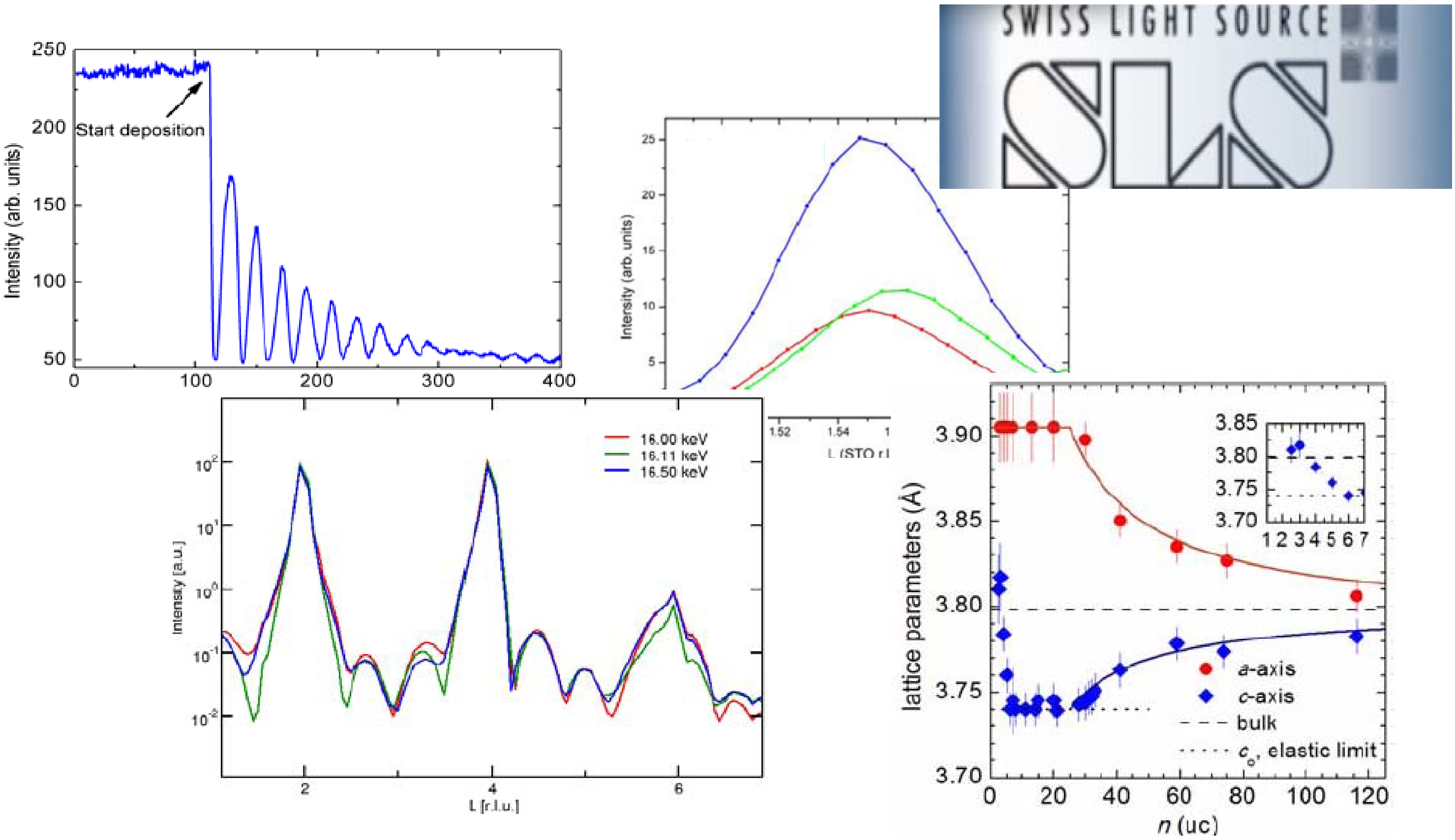

SLS Symposium on

Surface X-Ray Diffraction

Tuesday, April 5, 2011

10:00 to 12:15, WBGB/019

10:00 Structural studies of the metal-insulator transition in $LaNiO_3$ thin films
Steven J. Leake, S. A. Pauli, M. Schmitt, I. Kalichava, C. Cancellieri, M. Garcia-Fernandez, P. Aebi, R. Scherwitzl, P. Zubko, J-M. Triscone, and P. R. Willmott

