The Extreme Surface Sensitivity of TRHEPD Demonstrated

This work shows that total-reflection high-energy positron diffraction (TRHEPD), the positron counterpart of reflection high-energy electron diffraction (RHEED), is extremely sensitive to the structure (atomic configuration) of the topmost surface and the immediate sub-surface. The total reflection happens when the positrons are directed to a surface with a glancing angle smaller than a critical value; the TRHEPD patterns in this condition depend only on the surface atoms. When the glancing angle is larger than the critical value, the positrons penetrate the crystal making it possible to obtain information about the immediate subsurface. These features are demonstrated using the TRHEPD patterns from a Si(111)-(7×7) surface.

Crystal structure determination by X-ray diffraction has played a crucial role in the materials research and the life sciences. For the surface science, however, an equally effective single method to determine the top- and near-surface atomic configurations has yet to be established. This work shows that total-reflection high-energy positron diffraction (TRHEPD) is the ideal technique for this purpose [1]. The method is the positron counterpart of reflection high-energy electron diffraction (RHEED) and formerly called reflection high-energy positron diffraction (RHEPD). But now it has been renamed as TRHEPD with an intention to emphasize the important role of the total-reflection in this unique method.

Many aspects of the positron diffraction are superior to those of its electron counterpart. The only possible drawback with using positrons, that is, the difficulty in obtaining a beam of high intensity, has been resolved by using the high-intensity slow positron beam at the Slow Positron Facility (SPF) of the Institute of Materials Structure Science.

First proposed in 1992 [2], TRHEPD (then called RHEPD) uses positrons with energy of around 10 keV and directs them to a crystal surface with a glancing angle smaller than about 5°. The usefulness of this technique was proven in 1998 [3], with an apparatus using a ^{22}Na positron source to provide a beam of 10^3-10^4 slow-positrons/s.

The total reflection of the positrons takes place when the positron beam is directed to a material surface with a glancing angle smaller than a certain critical value because the crystal potential in every crystal is positive. When the glancing angle is larger than the critical value, the positrons penetrate the crystal, being refracted toward the surface. In contrast, the electrons penetrate the crystal being refracted off the surface regardless of the value of the glancing angle.

Recently a TRHEPD station was developed at the SPF in collaboration with the researchers of Japan Atomic Energy Agency [4]. The intense slow positron beam of the SPF is produced using a dedicated electron linac (55 MeV, 600 W) and a production unit consisting of a Ta target and a W foil moderator. A transmission-type brightness enhancement unit was installed upstream of the TRHEPD chamber, providing a beam of 5×10^5 slow-positrons/s.

The present study shows that the TRHEPD patterns from a Si(111)-(7×7) surface in the total reflection condition actually contain contributions only from atoms on the topmost surface. The critical glancing angle for the total reflection of 10 keV positrons from Si(111)-(7×7) surface is 2.0°. Figure 1 is a schematic drawing of this surface, in which the circles indicate Si atoms. It shows (a) the arrangement of the adatoms (colored red), (b) the adatoms and the first surface layer (colored green), and (c) down to the second surface layer (colored blue). Figure 1(d) shows the side view of this surface structure.

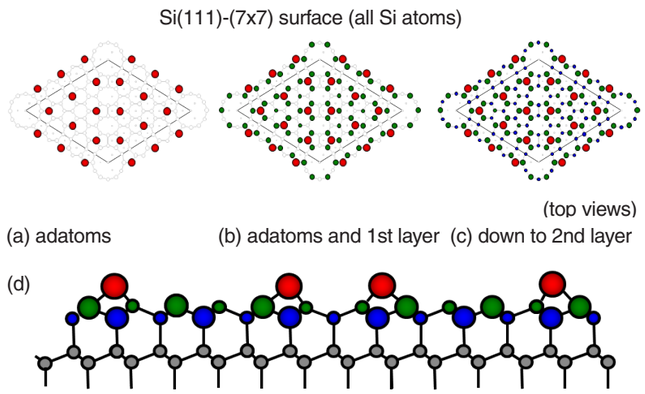

Figure 1: Schematic diagram of the Si(111)-(7×7) structure. The circles indicate silicon atoms.

