered reflection high-energy electron diffraction (RHEED) [3, 4]. They demonstrated that the glancing angle dependence of the mean excitation number of surface plasmon can be explained by Eq. (1) [4]. Recently, the energy loss process in the RHEED has been extensively investigated by many researchers [4–6]. However, the energy loss process of positrons at surfaces still remains unresolved.

Since positrons have a positive charge, crystals act as repulsive potentials for positrons. Therefore, when positron beam is incident on the crystal surface at small enough glancing angles, the total reflection takes place [7, 8]. Under the total reflection condition, the penetration depth of positrons into the crystal is sufficiently suppressed (∼ 2 Å). Thus, the positron diffraction intensity is very sensitive to the structure of the first surface layer. We developed the reflection high-energy positron diffraction (RHEPD) for detailed study of surface structures and properties [9, 10].

Due to the total reflection, the energy loss process for positrons is expected to be different from that for electrons. To investigate the energy loss process of totally reflected positrons at surfaces, we developed an energy-filtered RHEPD apparatus [11]. In this study, we measured the energy loss spectra by positrons from the Si(111)-7 × 7, Al(111)-1 × 1, and Bi(001)-1 × 1 surfaces. We will show the enhanced surface plasmon excitation by positrons under the total reflection condition.

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II. EXPERIMENTAL PROCEDURE

Positron energy loss spectra were measured in a ultra-high vacuum (UHV) chamber equipped with a positron source of ^{22}Na (370 MBq) with a base pressure of less than 6×10^{-8} Pa [12]. Electron energy loss spectra were measured using a conventional electron gun. The incident energies (E) were 10 keV in both cases. To measure the energy loss spectra for the diffracted spots, the energy analyzer consisting of two mesh electrodes was installed in the chamber. The energy resolution of the analyzer was 4.6 eV. All the measurements were conducted at room temperature.

The substrates (15 × 5 × 0.5 mm^3) were cut from a mirror-polished n-type Si(111) wafer with a resistivity of 1-10 Ωcm. They were heated at 400°C in several hours and flashed at 1200°C a few times in the UHV chamber to produce a well-ordered 7 × 7 structure. By depositing 1/3 monolayers (ML) of Al atoms onto the Si(111)-7 × 7 surface at 670°C, a $\sqrt{3} \times \sqrt{3}$-Al structure was formed (1 ML corresponds to the atomic density of 7.83×10^{14} cm^{-2}). Subsequently, 3 ML of Al atoms were deposited onto the Si(111)-$\sqrt{3} \times \sqrt{3}$-Al surface at 350°C. Eventually, surface periodicity transformed to a 1×1 [13]. Well-ordered Bi(001)-1 × 1 surfaces were prepared by depositing 8 bilayer (BL) of Bi atoms onto the Si(111)-7×7 surface at room temperature, followed by the annealing at 100°C [14] (1 BL corresponds to the atomic density of 1.14×10^{15} cm^{-2}). Since the mean inner potentials of Si and Al are 12 eV [15], the critical angle of the total reflection for $E = 10$ keV is estimated to be 2.0° via the Snell's equation [7]. In the case of Bi, the critical angle of the total reflection condition is evaluated to be 2.6° because of the mean inner potential of 20 eV [16].

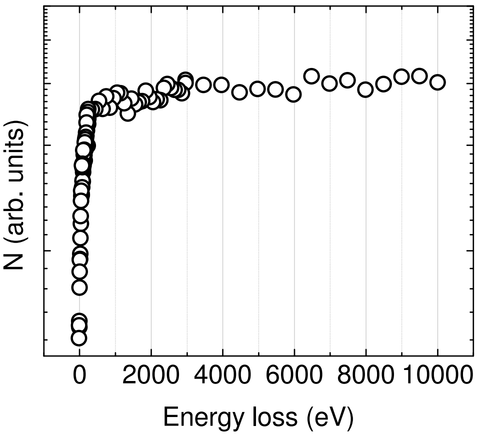

FIG. 1: Intensity (N) of the specular spot for positrons from the Si(111)-7 × 7 surface as a function of the energy loss. The glancing angle of the incident positron beam is set at 1.0° under the total reflection condition. The incident azimuth corresponds to the [11$\bar{2}$] direction.

