Inelastic scattering processes in reflection high-energy positron diffraction from a Si(111)-7×7 surface

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To analyze the inelastic processes of totally reflected fast positrons at solid surfaces, we measured the absolute reflectivity, specular beam profile, and energy-loss spectrum from a Si(111)-7×7 surface during the reflection high-energy positron diffraction. The absolute reflectivity of positrons was more than 1 order of magnitude greater than that of electrons; however, it was well below 100% even under the total-reflection condition. The specular beam profile of positrons exhibited a broad feature. The energy-loss spectrum showed that more surface plasmons are excited by positrons as compared to electrons. The degraded absolute reflectivity and the broadening of specular beam profile are explained by the multiple surface-plasmon excitations by positrons.

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Elastic and inelastic scattering processes of low-energy positrons at solid surfaces are markedly different from those of electrons due to the absence of exchange interaction and the presence of Coulomb repulsion from ion cores and no restriction from the Pauli's exclusion principle.[1] Consequently, low-energy positron diffraction (LEPD) has advantages in surface-structure analysis as compared to low-energy electron diffraction.[2] For fast positrons (>1 keV), the crystal potential may be simply represented by the Hartree-Fock potential because the image potential arising from the correlation interaction is negligible. When the surface normal energy is less than the crystal potential, positrons are totally reflected at the first surface layer.[3,4] Owing to this property, reflection high-energy positron diffraction (RHEPD) is an excellent method for surface analysis.

Similar to electrons, phonon and electronic excitations may be important inelastic processes for fast positrons. Under the total-reflection condition, fast positrons will excite phonons and electrons solely at the first surface layer. The previous RHEPD study showed that the surface Debye temperature is significantly less than that anticipated from electron diffraction.[5] As for electronic excitations, the collective (plasmon) excitation may be important in addition to single-electron (both core and valence electrons) excitations.[6,7] Reflection high-energy electron diffraction studies demonstrated the occurrence of multiple surface-plasmon excitations.[8–11] Through the observation of surface-plasmon excitation and its dispersion relationship, the surface electronic state concerning the metal-insulator transition and the formation of one-dimensional metals could be studied.

In this study, we investigated the inelastic processes of fast positrons. For this purpose, we measured the absolute reflectivity, specular beam profile, and energy-loss spectrum from a Si(111)-7×7 surface in RHEPD experiments and compared the results with the case of electrons.

Samples ($15\times5\times0.5$ mm^3) were cut from a mirror-polished n-type Si(111) wafer with a resisitivity of 10 Ω cm. After degassing at 400 °C, the samples were heated at 1200 °C in a ultrahigh vacuum (UHV) with a base pressure of less than 6×10^{-8} Pa to produce the 7×7 reconstructed surface. A positron beam with an energy of 10 keV was generated using a ^{22}Na source and electromagnetic lenses. The details of the apparatus have been described elsewhere.[12,13] An electron beam with an energy of 10 keV was generated by a conventional electron gun. The positron or electron beam was irradiated onto the sample surfaces at small glancing angles and the specular beams were observed using a multichannel plate assembly with a phosphor plane. The azimuthal angle was parallel to the $[11\bar{2}]$ direction or 7.5° away from the $[11\bar{2}]$ direction. In the latter condition, the specular beam is predominant because of the suppression of the simultaneous reflections parallel to the surface (one-beam condition).[14] The absolute reflectivity was determined from the ratio between the incoming and the specular beam intensities. An energy analyzer consisting of two mesh electrodes was installed to measure the energy-loss spectrum of the diffraction spots. The energy resolution of the analyzer was 4.6 eV. This is sufficient to observe both the surface plasmon (11.5 eV) and bulk plasmon (16.5 eV) of Si. To observe the broadening of the specular beam profile, the profile was deconvoluted by the incoming beam profile.