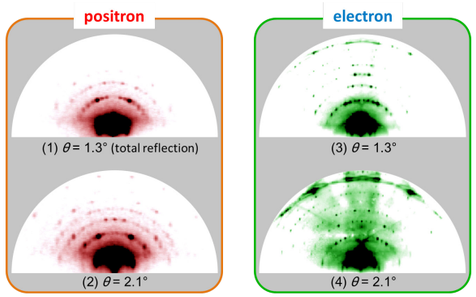

Figure 2. RHEPD (positron) and RHEED (electron) patterns of Si(111)-(7×7) surface for galncing angles θ =1.3° and 2.1°. The critical glancing angle for the total reflection of the positron is 2.0°.

Figure 2 shows the TRHEPD patterns observed for (1) a glancing angle, θ, of 1.3° (within the total reflection condition) and (2) 2.1° (slightly off the total reflection condition). The results of RHEED at the same energy and glancing angles, θ=1.3° (3) and 2.1° (4), are also shown. The TRHEPD and RHEED patterns are quite different for both angles. This is explained by the different depth of crystal penetration by the positrons and the electrons.

Figure 3(1-d) shows the calculated TRHEPD pattern for θ=1.3°. The established detailed positions of the atoms of this surface were assumed in the calculation. It can be clearly seen that the agreement of Fig. 3(1-d) with the experimental data for θ=1.3° in Fig. 2(1) is good. Figure 3(1-a) shows the TRHEPD pattern expected for a two-dimensional single sheet of the Si(111)-(7×7) adatom configuration [Fig. 1(a)]. Such a sheet cannot support itself in reality, but it is possible to assume its existence for the purpose of calculation. This calculated pattern already displays most of the features of Fig. 3(1-d) and Fig. 2(1) and strongly indicates that the main features of the latters for the whole sample are essentially determined by the contribution from the adatoms on the surface. If we look more closely at Fig. 3(1-a), (1-b), and (1-d), the inclusion of the contributions from the atoms in the first surface layer provides a better agreement. This is reasonably understood because 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 also exposed, as it can be seen in Fig. 1(b and d). Fig. 3(1-c) is indistinguishable from (1-b) and (1-d), indicating that the atoms in the second surface layer make almost no contribution.

Figure 3(2) shows similar results for a glancing angle of θ = 2.1°. This indicates that the pattern at this glancing angle includes contributions also from the second layer, but no more. In contrast, the results of the calculations for RHEED at θ = 1.3° and θ = 2.1° shown in Fig. 3(3, 4) indicate that the pattern from the bulk

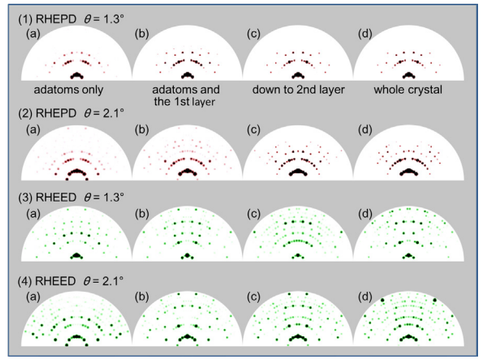

Figure 3. TRHEPD and RHEED patterns for glancing angles θ = 1.3° and θ = 2.1° calculated on the basis of the dynamical diffraction theory. The results are shown for the assumed two-dimensional crystal sheet of the Si adatoms (a), as in Fig. 1(a); the set of the sheets of adatoms and the first surface layers (b), as in Fig. 1(b); the set of sheets of layers down to the second layer (c), as in Fig. 1(c); and for the whole crystal (d).

sample does not include only the exposed atoms on the surface, nor down to the second layer surface – a feature that is expected from the positive crystal potential, which leads to negative potential energy of the electron in the bulk.

The conclusion is that the TRHEPD pattern in the total reflection condition for the Si(111)-7×7 surface *observed from a bulk sample* includes only the contributions from *the atoms exposed on the surface*. Surface science now has a method of observing a diffraction pattern formed only by the atoms on the surface by means of a straightforward measurement on a bulk sample. Moreover, analyzing TRHEPD patterns with increasing glancing angle across the critical angle for total reflection provides a powerful technique to characterize the surface and the near-surface crystal structure from the top downwards.

We expect the application of this technique to play a role as significant in surface science as the one played by the X-ray diffraction in the solid state physics and molecular biology.

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