III. RESULTS AND DISCUSSION

A. Surface plasmon excitations by positrons under total reflection condition

Figure 1 shows the specular spot intensity of positrons from the Si(111)-7×7 surface in the energy loss range from 0 to 10 keV. The glancing angle of the incident positron beam was 1.0° under the total reflection condition. Except for the energy loss around 0 eV, the intensity is nearly constant against the energy loss. The intensity steeply decreases when the energy loss approaches 0 eV. That is, the totally reflected positrons from the Si(111)-7 × 7 surface mainly lose the energy of 0-100 eV. This result indicates that the major energy loss process of the totally reflected positrons is due to the surface plasmon excitations.

To highlight the change of the intensity in the low energy loss region, we measured the positron energy loss spectrum from the Si(111)-7 × 7 surface with a finer energy step as shown in Fig. 2(a). The intensity decreases sequentially with decreasing the energy loss. Figure 2(b) displays the differential spectrum. Five distinct energy loss peaks are seen. The averaged interval of the loss peak positions corresponds to 11 eV. Thus, these peaks are responsible for the surface plasmon excitations of Si. The loss peak intensity of two-fold surface plasmon excitation ($2\hbar\omega_s$) is the highest among the observed loss peaks. The elastic (no-loss) peak intensity is relatively small as compared to the loss peak intensities.

According to the previous study of electron energy loss spectra [2], the intensity distribution of the loss peaks due to the surface plasmon excitations is given by the Poisson distribution

$$P(n) = \frac{n_s^n}{n!}\exp(-n_s). \quad (2)$$

Using this equation, we determined the mean excitation number of surface plasmon for totally reflected positrons. The mean excitation number (2.4±0.3) of surface plasmon by positrons is nearly independent of the glancing angle,

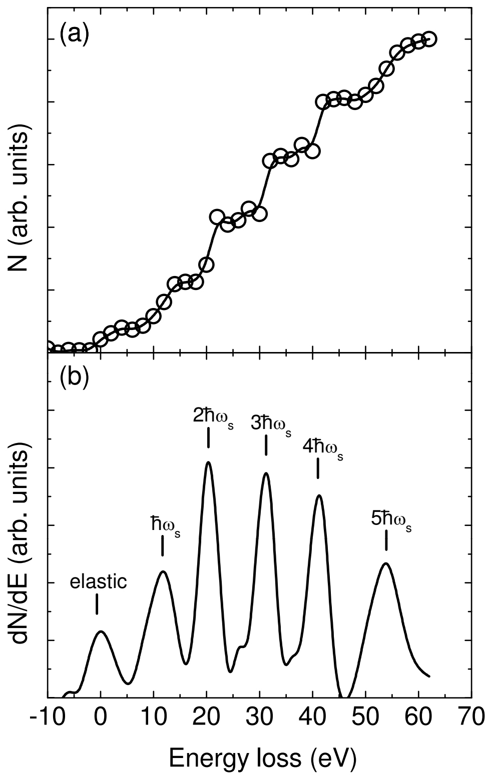

FIG. 2: (a) Intensity (N) and (b) differential intensity (dN/dE) of the specular spot for positrons from the Si(111)-7 × 7 surfaces as a function of the energy loss. The glancing angle of the positron beam is 1.5°, which satisfies the total reflection condition. The incident azimuth corresponds to the [11$\bar{2}$] direction. The curve in (b) was obtained by differentiating the smoothed data using the adjacent averaging.

as shown in Fig. 3. This is in contrast to the case of electrons where the mean excitation number of surface plasmon is proportional to 1/sinθ [2]. Under the total reflection condition, the penetration depth of positrons is very limited (∼ 2 Å) and hence most positrons travel in the first surface layer. Therefore, the mean excitation number of surface plasmon by totally reflected positrons is nearly independent of the glancing angle.

B. Comparison of energy loss spectra between positron and electron

Figures 4(a) and 5(a) show the energy loss spectra of specular spots by positrons from the Al(111)-1 × 1 and Bi(001)-1 × 1 surfaces as a function of the energy loss, respectively. The glancing angles of the incident positron beam were 1.0° for the Al(111)-1 × 1 surface and 2.0° for the Bi(001)-1 × 1 surface. In both spectra, five energy loss peaks are observed. In the case of Al(111)-1 × 1 surface, the intervals of the loss peak positions are not constant. Using the electron energy loss spectroscopy, the energy loss peak due to the surface plasmons of Al was clearly observed from the Al(111)-1 × 1 surface fabricated by the above-mentioned procedure [13]. Thus, the deviation of the intervals is not considered to be due to the interface of Al(111)/Si(111). Although the loss peak positions are

