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The phonon contribution to high-resolution electron microscope images

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Abstract

The amount of phonon scattering as a function of specimen thickness is determined for a clean silicon sample, free from amorphous surface layers, by measuring the diffuse scattering in energy-filtered convergent-beam diffraction patterns. It is found that for a 25 nm thick sample, only 7.5% of the intensity scattered to less than 18 nm^{-1} is phonon scattered. This means that in a typical high-resolution sample most of the diffuse scattering is caused by surface amorphous layers rather than phonon scattering.

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1. Introduction

When compared quantitatively, the contrast in high-resolution electron microscope (HREM) images (where contrast here refers to the amplitude of the lattice fringes) has usually been found to be lower than that predicted by image simulations, e.g. Refs. [1–4]. This loss of contrast, often called the Stobbs factor, has also been found in lower-resolution images. For example, defocus series of images of a p–n junction in a foil of uniform thickness prepared by focused ion beam milling have shown a similar discrepancy when compared with off-axis electron holograms of the same sample [5]. Previous work has shown that the loss of contrast is the same over most spatial frequencies, so that in effect a constant additional background intensity is present in the experimental images [6]. Boothroyd [7] examined the cause of this constant background intensity by measuring the level of the diffuse background between the diffracted discs in energy-filtered convergent-beam patterns of GaAs. This diffuse background is associated with scattering from amorphous materials and phonon scattering. Although such scattering was observed, it was not possible to distinguish between scattering from amorphous contamination layers on the sample surfaces and phonon scattering.

In the present paper, the level of diffuse scattering between diffracted discs in a clean silicon sample that contains no amorphous surface layers is measured. Energy-filtered convergent-beam

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patterns and images were recorded using a Gatan imaging filter attached to a JEOL 2000 V ultra-high vacuum electron microscope ($C_s = 0.8$) with a point resolution of 0.21 nm and a LaB_6 filament. The base column pressure of 1.5×10^{-10} Torr and in situ specimen heating stage allow the silicon sample to be cleaned by high-temperature heating and ensures that no carbon contamination builds up during observation. The silicon sample was prepared by dimpling and chemically polishing a [0 0 1] Si wafer in 20% HF, 60% HNO_3, 20% H_2O before loading into the preparation chamber of the electron microscope. The specimen was heated to 400°C with a direct current for 3 h, in order to remove adsorbed hydrocarbons. It was then heated briefly to about 1200°C to evaporate any native oxide and to leave a clean Si surface. Any remaining carbon-containing material is converted to SiC, leaving occasional SiC crystals on the Si surface. The surface diffusion from this rapid anneal rounds off the edge of the sample, giving a sample whose thickness increases rapidly with distance from the sample edge.

Fig. 1 shows an energy-filtered lattice image acquired from the edge of such a silicon sample. The {2 2 0} lattice fringes, with $d_{220} = 0.192$ nm, are on the limit of the resolution of the microscope, which has an information limit of around 1.8 nm and therefore appear with very low contrast. Some amorphous contrast is still visible and no surface reconstruction was observed, indicating that the cleaning process was not completely successful. It can be seen, however, that the amorphous surface layer is no more than one or two monolayers thick. No change in this amorphous layer was seen during the experiment, indicating that there was no deposition of carbon contamination. Thus the observed diffuse scattering can be assumed to be almost entirely from phonon scattering.

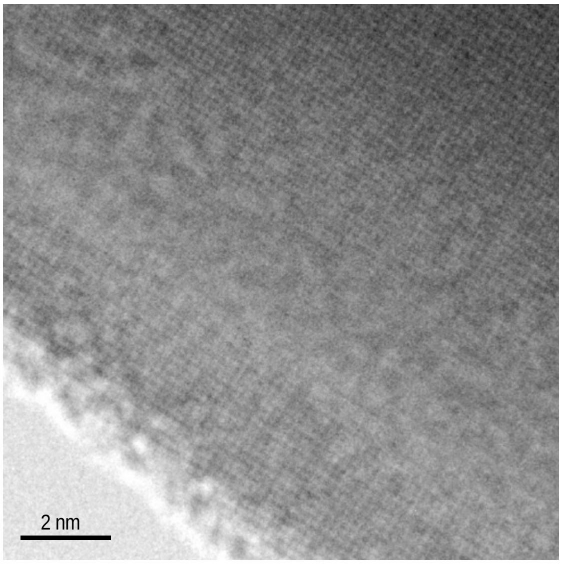

Fig. 1. HREM image of the edge of a silicon sample after cleaning showing {2 2 0} lattice fringes with $d_{220} = 0.192$ nm. The image was taken using a JEOL 2000 V 200 kV ultra-high vacuum electron microscope with $C_s = 0.8$ mm and a point resolution of 0.21 nm.

