Coherent X-ray Diffraction from Quantum Dot Arrays

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Introduction

Single molecule imaging is one of the holy grails of synchrotron radiation science. Single cluster imaging is therefore one of the intermediate goals. Small semiconductor clusters have important technological applications as “quantum dots.” They demonstrate a rich variety of structures containing the combined influence of strong strain and composition gradients. Their electrical properties, beyond the simple confinement effects, therefore deviate strongly from those of bulk semiconductors. Isolated quantum dots are hard to prepare because it is difficult to localize the source of material to a small region; however, methods have been devised for spacing the dots apart on a micrometer-length scale to create “quantum dot arrays.” The arrays are completely epitaxial, which means the diffraction from all the quantum dots in the field of view can interfere.

Sayre et al. [1] suggested that a combination of coherent x-ray diffraction with a so-called oversampling phasing method could be used to invert diffraction patterns from small nonperiodic objects to create images. In a first demonstration experiment, Miao et al. used that method to invert the soft x-ray forward-scattering pattern measured from a fabricated object [2]. More recently, the reconstruction of 2-D and 3-D crystalline and noncrystalline structures has been reported [3]. The latest results from Williams et. al [4], which demonstrate that the complete 3-D phase and shape information of a micrometer-sized gold crystal could be retrieved, impressively demonstrate the high potential of this nondestructive method.

Methods and Materials

In this experiment, we focus on the application of the above-mentioned technique to an ordered array of Si/Ge quantum dots (see Fig. 1). It has already been shown that imaging of such an ordered array of nano-objects by inverting coherent x-ray diffraction data is possible in principle [5]. Figure 2, for example, shows a calculated diffraction pattern as it would be expected in an experiment, where only a few dozen quantum dots from an ordered array are illuminated under (partly) coherent conditions [5].

The coherent x-ray diffraction experiments described in this report have been carried out at UNI-CAT.
beamline station ID-34-C at the APS. Monochromatic x-rays with an energy of 8.92 keV were selected by using a diamond (111) double-crystal monochromator.

In order to have both sufficient flux and the opportunity to select only a few quantum dots, a Kirkpatrick-Baez (KB) mirror system has been used to focus a $50 \times 50 \mu m^2$ x-ray beam to typically $1.0 \times 1.5 \mu m^2$ at the position of the sample [focal length of 50 mm (horizontal)/150 mm (vertical)]. The diffraction data were recorded by using a charge-coupled device (CCD) with a pixel size of 22.5 µm placed at a distance of 1092 mm away from the sample.

Results
Several different experimental geometries have been used (insert in Fig. 3b). In order to investigate the crystal structure, CCD images have been taken in the vicinity of the 202 Bragg peak of the GeSi lattice, which is in a distinct location with respect to the underlying silicon lattice. Figure 4 shows a 2-D contour plot of a typical result obtained from using that diffraction geometry. Several features can be clearly distinguished in the diffraction pattern. There are both broad diffuse and sharp Bragg-like diffraction features superimposed. The sharp peaks are the expected Bragg-like diffraction pattern of the ordered 2-D array structure of the quantum dots, while the existence of diffuse peaks may indicate some incoherence between the strains of individual neighboring dots. This whole pattern is somewhat distorted because of the inclined diffraction geometry. Miller index notation in the Si reciprocal lattice has been used to label the contour plot in Fig. 4, on the basis of the transformations of the “super” diffractometer program. The width of the array peaks suggest that there may be as many as 20 to 50 quantum dots illuminated, a number that is slightly at odds with the measured beam size. The broad and diffuse intensity distribution from the form factor of a single dot is modified by the 2-D grating pattern.

Furthermore, the CCD view in Fig. 4 corresponds to an Ewald-sphere slice that cuts approximately across the Ge Bragg peak directly below the Si(202) peak. The horizontal axis is a high-symmetry direction that should be a mirror plane, but the slice is slightly tilted, so the mirror symmetry is distorted, as indicated by the reciprocal lattice indices superimposed on the figure.

As a second approach and in order to investigate the feasibility of applying the methods of direct reconstruction, further measurements in a forward scattering geometry have been carried out. Fig. 3 shows results obtained in a grazing incidence small-angle x-ray scattering (GISAXS)-type geometry for two different values of the incidence angle $\alpha_i$. For values larger than the critical angle (Fig. 3a), the scattering pattern basically shows the diffuse surface diffraction (Yoneda wing) modified by the form factor of single quantum dots (broad distribution) and the 2-D grating pattern. For $\alpha_i < \alpha_c$ (Fig. 3b), the diffraction pattern, now taken above the strong specularly reflected beam, reduces to a simpler small-angle scattering pattern without the pronounced diffuse surface scattering effects that are found when $\alpha_i > \alpha_c$. 

![FIG. 3. Two-dimensional contour plots of the measured, forward scattered intensity for a fixed angle of incidence $\alpha_i$ of 0.6° (a) and 0.05° (b); see text.](image-url)
Discussion

The following major conclusions can be drawn from the experimental results. First and most important, the high-resolution diffraction patterns clearly demonstrate the feasibility of carrying out coherent x-ray diffraction experiments on an ordered array of quantum dots. Our results suggest that two experimental geometries (i.e., CCD measurements of the diffraction pattern both on the Bragg Peak of the quantum dots and in the forward scattering GISAXS geometry) should be used as the preferred methods. Finally, these results can now be used as a starting point for directly reconstructing the shape of single quantum dots.

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References


FIG. 4. Two-dimensional contour plot of the measured intensity in the vicinity of the GeSi 202 Bragg Peak.