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Total reflection high-energy positron diffraction (TRHEPD)

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Abstract. Total reflection high-energy positron diffraction (TRHEPD) promises to be an ideal technique for determining the atomic arrangement of solid surfaces. TRHEPD makes full use of the total reflection of the positron beam from a surface in reflection high-energy positron diffraction (RHEPD), the positron counterpart of reflection high-energy electron diffraction (RHEED). Owing to the development and use of a brightness-enhanced intense positron beam at KEK, it is now possible to obtain clear TRHEPD patterns. It is shown that the TRHEPD pattern from a Si(111)-7×7 reconstructed surface for the total reflection condition does not contain contributions from atoms in the bulk.

1. Introduction

In materials research, X-ray and neutron diffraction has long been the standard technique used to elucidate the bulk structure. Any interpretation of the phenomena found in bulk crystals have to be compatible with the structure determined with these techniques. This is not, however, necessary the case for the surface atomic structures.

Two types of techniques are used to investigate surface structures. One is the imaging technique. Scanning tunneling microscopy (STM) and atomic force microscopy (AFM) are the two most widely used methods. These are sensitive to the topmost surface, and probe the local structure within the area where the probe scanned. It cannot, however, get information on subsurface layers, and in general, it is difficult to distinguish different elements.

The other is the diffraction technique. Low energy electron diffraction (LEED) [1] and reflection high-energy electron diffraction (RHEED) [2] are the most commonly used techniques. As in the 3D diffractions, the symmetry of the patterns gives information on the symmetry of the surface atoms, and the intensity distribution of the spots gives the details of the positions. Unfortunately there is no way for these methods to observe only the topmost surface layers.

As positron counterparts of these electron diffraction techniques, low energy positron diffraction (LEPD) [3] and reflection high-energy positron diffraction (RHEPD)[4,5] have been developed. Here we show that total reflection high-energy positron diffraction (TRHEPD), which is nothing but a refined RHEPD with an intense, brightness-enhanced positron beam, has the promise to be an ideal technique for the surface structure analysis. TRHEPD makes full use of the total reflection of the

positron beam from a solid surface.

Though X-ray diffraction in conjunction with its total reflection is also used for the surface structure, the critical angle for X-ray is usually less than 0.2°, not easy to handle. Furthermore, though the penetration depth of X-ray into the sample in the total reflection condition is much smaller than that at a practical angle such as 2°, more than 1000 nm, it is still as large as 3 nm.

2. Total Reflection of a Positron

The positron wave function ψ outside and inside a crystal is described by

$$\left(-\frac{\hbar}{2m}\nabla^2 + \mathrm{qV}\right)\psi = E\psi, \qquad (1)$$

where potential V is 0 outside the crystal and V>0 inside in every crystal. The potential energy of a positron, qV, is positive since the charge of the positron $q = +e >0$, where e is the elemental charge. The abrupt increase in the potential energy at the crystal surface makes the positron reflected at the surface.

The work function of the positron in the same material is a different thing. The work function of the positron is related with the energy eigenvalue E of the ground state of the fully interacting positron (and the effect of the electric dipole layer in the case of a metal).

In a case where a positron beam is directed on a surface with an glancing angle smaller than a certain critical value, the beam is totally reflected [4]. This is not the case for an electron beam. This makes a positron much more sensitive to the topmost atomic layer of the surface than an electron. The wave vector of the incident positron can be decomposed into the component parallel to the surface, $k_{\parallel}$, and that perpendicular, $k_{\perp}$. It follows that its kinetic energy can be expressed with the component parallel to and perpendicular to the surface, $E_{0\parallel}$ and $E_{0\perp}$, respectively:

$$E_0 = \frac{\hbar^2 k_0^2}{2m} = \frac{\hbar^2 k_{0\parallel}^2}{2m} + \frac{\hbar^2 k_{0\perp}^2}{2m} = E_{0\parallel} + E_{0\perp}. \qquad (2)$$

For a beam incident with a glancing angle θ, the perpendicular component is expressed in terms of E_0 and θ as

$$E_{0\perp} = \frac{\hbar^2 k_{0\perp}^2}{2m} = \frac{\hbar^2 k_0^2 \sin^2\theta}{2m} = E_0 \sin^2\theta\ . \qquad (3)$$

