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To tilt or not to tilt: Correction of the distortion caused by inclined sample surfaces in low-energy electron diffraction

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ABSTRACT

Low-energy electron diffraction (LEED) is a widely employed technique for the structural characterization of crystalline surfaces and epitaxial adsorbates. For technical reasons the accessible reciprocal space is limited at a given primary electron energy *E*. This limitation may be overcome by sweeping *E* to observe higher diffraction orders decisively enhancing the quantitative examination. Yet, in many cases, such as molecular films with rather large unit cells, the adsorbate reflexes become less pronounced at energies high enough to observe substrate reflexes. One possibility to overcome this problem is an intentional inclination of the sample surface during the measurement at the expense of the quantitative interpretability of then severely distorted diffraction patterns. Here, we introduce a correction method for the axially symmetric distortion in LEED images of tilted samples. We provide experimental confirmation for micro-channel plate LEED and spot-profile analysis LEED instruments using the (7×7) reconstructed surface of a Si(111) single crystal as a reference sample. Finally, we demonstrate that the correction of this distortion considerably improves the quantitative analysis of diffraction patterns of adsorbates since substrate and adsorbate reflexes can be evaluated simultaneously. As an illustrative example we have chosen an epitaxial monolayer of 3,4,9,10-perylenetetracarboxylic dianhydride on Ag(111) that is known to form a commensurate superstructure.

1. Introduction

Low-energy electron diffraction (LEED) is a widely used technique for the structural examination of crystalline surfaces [1–3]. For the determination of lattice parameters the non-dynamical LEED theory is frequently employed. The achievable accuracy is naturally limited by the quality of the diffraction pattern which itself is in principal subject to radial and asymmetric distortions [4–9]. These distortions should generally be corrected as described in Ref. [5] if a *quantitative* analysis of the diffraction patterns is intended. An additional distortion occurs if the sample surface is not aligned orthogonal to the viewing direction, the latter being parallel to the primary electron beam leaving the electron gun in conventional (C-LEED) and micro-channel plate LEED (MCP-LEED) instruments. For spot-profile analysis LEED (SPA-LEED) [4,10,11] instruments the viewing direction is depicted in Fig. 1(a). Since the fixed viewing angle of the LEED detector limits the accessible reciprocal space at a given primary energy, tilting the sample surface during the measurement is a common practice in the characterization of unknown structures. Consequently, higher diffraction orders *unobservable at higher beam energies* can be evaluated which decisively enhances the quantitative analysis [4].

$I(V)$-LEED measurements may also benefit from intentionally tilted sample surfaces because more independent diffraction orders can be recorded simultaneously yielding more extensive datasets. Most methods measuring the intensity I of a specific spot versus the accelerating voltage V (and hence the primary electron energy $E=e\cdot V$) rely on the scaling of the reciprocal lattice with $1/\sqrt{E}$ [12]. Besides the typical energetic offset E_{off} which may be as large as several electron volts [5] this is especially difficult for tilted samples due to the axially symmetric distortion described here.

Alternatively, the use of an external electron gun with an experimental geometry as described for example in Refs. [4,13] is of particular interest since it allows real-time LEED studies of epitaxial growth fronts [14,15] or morphological changes [16,17]. As a matter of fact, these investigations are also hampered by axially symmetric distortions of a similar kind but with much stronger magnitudes.

Therefore, in this contribution we are concerned with correcting the distortion in LEED images of tilted samples. While the

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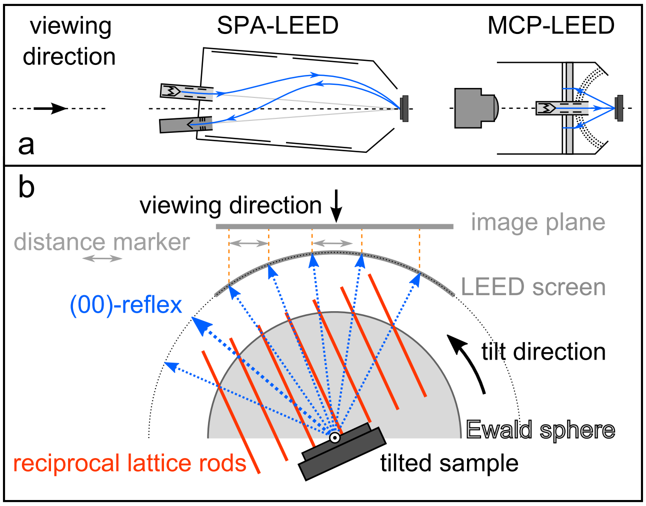

Fig. 1. (a) Definition of the viewing direction in SPA-LEED and MCP-LEED instruments (schematic). (b) Effect of a tilted sample surface on LEED images (schematic). In this case, the spherical LEED screen can be a real one (conventional LEED) or a virtual sphere (MCP-LEED, SPA-LEED).

distortions dealt with in Ref. [5] are caused by systematic errors of the hardware and lead to a radial and/or an asymmetric distortion, the distortion caused by a sample tilt is *axially symmetric with respect to the tilt direction*. One simplified description of this axially symmetric distortion was given already by Hoyer [18]. However, here we develop a more general description and provide experimental confirmation using MCP-LEED and SPA-LEED data. We also specify how to determine the various parameters required in the description as introduced in Section 3. Ultimately, this allows us to correct LEED images in terms of the axially symmetric distortion (Section 4). Since only the corrected images represent an unaffected view of the reciprocal space of the specimen's surface structure, only these images are usable to derive the angles and lengths of the lattice vectors *directly* from the pattern, or to determine the lattice parameters by fitting a theoretical lattice to the spot positions, e.g., by using the software *LEEDLab*.[1]

