New method for the analysis of reflection high-energy electron diffraction: α-Sn(001) and InSb(001) surfaces

I. Hernández-Calderón and H. Höchst
Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-7000 Stuttgart 80, Federal Republic of Germany
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It is shown that from the analysis of intermediate Laue zones of a reflection high-energy electron diffraction pattern it is possible to determine the surface reconstruction in a definite way. The method of analysis is illustrated with the study of α-Sn(001) and InSb(001) surfaces. The diamond-structured (α-) tin (001) surface exhibits a two-domain (2×1) reconstruction whereas for InSb(001) a $c(2\times8)$ reconstruction was obtained. These results are in contrast with the respective (2×2) and (2×4) reconstructions reported recently by Farrow *et al.* using the streak-separation method.

I. INTRODUCTION

The analysis of surface structures by diffraction of electrons is a well-established method in surface physics. The technique mainly used is low-energy ($\sim$100 eV) electron diffraction (LEED).[1] The method of reflection high-energy (5–100 keV) electron diffraction (RHEED) which shows some advantages as compared with LEED has been used so far mainly by material scientists to control and characterize *in situ* deposition of epitaxial films.[2,3] The information contained in the diffraction patterns of both methods is the following: (i) the symmetry and periodicity of ordered layers near the surface, (ii) the position of the atoms within the unit mesh, and (iii) the size of the scattering volume.

In relation to (ii), the weaker interaction of RHEED electrons with matter in comparison with those of LEED allows the use of simpler theories to explain the dynamical scattering process.[4–6] First attempts to analyze the intensity versus angle of incidence from RHEED experiments have been shown to contain similar information as I-V curves studied in LEED.[7,8]

The more restrictive condition of a RHEED experiment in relation to the order of the surface can be evidenced by the fact that at 50 keV the coherence area is about 100 times larger than that necessary for a well-resolved LEED pattern at 100 eV. Ordered regions on the surface with a diameter smaller than 100 Å will lead to a broadening of the diffracted LEED beam, whereas for RHEED a broadening in the pattern will be observed if the ordered regions are $\lesssim$1000 Å in diameter.[9]

The appearance of diffraction streaks instead of spots in RHEED patterns has been a matter of long discussion. Some explanations have been done in terms of refraction of electrons that are scattered from layers below the surface,[2,10] undulations in the surface,[11] flatness of the surface,[12] and thermal diffuse scattering.[13] So far a satisfactory explanation does not exist and this will not be further discussed here. What experience shows is that as the topography changes from a flat to a rough surface, the streaks change from (i) intense, short, and well defined to (ii) longer, diffused, and spotty, and finally (iii) to a pure spotted pattern due to transmission of electrons. Those characteristics are of great utility to monitor *in situ* changes in the topography of surfaces.

Quantitative analysis of RHEED patterns has been restricted so far to the measurement of streak separation under different azimuths in order to obtain the reconstruction of the surface. However, this procedure has shown several inconveniences: (i) the requirement of several diffraction directions,[9,12,14] (ii) great difficulty in distinguishing between diffraction from single and multiple domain reconstruction, and (iii) the possibility of confusion of streaks belonging to different but close intermediate Laue zones in the RHEED pattern. These disadvantages led to the widely accepted idea that LEED is the only method to determine, unambiguously, surface structures.

Since a RHEED pattern also contains features belonging to intermediate Laue zones of the reconstructed surface, we will show that a quantitative analysis of those features of one RHEED pattern along a given azimuth contains enough information to determine in a definite way the reconstruction of

the surface. The method will be illustrated with the reconstructions of InSb(001) and α-Sn(001) surfaces. These results are in contrast with those reported recently by Farrow *et al.*[15]

II. THE RHEED PATTERN

The fact that the diffraction features produced in a RHEED experiment along a given direction contain the information of the two-dimensional (2D) periodicity in k space was recognized by Ichikawa and Ino.[16] By using a spherical screen they obtained two different pictures of a RHEED experiment for a fixed azimuth, a conventional RHEED pattern, and a LEED-like pattern which allowed them to find intuitively the unit mesh of the surface. In this section it will be shown that the structure of the surface can be determined from the position of the spots from a conventional RHEED pattern.

