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Influence of total beam current on HRTEM image resolution in differentially pumped ETEM with nitrogen gas

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ABSTRACT

Environmental transmission electron microscopy (ETEM) enables the study of catalytic and other reaction processes as they occur with Angstrom-level resolution. The microscope used is a dedicated ETEM (Titan ETEM, FEI Company) with a differential pumping vacuum system and apertures, allowing aberration corrected high-resolution transmission electron microscopy (HRTEM) imaging to be performed with gas pressures up to 20 mbar in the sample area and with significant advantages over membrane-type E-cell holders. The effect on image resolution of varying the nitrogen gas pressure, electron beam current density and total beam current were measured using information limit (Young's fringes) on a standard cross grating sample and from silicon crystal lattice imaging. As expected, increasing gas pressure causes a decrease in HRTEM image resolution. However, the total electron beam current also causes big changes in the image resolution (lower beam current giving better resolution), whereas varying the beam current density has almost no effect on resolution, a result that has not been reported previously. This behavior is seen even with zero-loss filtered imaging, which we believe shows that the drop in resolution is caused by elastic scattering at gas ions created by the incident electron beam. Suitable conditions for acquiring high resolution images in a gas environment are discussed. Lattice images at nitrogen pressures up to 16 mbar are shown, with 0.12 nm information transfer at 4 mbar.

1. Introduction

The value of high-resolution transmission electron microscopy (HRTEM) images taken from samples while they are immersed in a gaseous environment is being increasingly recognized [1–5] and the recent availability of much improved dedicated environmental transmission electron microscopy (ETEM) instruments and holders is encouraging wider adoption. Many successful results have already been reported, particularly in the field of catalysis [1–13]. These researchers all used a differentially pumped ETEM system, which allows a pressure of about 20 mbar in the sample area while maintaining high vacuum in the region of the FEG [1,6,11,14], and without using gas-separation membranes. Several additional small apertures in the microscope column are used to separate regions at different gas pressure, which are pumped separately [5,6,11,14]. The alternative is an enclosed membrane holder to isolate the gas from the microscope vacuum. The design and function of a membrane E-cell holder is given in [15–17], the primary advantages being higher achievable gas pressure and lower system cost. Advantages of the differential pumping approach [1,14] include protection of the FEG, flexible use of normal holders, large field of view, usability at high temperature, high resolution imaging unobstructed by out of focus contrast from amorphous membranes, use in oxidizing environments without risk of membrane rupture, and improved reliability and repeatability of gas pressure measurement in the sample area. However, the achievable gas pressure is so far limited to around 20 mbar, and the gas path length (usually equal to the pole piece gap of the microscope) is normally 5.4 mm. E-cell holders with electron transparent membranes to contain the gas allow the use of higher gas pressures and the gas path length can be reduced [6,15–17].

In many ETEM applications the achievement of clear lattice contrast in the images greatly increases the value of the information obtained, and so one major goal of ETEM system development is to combine high gas pressure with high resolution imaging [2,3], which provides valuable information on crystal

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structure, shape and orientation and is much improved by image resolution close to 0.1 nm [4–6,10]. Image quality and resolution in ETEM experiments can be limited by various factors, including sample drift (which tends to be much worse during heating experiments or after changing the gas flow rate) and image noise (if fast acquisition time or low dose imaging is required). However, the basic system performance under ideal conditions is determined by the system design. It is widely acknowledged that achievement of high resolution TEM imaging becomes more difficult as the gas pressure increases due to scattering of the electron beam by the gas molecules [1–3,5,14,17–20]. However, until the study of Jinschek and Helveg [20], the relationship between achievable HRTEM image resolution and gas pressure had never been reported in detail with only a few studies reporting experimental results or calculations [17,19], and the mechanism for resolution loss by gas scattering is still unclear. We have attempted to measure and understand the main factors contributing to the achievable image resolution on our ETEM system using a standard gas (nitrogen), and we here report the effect of varying the total electron beam current (at constant beam current density), which has not been reported previously.

2. Experimental procedure

The microscope used is a spherical aberration corrected ETEM (Titan ETEM 80-300, FEI Company) with a modified S-Twin objective lens (pole piece gap and gas path length of 5.4 mm), and SFEG gun. It was operated at 300 kV. The gas used was nominally 99.99999% pure nitrogen, the bottle located about 15 m from the system. The ETEM uses a differential pumping system to allow up to 20 mbar of gas in the sample area while maintaining about 8×10^{-9} mbar of vacuum in the FEG gun area.