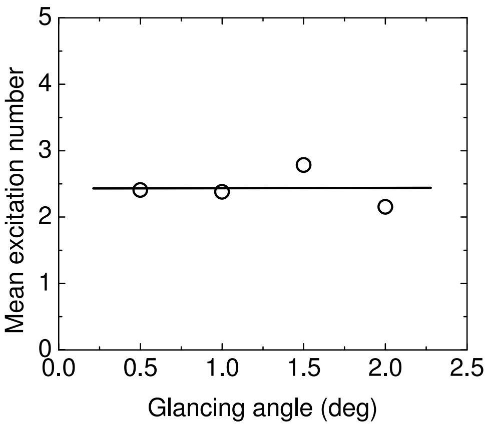

FIG. 3: Mean excitation number of surface plasmon by positrons as a function of the glancing angle under the total reflection condition.

slightly fluctuated due to the energy resolution, the averaged interval of the peak positions is estimated to be 12 eV for the Al(111)-1×1 surface and 11 eV for the Bi(001)-1 × 1 surface. Since the surface plasmon energy of Al is 11.3 eV, these loss peaks can be assigned to a sequence of the surface plasmon losses. Similarly, the loss peaks in Fig. 5(a) are also assigned to the surface plasmon losses of Bi because of the surface plasmon energy of 10 eV. In both cases, the loss peak intensity due to two- or three-fold surface plasmon excitations is higher than the other loss peak intensities.

Figures 4(b) and 5(b) represent the energy loss spectra of specular spots by electrons from the Al(111)-1 × 1 and Bi(001)-1 × 1 surfaces, respectively. The glancing angles of the incident electron beam were 1.5° for the Al(111)-1 × 1 surface and 1.3° for the Bi(001)-1 × 1 surface. The peak positions in Figs. 4(b) and 5(b) are almost the same as those by positrons in Figs. 4(a) and 5(a), respectively. However, the distributions of the peak intensities are considerably different from those by positrons. In both cases, the peak intensity due to no-loss or single surface plasmon excitation is higher than the other loss peak intensities.

Using Eq. (2), the mean excitation numbers of surface plasmon by positrons are estimated to be 2.8 ± 0.3 for the Al(111)-1 × 1 surface and 2.4 ± 0.2 for the Bi(001)-1 × 1 surface. These values are compatible with that for the Si(111)-7 × 7 surface, as described before [11]. Similarly, the mean excitation numbers by electrons are determined to be 1.8 ± 0.3 for the Al(111)-1 × 1 surface and 1.4 ± 0.4 for the Bi(001)-1×1 surface. These values are also close to that for the Si(111)-7×7 surface [4–6, 11]. We found that the mean excitation number by positrons is approximately twice as large as that by electrons. Thus, positrons under the total reflection condition excite more surface plasmons as compared to electrons.

The mean excitation number of surface plasmon is given by

$$n_s = \frac{t}{l}, \qquad (3)$$

where l and t denote the inelastic mean free path of positrons or electrons due to the surface plasmon excitation and the nominal interaction length with the crystal surface, respectively. In the high-energy region, the mean free path is theoretically the same for positrons and electrons [17]. The inelastic mean free paths and the nominal interaction lengths calculated using the so-called TPP-2M formula developed by Tanuma et al. [18] and Eq. (3) are summarized in Table I. In any cases, the interaction length (∼ 480 Å for the averaged value) of positrons is much longer than that of electrons (∼ 290 Å for the averaged value). The difference of the interaction lengths between positrons and electrons is ∼ 190 Å.

Both of positrons and electrons approaching the surface from the vacuum region can excite the surface plasmon. The trajectories of the incoming and outgoing beams are considered to be identical for positrons and electrons. Thus, since the excitation processes by positrons and elec-

