Reflection High Energy Positron Diffraction from a Si(111) Surface

A. Kawasuso and S. Okada
Japan Atomic Energy Research Institute, Takasaki Establishment, 1233 Watanuki, Takasaki, 370-12, Japan
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This report details the first observation of reflection high energy positron diffraction. A 20 keV positron beam was irradiated onto a hydrogen-terminated Si(111) surface from the $[1\bar{1}0]$ and $[11\bar{2}]$ directions. The zeroth Laue zone consisting of a specular and diffraction spots with a shadow edge was clearly observed using a microchannel plate at relatively high glancing angles ($\theta > 3.0°$). The rocking curve of the specular spot showed the primary Bragg peak and the total reflection of positrons. [S0031-9007(98)07178-6]

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In this paper, we report the first observation of reflection high energy positron diffraction (RHEPD) using a Si(111) surface terminated with hydrogen atoms. Previously predicted specific characteristics of RHEPD experiment, such as the total reflection and the primary Bragg peak [1,2], were experimentally shown in this research.

Since the first observation of low energy positron diffraction (LEPD) by the Brandeis research group [3], several advantages of LEPD over low energy electron diffraction (LEED) have been shown [4–6]. For example, the intensity versus energy (I-V) plot in the LEPD experiment complements the LEED experiment. This is due to the Coulomb repulsion from ion cores and the absence of exchange interaction for positrons in solids. Because of the high inelastic scattering cross section of positrons, the multiple scattering is fairly reduced, and hence I-V data can be analyzed in terms of the kinematic theory.

Reflection high energy electron diffraction (RHEED) is an established tool in the surface study today. This method is useful for *in situ* monitoring of the surface state under crystal growth and heat treatment taking advantage of the space available above the specimen. By replacing electrons with positrons, RHEPD experiment is possible. Oliva first proposed that total reflection of high energy positrons ($\sim$30 keV) may be observed in RHEPD experiment [1]. Ichimiya also suggested the appearance of the primary Bragg peak and total reflection from the simple kinematic description as follows [2]. The Bragg condition is given by $E_0 \sin^2\theta = 37.5n^2/d^2 + eV_0$ (n: integer), where E_0 is the energy of incoming particles, θ is the glancing angle, d is the distance between lattice planes, and eV_0 is the inner potential felt by the incoming particles. The difference of the signs of the inner potentials for electrons and positrons plays a critical role in the diffraction behavior. In the case of a Si(111) surface ($d = 3.14$ Å), eV_0 is -12 eV for electrons, and hence $E_0 \sin^2\theta$ should be negative for $n = 1$. That is, the primary Bragg reflection could not be observed in a RHEED experiment. From Snell's law, the glancing angle θ and the Bragg angle are correlated by $\cos\theta_B/\cos\theta = (E_0/E)^{1/2}$, where E is the energy of incoming particles in crystal. Incident electrons are accelerated due to the negativity of eV_0 so that the Bragg angle is greater than the glancing angle; i.e., the refractive index is greater than unity. This makes electrons penetrate into crystal with a steeper angle than the glancing angle. On the contrary, for positrons, the primary Bragg reflection could be observed since eV_0 is positive. The appearance of the primary Bragg reflection for positrons causes more intense diffraction spots than in the case of electron diffraction. Incident positrons are decelerated; i.e., the refractive index should be less than unity. This implies a possibility that positrons are totally reflected below a critical angle θ_C, where θ_C is given by $\sin\theta_C = (eV_0/E_0)^{1/2}$. Thus, it is expected that positrons have a tendency to survey a shallower layer than electrons.

Using the total reflection of positrons the surface Debye temperature could be determined in the RHEPD experiment without any disturbance from the bulk of the specimen [2]. It was also suggested that total reflection provides a direct measurement of the surface dipole potential [1]. It is expected that RHEPD holds great potential to reveal detailed kinetics of surface-related solid phenomena such as the surface melting. Despite several trials [7], clear RHEPD patterns have not yet been obtained. This report describes how the RHEPD experiment was successfully accomplished during this research.