Figure 1 shows the absolute reflectivities of positrons and electrons from the Si(111)-7×7 surface under the one-beam condition as a function of glancing angle. The upper horizontal axis denotes the surface normal energy of the beam, i.e., $E_\perp = E\sin^2\theta$. The reflectivity of electrons is only less than 4% in all the observed angles. In contrast, the reflectivity of positrons is more than 20% in the total-reflection region ($\theta < 2.0°$). It tends to reach nearly 100% as the glancing angle approaches zero. In the case of electrons, the penetration depth is more than 10 Å because of the negative crystal potential. Many electrons are therefore multiply scattered inside crystals and are lost without contributing to the specular beam. Contrarily, when the surface normal energy of positrons is less than the crystal potential energy of Si ($E_\perp < 12$ eV), the positrons are totally reflected at the surface with negligible penetration into the bulk. The reflectivity of positrons is therefore markedly higher than that of electrons.

Solving the simple Schrödinger equation with an ideal slab potential, the reflectivity of positrons should be 100% in the total-reflection region as represented by the solid line in Fig. 1. However, the measured reflectivity of positrons is

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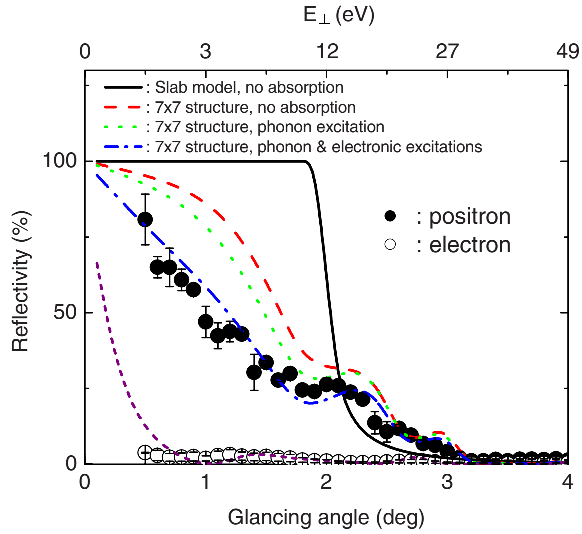

FIG. 1. (Color online) Absolute reflectivities of positrons (closed circles) and electrons (open circles) from the Si(111)-7 $\times 7$ surface as a function of the glancing angle. The lines represent the reflectivities of positrons calculated with the ideal slab potential (solid), the 7×7 structure (broken), the 7×7 structure and the phonon absorption potential (dotted), and the 7×7 structure and the phonon plus electronic absorption potentials (dash-dotted). The short-dashed line indicates the reflectivity of electrons calculated with the 7×7 structure and all the absorption potentials. The upper horizontal axis denotes the surface normal energy of the incoming beam.

far-below 100% even in the total-reflection region. One reason may be the effect of the surface structure on the reflectivity. The broken line in Fig. 1 represents the positron reflectivity calculated on the basis of the dynamical-diffraction theory[15] with the 7×7 structure.[16] However, the measured positron reflectivity is not reproduced by considering only the surface structure. This means that the loss of positrons through the inelastic processes, such as the phonon and electronic (both single-electron and plasmon) excitations should be taken into account. The dotted and dash-dotted lines in Fig. 1 represent the reflectivity of positrons calculated with the absorption potentials of the phonon excitation (V_{ph} $=0.71$ eV) (Ref. 5) and the phonon plus electronic (V_{el} $=1.76$ eV) excitations, respectively. The reliability factor between the measured and calculated (dash-dotted) curves is 2.9%. The measured reflectivity is not reproduced unless all the excitation processes are considered. Although the reflectivity of electrons is also reproduced by the calculation considering all the absorption potentials,[14] the effect of surface excitations is clearly observed in the reflectivity of positrons. The absorption potentials are listed in Table I. As for the phonon excitation, the absorption potential for positrons is significantly greater than that for electrons. This is because positrons are mainly reflected at the first surface layer where the thermal vibration amplitudes of atoms are considerably greater than those in the bulk. For the electronic excitations, the absorption potential for positrons appears to be greater than that for electrons. Plasmons are excited through the Coulomb interaction between charged particles and the electron cloud of media. Thus, the absorption potential of the plasmon excitation should be theoretically identical for positrons and electrons.[17] The cross section of single-electron excitations of positrons are generally greater than those of electrons since incident positrons and excited electrons are distinguishable, while incident electrons and excited electrons are not. The difference between positrons and electrons in the valence-electron excitation may be considered only when the incident energy is less than the Fermi energy.[18] The further study is required to elucidate the difference of absorption potentials of electronic excitations for positrons and electrons.

