Reflection high-energy positron diffraction: the past 15 years and the future

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Reflection high-energy positron diffraction: the past 15 years and the future

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Abstract. Reflection high-energy positron diffraction (RHEPD) is the positron counterpart of reflection high-energy electron diffraction (RHEED). Owing to the positive charge of the positron, RHEPD provides a powerful tool with which to determine the structure of the first surface layer. We have been investigating important surface systems concerning their unique electric and magnetic properties and also phase transition phenomena using positron beams (flux: 10^3~10^4 e^+/sec) with ^{22}Na sources. Currently, we are developing a new RHEPD apparatus with a bright and intense positron beam (flux: 10^5 e^+/sec) based on the LINAC at the Slow Positron Facility, KEK. Here, we summarize the past results and the future prospects of the RHEPD study in the surface science.

1. Introduction

In early 1980s, low-energy positron beam was utilized to study surface structures [1]. As the results of absence of exchange interaction for positrons and the Coulomb repulsion between positrons and ion cores, in the low energy region, the positron-solid interaction becomes much less complex as compared to the electron-solid interaction. LEPD has been applied to the structure analysis of surfaces including Cu and CdSe [2,3] and has proven to be superior to low-energy electron diffraction (LEED) in a number of important cases.

Ichimiya proposed that reflection high-energy positron diffraction (RHEPD) could also have a great advantage in the surface structure analysis [4] as compared to its electron counterpart, RHEED. Positrons are repelled from crystal surface. When a high-energy (10-20 keV) positron beam is incident on the crystal surface at grazing angle, the total reflection takes place below the critical angle. Under the total reflection region, the positron beam is diffracted at the top-most surface layer. This is a distinct feature for positron diffraction, which is never observed in electron diffractions. The critical angle for total reflection is typically larger than a few degrees.

So far, we have investigated the structures, thermal vibrations, electronic excitations and phase transitions of important surfaces by using the RHEPD. In this study, we report on some representative results obtained to date and on ongoing research using an intense positron beam generated from a LINAC at the Slow Positron Facility in KEK.

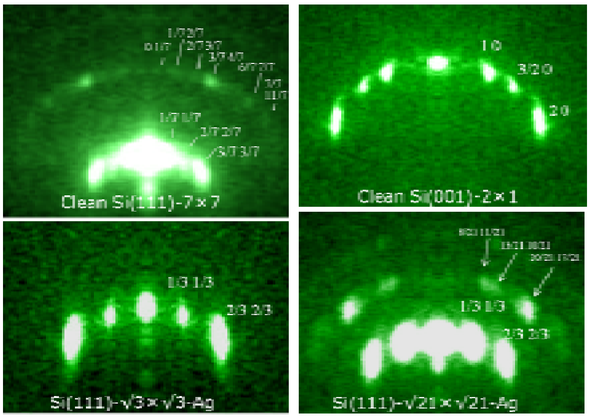

Figure 1. RHEPD patterns from various surface structures on crystals obtained by using a positron beam with a ^{22}Na source. The indexes in the pattern indicate the fractional- and integer-order spots accompanied with the surface reconstruction structures.

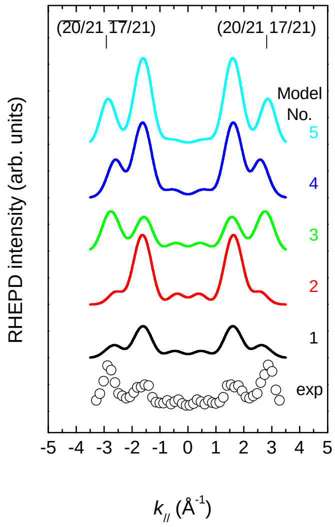

Figure 2. Intensity distribution in the 1/7th-Laue zone of the RHEPD pattern from the Si(111)-√21×√21-Ag surface as a function of wave number ($k_{//}$) parallel to the surface [17]. Open circles indicate the experimental line profile (Fig. 1). The line profiles denoted as solid lines represent the calculated ones for various structure models.

2. RHEPD study in the past 15 years

2.1. Early RHEPD study

To observe clear RHEPD patterns, it is critically important that a positron beam with a sufficient long coherence length is used. In 1998, Kawasuso and Okada developed a RHEPD apparatus employing a ^{22}Na source and electrostatic lenses, and for the first time succeeded in observing the RHEPD patterns from a Si(111)-1×1-H surface and the total reflection of positrons [5]. Afterwards, the positron beam apparatus was improved by adopting the electromagnetic lenses [6,7]. Clear RHEPD patterns from various surface structures were observed, as shown in Fig. 1.

2.2. Surface structure analysis

So far, RHEPD has been applied to various surface systems such as Si(111)-√3×√3-Ag [8], Si(111)-4×1-In [9], Ge(111)-√3×√3-Sn [10], and Ge(001)-2×1-Pt [11]. Among them, the √21×√21 superstructure is reported below as one of the complicated surface structures.

