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Diffraction from Oxide Superlattices - Stacking Sequence Effects in Plan View.

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As nanoscience moves from observation to control, and electron microscope image interpretation becomes simplified by the new corrected TEMs, there is now a more realistic prospect than ever of both fabricating, with control at the atomic level, structures with wanted properties, and interpreting their bulk properties in terms of the atomic structures seen in the (S)TEM. Oxide superlattices, for example, may now be tailor-made by atomic-layer deposition to produce both conductive and ferromagnetic or antiferromagnetic interfaces by choice of stoichiometry and stacking sequence [1].

"Termination" or forbidden Bragg reflections (such as the (-422)/3 in silicon) have been used to image stacking faults in single-beam dark field TEM mode [2], since they are generated only by this additional plane of atoms in a thin sample with atomically rough surfaces. (Sharp termination can also generate these spots under UHV conditions). The perovskite structure , popular for digital oxide superlattices, has no such termination reflections, however the "difference" reflection (1,0,0) is extremely sensitive to stacking sequence, as shown in figure 1 . Here we show, with incident beam [001] normal to interfaces, the calculated intensity of the (100) reflection from the superlattice shown in figure 2 [3], compared with that from the ideal LaMnO3 structure. The superlattice contains an offset layer of SrMnO3. (A=LaO layer, a = MnO2 layer, B=SrO layer, prime indicates $a_o/2$ shear along [100]). The upper curve does not contain the sheared layer whose interfaces AB' and B'A may be responsible for observed ferromagnetism [1]. Plan view TEM single-beam dark-field images using this reflection would thus show large contrast differences between sheared and unsheared regions, and, if quantified, could provide a useful measure of both interfacial abruptness and compositional variation, provided samples of constant thickness could be prepared.

Spot diffraction patterns recorded with the beam in the plane of the interfaces show superlattice reflections as expected (figure 3) , indicating the high quality of the superlattice. We plan to collect these around 100K, where a peak is seen in magnetization ([3], fig 3(f)) to identify any associated structural changes. A quantititive analysis of digital superlattice reflections by QCBED [4], in order to map out the charge-ordering in real-space, is complicated by the large number of atoms involved in the supercell, and the small Bragg angles, causing overlap of orders. This may be addressed by modelling all but the few atoms at the electronically unique interface, and by recording Kossel-like diffraction patterns, in which a large overlap of orders is permitted, and simulations performed for those experimental conditions. The use of incoherently superimposed orders is preferable, since it renders the intensity distribution insensitive to the location of the probe [5, 6].

References

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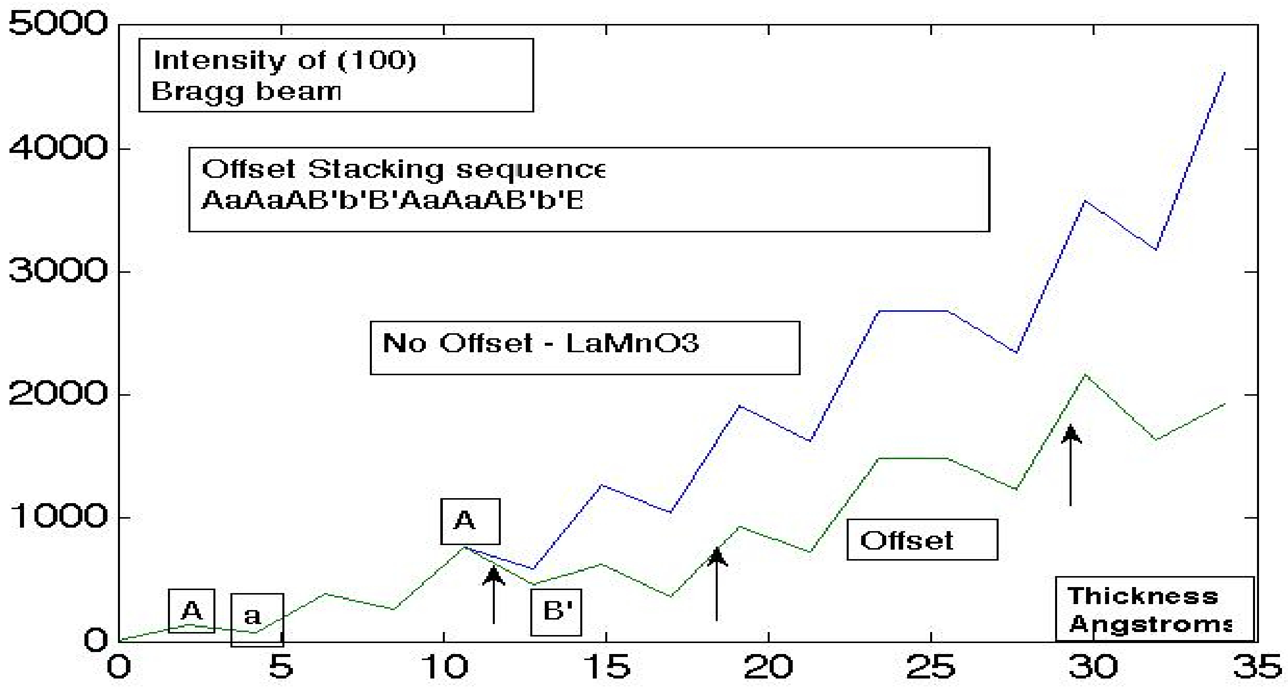