2. Results

The approach used to measure the amount of phonon scattering has been described by Boothroyd [7]. Two series of 20 energy-filtered convergent-beam electron diffraction patterns were collected with the beam stepped in units of ~10 nm from outside the edge of the sample up to a sample thickness of about 240 nm. Condenser apertures of radii 3.86 (1.53 nm^{-1}) and 1.11 mrad (0.44 nm^{-1}) were used for the two series. Four of the diffraction patterns from the large condenser aperture series are shown in Fig. 2. Comparison of the diffraction patterns with simulations calculated using the EMS Bloch wave program cb2 and measurements of the inelastic/elastic scattering ratio calibrated from weak beam dark-field thickness fringes both allowed the specimen thickness to be determined.

The intensity of phonon scattering in the diffraction patterns was measured between the convergent-beam discs and interpolated beneath them on the assumption that phonon scattering is uniform under the diffraction maxima. It was assumed that this diffuse intensity does not contribute to lattice fringe contrast, but just adds a uniform background to lattice images. It is arguable that some phonon intensity could produce lattice fringe contrast, especially if it is

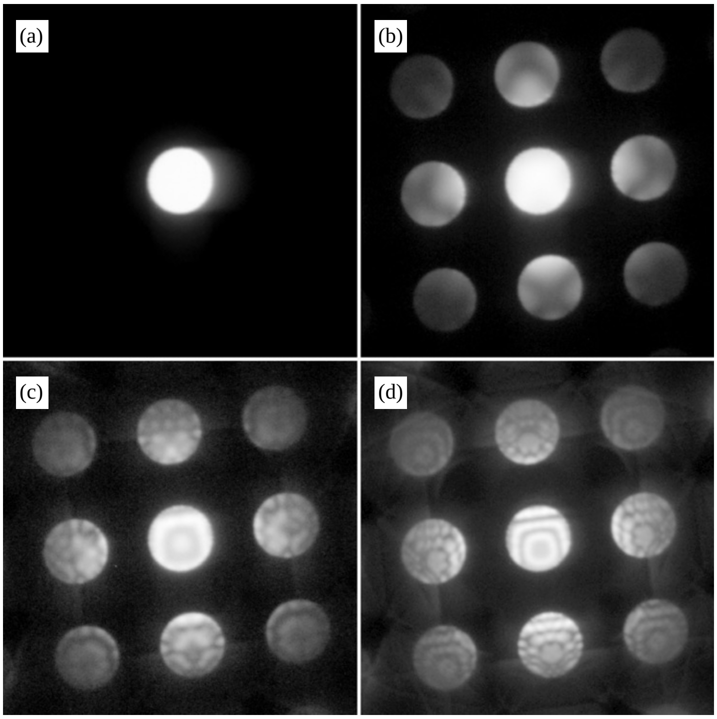

Fig. 2. Four energy-filtered convergent-beam diffraction patterns of clean silicon from a series of 20, taken with a 3.86 mrad condenser aperture. The patterns in the series were taken by moving the beam in steps of 10 nm and the thickness was determined from a combination of inelastic to elastic scattering ratios and simulations. The sample thicknesses shown are (a) 0 nm, (b) 49 nm, (c) 126 nm, and (d) 205 nm.

peaked close to diffraction maxima. However, under the present assumption the measurements represent the maximum effect that phonon scattering can have in reducing the contrast in a lattice image.

Fig. 3 shows the total scattering and the measured phonon scattering for the two condenser aperture sizes for an incident beam intensity of 1, plotted as a function of sample thickness. Total scattering includes all intensities in the area of the diffraction patterns out to a scattering angle of about $18\,\mathrm{nm}^{-1}$, while the measured phonon scattering is the diffuse intensity between the beams plus that estimated under the diffraction maxima by interpolation. The decrease in the total scattering with increasing thickness is a measure of the intensity that is either inelastically scattered or scattered to angles greater than the edges of the diffraction patterns. At all sample thicknesses, the phonon scattering measured with the small condenser aperture is larger than that measured with the large condenser aperture. This is because with the smaller condenser aperture, measurements can be made closer to the diffracted beams. As the phonon scattering is peaked at the diffracted beams, a greater proportion of the electrons scattered to small angles will be recorded. Even using the small condenser aperture not all the phonon scattering will be measured, so the total phonon scattering will always be underestimated. However, to a rough approximation, the small difference between the measurements for the small and large condenser apertures suggests that this underestimate is not great.

