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IM.4.076 FTEM at atomic resolution in the chromatic-aberration corrected transmission electron microscope

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Energy-filtered transmission electron microscopy (EFTEM) is a technique that images a specimen using inelastically scattered electrons that have undergone a specific range of energy losses within the specimen $\Box -4\Box$. By selecting energy windows that cover a range of energies pertinent to inner-shell ionization of elements present in the sample, it is possible to obtain chemical maps and bonding information of different atomic species. However, up to now it was not possible to realize atomic resolution in EFTEM. Chromatic aberration degrades the image formed, as electrons that have lost different amounts of energy within an energy window will be focused in different image planes. This effect can be reduced by decreasing the width of the energy window. However this also leads to a reduction of the signal to noise ratio. Due to these competing effects, the resolution of energy-filtered images has been limited to about 4 Å $\Box 5,6\Box$. Recently chromatic aberration (C_C) correction has been implemented $\Box 7\Box$. We have used the FEI Titan 60-300 'PICO' instrument to demonstrate that C_C correction allows wide energy windows to be used and atomic resolution to be realized $\Box 8\Box$. Experimental EFTEM images of silicon were obtained using signal from the L_{2,3} ionization edge

(threshold energy for ionization ~100 eV). The microscope was operated at 300 keV with a semiconvergence angle for the incident beam of 1.7 mrad. The specimen, a wedge whose thickness varied between 10 and 40 nm, was imaged along the [110] zone axis. The three-window technique $\Box 3,4\Box$ was used with the pre-edge images centred at energy losses of 55 eV [Fig. 1(a)] and 75 eV [Fig. 1(b)] to do a background subtraction on the raw post-edge data, for the energy window centred at 120 eV [Fig. 1(c)], thus obtaining the Si elemental map shown in Fig. 1(d). Energy slit widths of 40 eV were used, which was only possible using the achromatic objective lens ($C_c = 0$). Overlapping windows were chosen in the pre-edge region in order to avoid positioning the first pre-edge window in the strongly oscillating plasmon-loss region. The atomic 'dumbbell' structure of Si in [110] projection is resolved, where the centres of the two atoms in each dumbbell pair are separated by 1.35 Å.

To understand the complex physics and electron optics underpinning the formation of the elemental map in Fig. 1(d) it is necessary to model the elastic and inelastic scattering of the incident electrons in the specimen. To do this we supplement the approach in Ref. 90 with the use of the quantum excitation of phonons (QEP) model 10, which calculates the underlying elastic and thermal diffuse scattering (TDS) of the incident electrons and provides the basis for modelling the ionization of the silicon atoms by both elastically and thermally scattered electrons. The transition potential approach allows to understand why the contrast of the 'inelastic' images is similar to that of the ordinary 'elastic' images ('preservation of elastic contrast'). For high-energy incident electrons causing ionization, the transition potentials become the more extended ('delocalized') the lower the energy loss. As a consequence features of the elastic and thermal scattering prior to and after the ionization event contribute directly and significantly to the energy-filtered image.

Our work has demonstrated experimentally that by using spherically and chromatically corrected electron optics the resolution of EFTEM can be improved to atomic. This allows unambiguous identification of the chemical nature of individual atom columns in the transmission electron microscope on the basis of images produced by electrons that have experienced a characteristic inner-shell excitation energy loss. However, the theoretical work clearly shows that carrying out atomic-resolution EFTEM can only be considered a technique satisfying modern standards of quantitative and quantum-mechanically sound image interpretation if the experimental work is accompanied by state-of-the-art image calculations including a proper treatment of the inelastic scattering. This confirms earlier objections against the intuitive interpretation of EFTEM results as (quasikinematical) maps of elements in samples imaged under spectroscopic conditions $\Box 11 \Box$

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Figure 1. EFTEM images used to construct the Si L_{2,3} elemental map by means of the three-window technique in the chromatic- and spherical-aberration corrected electron microscope. All images were taken using a 40 eV wide energy window. (a) Pre-edge image centred at an energy loss of 55 eV. (b) Pre-edge image cantered at 75 eV. (c) Post-edge image cantered at 120 eV. (d) Resulting atomic-resolution elemental map; a standard 'average background subtraction filter' $\Box 12\Box$ was applied. Standard conditions for the negative-spherical aberration imaging mode $\Box 13,14\Box$ were applied in all images, i.e. a C_s of -8.27 µm and a defocus of +40 Å