The adsorption of a small amount of the metal (Ag, Au, and Cu) atoms on the Si(111)-√3×√3-Ag surface leads to the superstructure having a √21×√21 periodicity. This surface shows a high electric conductivity as compared to the Si(111)-√3×√3-Ag surface [12]. The surface structure is still controversial although it has been studied by using various surface techniques [13-16]. The Si(111)-√21×√21-Ag and the Si(111)-√21×√21-(Ag,Au) surfaces give rise to clear fractional-order spots related to the √21×√21 periodicity (see also Fig. 1). The intensity distributions from these surfaces are very similar to each other. In the structure analysis of these surfaces, we may determine the positions of the additional metal atoms on the Si(111)-√3×√3-Ag surface. However, this work was very problematic since there exist other possible ${}_{21}C_4$ structures considering the distribution of the additional metal atoms (3-4) to available adsorption sites (21). To accomplish the above work, we followed the screening method so that the true structure should reproduce both the observed rocking curves and patterns. The solid lines in Fig. 2 represent the experimental line profile of the 1/7th-Laue zone and those calculated assuming the finally remaining structural models after the screening in the rocking curve analysis. Only one structural model (#3), where the center of three adatoms corresponds to the Si trimer, reproduces the experiment [17]. The atomic positions of the Si(111)-√21×√21 surface are lately explained in terms of cohesive nature of noble metals [18,19].

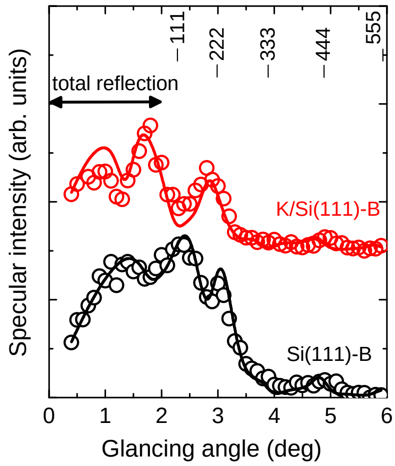

Figure 3. RHEPD rocking curves of specular spots from the Si(111)-√3×√3-B surface (black) and the K/Si(111)-√3×√3-B surfaces (red) under the one-beam condition. The circles indicate the measured curves. The solid lines denote the calculated curves using the optimum heights of K atoms.

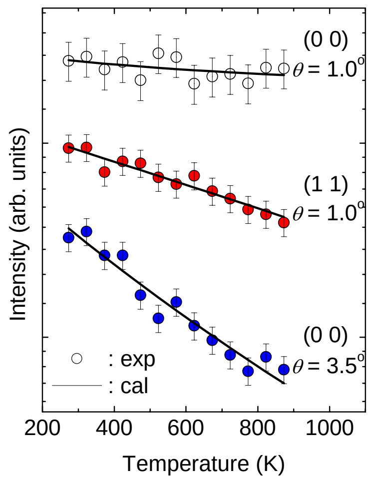

Figure 4. Temperature dependences of the RHPED intensities from a Si(111)-7×7 surface (circles). Solid lines represent the calculated intensities using Θ_S = 290 K and Θ_B = 600 K. The accelerating voltage of the incident positron beam is 20 kV.

2.3. Surface charge transfer

Recently, the RHEPD was found to be also useful for the study on the charge transfer near the surface. When a highly B-doped Si(111) surface is saturated by the K atoms, the K/Si(111)-√3×√3-B surface is formed. This surface is known as a typical example of Mott-type insulator surfaces [20]. The electronic states of the K/Si(111)-√3×√3-B surface have been extensively investigated using angle-resolved photoemission spectroscopy [21].

The red circles in Fig. 3 represent the RHEPD rocking curves of the specular spots from the K/Si(111)-√3×√3-B surfaces at room temperature under the one-beam condition. In this condition, the rocking curve is mainly sensitive to the atomic positions perpendicular to the surface. In the total reflection region, the dip structure resulting from the adsorption of K atoms is clearly observed at 1.3°. As compared with the Si(111)-√3×√3-B surface (denoted as black circles), the position of the 111 Bragg peak is shifted to lower angle. This indicates that the mean inner potential of the Si(111)-√3×√3-B surface is dramatically changed after the adsorption of K atoms. The analyses on the basis of the dynamical diffraction theory [22] demonstrated that the above result is explained considering the charge transfer of 1.0e^- per K atom to Si atoms [23].