For a typical specimen thickness of 25 nm, the phonon scattering measured using the large condenser aperture is 0.049, compared to 0.060 using the small aperture. These correspond to 6% and 7.5%, respectively, of the intensity scattered to less than $18\,\mathrm{nm}^{-1}$ in an energy-filtered image being due

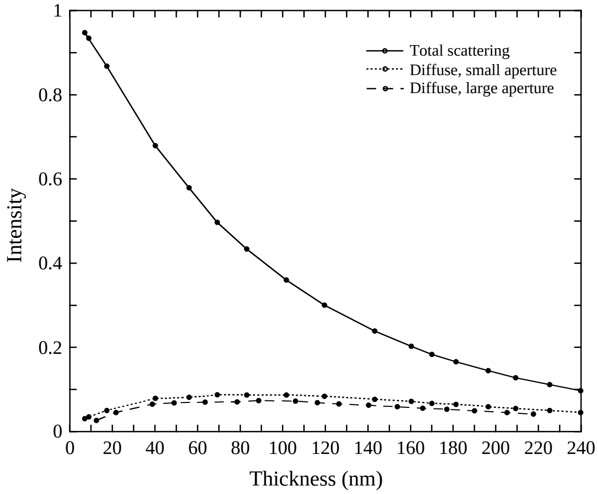

Fig. 3. Intensities measured from a series of convergent-beam diffraction patterns of silicon, like those in Fig. 2, as a function of specimen thickness. All intensities are normalised so that the incident beam has an intensity of 1. 'Total scattering' is the sum of all intensities in the energy-filtered convergent-beam pattern, up to a radius of about 18 nm^{-1}. 'Diffuse scattering' is the sum of all diffuse scattering between the diffracted beams plus that estimated to lie under the diffracted beams. The diffuse scattering was measured from diffraction patterns taken with both a small (1.11 mrad radius) and a large (3.86 mrad radius, as in Fig. 2) condenser aperture.

to phonon scattering. About 7.5% phonon scattering, if all contributing a constant background to a lattice image, would reduce the contrast to 92.5%, much less of a reduction than needed to account for the typical measured reduction in contrast, or Stobbs factor, of 2 or 3. It is not until the thickness is over 200 nm that sufficient phonon scattering is measured to produce a contrast reduction of a factor of 2. These figures should be compared to the 33% of the electrons that were found to be scattered diffusely by either phonons or amorphous materials for 25 nm of GaAs by Boothroyd [7].

3. Discussion and conclusions

The measurements of phonon scattering reported here are much lower than needed to account for the observed low contrast in experimental lattice images, suggesting that for a clean sample with no damaged or amorphous surface layers phonon scattering does not result in a significant contrast reduction. However, there are a number of complications to this conclusion.

Firstly, as mentioned earlier, phonon scattering is peaked at the diffraction maxima, making these measurements an underestimate of the true contribution of phonon scattering to lattice images. A measure of the amount of this underestimate based on the two condenser apertures used suggests that the underestimate is small, provided phonon scattering is not highly peaked at the diffraction maxima.

The assumption has been made that phonon scattering produces a constant background to a lattice image. This is a worst possible assumption and it is quite likely that at least some of the phonon scattering produces lattice images in the same way as for plasmon scattering, making the contrast reduction from the observed phonon scattering even less.

The microscope used for these measurements on clean silicon was not a high-resolution microscope, so lattice images obtained from the same area (Fig. 1) had a very low lattice fringe contrast. Quantitative comparison with simulations is therefore difficult because the lattice fringe contrast depends mainly on the effects of beam convergence, lens and high-voltage instabilities and specimen vibration, all of which (except beam convergence) are difficult to measure quantitatively. Ideally a higher-resolution microscope with a field emission source would have been used to minimise these effects, but this would have involved removing the sample from the high vacuum thus exposing it to the formation of amorphous surface layers. Thus it was not possible to prove that the Stobbs factor for images taken under clean conditions was still as high as 2 or 3. Indeed there is some evidence that the Stobbs factor is lower for clean specimens, such as annealed sapphire [8].

Most samples examined by high-resolution microscopy have surface amorphous material either through surface damage from sample preparation (e.g. ion milling), or from oxidation, or from carbon contamination during observation. Since high-resolution imaging is performed in the thinnest regions where surface amorphous layers are proportionally greatest, their effects will be seen mostly at low sample thicknesses. We have shown that the proportion of the electrons scattered by phonon scattering in a clean specimen is quite low (7.5% for 25 nm thick clean Si) compared to all diffuse scattering (33% for 25 nm thick GaAs) meaning that most diffuse scattering in typical slightly contaminated high-resolution specimens is due to amorphous surface layers and not phonon scattering.

On the other hand, Howie [9] has pointed out that thermal excitation of low-frequency vibration modes and rigid unit modes may be significant. These modes produce scattering at low q and are sharply peaked at the diffraction maxima, and thus would not be measured by the method used in this paper. It is thus possible that they may be causing enough diffuse scattering to significantly reduce the image contrast while not being measured by the method used here.

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