Surface plasmon excitation at metal surfaces studied by reflection high-energy positron diffraction

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Surface plasmon excitation at metal surfaces studied by reflection high-energy positron diffraction

Y Fukaya, A Kawasuso and A Ichimiya

Advanced Science Research Center, Japan Atomic Energy Agency, 1233 Watanuki, Takasaki, Gunma 370-1292, Japan

fukaya.yuki99@jaea.go.jp

Abstract. We investigated the excitation process of surface plasmon at metal surfaces by fast positrons using an energy-filtered reflection high-energy positron diffraction. We measured the positron and electron energy loss spectra for the specular spots from Al(111)-1×1 and Bi(001)-1×1 surfaces at grazing angles. 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. The element dependence of the surface plasmon excitation process is not significant from the comparison between the Al(111)-1×1 and Bi(001)-1×1 surfaces.

1. Introduction

When high-energy electrons are incident on crystal surfaces, various energy loss processes such as phonon, plasmon, and core-level electron excitations occur. Among the excitations, the surface plasmon excitation is known as a dominant process at grazing incidence [1]. The surface plasmon is a collective excitation of electrons at crystal surfaces. Its energy is reduced by a factor of $\sqrt{2}$ as compared with the volume plasmon [2]. Generally, the excitation process of surface plasmon has been studied by using (low-) energy electron loss spectroscopy (EELS). Recently, the energy loss process of electrons due to surface plasmon excitations has been extensively investigated through an energy-filtered reflection high-energy electron diffraction (RHEED) [3-6]. On the other hand, the energy loss process of positrons at surfaces has not been well-studied so far.

Reflection high-energy positron diffraction (RHEPD) is a surface-sensitive tool for analyzing the surface structures owing to the occurrence of the total reflection [7-10]. Since positrons have a positive charge, opposite to electrons, they are totally reflected at the first surface layer when the glancing angle is small enough. In the total reflection region, the incident positron beam does not penetrate the bulk. This is different from the case of electrons. Therefore, under the total reflection condition, the energy loss process by positrons at surfaces is expected to be different from that by electrons.

Very recently, we fabricated an energy-filtered RHEPD to measure the energy loss spectrum from a Si(111)-7×7 surface [11]. We found that under the total reflection condition the mean excitation number of surface plasmon by positrons is larger than that by electrons [11]. In this study, to investigate the excitation process of surface plasmon by positrons at metal surfaces, we measured the positron energy loss spectra from Al(111)-1×1 and Bi(001)-1×1 surfaces under the total reflection condition. To compare the excitation process by electrons, we also measured the electron energy loss spectra from the Al(111)-1×1 and Bi(001)-1×1 surfaces.

2. Experimental procedure

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 introduced into a ultra-high vacuum (UHV) chamber with a base pressure less than 6×10^{-8} Pa. They were annealed at 400 °C and flashed at 1200 °C in a few seconds several times to produce a 7×7 reconstruction. 1/3 monolayer (ML) of Al atoms were deposited on the Si(111)-7×7 surface at 670 °C using a crucible to produce a √3×√3-Al structure (1 ML corresponds to 7.83×10^{14} cm^{-2}). To prepare Al(111)-1×1 surfaces, 3 ML of Al atoms were deposited on the √3×√3-Al structure at 350 °C [12]. Since the mean inner potential (V_0) of Al crystals is 12 eV [13], the critical angle of the total reflection for the incident positron energy (E) of 10 keV is estimated to be 2.0° via Snell's equation [7]. Bi(001) surfaces were prepared by the deposition of 8 BL Bi atoms on the Si(111)-7×7 surface at room temperature (1 BL corresponds to 1.14×10^{15} cm^{-2}). Subsequently, they were annealed at about 100 °C to produce well-ordered Bi(001)-1×1 surfaces [14]. In the case of Bi, the critical angle of the total reflection for E = 10 keV and V_0 = 20 eV [15] is estimated to be 2.6°.

