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DIRECT STRUCTURE DETERMINATION BY ATOMIC-RESOLUTION INCOHERENT STEM IMAGING

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Direct structure determination by atomic-resolution incoherent STEM imaging

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ABSTRACT: Use of a large, annular dark-field (ADF) detector in a scanning transmission electron microscope is shown to give images that can allow direct structure determination, being a convolution between the illuminating probe intensity and an object function localised at the atomic column positions. The ADF image is also shown to resolve crystal spacings more than twice smaller than the phase contrast point resolution limit of the microscope used, with sub-angstrom structural information being retrieved.

1. INTRODUCTION

In the determination of atomic structures by conventional high-resolution transmission electron microscopy (HRTEM), there are two important difficulties. First, because the electron interacts strongly with the electric potential within the crystal, strong multiple scattering occurs that can complicate the image intensity and in general renders it impossible to interpret the image intuitively in terms of the projected structure. Second, spherical aberration limits the angle of scattered electrons that can be focussed by the objective lens, giving a resolution limit that can limit the accuracy to which the positions of atomic can be determined, and can also give rise to artefacts in the image.

It is now more than 100 years since Lord Rayleigh (1896) considered the incoherent mode of imaging for light optics. He noted that illuminating the specimen with a large incoherent source renders the specimen effectively self-luminous, destroying interference effects between spatially separated parts of the specimen. In addition, he noted that the incoherent mode could resolve features with half the spacing of that for axial plane wave illumination. By the principle of reciprocity a large detector in a scanning transmission electron microscope (STEM) is equivalent to a large incoherent source in a conventional microscope. Because few electrons are absorbed in a thin specimen, an infinite detector that collected all transmitted electrons would give no contrast, so we add contrast by introducing a hole, giving an annular dark-field (ADF) detector (Fig. 1). An ADF image from a $\{1\bar{2}10\}$ domain boundary in wurtzite GaN viewed along $\langle 0001 \rangle$ is shown in Fig. 2. The positions of the atomic columns are clear in this image and agree with the structure proposed by Xin et al. (1997). Here we describe briefly how such structure images arise, and how quantitative information may be retrieved from them.

2. INCOHERENT IMAGING FROM COHERENT SCATTERING

We start by considering the situation for a stationary lattice, so that the scattered electron wavefield can be assumed to be completely coherent, with purely Bragg diffraction for a crystalline specimen giving Bragg diffracted discs as depicted in Fig. 1. The Fourier transform of the ADF image intensity can be written in terms of the Fourier components of the

Fig. 1. A schematic of ADF image formation in a STEM

Fig. 2. A $\{1\bar{2}10\}$ domain boundary in wurtzite GaN viewed along $\langle 0001 \rangle$. The atom columns are 0.19 nm apart.

Bloch wave solutions of the Schrödinger equation (Nellist and Pennycook, 1997a). The advantage of such a formulation is that it is possible to perform the summation over the Bragg beams incident on the ADF detector *before* summing over the Bloch waves. It is then possible to determine which Bloch states contribute to the ADF image. A calculation for GaAs viewed along $\langle 110 \rangle$ at 300 kV shows that for a 30 mrad inner detector radius, the image contrast is dominated by the contribution from the tightly bound 1s-type states (Fig. 3). It is only the 1s states that have the sufficient transverse momentum to scatter the electrons to the detector. Even if we now include the effect of lattice vibrations, phonon scattering is not able to provide the high-transverse momentum required, and only helps to destroy interference effects between the high-angle elastic scattering from different heights within a single column. We can therefore think of the ADF detector as a Bloch state filter.

Since the excitation and form of the 1s-type states, which dominate the ADF image contrast, vary only slowly as a function of the incident beam angle, all the coherent partial plane waves in the probe-forming incident cone of illumination can contribute with similar weight. In this

situation it is straightforward to show (Nellist and Pennycook, 1997b) that the image can be treated as a convolution between an object function formed from the highly localised 1s type states and the probe intensity,

$$I(\mathbf{R}) = \int |P(\mathbf{R}' - \mathbf{R})|^2 O(\mathbf{R}') d\mathbf{R}' \quad (1)$$

where $P(\mathbf{R})$ is the complex amplitude of the illuminating probe, and $O(\mathbf{R})$ is the object function. Since the intensity of the probe is a real and positive quantity, it can be immediately deconvolved from the image intensity, in contrast to the coherent imaging process where in general it is necessary to solve for the phase of the image-plane before a deconvolution can be performed.

3. DECONVOLUTION AND RESOLUTION

Equation (1) can be used in two ways: If the probe intensity function is known, then it can be deconvolved to give information on the object function that allows greater accuracy in structure determination. This is useful because in any resolution limited imaging system, the peak intensity positions will not necessarily be located at the atomic column positions. Alternatively, if the object function is known, which often happens in practice because a typical image will contain a defect surrounded by regions of perfect crystalline matrix, then it is possible to find an estimate for the probe intensity (Nellist and Pennycook, 1997b). Having determined the probe intensity, we need to consider how it may be deconvolved from the experimental image intensity. Fig. 4 shows raw data from an image of GaAs<110> and the result when the probe intensity function is deconvolved from it using a Wiener filter. Although the deconvolution has reduced the noise level in the image, the resulting object function is not localised, and there are stronger artefacts in the tunnels between the dumbbells. The reason that an object function with localised peaks at the atoms columns has not been reconstructed is because the deconvolution has not been able to reconstruct information beyond the resolution limit of the microscope. There is not one unique object function that fits the data, and to find the atomic locations accurately a further constraint is required (Nellist and Pennycook, 1997).

For the accurate determination of structures, therefore, the achievement of higher resolution in TEM is an important goal, even when the projected interatomic spacings are within the resolution limit of the microscope. For the same imaging conditions, the ADF image is able to resolve spacings half as small as conventional bright-field imaging, as indicated in Fig. 1 where the bright-field image would not show any contrast. For ADF imaging, using a large objective aperture with a highly underfocussed lens can pass information well beyond the conventional Scherzer incoherent resolution limit, albeit with some loss in the intuitive nature of the image. Fig. 5 shows the magnitude of the Fourier

Fig. 4. (a) Raw image data of GaAs<110> and (b) the image when the probe intensity function has been deconvolved using a Wiener filter.

transform of an image of $\text{Si}\langle 110 \rangle$ taken using an objective aperture 180% larger than the optimum Scherzer size, and with the objective lens approximately 130 nm underfocussed. A (444) spatial frequency may be seen, which indicates information transfer to 0.078 nm. Data taken from a sample of $\text{CdTe}\langle 11\bar{2} \rangle$ under similar conditions is shown in Fig. 6. A (444) spatial frequency can be seen in both the image intensity profile and the magnitude of the Fourier transform, thus resolving the 0.093 nm spacing between the Cd and Te columns in this projection. The phase relationship between the (111) and (444) image spatial frequencies should allow the positions and polarity of the Cd and Te atomic columns to be determined.

Fig. 5. The magnitude of the Fourier transform of a highly underfocussed ADF image of $\text{Si}\langle 110 \rangle$ using an oversized objective aperture.

0.093 nm spacing resolved,
and showing polarity information

Fig. 6. The magnitude of the Fourier transform of an image of $\text{CdTe}\langle 11\bar{2} \rangle$ taken using similar conditions to Fig. 5 along with an image profile along the $[111]$ direction..

Lord Rayleigh 1896 Phil. Mag. (5) 42, 167

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