TABLE I. Absorption potentials due to the phonon (V_{ph}) and electronic (V_{el}) excitations, which are used in the calculations of the reflectivities for positrons and electrons from the Si(111)-7×7 surface. The mean excitation numbers of surface plasmon are also listed.

	Positron	Electron
V_{ph} (V)	0.71	0.20
V_{el} (V)	1.76	1.30
n_s	2.6	1.4

Figure 2(a) shows the specular beam profile of positrons under the one-beam condition at $\theta=2.0°$. The broader tails at $k_{\parallel}>0.1$ and $k_{\parallel}<-0.1$ Å^{-1} are observed in addition to a relatively narrow central component. The broadening of the specular beam profile is also probably due to the surface excitations. To reveal the effect of surface excitations in detail, we measured the energy-loss spectra of positrons and electrons as follows.

Figure 3(a) shows the intensities of the specular beams for positrons and electrons as a function of lost energy (E_{loss}) of 0–70 eV. In this energy region, the plasmon excitation is predominant. The difference between positrons and electrons is clearly observed. The intensity of electrons increases steeply from $E_{loss}=0$ eV and nearly continuously increases with E_{loss}. However, the intensity of positrons increases gradually from $E_{loss}=0$ eV and exhibits distinct steps. Figure 3(b) shows the differential curve of the energy-loss spectrum of positrons as a function of lost energy. The elastic peak intensity is rather weak. Five prominent loss peaks are observable. Having the surface-plasmon energy of Si (11 eV), these peaks are assigned to single ($\hbar\omega$) through fivefold ($5\hbar\omega$) surface-plasmon losses, respectively. The peak intensities of $E_{loss}=2\hbar\omega$ and $E_{loss}=3\hbar\omega$ are higher than the others. The positron energy-loss spectra were also measured at various glancing angles in the total-reflection region (0.5–2.0°). Although the theoretical study suggests that more surface plasmons are excited with decreasing glancing angle,[19] there are only small differences, i.e., generally the peak intensity of 2 or $3\hbar\omega$ is stronger than the others. Figure 3(c) shows the differential curve of the electron energy-loss spectrum. In the electron energy-loss spectrum, the peak intensity of E_{loss} $=\hbar\omega$ is the highest. The elastic peak intensity is also relatively high. Such features are consistent with those obtained in the previous studies[8–11] but differ from the positron energy-loss spectrum in Fig. 3(b).