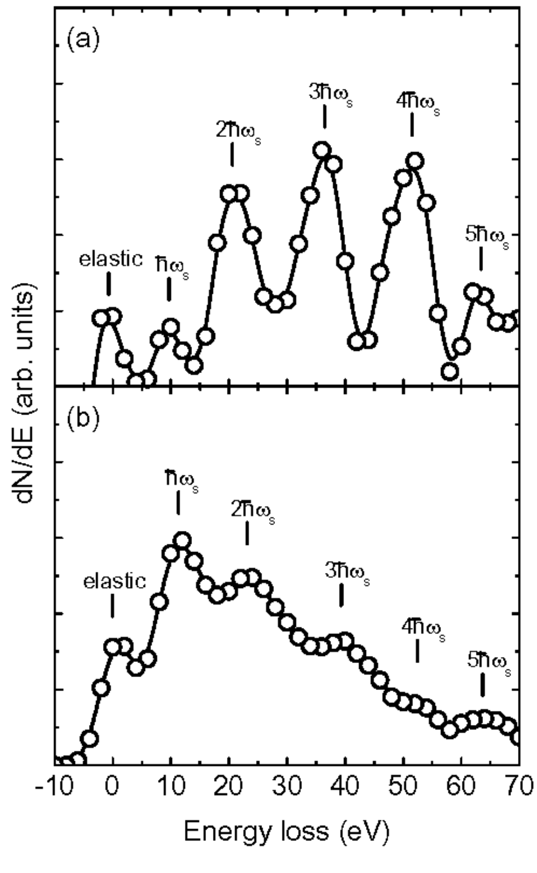

Figure 1. Energy loss spectra of the specular spots for (a) positrons and (b) electrons from the Al(111)-1×1 surfaces. The glancing angle of the positron beam is set at 1.0° (total reflection condition). The glancing angle of the electron beam is 1.5°. The incident azimuths of the positron and electron beams correspond to the $[11\bar{2}]$ direction and 7.5° away from the $[11\bar{2}]$ direction, respectively.

The positron beam was generated with a ^{22}Na positron source (370 MBq) and electrostatic lenses. The details of the apparatus were described elsewhere [11,16]. Electron energy loss spectra were measured using a conventional electron gun. The incident energies were 10 keV in both cases. The retarding-field-type energy analyzer was installed into the UHV chamber to measure the energy loss spectra. The energy resolution of the analyzer was estimated to be 4.6 eV. All the measurements were conducted at room temperature.

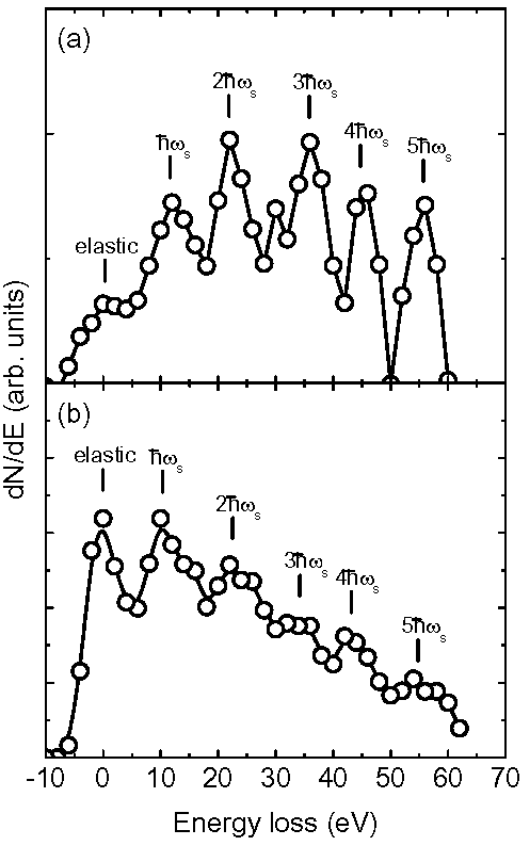

Figure 2. Energy loss spectra of the specular spots for (a) positrons and (b) electrons from the Bi(001)-1×1 surfaces. The glancing angle of the positron beam is set at 2.0° (total reflection condition). The glancing angle of the electron beam is 1.3°. The incident azimuths of the positron and electron beams correspond to 7.5° away from the $[11\bar{2}]$ direction.

