Direct structure determination of systems with two-dimensional periodicity

Y. Yacoby
Racah Institute of Physics, Hebrew University, Jerusalem, Israel, 91904
R. Pindak, R. MacHarrie, and J. Pitney
Bell Laboratories, Lucent Technologies, 700 Mountain Ave., Murray Hill, NJ 07974 USA
R. Clarke
Randall Laboratory of Physics, University of Michigan, Ann Arbor, MI 48109 USA

Introduction

Systems which can be structurally characterized as being fully or partially periodic in two dimensions, aperiodic in the third, and positionally correlated to an underlying substrate crystal constitute a large class of scientifically and technologically interesting structures. Examples of such 2D structures include reconstructed crystal surfaces, layered heterostructures, crystalline-amorphous interfaces and proteins crystallized on a substrate. Establishing techniques to measure these 2D structures is crucial for the development of new devices and materials processing methods. Here we report a new method for the direct measurement of such 2D structures that utilizes the coherence found in the high-brilliance APS synchrotron x-ray source to generate phase-preserving diffraction-interference patterns. The method provides real space positions with atomic resolution and does not require an *a-priori* conjecture of the structure.

Method

The method we have developed is quite general but it will be easier to explain using a specific example. We consider a system composed of a three-dimensional semi-infinite crystal, GaAs, containing embedded crystalline monolayers of AlAs. The electron density of the unknown 2D structure is the difference between the semi-infinite GaAs and the actual sample that contains a few monolayers of AlAs. Note that if the GaAs is distorted relative to its ideal structure, the difference between the ideal and the actual distorted structure is also part of the unknown 2D structure.

The 3D crystalline lattice defines a two-dimensional grid on x-y planes parallel to the substrate surface. At first we assume that the 2D structure is fully periodic with respect to this grid. The complex scattering factor (CSF) of the ideal semi-infinite substrate crystal, $S(k_z)$, is known and non-zero along a set of Crystal Truncation Rods (CTRs) which are oriented parallel to the surface normal (z axis). The CSF of the unknown 2D structure, $U(k_z)$, also contributes to the scattering intensity along these CTRs. The absolute value of the total CSF $|T(k_z)|$, the quantity measured experimentally, is the modulus of the complex sum of the two.

For the case of a buried interface where the thickness of the unknown 2D structure is small compared to its distance, z•, from the surface, it is possible to determine $U(k_z)$ by the following efficient iterative procedure. We first consider two points along a CTR at at $k_1=k_z$ and $k_2=k_z+\delta k_z$. The total CSFs at the two points can be expressed in terms of the known substrate CSF and unknown CSF as follows:

$$|T_1| \exp(i \phi_1) = S_1 + U_1 \qquad (1)$$
$$|T_2| \exp(i \phi_2) = S_2 + R\, U_1 \qquad (2)$$

Here, indices 1 and 2 correspond to k_1 and k_2 respectively and $R = U_2/U_1$. In these equations the real space zero point of the electron density function is taken to be approximately at the location of the buried 2D structure. This choice insures that the phase of $U(k_z)$ will vary slowly with k_z. The zero point distance from the surface needs to be chosen with only a 20% accuracy. For the first iteration, take $R = U_2/U_1 \approx 1$, then solve equations

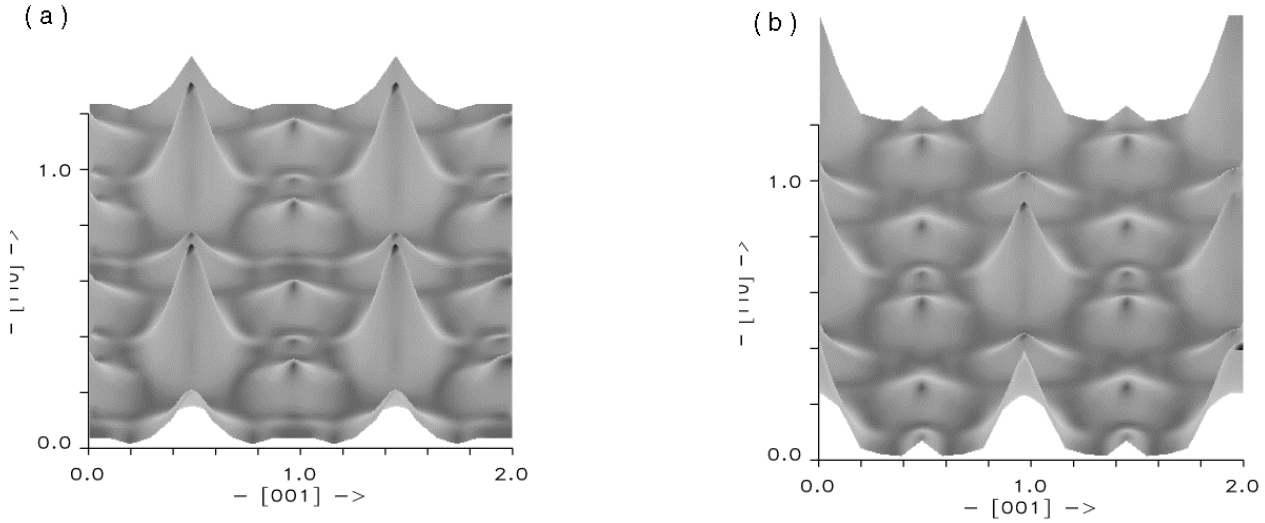

Figure 1. Surface plots for the in-plane Al concentration (a) in the AlAs monolayer closest to the surface and (b) in the next AlAs monolayer. As expected, the structure in (b) is the same as (a) but shifted by 0.5 and 0.353 unit cell units in the <001> and <110> directions, respectively.