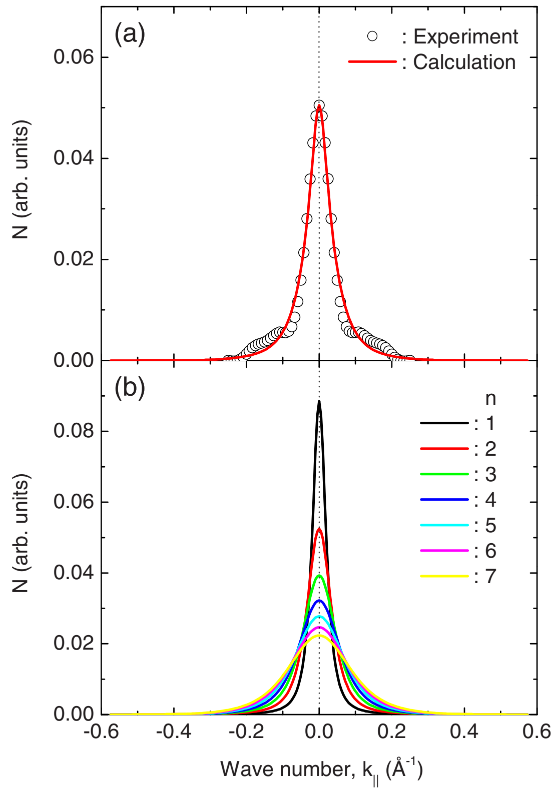

FIG. 2. (Color online) (a) Specular beam profile of positrons that is deconvoluted by the incoming beam profile from the Si(111)-7×7 surface at the glancing angle of 2.0°. The solid line denotes the profile calculated with Eq. (2) and n_s=2.6. (b) Calculated specular beam profile for various excitation numbers of the surface plasmon. The horizontal axis is transformed from the angle ($\theta_{\parallel}$) to the wave number ($k_{\parallel}$) using the relationship $k_{\parallel}=K \sin \theta_{\parallel}$, where K is the incident wave number.

The above-mentioned energy-loss spectra are approximated by the Poisson distribution $P(n)=n_s^n \exp(-n_s)/n!$, where n_s is the mean excitation number of surface plasmon.[19] By fitting this equation to the energy-loss spectra in Figs. 3(b) and 3(c), we obtained n_s=2.6 for positrons and 1.4 for electrons. The latter number agrees with the numbers obtained in the previous studies.[8–11] Thus, positrons excite surface plasmons efficiently as compared to electrons.

When a single plasmon is excited, the specular beam profile in cone angles is expressed by

$$I_1(\theta_{\parallel},\theta_{\perp}) \propto \sqrt{(\theta_{\parallel}/\theta_E)^2+1}/\{(\theta_{\parallel}/\theta_E)^2+(\theta_{\perp}/\theta_E)^2+1]^2\}, \tag{1}$$

where $\theta_E=\hbar\omega/2E$ (E: incident beam energy) and $\theta_{\parallel}$ and $\theta_{\perp}$ denote the cone angles from the beam center to the azimuthal and glancing angle directions, respectively.[11,20] For a double plasmon excitation, the beam profile becomes a self-convolution of this profile, i.e., $I_2(\theta_{\parallel},\theta_{\perp})=I_1(\theta_{\parallel},\theta_{\perp}) \otimes I_1(\theta_{\parallel},\theta_{\perp})$. Similarly, for the n-fold plasmon excitation, the expression is $I_n(\theta_{\parallel},\theta_{\perp})=I_{n-1}(\theta_{\parallel},\theta_{\perp})\otimes I_1(\theta_{\parallel},\theta_{\perp})$. The wave numbers are given by $k_{\parallel}=K \sin \theta_{\parallel}$ and $k_{\perp}=K \sin \theta_{\perp}$, where K is the wave number of the incident beam. The overall beam profile is given by