2. Experimental

For the experimental investigation of the axially symmetric distortion we have chosen the (7×7) reconstructed surface of a Si(111) single crystal due to its well-known structure and high density of diffraction spots. The Si(111) samples were thoroughly outgassed in an ultra high vacuum (UHV) chamber with a base pressure better than 2×10^{-10} mbar for a few hours. Direct current sample heating was used to remove the native oxide by flash annealing to 1500 K in Duisburg. Several cycles of flash heating (typically 10–15 times, ca. 30 s each) to approximately 1500 K were done by electron bombardment in Jena. These procedures are described in more detail in Refs. [19,20]. The Ag(111) substrate was prepared by repeated Ar^+ sputtering (600 eV, 1–3 $\mu A/cm^2$) and annealing cycles (700–800 K) [19]. 3,4,9,10-Perylenetetracarboxylic dianhydride (PTCDA) obtained from Sigma-Aldrich was purified by temperature gradient sublimation [21] and thoroughly degassed for several hours after being transferred to the UHV chamber. Subsequently, the molecules were evaporated from home-built effusion cells comprising boron nitride crucibles while the Ag(111) surface was kept at room-temperature.

The measurements discussed in the following were carried out in Jena with a 4-grid MCP2-SPECTALEED with two micro-channel plates (MCP-LEED) available from Omicron and in Duisburg with a spot-profile analysis LEED (SPA-LEED) from Leybold. All *in situ* substrate treatments were performed with a five-axial $(x, y, z, \vartheta, \phi)$ manipulator. The sample tilt angles were adjusted using the polar rotation (axis perpendicular to the viewing direction) allowing a reproducibility within $\pm 0.1^\circ$. Additionally, the sample alignment was assisted in Duisburg by the specular reflex of a laser firmly mounted to the UHV chamber. The algorithm correcting the LEED images has been implemented in MATLAB from MathWorks.

3. Method and theory

In the following descriptions it is always assumed that besides the axially symmetric distortion considered here no other distortions remain. To ensure this, the LEED images used were corrected beforehand for distortions occurring systematically by means of the software *LEEDCal* [5].

The symmetry of the whole diffraction pattern of the surface structure is independent of the angle of incidence of the primary electron beam. The crucial difference during the measurement with a tilted surface is the change in orientation of the *reciprocal lattice rods* with respect to the *viewing direction*. Due to the tilt, the lattice rods are no longer parallel to the viewing direction (cf. Fig. 1(b)).

Thereby the projection of the reciprocal space is distorted along the tilt direction. This axially symmetric distortion can be thought of as a planar projection of a tilted striped sphere. Originally evenly spaced stripes are no longer equidistant and additionally curved (cf. Fig. 2).

In order to derive a corrected LEED image, we need a relation between the position (x_{id}, y_{id}) of a pixel in the ideal corrected image and the corresponding position (x_{or}, y_{or}) in the original distorted LEED image. One then obtains the corrected image by filling an empty pixel map with the correct gray values found in the distorted image. This procedure is described in more detail in Ref. [5]. According to general conventions in image processing the coordinate systems of the images are defined as left-handed with the origin located in the upper left corner. Therefore the following relationship results split into several equations for the sake of clarity:

$$\begin{pmatrix} x_{\mathrm{I}} \\ y_{\mathrm{I}} \end{pmatrix} = \begin{pmatrix} x_{id} \\ -y_{id} \end{pmatrix} + \begin{pmatrix} -x_C + \sin(\beta)\cdot K\cdot \sin(\kappa) \\ y_C + \cos(\beta)\cdot K\cdot \sin(\kappa) \end{pmatrix} \tag{1}$$

$$\begin{pmatrix} x_{\mathrm{II}} \\ y_{\mathrm{II}} \end{pmatrix} = \begin{pmatrix} \cos(\beta) & -\sin(\beta) \\ \sin(\beta) & \cos(\beta) \end{pmatrix} \cdot \begin{pmatrix} x_{\mathrm{I}} \\ y_{\mathrm{I}} \end{pmatrix} \tag{2}$$

$$z_{\mathrm{II}} = \left|\sqrt{K^2-(x_{\mathrm{II}})^2-(y_{\mathrm{II}})^2}\right| \tag{3}$$

$$y_{\mathrm{III}} = y_{\mathrm{II}}\cdot\cos(\kappa) - z_{\mathrm{II}}\cdot\sin(\kappa)\;;\quad x_{\mathrm{III}} = x_{\mathrm{II}} \tag{4}$$

$$\begin{pmatrix} x_{or} \\ y_{or} \end{pmatrix} = \begin{pmatrix} \cos(\beta) & \sin(\beta) \\ \sin(\beta) & -\cos(\beta) \end{pmatrix} \cdot \begin{pmatrix} x_{\mathrm{III}} \\ y_{\mathrm{III}} \end{pmatrix} + \begin{pmatrix} x_C \\ y_C \end{pmatrix} \tag{5}$$

Here, the coordinates x_C and y_C describe the position of the (00)-reflex in the image if the surface is not tilted (in the case of a conventional LEED instrument: the center of the LEED screen) and can be read directly from the images. The angle κ represents the tilt angle of the sample surface with respect to the non-tilted position. The angle β is defined between the x-axis and the tilt axis which is parallel to the x-y-plane of the images. The positions of all reflexes at different tilt angles form straight lines in the x-y-plot (cf. Fig. 3(a)). The tilt axis is orthogonal to that line, hence the

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[1] See http://www.omicron.de/en/home for further information about the software provided in the 'Pico-Newsletter' vol. 16, no. 1 on page 12, or find a demoversion in the section 'software-downloads' soon.