Information limit (which is equal to the point resolution because of the image C_s corrector) was measured at 860kx magnification using a CCD camera (US1000, Gatan) on a Tridiem 863 energy filter (zero loss energy filtering had little effect on the results). The sample is a widely available cross grating sample consisting of a high density of AuPd metal crystallites on an amorphous carbon film (S106 cross grating, Agar). We used a 1 s exposure time, shifting the image by 1 nm halfway. Fast Fourier Transforms (FFTs or diffractograms) of the resulting images were taken using the Gatan Digital Micrograph software and the extent of Young's Fringes was measured. Information transfer was found or is known to depend on many factors, including the focus setting, sample type, e-beam damage, microscope alignment status, energy spread of the electron beam, chromatic aberration, beam intensity, image magnification, exposure time, and sample drift. These were controlled as much as possible to achieve useful comparison measurements.

Most users of HRTEM in materials science image crystalline material lattices, but there is no widely agreed way of evaluating the resolution or quality of such lattice images. Still, the extent of the FFT of a (well oriented) single crystal lattice sample does provide a useful guide to the image resolution, and the data is more repeatable than Young's fringes when gases are present in the chamber. Silicon lattice images were taken with similar settings to the information limit but without image shift.

Nitrogen gas pressure in the chamber was adjusted with needle-valves and measured using a gas-independent Barocell pressure gauge nearby. Software maintains the gas pressure at a constant level during the measurements. Current flowing through the fluorescent screen was used to measure the electron beam current (this reading is calibrated in the factory), and the beam current was adjusted by changing the condenser aperture size and first condenser lens (spot size).

'Total beam current' means the current measured on the fluorescent screen when (in the absence of gas or sample) all the electrons passing down the column hit it. The 2nd condenser lens (C2) was used to spread and converge the beam, resulting in a change in current density on the specimen, but almost no change in the total beam current. Increasing the C2 aperture size increases the illumination area on the fluorescent screen but results in no change in current density at the specimen (only the total current increases). Thus, total beam current (nA) and beam current density (A/cm^2) at the sample can be controlled independently, and their relative importance was investigated. The study of Jinschek [20] refers only to current density, which is the usual measure of beam intensity. The beam current density is equal to the total beam current divided by the illumination area on the sample, and this area was measured on the bottom mounted CCD camera (after this has been done once, the relative beam current densities can be calculated using the counts per pixel on the CCD).

3. Results and discussion

The reduction in TEM image resolution as the chamber gas pressure rises is known, but probably not well understood. Fig. 1 shows the effect of increasing nitrogen gas pressure on the TEM image resolution, using a constant total beam current of 1.3 nA as measured without gas or sample. Both information limit on AuPd/carbon (extent of Young's Fringes) and the extent of a diffractogram of a silicon $<011>$ HRTEM lattice image are shown, as measures of resolution (a reference HRTEM image obtained from the AuPd/carbon sample is available as Fig. S1 in Supplementary material). The trends are similar, although the FFT extent for silicon is rather greater than the information limit (images are provided as Supplementary material, Fig. S2). The difference is notably larger at 16 mbar, perhaps because of the difficulty of accurately measuring the information limit at this pressure. Beam current density was adjusted to similar levels in each case so that CCD noise levels were about the same for all data points. Note that displayed screen current falls as gas pressure rises due to increased scattering or absorption [14], but the beam conditions were left unchanged. The information limit remains close to 0.10 nm up to 4 mbar, then drops to 0.13 nm at 8 mbar and about 0.23 nm at 16 mbar of nitrogen pressure, and silicon lattice information shows a corresponding drop, from 0.091 nm (the 244 reflexion) without gas to 0.15 nm with 16 mbar of nitrogen. So far this is as expected.

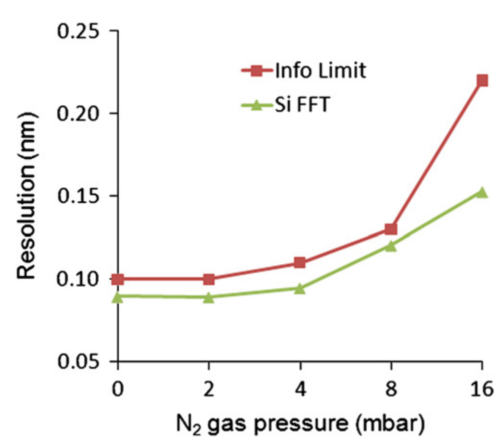

Fig. 1. Variation in TEM image resolution with increasing nitrogen gas pressure using a 1.3 nA total beam current. The upper line shows variation in information limit (Young's fringe extent) taken on a AuPd/carbon cross-grating sample. The lower line shows the extent of the diffractogram of a Si $<011>$ single crystal lattice image.