Holographic Image Reconstruction from Electron Diffraction Intensities of Ordered Superstructures

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We report on a novel holographic reconstruction of well resolved atomic images from discrete spot intensities appearing in low-energy electron diffraction (LEED) from crystalline surfaces. This opens holographic LEED to the wide field of ordered systems giving access to rather complex surface structures.   [S0031-9007(97)04833-3]

The power of low-energy electron diffraction (LEED) for the quantitative analysis of surface structures is well known. As the method is based on a trial-and-error scheme to retrieve the correct atomic arrangement, structural search procedures combined with perturbative intensity calculations (see, e.g., Ref. [1]) have been developed in the past and quite complex structures were solved [2]. Yet, in spite of this progress, the accessible complexity is limited by the need to initiate the search from a promising starting structure—a task our imagination frequently is unable to fulfill. This situation necessitates a direct precursor method, which provides some rough idea of the essential features of the structure as input to a subsequent structural refinement.

Holographic reconstruction methods as originally proposed by Szöke [3] seem to have the potential to meet this demand. Yet reconstruction of atomic images from photoelectron diffraction (PED) patterns as introduced by Barton [4] requires the use of dedicated, large facility x-ray sources. The extension of the holographic method to diffuse LEED (DLEED) intensity distributions as proposed by Saldin and De Andres [5] brings the method back to the home lab, but is only applicable to the restricted class of systems, where a species is adsorbed in lattice gas disorder on a crystalline substrate. This prohibits the application to the remaining large majority of superstructure systems, i.e., such in which adsorbates assume long range order or in which long range order is intrinsic by reconstruction of a crystalline surface. Also, despite a few successful holographic reconstructions using DLEED intensities [6–9], one has to admit that the measurement of diffuse intensity distributions is a delicate task, in contrast to the measurement of discrete spots. Even though the theoretical possibility to extend DLEED holography to ordered systems was already recognized earlier [10,11], a real proof in form of a convincingly resolved structure could never be given. Therefore, we feel to achieve real progress when presenting in this Letter the first holographic reconstruction of well resolved atomic images from discrete LEED spot intensities generated by an ordered and complex superstructure.

The holographic technique itself consists of a numerical inversion method of the diffraction data acquired in a standard LEED experiment. It makes use of the existence of one prominent atom in the unit cell acting as a beam splitter on the incoming electron wave: electrons scattered back directly to the detector form a reference wave in the holographic sense. In contrast, those electrons undergoing an additional scattering process in the substrate before reaching the LEED screen can be considered as an object wave. Hence, the recorded interference pattern contains the complete crystallographic information on the atomic environment of the beam splitter, which can be extracted by numerical inversion [4,5]. Disturbing contributions due to strong multiple scattering of the low-energy electrons can be suppressed combining data taken at different energies, whereby different methods have been proposed so far [12–15].

Since the inversion algorithm involves an integral over all angles of the outgoing electron beam, all of the above outlined reasoning was based on diffuse LEED. In this case, the beam splitter is a single atom adsorbed on a crystalline substrate and, due to the lacking periodicity, the diffraction intensities generated by the latter are diffuse, i.e., distributed over the full screen. With several such adatoms adsorbed in the same local structure but in lattice gas disorder, the situation does not change in the low coverage regime and the intensities simply add up leaving the diffuse intensity distribution practically unchanged (for a recent review of DLEED see Ref. [16]). For ordered structures, the prevailing periodicity on the surface leads to the complete extinction of intensities in almost all directions due to destructive interference and causes the formation of sharp superstructure spots. However, these spots have been proven to still contain the same crystallographic information on the local surrounding of the beam splitter. They exhibit exactly the same energy dependence as the diffuse intensity emerging into the same direction in the disordered case [17–19]. In other words, the spots simply sample the anterior diffuse distribution on a finite grid. We notice that, with this understanding, the only remaining difference between DLEED and LEED holography is the accessible data density in the diffraction pattern.

