Time resolved Bragg Coherent Diffraction Imaging and Fluctuation Spectroscopy of Oxide Thin Films

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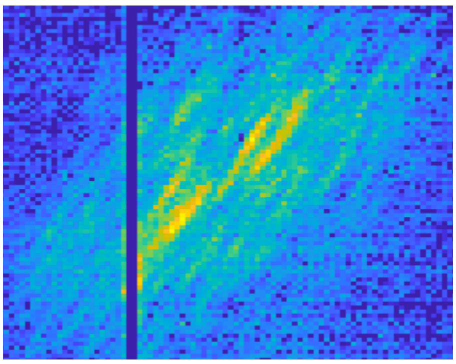
One of the great hopes for a high repetition rate XFEL is the possibility to "see" the critical fluctuations expected near a classical phase transition. In the early availability of the MID station, there is only limited temperature control. So we chose to look at a sample of $La_{2-x}Sr_xCuO_4$ (LSCO) for which the phase transition between the high temperature tetragonal (HTT) phase and the low-temperature orthorhombic (LTO) phase can be adjusted with the composition variable, x. We found in preliminary experiments that an epitaxial thin film of LSCO, x=0.10, had its HTT-LTO transition at 16C. When illuminated with a coherent X-ray beam, the sample showed strong speckles in its 103 Bragg peak, indicating the presence of a domain structure with ~20nm domains. Some of these arise from misfit with the $LaSrAlO_4$ (LSAO) substrate, but the speckle pattern changed at the transition, indicating the presence of some thermally activated domains as well.

I will present the results of our short beamtime at the MID instrument in July 2019. We planned to look for fluctuations of these domains near the phase transition, but technical interruptions prevented us from actually looking at the x=0.10 sample. We looked instead at an x=0.07 thin film of LSCO on LSAO, for which the transition is expected to be around 70C. We found the sample was undamaged by the full SASE2 monochromatic beam, even close to the 200nm focus of a CRL stack, so long as at least 30% attenuation was retained. This surprising radiation immunity may have arisen from the presence of a thin 200nm gold film covering the sample, which could redistribute photoelectrons and reduce charging effects; previously ferroelectric thin films were found to damage easily in a focussed XFEL beam.

The diffraction patterns are clearly recorded in single shots and seem to be reproducible over the 140 pulse trains, so could be averaged to give high-contrast diffraction patterns, like the one shown below. Train-to-train reproducibility was somewhat less. Domain counting arguments [1] can be used to construct models of the domain configuration and the asymmetric illumination geometry. Even though the beam profile is contained in the diffraction patterns, we have made some progress fitting the data to an array of small domains. We note that this phase problem may be amenable to machine learning approaches.



I am grateful for the contributions of a large team of researchers who attended the experiment and contributed to the progress towards the goal of visualising domain fluctuations [2].

[1] Domain Texture of the Orthorhombic Phase of $La_{2-x}Ba_xCuO_4$ Ian Robinson, Tadesse Assefa, Yue Cao, Genda Gu, Ross Harder, Evan Maxey, Mark Dean, Journal of Superconductivity and Novel Magnetism (2019)

[2] L. Wu, T. A. Assefa, M. P. M. Dean, E. S. Bozin, Y. Cao, H. You, D. Sheyfer, S. Rosenkranz, P. G. Evans, S. Marks, J. Diao, D. A. Keen, J. Hallmann, A. Madsen and I. K. Robinson to be published