If $E_{0\perp}$ is smaller than the average potential energy of the positron inside the crystal, eV, the beam will be totally reflected. The critical angle for the total reflection, θ_c, is expressed as

$$E_0 \sin^2\theta_c = eV \rightarrow \ \theta_c = \sin^{-1}\sqrt{eV/E_0}. \qquad (4)$$

3. Improvements in Intensity and Brightness

RHEPD was proposed by Ichimiya [4] and its usefulness was proved by Kawasuso and Okada [5] using apparatus based on a ^{22}Na positron source. The intensity of the beam was 10^3-10^4 slow- e^+/s. Unfortunately the intensity of the beam was not satisfactory for RHEPD experiments. It was not even possible to orient the sample with the positrons in real time. Yet fruitful determinations of the structures of metal-deposited surfaces of Si and Ge crystals, some of which were impossible or very difficult with other techniques, were performed [6].

Principally used technique in RHEPD is the measurement of a rocking curve of the specular reflection spot in which the intensity of the specular spot is plotted as a function of the incident glancing angle. The experimental pattern is compared with a calculated pattern for an assumed atomic arrangement consistent with the symmetry of the RHEPD pattern. The goodness of fit is expressed by

using a reliability factor, or R-factor and the assumed atomic arrangement is modified. The process is repeated until satisfactory fit is obtained.

Now the only one RHEPD station in the world has been moved to the Slow Positron Facility at KEK and connected to the intense slow positron beam there. The facility provides a high-intensity, pulsed slow-positron beam produced by using a dedicated linac (55 MeV, 600 W) [7]. The moderator is kept at high voltage (variable up to 35 kV) that the energy of the positrons is controlled easily. The long (width 1 μs) and short (width 1-10 μs) pulse modes provide 5×10^7 slow-e^+/ s and 5×10^6 slow-e^+/ s, respectively.

The one-dimensional structure of Pt-deposited Ge(001) surface, with which a number of different structure models had been proposed, was determined with the RHEPD rocking curve [8], showing that one of the proposed models was essentially correct.

Then a transmission-type brightness-enhancement unit [9, 10] was installed upstream of the RHEPD chamber. This has resulted in an enhancement of the brightness by ~5000 times over the initial magnetically guided beam in the long pulse mode, with an expense of the beam intensity reduction to 1/100 through the release into the magnetic-field free region and the annihilation in the remoderator foil. This refined beam of energy 10 keV and 5×10^5 slow e^+/s, having a beam diameter ~0.5 mm (FWHM), an energy spread ~40 meV, an angular divergence ~12 mrad, and a normalized emittance ~0.01 cm rad $eV^{1/2}$ [11], yields clear positron diffraction patterns from a crystal surface. The orientation of the sample is now performed in real time observing the phosphor screen behind MCP. A RHEPD pattern can be taken within 1 hour and a full single rocking curve for the specular spot within 3 hours.

The brightness-enhanced beam was recently used to confirmed that silicene constructed on Ag(111) surface has a buckling structure [12] unlike graphene.

4. Form RHEPD to TRHEPD

The pattern in Fig. 1 (1)-(5) show the RHEPD patterns from the Si(111)-7×7 reconstructed surface observed for the indicated glancing angles, θ, observed with the refined beam [13]. The value of the critical glancing angle for the total reflection of 10 keV positrons from this surface is 2.0°. Thus the pattern (1) corresponds to the glancing angle within the total reflection condition, i.e., TRHEPD in a narrow sense. It is to be noted that the integer order spots due to bulk diffraction, commonly seen on