The structure of the RHEED pattern can be easily understood with the help of the Ewald sphere. Owing to the absence of translational symmetry in the direction normal to the surface caused by the solid-vacuum interphase, the discrete points present in the three-dimensional (3D) reciprocal space collapse into continuous rods which are normal to the surface and which possess a 2D translational symmetry in that plane. The Ewald sphere intersects all rods contained in the projection of this sphere onto the surface. Diffracted beams are observed in the angular directions α and β as schematically shown in Fig. 1.

In a typical RHEED experiment with an electron wave vector $k_0 \sim 10^2$ Å^{-1} and a reciprocal surface vector g, we have a characteristic value of $g/k_0 \sim 10^{-2}$. This means that the Ewald sphere will touch a few rods at both sides of the (00) rod [Fig. 1(b)]. The angles between these reflections will practically be the same and are given by

$$\beta_0 = \tan^{-1}\left[\frac{g_\perp}{(k_0^2 - g_\perp^2)^{1/2}}\right] = \tan^{-1}(t/L)\,, \qquad (1)$$

where t is the distance between streaks and L is the sample-to-screen distance. Since $g_\perp = 2\pi/d_{||}$, where $d_{||}$ is the distance between equivalent rows of atoms parallel to the incident beam, and $g_\perp \ll k_0$, the relation

$$d_{||} t = L\lambda_0 \qquad (2)$$

is obtained from Eq. (1), where λ_0 is the wavelength of the electron in the relativistic approximation. Working in a fixed energy and geometry configuration, the camera length $L\lambda_0$ can be calibrated with a sample whose lattice constant is well known. In this way a precision in the determination of lattice constants of 10^{-3} can be easily obtained. As mentioned before, the analysis of RHEED patterns has been restricted to the application of Eq. (2).

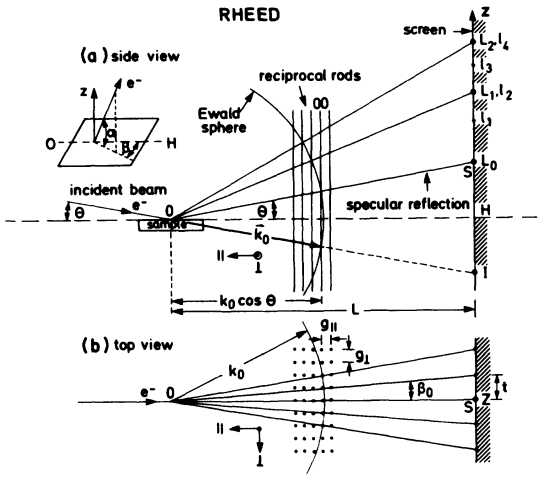

FIG. 1. Schematic picture of a typical RHEED experiment (not to scale). (a) L_n corresponds to the Laue zones of the ideal surface. As illustration are shown the spots on intermediate, l_n, zones that would be present in the case of $g_{||} = \frac{1}{2} g_{||}^i$. (b) Projection of the Ewald sphere on a plane parallel to the surface.

The intersections of each row of rods perpendicular to the incident beam will be projected on a screen over arcs of circumference. The distances between these arcs will be determined by the periodicity in the $g_{||}$ direction. The separation between spots on the same arc will give the information of the periodicity in the $g_\perp$ direction (Fig. 1). The magnitude of the $\vec{g}_\perp$ vectors is obtained from Eq. (1) and the magnitude of $\vec{g}_{||}$ can be obtained following the procedure described below.

Consider a plane passing through the center of the Ewald sphere and containing the (00) rod. For an incident angle $\theta \neq 0$, the (00) rod will intersect the Ewald sphere two times. The intersection at an angle 2θ with respect to the incident beam will correspond to the specular reflected beam shown by the spot S on the screen. Further intersections of the Ewald sphere with rods in the plane of the figure will be indicated by spots along the z axis. Successive projections of those intersections along $\overline{OH}$ will give the magnitude of $g_{||}$ [Fig. 1(a)].

The distance $\Delta g_{||}$ in k space between the perpendicular row containing the (00) rod and a given rod can be calculated by using

$$\Delta g_{||} = k_0 \left\{ \cos\theta - \left[\left[\frac{D}{L} - \tan\theta \right]^2 + 1 \right]^{-1/2} \right\} \qquad (3)$$

obtained from the geometry of Fig. 1(a). Here D is the distance between the spot I of the incident beam and the intersection of the z axis with the arc passing through the spot being analyzed.

