8 Surface Structure and Electronic State

– Surface studies by using total-reflection high-energy positron diffraction (TRHEPD) and other methods –

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8-1 Introduction

Definitive knowledge of the crystal structure of materials is key to understanding their physical and chemical properties and their technological applications. Crystal structure determination by X-ray diffraction has played a crucial role in materials research and life science. For surface science, however, an equivalent method for determining the top- and near-surface atomic configurations has yet to be established. It has been shown that total-reflection high-energy positron diffraction (TRHEPD) is the ideal technique for this purpose [1], where full use is made of the total reflection of the positron beam from a solid surface in the method formerly called reflection high-energy positron diffraction (RHEPD), the positron counterpart of reflection high-energy electron diffraction (RHEED). A high-intensity positron beam whose intensity had been increased 10 times in 2010 was used to obtain clear diffraction patterns from the (111) reconstructed surface of a silicon single crystal.

8-2 Brightness enhancement of a linac-based intense positron beam for TRHEPD

The positron is the antiparticle of the electron. It has the same mass and spin as an electron, but has a positive charge equal in magnitude to that of the electron's negative charge. An experiment station for TRHEPD was installed in 2010 [2,3] at the Slow Positron Facility, High Energy Accelerator Research Organization (KEK). Taking advantage of the positron total-reflection from the surface with Bragg's condition, the TRHEPD experiments are delivering new information on topmost and immediate sub-surface atomic configurations.

A pulsed 50-Hz electron beam (55 MeV and 0.6 kW) from a dedicated linear accelerator (linac) is injected on a Ta converter and causes the creation of fast positron-electron pairs through bremsstrahlung. The fast positrons are thermalized in W foils and then emitted spontaneously from the foils with an energy of 3 eV determined by the negative work function. The moderated positrons are accelerated by an electric field to an energy of 15 keV to the grounded beam line. The positrons are guided magnetically to the TRHEPD experiment station, and released into a nonmagnetic region. Then a brightness-enhancement unit with a second moderator is effectively used to achieve a small-diameter and highly-parallel 10-keV beam with a sufficient flux.

The present system provides a final beam of almost the same quality as the previous one with a ^{22}Na-based positron beam [4] but greatly increased flux, i.e., almost the same emittance but much higher brightness. It gave a 60-fold intensified diffraction pattern from a Si(111)–(7×7) reconstructed surface compared to the previous result. An improved signal-to-noise ratio in the obtained pattern due to the intensified beam allowed observation of clear fractional-order spots in the higher Laue zones, which had not been observed previously. Several results [1,5-9] have been obtained efficiently using this system.

8-3 Demonstration of extreme surface sensitivity of TRHEPD

The silicon (111) topmost surface that was measured has a so-called reconstructed Si(111)-(7×7) DAS structure (Fig. 1), in which silicon atoms are reordered from the structure of the (111) planes inside the crystal. Total reflection of the positrons

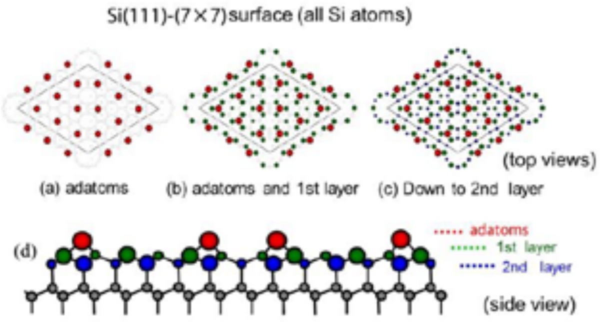

Fig. 1: Schematic diagram of Si(111)-(7×7) DAS (dimer-adatom-stacking-fault) structure. The circles are all silicon atoms.

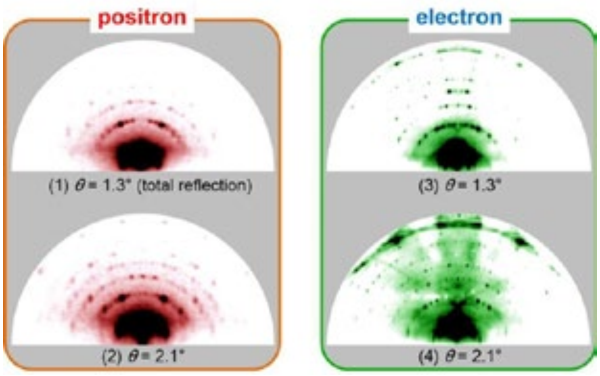

Fig. 2: Positron and electron diffraction patterns obtained for glancing angles of 1.3° and 2.1°.

takes place when incident on a material surface at 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 into the crystal, being refracted toward the surface. In contrast, electrons penetrate into the crystal being refracted off the surface regardless of the value of the glancing angle. The 10-keV positrons used in the measurements are totally reflected when incident on the surface at a glancing angle below the critical angle of 2.0°. The diffraction pattern at a glancing angle of 1.3° (Fig. 2) corresponds to the topmost atomic layer only, while that at a glancing angle of 2.1° includes reflections from the second atomic layer.