10:30 Evolution of the Interfacial Structure of $LaAlO_3$ on $SrTiO_3$
Stephan A. Pauli, S.J. Leake, B. Delley, C.W. Schneider, M. Björck, D. Martoccia, S. Paetel, J. Mannhart, C.M. Schlepütz, and P.R. Willmott

11:00 Coffee

11:15 Electrostriction at the $LaAlO_3/SrTiO_3$ interface
Claudia Cancellieri, D. Fontaine, S. Gariglio, N. Reyren, S. J. Leake, S. A. Pauli, M. Stengel, P. Ghosez, P. R. Willmott, J.-M. Triscone

11:45 Better understanding of the LAO/STO interface conductivity: contribution of intermixing – study of a $La_{0.5}Al_{0.5}Sr_{0.5}Ti_{0.5}O_3$ thin film
Mathilde L. Schmitt, C. Cancellieri, S.A. Pauli, C.W. Schneider, S.J. Leake and P.R. Willmott

Structural studies of the metal-insulator transition in $LaNiO_3$ thin films

***S. J. Leake*[1], *S. A. Pauli*[1], *M. Schmitt*[1], *I. Kalichava*[1], *C. Cancellieri*[1], *M. Garcia-Fernandez*[2], *P. Aebi*[2], *R. Scherwitzl*[3], *P. Zubko*[3], *J-M. Triscone*[3], and *P. R. Willmott*[1]**

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The *R*NiO_3 perovskites are an exciting family of rare earth (R) nickelates exhibiting a sharp metal-insulator (M-I) transition with resistance increases of several orders of magnitude [1,2]. However, $LaNiO_3$ has proved to be an exception, it is the only nickelate not to exhibit a M-I transition in its bulk form remaining a metal. Scherwitzl *et al.* recently observed a M-I transition (20-40K) when grown as a thin film on $SrTiO_3$ below a critical thickness (t_c) of 8u.c (unit cells), and insulating behaviour at room temperature below 5u.c [3]. Physical properties of complex metal oxides can vary dramatically with structural change, here the lattice mismatch between film and substrate is ~1.7% so the film is strained. A detailed structural understanding is thus required if we are to shed light on the nature of the M-I transition in $LaNiO_3$ thin films.

Surface X-Ray Diffraction (SXRD) can resolve the entire atomic structure of the film under investigation with unparalleled sub-Angstrom resolution. The SXRD technique will be introduced, the direct methods [4] used to solve the phase problem discussed and the results of the film structure of $LaNiO_3$ described as a function of thickness around t_c. Superstructure signal arising from the tilting of oxygen octahedra was measured [5,6]. The film of thickness above t_c was found to reconstruct completely, just below t_c a change in the symmetry of the reconstruction was observed, see Figure. The insulating film did not exhibit a reconstruction.

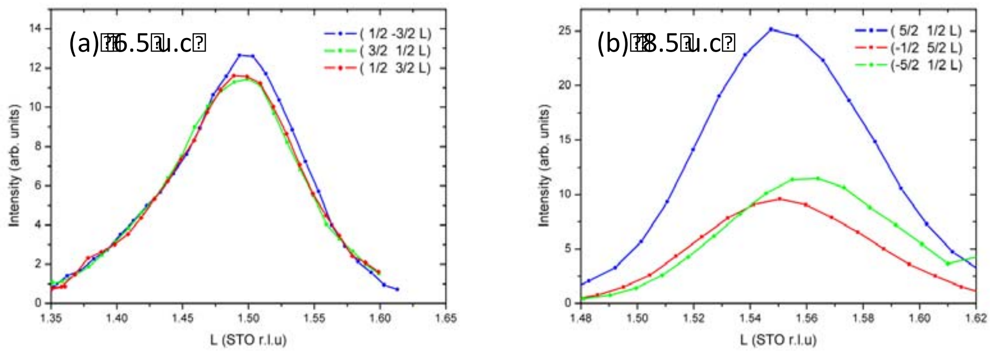

Figure: Expected symmetrically equivalent superstructure Bragg peaks for $LaNiO_3$ films with thickness (a) below t_c and (b) above t_c