We observed a strong modulation of the intensities of the spots as a function of the angle of incidence. Therefore, not all the expected spots had enough intensity to be analyzed. However, since a RHEED pattern maps out several rectangles $g_{||} \times g_{\perp}$ containing spots related to equivalent positions in k space, the reciprocal unit mesh is easily determined. By using the relation $\vec{a}_i \cdot \vec{b}_j = 2\pi\delta_{ij}$ between real and k-space primitive vectors, we obtain the unit mesh of the surface. The procedure described above will be illustrated in Secs. IV and V with the determination of the InSb(001) and α-Sn(001) reconstructions.

III. EXPERIMENTAL SETUP

RHEED studies were carried out employing a Riber gun model CER 550. The patterns were photographically recorded from a fluorescent screen.

Crystal plates of InSb with the required orientation were brought to a mirror finish by mechanical polishing. A procedure consisting of simultaneous Ar^+ bombardment (500 eV) and annealing ($T = 300$ °C) during 4 h was sufficient to clean the substrates.

Molecular beam epitaxy (MBE) films of α-Sn(001) were grown following the method of Farrow *et al.*[15] A deposition rate of 2.5 Å/sec was used. The pressure during deposition was 2×10^{-9} Torr. The cleanliness of InSb substrates and α-Sn films was indirectly controlled by photoemission spectroscopy.[17]

IV. RECONSTRUCTION OF InSb

Figure 2(a) shows the visible features from a InSb(001) RHEED pattern obtained with 30-keV electrons along a ⟨110⟩ azimuth. For this direction $g^i_{||} = g^i_{\perp} = \sqrt{2}(2\pi/a)$ for the ideal surface. From the streak separation t and using Eq. (1) it is found that $g_{\perp} = g^i_{\perp}$. From the measurements of spots on l_2, l_4, l_6, and $l_8 = L_1$ and using Eq. (3) it is found that those spots correspond to rods separated by $g_{||} = 0.25 g^i_{||}$. The vertical separation between spots in the l_1, l_3, l_5, and l_7 arcs correspond also to rod separations of $0.25 g^i_{||}$. These rods are located in the center of the rectangles $g_{||} \times g_{\perp}$. The resulting distribution of the reciprocal rods is shown in Fig. 2(b) and the corresponding unit mesh of the $c(2\times8)$ reconstructed surface is illustrated in Fig. 2(c).

A false picture of this reconstruction would be obtained by the simple analysis of the separation of the streaks. A $2\times$ reconstruction would appear to exist in one ⟨110⟩ azimuth, and a $4\times$ reconstruction in

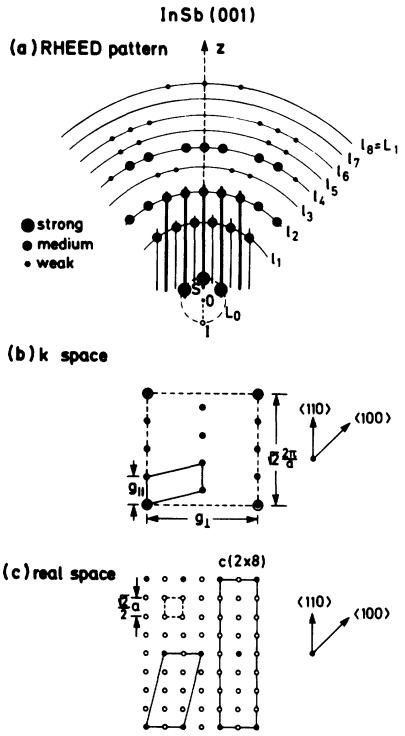

FIG. 2. (a) InSb(001) RHEED pattern with a 30-keV beam along a ⟨110⟩ azimuth. S, specular spot; O, center of the zeroth Laue zone; I, incident beam spot. (b) and (c) are unit cells in k and real space, respectively. Dashed lines: ideal unit cell; solid line: reconstructed surface unit cell. In (c) are shown the primitive and conventional unit cells of the reconstructed surface.

the orthogonal azimuth, and so an incorrect (2×4) reconstruction could be concluded. In particular, we failed to obtain the (2×4) reconstruction claimed by Farrow *et al.*[15] It is possible that it was the $c(2\times8)$ described before. A careful examination to distinguish between these two reconstructions has to be made, especially with the isomorphic III-V compounds. For instance, for GaAs(001), (2×4) structures,[18,19] as well as $c(2\times8)$ structures, have been reported.[12]