(1) and (2) for U_1, ϕ_1, and ϕ_2. Next, using the calculated values for $U(k_z)$, obtain a better estimate for R and continue with the iteration. In all of our simulations this procedure converged after less than 6 iterations.

This procedure provides $U(k_z)$ for all relevant CTRs in reciprocal space. Its three-dimensional back-Fourier transformation into real space provides the 3D real space x-ray dielectric function, which is related in a known way to the electron density function. In cases where the 2D structure is close to the physical sample surface, and hence, the phase of $U(k_z)$ will not necessarily vary slowly, we have developed an alternate two-beam approach [1-2]. In this approach a reflecting overlayer is added to the sample and the beam enters the sample from the side at glancing incidence. Under these conditions, the incident beam is reflected from the overlayer and the incident and reflected beam are diffracted in pairs of coherent and exactly parallel beams. The interference between these beams allows for a determination of the phase derivative along the CTR.

Results

Previously, we tested this iterative procedure using both simulated data as well as experimental results from a GaAs/AlAs multilayer [3]. Specifically, the z-dependence of the in-plane averaged electron density was determined from the analysis of a single CTR that passed through 4 Bragg peaks. Both single beam and two-beam approaches were tested. At the APS MHATT-CAT undulator beamline, we will extend the technique to measure complete sets of CTRs to obtain the full 3D structure of the buried interfacial region. This will be made possible by the significant gain in flux from the higher source brilliance. We are also implementing several refinements in our experimental technique. First, we are using a 2+2+Kappa goniometer that allows us to do CTR scans by limiting sample motion to primarily a single rotation (phi) while moving the detector in two angular directions. Next, a CCD area detector is being used to handle shifts in the diffracted beam and simultaneously collect scattering from both the CTR and background. A LABVIEW based data acquisition program has been developed that incorporates code to correct for inaccuracies in the alignment of the axes of the goniometer circles. The code also includes 'cones of collision' that prevent collisions between the sample and detector arms – crucial for the safe operation of a kappa instrument. Finally, code is being developed to do on-line analysis of the image files for efficient data storage.

While the beamline hardware and software to accomplish the efficient CTR data collection is being commissioned, we decided to test the iterative procedure on a full set of simulated CTRs. The simulated data included noise typical for the relevant experimental implementation. The structure we used was GaAs with 4 monolayers of AlAs buried under 20 monolayers of GaAs. In these simulations we generated the diffraction data along 49 CTRs and analyzed the data of each rod as previously described. This provided us with the scattering factor $U(k_\xi,k_\eta,k_z)$, where k_ξ, k_η are the x and y components of the (ξ,η) discrete in-plane reciprocal lattice vectors. We then back Fourier transformed this function in three dimensions to obtain the three-dimensional dielectric function. An example of the results is shown in Fig. 1. In Fig. 1a we show the difference between the electron density of GaAs and AlAs as a function of position in the plane of the AlAs monolayer closest to the surface. The in-plane structure of the next AlAs monolayer is shown in Fig. 1b. As expected, the structure is the same but shifted by 0.5 and 0.353 unit cell units in the <001> and <110> directions, respectively. The additional substructure in between the dominant peaks, is an artifact of analyzing a limited number of CTRs.

Discussion

The method presented here provides three-dimensional structural information with atomic resolution. It does not require any a priori model of the structure (except for the approximate distance from the surface of the unknown 2D structure). It can handle structures with large 2D unit cells and large layer thickness, and, it is non-destructive. Our method will provide important structural information on epitaxially-grown buried thin films and on interfaces between a crystal and an amorphous substance such as an oxide. We expect this method to be useful in the study of surface reconstruction and surface structural phase transitions and in the investigation of self-assembled systems grown on single crystals. The basic principle of the method presented here may also be extended to other systems including large biological molecules.

Acknowledgments

This work was supported by Intel Corporation and the Quarter-Micron Consortium in Israel. Work on the MHATT-CAT beamline is supported in part by a Department of Energy grant DE-FG02-99ER45743.

Use of the Advanced Photon Source was supported by the U.S. Department of Energy, Basic Energy Sciences, Office of Science, under Contract No. W-31-109-Eng-38.

References

[1] Y. Yacoby, *Solid State Communications* **91**, 529 (1994).
[2] H. Baltes et al., *Phys. Rev. Lett.* **79**, 1285 (1997).
[3] Y. Yacoby et al., submitted to *J. of Phys. Condensed Matter*.