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[6] S. J. May *et al., Phys. Rev. B*, **82**, 014110 (2010)

Evolution of the Interfacial Structure of $LaAlO_3$ on $SrTiO_3$

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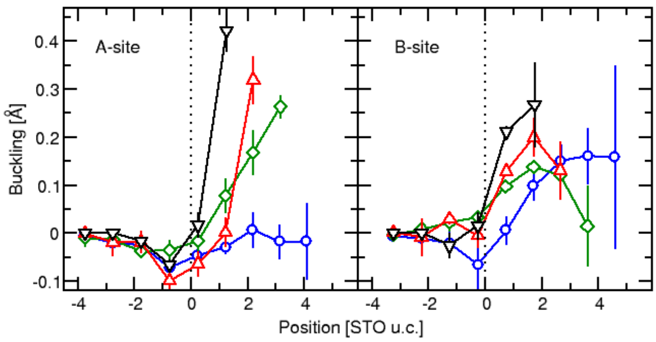
In complex metal oxides small atomic displacements can have a large influence of the physical properties. At surfaces and interfaces new phases and physical phenomena can even appear. Thus a knowledge of the atomic structure is a prerequisite for a deeper understanding of such processes. Surface x-ray diffraction with its sub-Angstrom resolution is an ideal tool to investigate such materials [1]. But, in all diffraction techniques, the lack of the phase information prevents a direct Fourier back-transformation of the measured intensities, which would reveal the atomic structure. In order to overcome the phase problem, direct-methods phase-retrieval algorithms have to be applied [2].

In this talk, the evolution of the atomic structure of $LaAlO_3$ grown on $SrTiO_3$ between two and five monolayers film thickness will be presented. This heterostructure between two insulators has attracted a lot of attention since the discovery of a quasi-2D electron gas at its interface [3]. The origin of the conductivity is however still under discussion. We find indirect evidence of an internal electric field in the polar $LaAlO_3$ from a depolarizing buckling between cation and oxygen positions, which decreases with increasing film thickness. We explain this in terms of competition between elastic strain energy, electrostatic energy, and electronic reconstructions [4].

The phase problem in the $LaAlO_3/SrTiO_3$ heterostructure was solved with an iterative algorithm. In a brief outlook, we will show the further development of our phase-retrieval attempts using the anomalous contribution to the atomic form factor around the Sr K-edge in $SrTiO_3/NdGaO_3$.



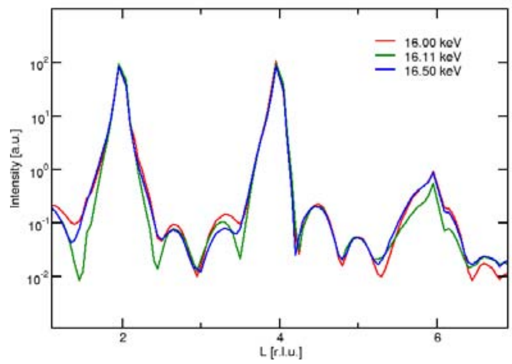


Figure: *Left:* The buckling (displacement) between the cation and oxygen position for the different film thicknesses. A- and B-site stands for the (Sr,La) and (Ti,Al) atomic layers, respectively. *Right:* The 22L crystal truncation rod of $SrTiO_3$ on $NdGaO_3$, measured at three different energies.

References:

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Electrostriction at the $LaAlO_3/SrTiO_3$ interface

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The appearance of a high-mobility electron gas at the interface between two band insulators, $LaAlO_3$ (LAO) and $SrTiO_3$ (STO) [1], has been the subject of intense research. The polar discontinuity at the interface between the non-polar (001) STO substrate and the polar LAO layer introduces an energetic instability associated with the divergence of the electrostatic potential. This can be solved by a transfer of charges to the interface [2]. This scenario is supported by the fact that metallic conduction is observed only when more than 3 unit cell of LAO are deposited on top of STO [3]. However, due the presence of defects like oxygen vacancies and atomic intermixing, experimentally measured at the interface, no general consensus on the origin of the doping has been yet achieved.

Here we study the lattice response of polar LAO layers to the screening configuration provided by the presence/absence of an electron gas at the LAO/STO interface, in order to probe the electric field inside the LAO layer.