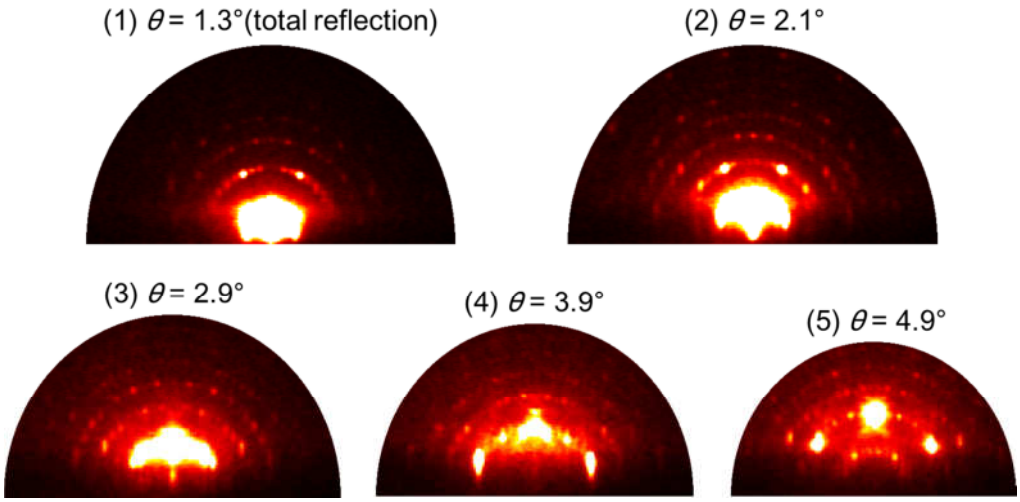


Figure 1. RHEPD patterns observed with a brightness-enhanced high-intensity positron beam.

the RHEED patterns are barely visible on the pattern, indicating that it does not have information on the bulk crystal. Another feature is that Kikuchi lines which are also commonly observed on RHEED patterns are not noticeable.

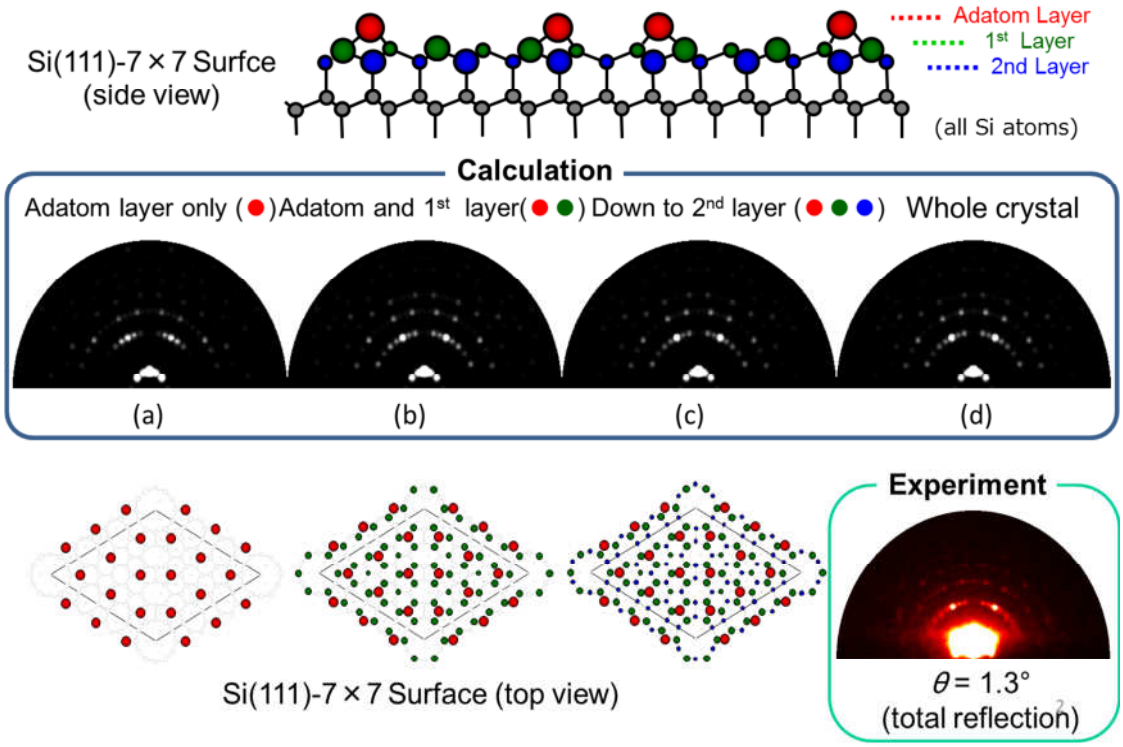
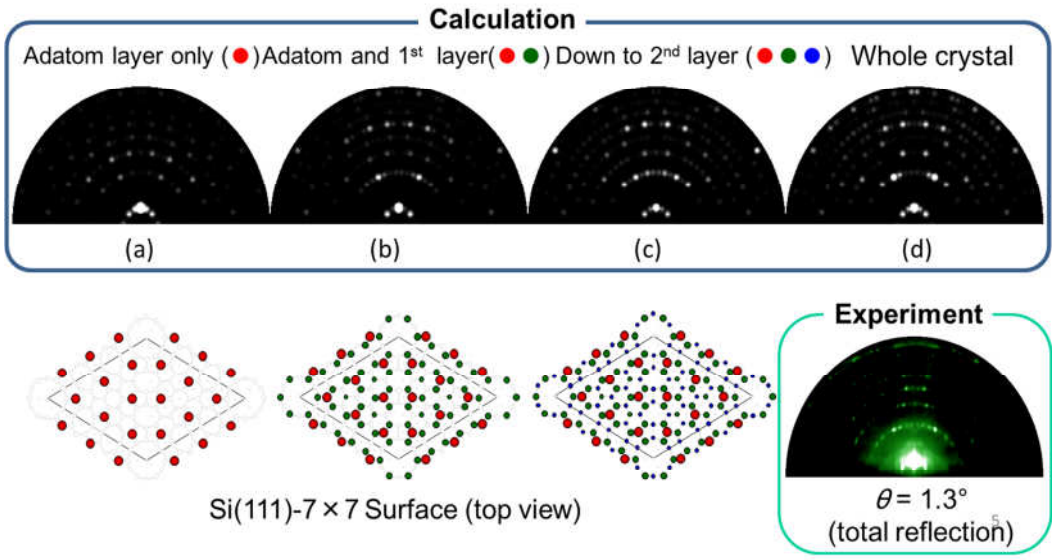