Figure 1. Intensity of (100) beam vs thickness with [001] incident beam for two stacking sequences.

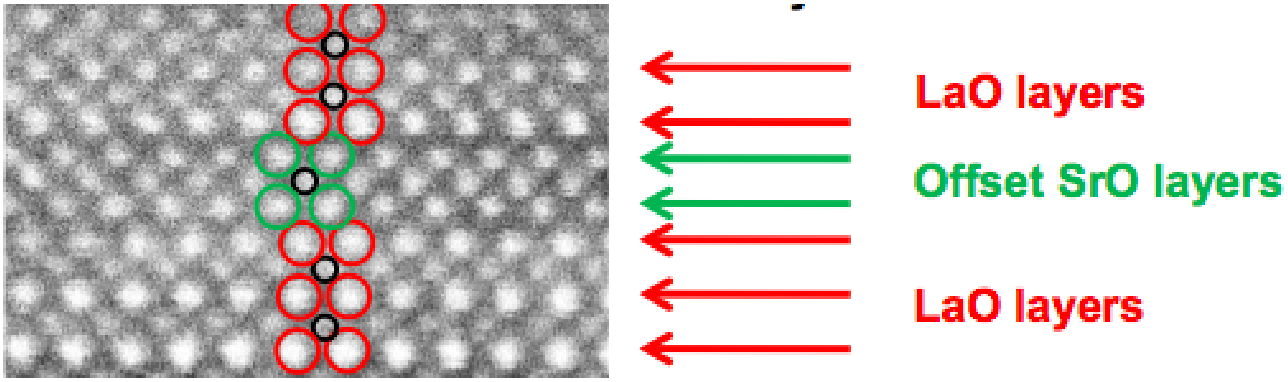

Figure 2. 200 kV ADF STEM image of perovskite digital superlattice from ref [3] (sample 0148) showing preliminary layer identification based on STEM contrast. [010] into page, [100] across.

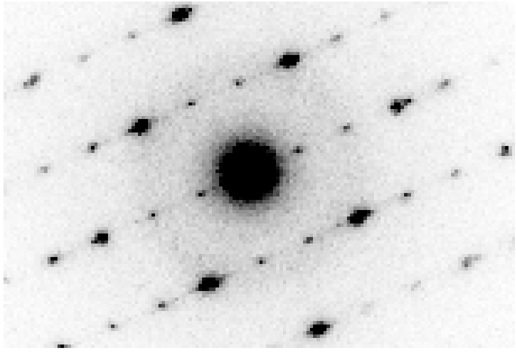

Figure 3. Transmission electron diffraction pattern at 200 kV down [010] from sample in figure 2.