We present a direct comparison between structural data obtained from experiments and ab-initio calculations for the electrostrictive effect on LAO thin films with different thicknesses grown on STO substrates. Our study shows that in polar materials the electrostrictive effect leads to a sizable lattice expansion which can be probed by x-ray diffraction. From the experimental data, a complete screening of the LAO dipole field is observed for film thicknesses between 6 and 20 uc. For thinner films, an expansion of the *c*-axis matching the theoretical predictions for an electrostrictive effect is measured (Fig.1).

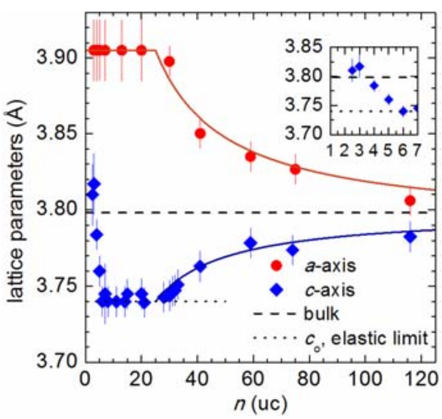

Fig.1: LAO *a* and *c*-axis parameters plotted as a function of the film thickness. The solid lines are a guide to the eye. The dashed horizontal line indicates the quasicubic LAO bulk lattice constant. The inset shows in detail the expansion of the c-axis for low LAO thicknesses.

References

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Better understanding of the LAO/STO interface conductivity: contribution of intermixing – study of a $La_{0.5}Al_{0.5}Sr_{0.5}Ti_{0.5}O_3$ thin film

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A quasi-2D electron gas was discovered in 2004 at the interface between the two insulating perovskites $LaAlO_3$ (LAO) and $SrTiO_3$ (STO) [1]. The conductivity only appears for an LAO film thicker than 3 monolayers. Several possible explanations for the origin of the electron gas have been given, among them: lattice distortions, electronic reconstruction, oxygen vacancies in STO, and cationic intermixing. Depending on the interface state and growth conditions, several of these mechanisms could influence the conductivity. Therefore the origin of the conductivity is still strongly debated.

Recent work presented in the previous talk support the electronic reconstruction scenario [2]. However, studies such as those by Willmott [3], Qiao [4] and Kalabukhov [5] concluded that intermixing also plays an important role regarding interface conductivity.
In order to enlighten the role of the intermixing for the electronic properties of LAO/STO interfaces, we studied $La_{0.5}Al_{0.5}Sr_{0.5}Ti_{0.5}O_3$ (LASTO) thin films grown by pulsed laser deposition (PLD) on STO substrates. Different parameters were modified such as O_2 pressure, laser fluence and substrate termination. Other LASTO thin films with different stoichiometries as well as conductivity and RBS measurements over a large deposition area are planned to be performed to link electronic properties with atomic structure.

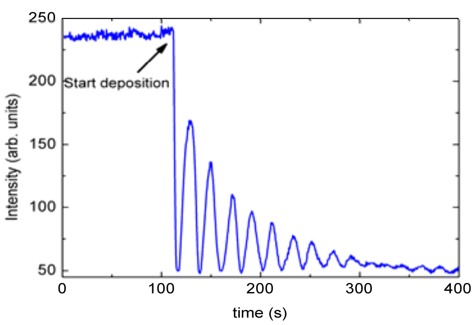

Figure 1

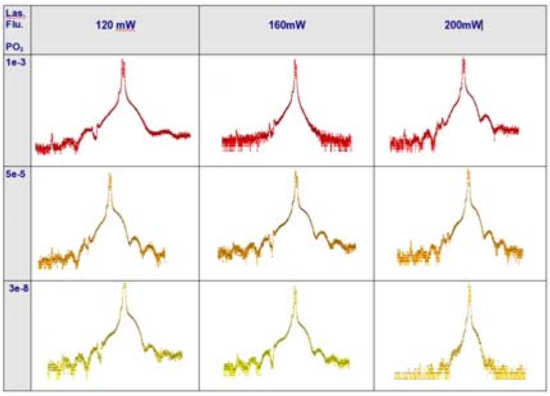

Figure 2

Figure 1: RHEED oscillations showing a layer-by-layer growth
Figure 2: comparison of the XRD Bragg peaks for different growth conditions

References:

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