It should be mentioned that a (4×1) reconstruction of a contaminated InSb(001) surface was observed in the early stages of the cleaning procedure. The analysis of the other surfaces showed InSb(110)-(1×1), InSb(111)-(2×2), and InSb($\bar{1}\,\bar{1}\,\bar{1}$)-$(3\times3)$ reconstructions in agreement with previous results.[20,21]

V. RECONSTRUCTION OF α-Sn(001)

Grey (α-) Sn is a zero–band-gap diamond-type semiconductor with lattice constant $a = 6.489$ Å. MBE heteroepitaxial layers of α-Sn were grown on top of InSb(001)-$c(2\times8)$ surfaces with a thickness

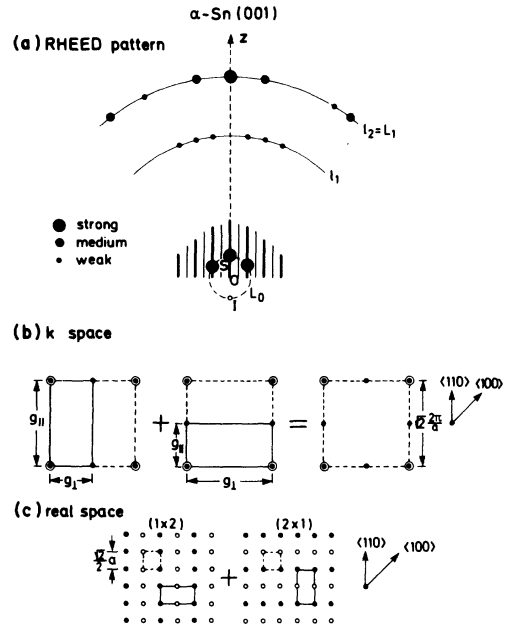

FIG. 3. α-Sn(001) RHEED pattern with a 30-keV beam along a ⟨110⟩ azimuth. In (b) and (c) are shown the unit cells for a two-domain reconstructed surface.

up to 2000 Å. A 30-keV RHEED pattern recorded along a ⟨110⟩ azimuth is shown in Fig. 3(a). No change was observed with the thickness of the film. This pattern presents a peculiarity: From the measurement of the streak separation t for the zeroth Laue zone and the analysis of the spots in l_1 is obtained in the fact that $g_\perp = g_{||} = \frac{1}{2} g^i_{||,\perp}$. This could mean that we have a (2×2) reconstruction as reported by Farrow *et al.*, but then the systematic absence of a spot corresponding to the center of the ideal recriprocal unit mesh [Fig. 4(b)] cannot be simply explained. This pattern can be understood if we suppose that it is originated by the superposition of spots produced by two (2×1) domains, one rotated 90° with respect to the other [Fig. 3(c)]. Further evidence for this structure was obtained by a RHEED pattern recorded along a ⟨100⟩ azimuth. The absence of $\frac{1}{2}$-order streaks in L_0 eliminated the possibility of a (2×2) reconstruction. Since consecutive (001) layers of α-Sn present a 90° rotation of bonds, a surface with an incomplete top layer could explain the observed two-domain reconstruction. It should be noted that for the isomorphic Si(001) surface the same two-domain (2×1) reconstruction was observed.[22]

VI. SUMMARY

Traditionally, the analysis of RHEED patterns has been restricted to streak-separation measurements. This might lead to an incorrect picture of the surface structure. We have shown that from the structural analysis of the features in one RHEED pattern the reconstruction of the surface can be determined in a definite way.

InSb(001) showed a $c(2\times8)$ reconstruction, whereas α-Sn(001) exhibited a two-domain (2×1) reconstruction. These results are in contrast to the respective (2×4) and (2×2) reconstructions published recently by Farrow *et al.*, obtained by the streak-separation method. Since InSb(001) and GaAs(001) surfaces exhibit $c(2\times8)$ reconstruction and α-Sn(001) and Si(001) surfaces present a two-domain (2×1) reconstruction, these structures seem to be characteristic of zinc-blende III-V compounds and diamond-type group-IV elements, respectively.

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