The specimen used in this study was a commercial Si(111) wafer ($15 \times 20 \times 0.5$ mm^3) oriented to the $[11\bar{2}]$ and the $[1\bar{1}0]$ directions. It was important to choose the surface which was proof against oxidation and contamination during RHEPD experiment (more than a few hours). The specimen surface was therefore terminated with hydrogen atoms as follows. After the degreasing treatment, the specimen surface was oxidized in a dry oxygen ambient at 1100 °C for 25 min. About 1000 Å oxide layer was grown in this procedure. The oxide layer was removed by a HF (1%) solution, and subsequently the specimen was rinsed in flowing ultrapure water for 5 min. Then, the specimen was immediately transported into a vacuum chamber (8×10^{-7} Pa). It is established that the Si(111) surface was terminated with hydrogen atoms by the above procedure. However, not only monohydrides but also dihydrides and trihydrides may coexist on the surface. The (111) terraces terminated by hydrogen atoms may also be produced [8–10]. We therefore carried out the infrared absorption measurement. From the observation of the local vibrational mode of atomic hydrogen at 2084 cm^{-1}, it was confirmed that monohydride phase was predominant. White positrons emitted from a ^{22}Na source (~185 MBq) were first moderated by a tungsten single crystal with a thickness of 5000 Å, and then thermalized positrons were accelerated to 20 keV in an electrostatic positron beam line which will be described elsewhere. The primary beam diameter was about 3 mm with an angular divergence of less than 1°. To obtain a clear diffraction pattern, it was critical to make a fine and highly parallel beam. Then, the beam was collimated by a 60-mm-length slit with a 1-mm-diam hole. The final beam intensity was approximately 5000 positrons/sec. The RHEPD experiment was carried out by irradiating the beam onto the specimen from the $[1\bar{1}0]$ and $[11\bar{2}]$ directions. Reflected positrons were monitored by a Hamamatsu F2226 microchannel plate assembly (MCPA) with a phosphor plane. The image was observed by a charge-coupled device camera in a dark room and digitally accumulated more than 4 h using a personal computer. The specimen and the MCPA were separated by 170 mm. The beam exit, the specimen, and the MCPA entrance were all electrically grounded. The glancing angle was changed by an electrostatic deflector and a mechanical rotator with a precision of ±0.25°. We also performed the RHEED experiment using a 15 keV electron beam.

Figure 1 shows RHEED patterns for the $[1\bar{1}0]$ and $[11\bar{2}]$ incidences. For the $[1\bar{1}0]$ incidence, the specular (00) with (01) and $(0\bar{1})$ diffraction spots are seen. Although the (02) and $(0\bar{2})$ spots are not clearly observed, weak streak patterns exist outside of the (01) and $(0\bar{1})$ spots. For the $[11\bar{2}]$ incidence, the zeroth Laue zone composed of the specular, (11), and $(\bar{1}\bar{1})$ diffraction spots and also the first Laue zone with (01) and $(\bar{1}0)$ spots are observed. No fractional-order spots appear for the present specimen. This suggests that the hydrogen terminated Si(111) surface closely resembles an unrelaxed 1×1 structure [11].

Now we show the results of the RHEPD experiment. Figure 2 shows the RHEPD patterns for the $[1\bar{1}0]$ and $[11\bar{2}]$ incidences at the glancing angles of 4.5° and 4.0°, respectively. These results exhibit the zeroth Laue zone with direct spots and the shadow edges. The zeroth Laue zone is composed of the specular and diffraction spots. For the $[1\bar{1}0]$ incidence, the observed diffraction spots are assigned to be (01), $(0\bar{1})$, (02), and $(0\bar{2})$ as shown in the picture. Similarly, the diffraction spots for the $[11\bar{2}]$ incidence are assigned to be (11) and

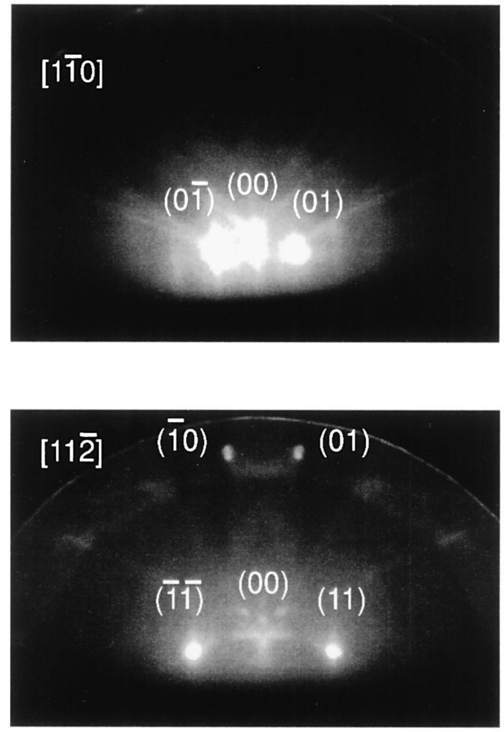

FIG. 1. RHEED patterns from a hydrogen-terminated Si(111) surface for the $[1\bar{1}0]$ and $[11\bar{2}]$ incidences.