Comparisons between simulations (Fig. 3) show that the positron diffraction patterns corresponding to a glancing angle of 1.3° include information on the adatoms and the atoms at the first layer only, and that corresponding to a glancing angle of 2.1° includes information on the atoms on the second layer. Furthermore, a similar comparison for electrons shows that they definitely penetrate below the second layer even at a glancing angle of 1.3°.

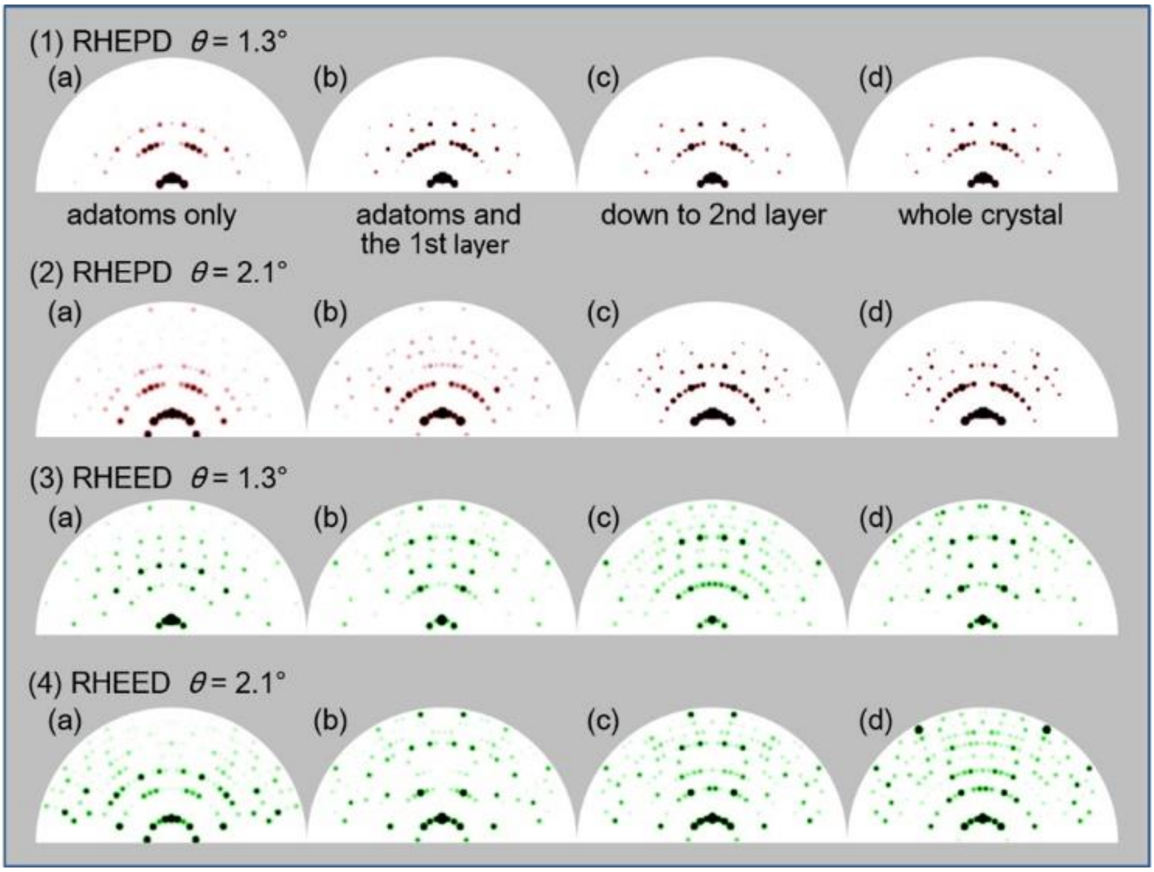

Fig. 3: Simulated diffraction patterns from the Si(111)-(7×7) surface.

The results indicate that positrons penetrate less into crystals than electrons, and that diffraction patterns only from atoms exposed at the topmost surface can be obtained by using a glancing angle below the critical angle. By measuring the diffraction patterns obtained at increasing glancing angles, it is possible to obtain data with increasing contributions from lower atomic layers. These demonstrate that TRHEPD is very sensitive and useful for highly accurate analysis of the arrangements of atoms at the topmost surface and subsurface of solids.

8-3 The uniqueness of the TRHEPD method

The above experiment shows some unique aspects of the TRHEPD method:

(1) Since electrostatic potential inside every solid is positive, a positron beam tends to be pushed out of a solid in contrast to an electron beam which tends to be pulled in.

(2) When the incidence glancing angle is smaller than a critical angle, the positrons are totally reflected.

(3) The range of the glancing angle for the total reflection overlaps well with that for the diffraction.

(4) By increasing the glancing angle across the critical angle, information on the immediate subsurface is also available and the depth of interest is adjustable.

(5) The diffraction patterns are free from the background from the deeper layers because the inelastic scatterings prevent a positron from contributing to the diffraction pattern.

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