Imaging Buried Monolayers at Atomic Resolution using Electron Channeling

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Electron channeling in a crystal can in principle enhance the probe intensity at the exit surface [1, 2], thus leading to increased visibility of adatoms [2] or dopant atoms [3]. Here we use this effect to tune the contrast of buried monolayers by selecting the thickness of the crystal substrate to be close to the channeling maximum where entrance-surface effects are suppressed and the visibility of the buried monolayers is enhanced. Our theoretical and experimental studies provide us with new tools for studying nucleation and the early stages of crystal growth, which we apply to the formation of thin oxide crystals on Si.

We have conducted multislice simulations to study the effect of beam focusing in Si(100) crystals. Fig. 1 shows the beam profile at the entrance and at the exit surface of a 15nm thick, [100] zone-axis Si crystal. As the probe propagates through the crystal, a sharp channeling peak forms and the FWHM decreases by a factor of three to ~0.4Å. Here, the thickness of the crystal was chosen, so that it coincides with the channeling maximum (Fig. 2). Due to the large intensity of the channeling peak, an atom placed on a Si column at the exit surface of the Si crystal should contribute significantly more (~4 times) to the annular dark field (ADF) signal than at the entrance surface. Indeed, the visibility of a single Sr atom on a 15nm thick Si crystal simulated by multislice is strongly enhanced if the probe propagates through the crystal first before scattering off of the Sr atom. However, if the Sr atom is misplaced from the atomic column its visibility quickly decreases, tracking the channeling probe profile (Fig. 3).

To explore this effect experimentally, we grew 2.5uc SrTiO$_3$ on Si(100), capped with 15nm a-Si using MBE. While these layers have been studied during growth, a serious question for such thin layers is if their structure remains unaltered after they have been overgrown. Fig. 4 shows plan view ADF-STEM images of the thin, buried SrTiO$_3$ film where the sample was oriented so that the crystalline substrate constituted the entrance surface. We find the formation of islands of the SrTiO$_3$ film after capping with a-Si. Fig. 4b shows perovskite lattice fringes in the bright region confirming that these patches are due to SrTiO$_3$. The contrast of the SrTiO$_3$ islands can be maximized by choosing a region of the sample where the thickness of the crystalline Si is about 15nm which corresponds to maximum channeling. The average island thickness of 4uc (Fig. 4c) is consistent with a non-uniform SrTiO$_3$ coverage. When the sample was turned up-side-down so that the beam passed through the a-Si first, we were not able to resolve the SrTiO$_3$ lattice. In this geometry, narrowing of the probe due to channeling will occur only after the beam has passed through the SrTiO$_3$ layer and will not lead to enhanced visibility of this layer.[4]

References

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Fig. 1. Sharpening of an electron probe due to channeling in a crystal. Probe profile at the entrance and at the exit surface of a 15nm thick, [100] zone-oriented Si crystal, respectively.

Fig. 2. Channeling pattern for Si [100], where the probe intensity $|\Phi(z)|^2$ at thickness z is proportional to the change of the ADF intensity with respect to z.

Fig. 3. Multislice simulations of the visibility of a single Sr atom misplaced for an atomic column of a 15nm (dots) and 55nm (squares) Si crystal, respectively. The visibility decreases as the probe intensity (solid line) at the exit surface. The inset shows the calculated visibilities according to the incoherent imaging model.

Fig. 4. ADF-STEM image of 2.5uc SrTiO$_3$ grown on Si(100) and capped with 15nm a-Si. (a), (b) Plan view STEM shows the formation of SrTiO$_3$ islands and a non-uniform coverage. (c) The average thickness of the islands is 4uc as found by cross sectional STEM.