$(\bar{1}\bar{1})$. The diffraction angles of (01) and (02) spots measured from the specular spot for the $[1\bar{1}0]$ incidence are 1.4° and 2.8°, respectively. Similarly, the angle of the (11) spot for the $[11\bar{2}]$ incidence are determined to be 2.6°. These correlate well to the estimated values with the lattice constant (3.84 Å) for the Si(111) surface [12]. Thus, the observed diffraction spots seem to be approximately consistent with those for the RHEED experiments suggesting that RHEPD gives comparable information to RHEED. However, several extra spots are found in RHEPD patterns. **(**For instance, it appears at the upper side of the (01) spot for the $[1\bar{1}0]$ incidence as seen in Fig. 2.**)** Considering the fact that the surface sensitivity of RHEPD is greater than that of RHEED, such extra spots in RHEPD patterns may reflect detailed surface states such as the microscopic roughness of surface.

Figure 3 shows the RHEPD patterns for the $[1\bar{1}0]$ incidence at different glancing angles. The diffraction patterns could be observed at $\theta > 3.0°$. At high glancing angles, the intensities of the spots are found to be weakened. With decreasing the glancing angle, it is found that the specular intensity fairly increases. The blurry background in pictures comes from inelastically scattered positrons. At $\theta = 0.5°$, such a background is found to be quite weak. This implies the effect of the total reflection as shown below. The shadow edge is usually located equidistant between the direct and specular spots, but, as can be seen, it is located nearer to the specular spot. This

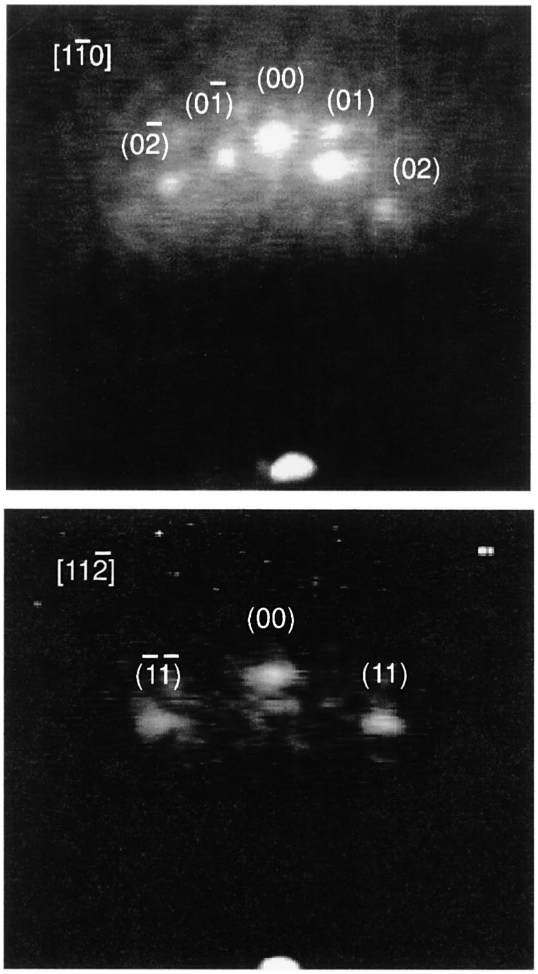

FIG. 2. RHEPD patterns from a hydrogen-terminated Si(111) surface for the [1$\bar{1}$0] and [11$\bar{2}$] incidences at glancing angles of 4.5° and 4.0°, respectively.