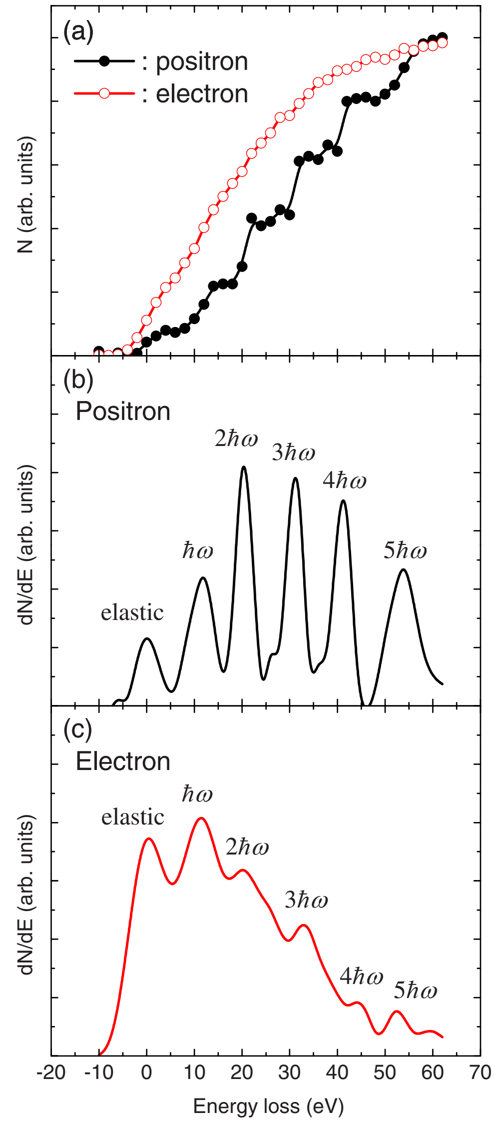

FIG. 3. (Color online) (a) Intensities of the specular beams for positrons (closed circles) and electrons (open circles), (b) the differential curve for positrons, and (c) that for electrons from the Si(111)-7×7 surface as a function of energy loss. The glancing angles are 1.5° for positrons and 1.3° for electrons. The incident azimuths are parallel to the $[11\bar{2}]$ direction for positrons and 7.5°-off oriented from the $[11\bar{2}]$ direction for electrons. The curves in (b) and (c) were obtained by the spline interpolation of the differential curves.

$$I(\theta_{\parallel},\theta_{\perp}) = \sum_{n=1}^{\infty} I_n(\theta_{\parallel},\theta_{\perp})P(n). \tag{2}$$

Here, we focus on the $\theta_{\parallel}$ component because the beam profile is more broadened in this direction. Figure 2(b) shows the beam profile calculated by Eq. (1). The beam profile is broadened with the increasing excitation number. The solid line in Fig. 2(a) represents the specular beam profile calculated by Eq. (2) with n_s=2.6 obtained for positrons. The experimental beam profile is reproduced considering the excitation of surface plasmons.

The mean excitation number of the surface plasmon is related to the nominal interaction length (t) and the mean free path (l); i.e., $n_s = t/l$. The mean free path is given by $l = 2a_0 E/\hbar\omega \ln(\theta_C/\theta_E)$, where $\theta_C = \hbar\omega k_F/(2E_F K)$; k_F and E_F are the Fermi wave number and the Fermi energy, respectively, of valence electrons. The mean free path is identical for positrons and electrons theoretically.[17] Having $k_F = 0.90$ Å^{-1}, $E_F = 3.12$ eV, $K = 51.2$ Å^{-1}, $\hbar\omega = 11$ eV, and $E = 10$ keV we obtain $l = 238$ Å. Consequently, we obtain $t = 620$ Å for positrons and $t = 334$ Å for electrons. The interaction length for positrons is approximately twice of that for electrons. Considering the fact that positrons do not penetrate the bulk region, the long interaction length implies that positrons are channeled in the first surface layer and they excite surface plasmons. Less screening of the Coulomb interaction between positrons and the surface valence electrons may also enhance the surface-plasmon excitation. Penetration of electrons into the bulk suppresses the nominal interaction length and hence the surface-plasmon excitation.

In conclusion, by using the Si(111)-7×7 reconstructed surface, we demonstrated that the absolute reflectivity of fast positrons at the grazing incidence is markedly higher than that of electrons owing to the effect of total reflection, while various inelastic scattering processes degrade it. The specular beam profile was also found to be broadened. The investigation of energy-loss spectra for both positrons and electrons revealed that positrons excite more surface plasmons as compared to electrons.

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