3. Results and discussion

3.1. Positron energy loss spectrum

Figures 1(a) and 2(a) show the energy loss spectra (dN/dE) of the specular spots for positrons from the Al(111)-1×1 and Bi(001)-1×1 surfaces, respectively. The energy loss means the difference between the incident beam and retarding energies. The glancing angles of the incident positron beams for the Al(111)-1×1 and Bi(001)-1×1 surfaces are 1.0° and 2.0°, respectively, which satisfy the total reflection condition. The incident azimuths of the positron beams for the Al(111)-1×1 and Bi(001)-1×1 surfaces correspond to the $[11\bar{2}]$ direction and 7.5° away from the $[11\bar{2}]$ direction [17], respectively. In this energy loss region, five prominent loss peaks are observed in both spectra. For the Al(111)-1×1 surface, the intervals of the loss peak positions are not constant. According to the previous study using the electron energy loss spectroscopy, the energy loss peaks due to the surface plasmons of Al were observed from thin Al(111) films fabricated on the Si(111) surface [12]. Thus, the deviation of the intervals is not considered to be due to the effect of Al(111)/Si(111) interfaces. Although the loss peak positions are 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 volume plasmon energy for Al crystals is 16 eV, the surface plasmon energy is estimated to be approximately 11 eV from the relation $\hbar\omega_s = \hbar\omega_v/\sqrt{2}$, where $\hbar\omega_s$ and $\hbar\omega_v$ are the surface and volume plasmon energies, respectively. In a similar way, the surface plasmon energy for Bi is estimated to be 10 eV. Thus, these peaks in figures 1(a) and 2(a) are assigned to a sequence of the surface plasmon losses. In both cases, the peak intensity of two- or three-fold surface plasmon losses is larger than the others. These results indicate that the totally reflected positron multiply excites surface plasmons.

3.2. Electron energy loss spectrum

Figures 1(b) and 2(b) show the energy loss spectra (dN/dE) of the specular spots for electrons from the Al(111)-1×1 and Bi(001)-1×1 surfaces, respectively. The glancing angles of the incident electron beams for the Al(111)-1×1 and Bi(001)-1×1 surfaces are set at 1.5° and 1.3°, respectively. The incident azimuth corresponds to 7.5° away from the $[11\bar{2}]$ direction [17]. Similar to the case of positrons, five loss peaks are observed in figures 1(b) and 2(b). These peaks in the energy loss spectra by electrons can be also assigned to a series of the loss peaks due to the surface plasmon excitations of Al and Bi. In the case of electrons, on the contrary, the peak intensity resulting from the elastic or single surface plasmon excitation ($\hbar\omega_s$) is higher than the other loss peaks. These features are in contrast to those for positrons. Therefore, multiple surface plasmon excitations by electrons are suppressed as compared with positrons.

3.3. Excitation number of surface plasmon

The intensity distributions in the energy loss spectrum due to the surface plasmon excitations can be well expressed by the Poisson distribution. From the analysis of the loss peak intensities in figures 1(a) and 2(a), we determined the mean excitation numbers as 2.8 for the Al(111)-1×1 surface and 2.4 for the Bi(001)-1×1 surface, as listed in Table 1. These values are close to that from the Si(111)-7×7 surface [11]. Therefore, it is considered that under the total reflection condition, the excitation processes of surface plasmon for positrons from the Al(111)-1×1 and Bi(001)-1×1 surfaces are nearly the same as the Si(111)-7×7 surface. Similar to it, the mean excitation numbers of surface plasmon for electrons are determined to be 1.8 for the Al(111)-1×1 surface and 1.4 for the Bi(001)-1×1 surface. The values are compatible to the previous study on the Si(111)-7×7 surface [11]. In a similar way of positrons, the excitation process of surface plasmon by electrons does not almost depend on the elements consisting of the surface structures.

Table 1. Mean excitation numbers of surface plasmon by positrons and electrons.

	Positron	**Electron**
Al(111)	2.8	1.8
Bi(001)	2.4	1.4
Si(111) [a]	2.6	1.4

[a] Ref. [11].

The mean excitation number (n_s) of surface plasmon is given in the form

$$n_s = t/l\,, \tag{1}$$

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 paths of positrons and electrons are nearly the same [18]. In the case of the Al(111)-1×1 surface, the mean free path is estimated to be 190 Å using the so-called TPP-2M formula [19] and $\hbar\omega_s$ = 11 eV. Thus, the nominal interaction length of positrons with the Al(111)-1×1 surface is determined to be 530 Å. Similar to it, the interaction length of electrons is determined to be 340 Å. In the case of the Bi(001)-1×1 surface, the mean free paths of positrons and electrons are evaluated to be 190 Å. Thus, the nominal interaction lengths of positrons and electrons for the Bi(001)-1×1 surface are determined as 460 Å and 270 Å, respectively. In both cases, the difference of the nominal interaction lengths between positrons and electrons is 190 Å. When positrons and electrons approach the surface from the vacuum region, both of them can excite the surface plasmon. Thus, the nominal interaction lengths of positrons and electrons are the same because the excitation process of surface plasmon is similar to each other. Positrons under the total reflection condition channel the first surface layer. On the other hand, electrons penetrate the first surface layer. Positrons channeling in the first surface layer excite the surface plasmon. However, electrons penetrating the first surface layer hardly excite the surface plasmon. Therefore, the difference of the interaction lengths between positrons and electrons can be explained by the channeling length of positrons in the first surface layer.

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