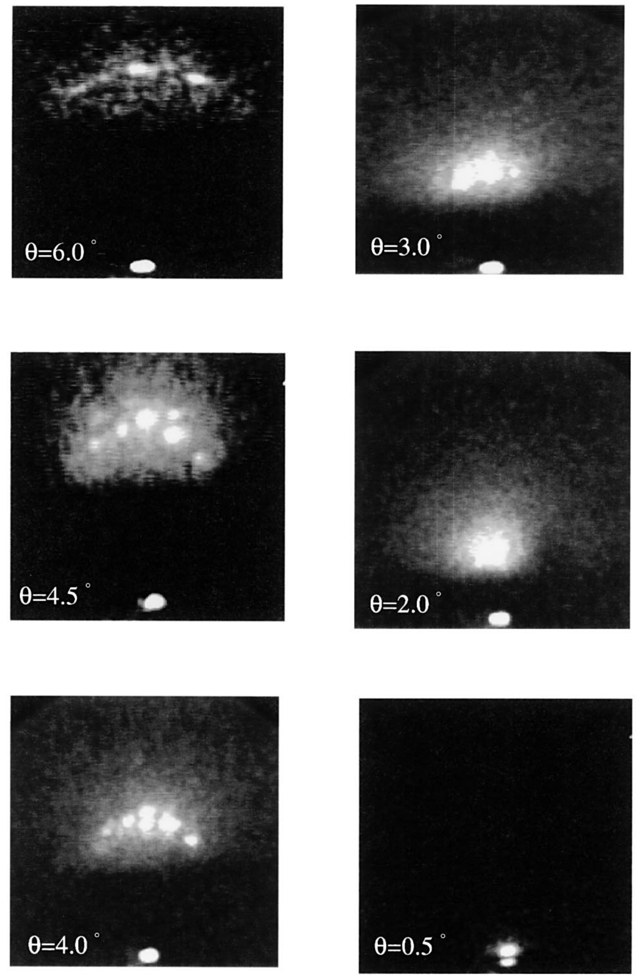

FIG. 3. RHEPD patterns for a hydrogen-terminated Si(111) surface for the [1$\bar{1}$0] incidence obtained at various glancing angles.

may be explained as the refractive index of positrons is less than unity as stated above.

Figure 4 shows the rocking curve for the specular spot (the plot of the specular intensity versus the glancing angle). The reflection intensity is quite strong at $\theta < 2.5°$, and shows a peak at $\theta = 1.5°$. According to Ichimiya's estimation the critical glancing angle for the total reflection is estimated to be $\theta_C = \arcsin(eV_0/E_0)^{1/2} = 1.4°$ with $eV_0 = 12$ eV [2]. Similarly, the first, the second, and the third Bragg peaks may appear at $\theta = 1.6°$, 2.1°, and 2.8°, respectively. Thus, the peak at $\theta = 1.5°$ and the high reflection intensity up to 1.5° may represent the primary Bragg peak and the effect of the total reflection. The small shoulder at $\theta = 2.5°$ may be due to the overlapping of the second and the third Bragg peaks. To reveal the higher order Bragg peaks, however, the angular resolution should be improved more. Thus, the existence of the total reflection region is indicated from the rocking curve. As mentioned above, at $\theta = 0.5°$, the blurry background caused by the inelastically scattered positrons is quite

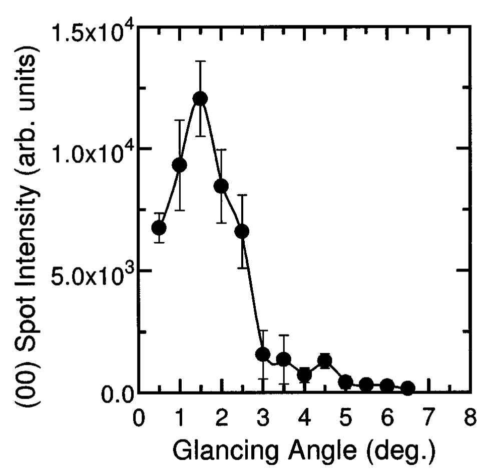

FIG. 4. Rocking curve for the specular spot in the RHEPD experiment.

reduced. This may be explained as most positrons are elastically scattered at the surface in the total reflection region. It was reported that the specular intensity for a copper (100) surface is fairly enhanced at relatively low glancing angles in the LEPD experiment [13]. This result implies the repulsion of positrons due to the surface dipole potential. However, since the inelastic scattering cross section of low energy positrons (50 to 450 eV) in the LEPD experiment is relatively high, it seems to be difficult to observe directly the reflection from the surface dipole [13]. Using total reflection of positrons in the RHEPD experiment it is expected that the surface dipole potential can be determined directly as suggested by Oliva [1]. Further study is presently in progress.

In summary, RHEPD patterns were first observed using a hydrogen-terminated Si(111) surface. The observed diffraction spots were successfully assigned as the surface has a 1×1 structure. The position of the observed shadow edge implied that the refractive index for positrons are less than unity; i.e., positrons are decelerated in the specimen. From the rocking curve for the specular spot, the effect of the total reflection and the primary Bragg peak for positrons were suggested. The results obtained in this research show the potential of RHEPD in the surface study.

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