Super Resolution Imaging of Complex Metal Oxides

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Summary: We report the results of super resolved image reconstruction of complex metal oxides. The complex reconstructed wavefunction provides information about both the cation and anion lattices at higher resolution than the conventional HREM image. Details of the methodology employed for data acquisition and processing are discussed.

1. Introduction

Indirect microscopy, wherein the complex specimen exit surface wavefunction is recovered from either a focal [1] or tilt azimuth [2, 3] series of images offers a number of advantages compared to conventional HREM. Firstly, the complex exit surface wavefunction can be recovered at higher spatial resolution. Secondly the wavefunction is recovered free of artifacts introduced by the objective lens aberrations. Finally, both phase and amplitude are recovered rather than the image intensity that is recorded by conventional HREM.

In this paper, restorations of complex oxides are presented with emphasis on the methodology used to recover the exit surface wavefunction from crystalline specimens and on the structural data obtained.

2. Experimental.

The oxides that have been investigated are $Nb_{16}W_{18}O_{94}$, which consists of pentagonal columns of metal cations projected along the [010] direction and the $A_nB_nO_{3n+2}$ layered perovskites $Nd_5Ti_5O_{17}$ and $Nd_4SrTi_5O_{17}$. In all cases, samples were prepared by crushing and dispersion of the raw material onto a holey carbon support film. The microscope (JEOL JEM 3000F FEGTEM, C_s=0.6mm at 300kV) was manually aligned to the coma-free axis, and the two-fold astigmatism was corrected with the aid of on-line power spectra. Both focal and tilt series were acquired on a 1024 x 1024 pixel CCD camera using Digital Micrograph scripts to automate the acquisition. For each data set 30 images were recorded at an initially overfocus condition with a nominal focal increment of -12nm between each image. A final image at the starting defocus was also recorded in order to assess the focal drift during the total collection time. Following the acquisition of each focal series a tilt azimuth data set from an adjacent area of amorphous carbon was additionally recorded, comprising initial and final axial images and four further images recorded with orthogonal beam tilts of 1 Gl/Sch. The residual odd aberrations present were measured via power spectra taken from this tilt series [4] and the defoci of the individual images used for the restoration were determined using a new method based on the measurement of the phase correlation function [5]. Importantly, this measurement does not require the presence of significant amounts of amorphous material and was thus well suited to reconstruction of these samples. The exit surface wavefunction was restored using a Wiener filter using methods previously reported [2, 3] and the blurring effects due to the MTF of the CCD detector were fully deconvolved [6].

3. Results and Discussion

Figure 1 shows the phase and modulus of the wavefunction restored from a [100] oriented crystal of $Nd_4SrTi_5O_{17}$ intergrown with $SrTiO_3$ in a well-ordered fashion. It is apparent that the modulus of the restoration contains high resolution information corresponding directly to the cation lattice sites which is relatively insensitive to the specimen thickness. The phase also gives similar information about the cation positions (recorded with reverse contrast) but also shows subsidiary maxima between the cation sites corresponding approximately to the anion positions. From both modulus and phase it is evident that the octahedral slabs are tilted (by *ca.* 5º) in a direction parallel to the ***b*** axis in a similar fashion to that reported for the analogous La substituted compound [7]. Within the region of $SrTiO_3$ several of the cation sites appear with reduced contrast in the modulus consistent with irregularly arranged cation vacancies in this material (figure 1(c)).

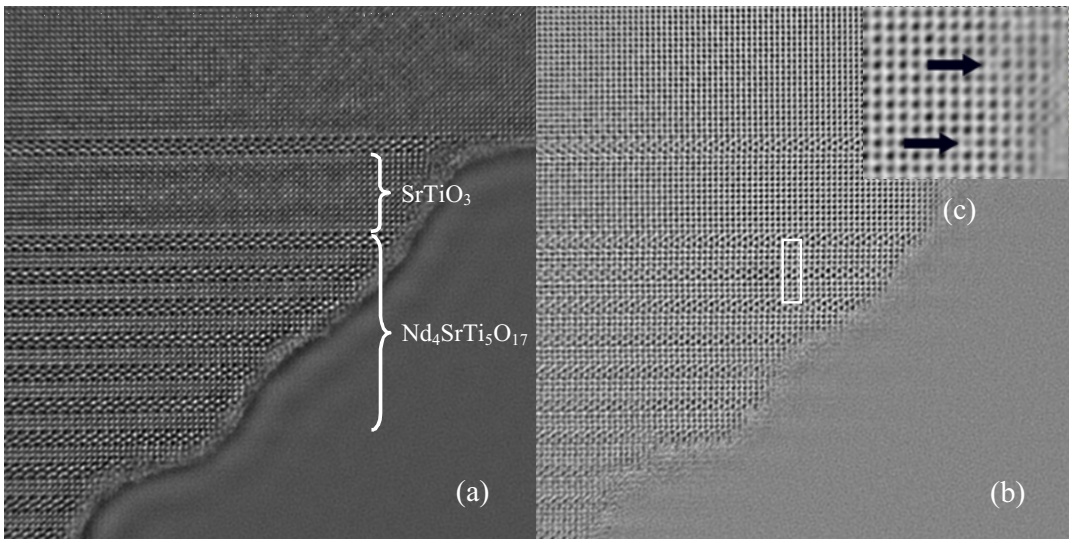

Figure 1: *(a) Phase and (b) Modulus of the restored wavefunction from a crystal of $Nd_4SrTi_5O_{17}$ in the [100] orientation. The $Nd_4SrTi_5O_{17}$ forms a well-ordered intergrowth with $SrTiO_3$ and the octahedral tilt mentioned in the text is clearly visible within the enclosed area marked in (b). (c) Enlarged region of (b) with local reductions in contrast at the cation sites indicated corresponding to cation vacancies within the $SrTiO_3$.*

4. Conclusions

Super resolved exit surface wavefunctions have been recorded from the complex metal oxide, $Nd_4SrTi_5O_{17}$ using a focal series of 30 images. The modulus of the restored wavefunction showed the cation lattice positions at high resolution over a wide range of specimen thicknesses. Local tilting of the slabs of octahedra within the perovskitic material is indicated together with the presence of ordered intergrowths of $SrTiO_3$. Within the $SrTiO_3$ regions, a number of irregularly arranged cation vacancies were also revealed. The phase of the restoration also revealed the cation lattice directly but in addition contained subsidiary contrast between the cation sites corresponding to the anion lattice.

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