Earlier investigations on the information content of diffuse intensities revealed that for a single adatom per hexagonal unit cell already a (3×3) sampling grid describes the continuous distribution in sufficient approximation, i.e., the information on a denser grid is largely redundant [18]. Also, recent work demonstrated that there is practically no image degradation when a restricted database is used in the holographic reconstruction of DLEED intensities [20]. Consequently, the data available for large ordered superstructures appears to match very well with the minimum base required for a reliable reconstruction, enabling the application of holography to these ordered phases. However, we should emphasize that this is only true if there is just one single beam-splitter atom per superstructure unit cell. In other cases the situation may be less favorable.

Having established the equivalency of DLEED and LEED holography, let us focus on the consequences of this finding. First, it is irrelevant whether the beam splitter is an adsorbate, as it is in the case of ordered adsorption phases, or belongs to the substrate itself, bringing in reach also ordered substrate reconstructions. As mentioned, the only restriction in this respect is that there has to be only one such atom per surface unit cell in order to prevent intermixing of images. The larger the unit cell, the more fractional order spots are available for a more complete sampling and consequently for a better working numerical inversion. As large unit cells are more difficult to be analyzed by conventional LEED, it seems that the latter and LEED holography complement each other in a nearly ideal way to tackle complex structures: The more difficult the realization of one method, the better applicable the other. Also, as already indicated, the acquisition of $I(E)$ spectra is experimentally much easier for discrete spots than for diffuse intensities. For discrete spots the signal-to-noise ratio is much higher; contributions from thermal diffuse scattering are less important and easy to subtract. Data at much higher energies are accessible and there is less danger that the intensities of interest are overscreened by bright substrate spots. Higher energy diffuse data always suffer from this circumstance [19] and, consequently, the proper working of the inversion algorithm can be put in jeopardy as data of a certain minimum energy range are needed to efficiently suppress the unwanted multiple scattering contributions. Of course, the increase of the usable energy database provided by the LEED experiment (as compared to DLEED) is accompanied by a drastic reduction in angular input due to the finite sampling. It should be emphasized that the use of too coarse a grid, as present for smaller superstructures, leads to the breakdown of the algorithm due to insufficient sampling of the intensity distribution. The lateral extension of real space that can reliably be imaged is in any case correlated to the density of the grid used, since aliasing effects are to be expected in the Fourier-like transform outside a certain region [20].

In order to verify our reasoning we applied holographic LEED to the (3×3) reconstruction phase of SiC(111).

SiC is a material with most promising electronic properties, which have triggered numerous research activities for high power and high frequency devices. However, its commercial application has been hampered by difficulties in growing wafers of sufficient quality. In this respect it is most interesting that recently achieved high quality homoepitaxial growth was interpreted to be related to the (3×3) phase which forms under silicon rich conditions [21]. Yet, despite many efforts, the atomic structure of this phase could not be revealed up to now as it seemed simply too complex to be solved by trial-and-error methods. Since we know from separate work using scanning tunneling microscopy that each (3×3) unit cell contains only one prominent atom to serve as a beam splitter [22], the (3×3)-SiC structure appeared to be the ideal candidate for an image reconstruction through holographic LEED. The upper panel of Fig. 1 shows a corresponding LEED pattern at 186 eV for normal incidence of the primary beam. Normal incidence was chosen because it allows high quality measurements: The angle of incidence can easily be adjusted by comparison of symmetrically equivalent beams avoiding errors by sample misalignment to which LEED intensities are rather sensitive. Also, symmetrically equivalent beams can be averaged to increase the quality of the database. The intensities of the fractional order beams in the range of 50–300 eV were taken using a video based method operated under computer control [23] (more details of the sample preparation and measurement will be published elsewhere [24]).