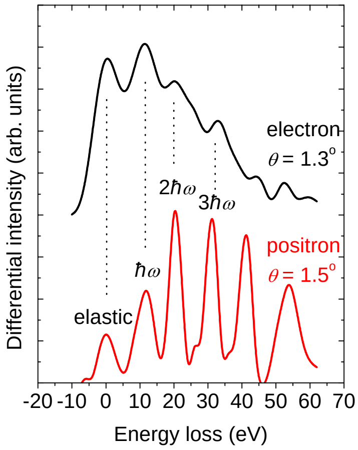

Figure 5. Differential curves for positrons (red line) and for electrons (black line) from the Si(111)-7×7 surface as a function of energy loss. The glancing angles are 1.5° for positrons and 1.3° for electrons. The incident azimuths are parallel to the $[11\bar{2}]$ direction for positrons and 7.5°-off oriented from the $[11\bar{2}]$ direction for electrons.

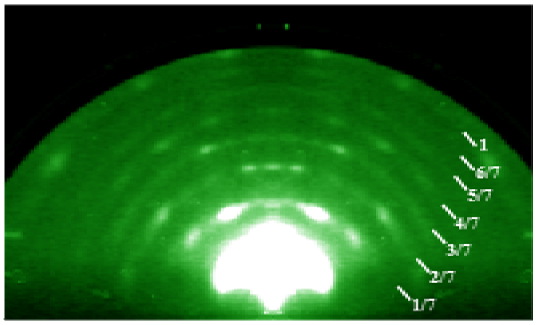

Figure 6. RHEPD pattern from the Si(111)-7×7 surface. The incident azimuth of the positron beam corresponds to the $[11\bar{2}]$ direction. The glancing angle is set at 2.2°. The indexes at the right hand side indicate the Laue zone. The pattern shows the superimposition of the left and right parts.

2.4. Surface thermal vibration

To understand the surface electrical conductivity, surface phase transition, and adsorption, the thermal vibrations of surface atoms is one of the important factors. Figure 4 shows the temperature dependence of the RHEPD intensity measured from a Si(111)-7×7 surface (see also Fig. 1). Since the incident positron energy was 20 keV, the critical angle for total reflection is estimated to be 1.4°. The RHEPD intensities of (0 0) and (1 1) spots at θ = 1.0° and (0 0) spot at θ = 3.5° satisfy the total reflection and the 444 Bragg reflection conditions, respectively. The calculations based on the dynamical diffraction theory confirmed that the RHEPD intensity in the total reflection region is not affected by the bulk Debye-temperature (Θ_B). Thus, the Debye-temperature of the adatoms is solely determined under the total reflection condition. Consequently, the surface Debye-temperature (Θ_S) of the adatoms on the Si(111)-7×7 surface is determined to be 290 K, which is significantly smaller as compared with that obtained by the first-principles calculations and electron diffraction. The result demonstrated that the thermal vibrational amplitude of the adatoms is much larger than those expected before [24].

2.5. Surface plasmon

When charged particles are incident on the crystal surface, various energy loss processes such as phonon, plasmon, and core-level electron excitations occur. Among these excitations, the surface plasmon excitation is known as a dominant process for electrons [25-28]. On account of the positive charge and the total reflection, the surface plasmon excitation due to positrons is probably different from that of electrons. To reveal this, the energy spectra of diffracted positron beam were measured.

Figure 5 shows the positron and electron energy loss spectrum of the specular spot from a Si(111)-7×7 surface. Five prominent loss peaks are observed, which are assigned to single ($\hbar\omega$) through fivefold ($5\hbar\omega$) surface plasmon loss of Si (11 eV), respectively. For positrons, the elastic peak intensity is rather weak and the peak intensities of $2\hbar\omega$ and $3\hbar\omega$ are higher than the others. On the other hand, for electrons, the peak intensity of $\hbar\omega$ is the highest and the elastic peak intensity is also relatively high. The energy loss spectra are approximately expressed by the Poisson distribution: $P(n) = n_s^n \exp(-n_s)/n!$, where n_s is the mean excitation number of surface plasmon [25]. By fitting this equation to the energy loss spectra, n_s = 2.6 for positrons and n_s = 1.4 for electrons are obtained. Thus, the totally reflected positron beam excite more surface plasmons at surfaces than electrons [29].

3. RHEPD with an intense beam and future prospect

At the Slow Positron Facility in KEK, the intensity of the initial positron beam was increased to 10^7 e^+/sec by the modification of converter/moderator assembly [30]. In 2011, a RHEPD apparatus was connected to this KEK positron beamline. A remoderator composed of 100 nm-thick W foil and magnetic lens were installed in front of the RHEPD chamber. The energy spread of the incident positron beam was dramatically reduced to ~8 eV and the beam radius is decreased to ~1.0 mm. As a result, we succeeded in measuring a RHEPD pattern from a Si(111)-7×7 surface (Fig. 6) which showed a significant improvement in clarity in comparison to those obtained before the use of the remoderator. In future studies, the RHEPD method utilizing the intense positron beam system will be applied to advanced surface systems that are important in future spintronics, green technology and so on. A method to directly determine the surface structure will also be developed.

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