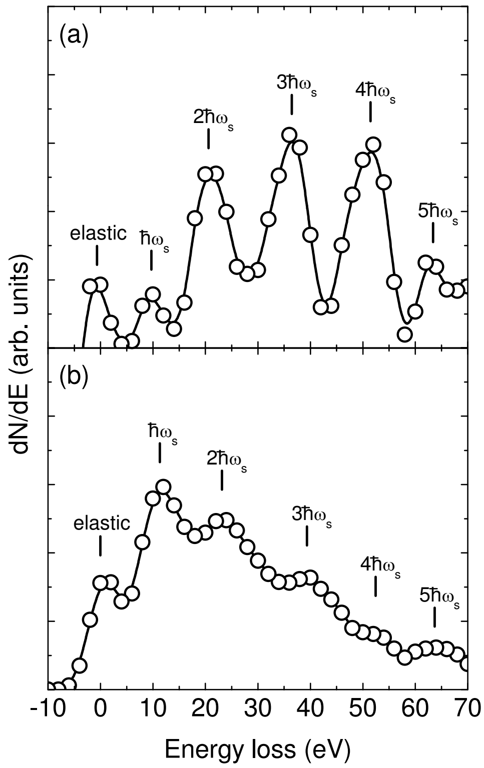

FIG. 4: Energy loss spectra (dN/dE) (a) by positrons and (b) by electrons from the Al(111)-1×1 surfaces as a function of the energy loss. The glancing angle of the positron beam is 1.0°, which satisfies the total reflection condition. The glancing angle of the electron beam is 1.5°. The incident azimuths of the positron and electron beams correspond to the [11$\bar{2}$] direction and 7.5° away from the [11$\bar{2}$] direction, respectively.

TABLE I: Inelastic mean free paths of positrons and electrons due to the surface plasmon excitation and nominal interaction lengths with the Si(111)-7×7, Al(111)-1×1, and Bi(001)-1×1 surfaces.

	Positron		Electron	
	l (Å)	t (Å)	l (Å)	t (Å)
Si(111)	180	460	180	250
Al(111)	190	530	190	340
Bi(001)	190	460	190	270

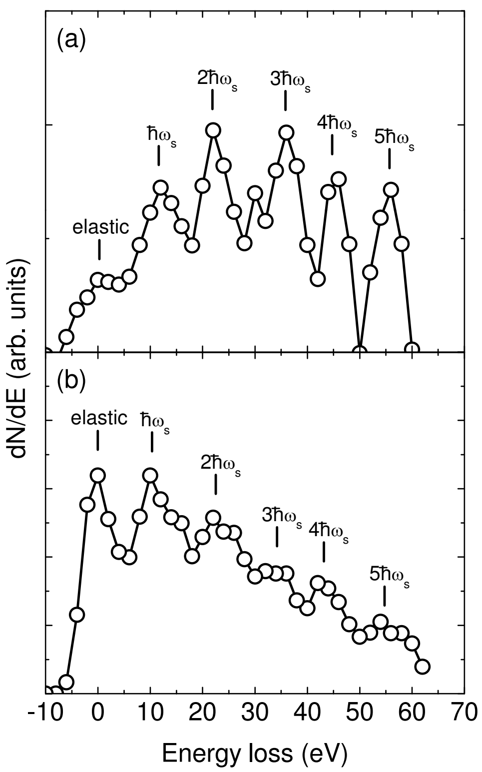

FIG. 5: Energy loss spectra (dN/dE) (a) by positrons and (b) by electrons from the Bi(001)-1 × 1 surfaces as a function of the energy loss. The glancing angle of the positron beam is 2.0° under the total reflection condition. The glancing angle of the electron beam is 1.3°. The incident azimuths of the positron and electron beams correspond to 7.5° away from the [11$\bar{2}$] direction.

trons approaching the surface from the vacuum region are considered to be identical, the interaction lengths by positrons and electrons should be also the same. Positrons channel the first surface layer. The channeling length is the same order of magnitude as the elastic mean free path [19]. Such positrons are able to excite the surface plasmon. On the other hand, electrons penetrate the first surface layer. Such electrons hardly excite the surface plasmon. Therefore, the difference (190 Å) of the interaction lengths between positrons and electrons can be explained by the channeling length of positrons in the first surface layer.

IV. SUMMARY

We investigated the excitation process of surface plasmon at the Si(111)-7 × 7, Al(111)-1 × 1, and Bi(001)-1 × 1 surfaces by fast positrons and electrons. We found that the mean excitation number of surface plasmon by positrons is approximately twice as large as that by electrons. Owing to the existence of the total reflection, the surface plasmons are much excited by positrons as compared with electrons. Furthermore, we found that the element dependence of the plasmon excitation process is not significant from the comparison between the Si(111)-7 × 7, Al(111)-1 × 1, and Bi(001)-1 × 1 surfaces.

Acknowledgments

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