The restriction to just normal incidence of the primary beam requires the use of a holographic reconstruction scheme which successfully works with such a single data set, i.e., does not need data sets taken at different angles of incidence. We therefore applied the corresponding method proposed by Saldin and Chen [15], which had worked reliably already for simple disordered adsorbate systems [8,9]. The resulting 3D real-space image is shown in the middle panel of Fig. 1 displaying the atomic surrounding of the beam splitter. The atomic positions retrieved are schematically reproduced in the bottom panel including a visualization of chemical bonds. In spite of the low cutoff level chosen (25%) there are practically no artifacts in the reconstructed image. The noise is at an unprecedented low level and the atoms show up with unambiguous high signal. The beam splitter is the topmost atom in a tetramer unit typical for hexagonal semiconductor surfaces. Yet the full structure is even more complex: The imaged structural element has been used as input for subsequent refinement using conventional LEED structure analysis and first principle calculations by density functional theory [22]. On the basis of the holographic result the majority of the previously existing structural models could be ruled out directly. This enabled the efficient search of both methods in a then drastically reduced parameter space. Figure 2 shows the finally determined full unit cell with the structural element retrieved in the
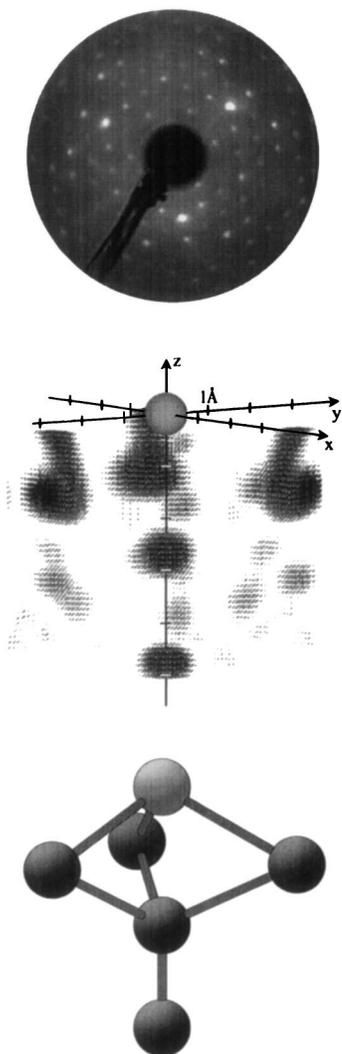
FIG. 1. (3×3) LEED pattern of SiC(111) at 186 eV (upper panel) and atomic image as reconstructed (middle) and as schematically reproduced including chemical bonds (bottom). In the reconstructed image the beam-splitter atom is added (= origin of coordinate system). The image reconstruction is performed on a 3D grid. Small spheres whose diameters scale linearly with the reconstruction intensity are drawn at the grid points (noise cutoff: 25%).

holographic reconstruction displayed with white balls for clearness, i.e., a tetramer at the surface and two silicon atoms in deeper layers. A more detailed description of the structure including the structural refinement by conventional LEED analysis, which is not the aim of the present Letter, will be given elsewhere [25]. It is worth emphasizing that LEED holography actually provided the key information in the determination of this complex and previously *unknown* phase.

In conclusion, we have shown that conventional LEED superstructure spot intensities can be transformed into well resolved atomic images by holographic inversion. The necessary condition for this procedure is that the unit cell is large enough to provide a sufficiently high
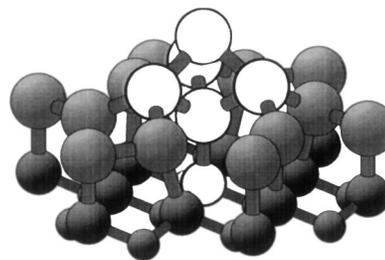

FIG. 2. Full unit cell of the SiC(111)-(3×3) superstructure as resulting from a structural refinement by conventional LEED [25] with the reconstructed structural element drawn as white balls.

density of superstructure spots and that there is only one prominent atom acting as the holographic beam splitter. Despite these restrictions, our result opens the direct inversion of electron diffraction data to the wide field of ordered superstructures, independent whether they form by adsorbates or by reconstruction of a crystalline substrate. The new method of LEED holography profits from much better experimental conditions than those applying to DLEED holography: The measurement of discrete spot intensities is much safer and more routine than that of diffuse intensities and can easily be extended to higher energies, i.e., shorter wavelengths, in order to improve spatial resolution. Additionally, the successful application to SiC(111)-(3×3) also proves the long claimed capability of holography to provide substantial direct information on complex and unknown surfaces and to act as a guiding precursor for a structural refinement.

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