Figure 2. Experimental TRHEPD pattern for the Si(111)-7x7 reconstructed surface structure observed with the glancing angle θ =1.3° (bottom right). The patterns (a)-(c) are those calculated for the adatom sheet, adatom sheet and the first layer, and adatom, first and second layers. The pattern (d) shows that calculated for the bulk sample with the reconstructed surface. The schematic diagram at the top shows the side view of the Si(111)-7x7 structure, while those at the bottom show the top view of the sets of layers corresponding to the calculated patterns. The circles are all silicon atoms.

In order to unambiguously show that the TRHEPD pattern, Fig. 1(1), has only information about the atoms on the surface and not those in the bulk, we calculated contributions of the sheet(s) of the Si(111)-(7×7) surface atoms to the pattern. Figure 2 (a)-(d) show RHEPD patterns for a two dimensional single sheet of the adatom (atoms colored red), the adatoms and first surface layer (atoms colored green) and down to the second surface layer (atoms colored blue) and the whole crystal, respectively. Code developed by Ichimiya for the analysis of RHEED patterns based on dynamical electron diffraction theory [2, 14] was adapted to be applicable to RHEPD by changing the electric charge of the incident particle from negative to positive. Detailed positions of the atoms were taken from the ab initio calculation of this surface, i.e., the rumpling model, by Brommer et al. [15]. It is immediately noticed that the pattern (b) and (d) are in good agreement. This indicates that only the atoms exposed on the surface contribute to the pattern even a bulk sample is observed. Note that the distribution of the Si adatoms in the 7×7 super structure is rather sparse so that most of the atoms in the first surface layer (colored green) are exposed.

Figure 3 shows the experimentally observed RHEED pattern (bottom right) with the same condition as in the TRHEPD pattern (Fig. 2, bottom right) and the calculated patterns for the sheets as for Fig. 2 (a)-(d). The code developed by Inchimiya was used. It is seen that the calculated pattern (c) is appreciably different from the pattern (b) and (d), indicating that (c) contains contributuion from the atom on the layer below the second surface layer.



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In summary it has been shown that TRHEPD yields diffraction patterns separating out the contributions from the atoms exposed on the surface alone by simple measurements on a bulk sample. In addition, by analysing the RHEPD patterns across the total reflection range as shown in Fig. 1, it should be possible to determine accurately the crystal structure of the surface from the topmost layer downward. We propose to call hereafter such methods of analyse including the RHEPD data taken in total reflection condition as Total Reflection High-Energy Positron Diffraction (TRHEPD) in a broad sense.

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