EPJ Web of Conferences **41**, 03003 (2013)
DOI: 10.1051/epjconf/20134103003

Measuring 3D magnetic correlations during the photo-induced melting of electronic order in $La_{0.5}Sr_{1.5}MnO_4$

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Abstract. Time-resolved x-ray diffraction measures the dynamics of antiferromagnetic correlations by reconstructing the reciprocal-space scattering volume for the magnetic Bragg peak. Modifications in the scattering line shape along the three principal reciprocal lattice directions are measured.

Ultrafast dynamics in materials with strongly interacting electrons is a burgeoning field. Owing to the inherent electronic correlations in the materials, transient perturbations to the system result in strongly non-linear behaviour and the subsequent evolution into transient, metastable, states that persist for up to nanoseconds. Understanding and controlling the dynamics in these materials has therefore been envisaged as a platform for next generation magneto-optic, electro-optic and photonic devices. In nearly every case, however, the underlying microscopics of these transitions is complicated by the very interactions that make them possible: Electronic correlations. To make progress in this field, specialized techniques must be employed to unravel the effects of these correlations and to understand the evolution of these systems into the metastable states.

To gain further insight into the dynamics of correlated electrons during a photo-induced ultrafast electronic melting process, we employ Time Resolved Resonant Soft X-ray Diffraction (TR-RSXD)

to reconstruct the three-dimensional reciprocal-space scattering volume of the antiferromagnetic superlattice reflection in $La_{0.5}Sr_{1.5}MnO_4$ during a photo-induced electronic melting process. By measuring the full 3D scattering volume, we extract correlation lengths along the three principal lattice directions and monitor the evolution of the magnetic sublattice when the system is driven out of equilibrium. We show that the scattering volume does not evolve homogeneously, and therefore our technique provides information on the evolution of correlations over a span of length scales.

The material chosen for this study, $La_{0.5}Sr_{1.5}MnO_4$, is the canonical charge, orbital and spin ordered manganite [1]. In the present study, we focus on the magnetic degree of freedom, which can be probed using RSXD at a wavevector of (0.25 -0.25 0.5) below the Neel temperature, T_N= 120 K. The sample was mounted on a five-circle diffractometer in the Soft X-Ray (SXR) hutch of the Linac Coherent Light Source (LCLS), and cooled to 25 K. Soft X-rays, resonant with the Mn L_3-edge (E=642 eV, DE=1.5 eV), were focused to a spot of 250 mm diameter on the sample [2]. The pump beam (1.5 eV) and x-rays were brought together upstream of the diffractometer and collinearly propagated to the sample position. The ultimate temporal resolution is estimated to be about 250 fs and is limited by FEL – laser timing jitter. Both pump and probe impinge on the sample at a near-normal angle of incidence with p-polarization. The arrival time of the pump beam, relative to the x-rays was varied with a mechanical delay stage. Scattered x-rays were detected with an in-vacuum CCD camera capable of recording individual shots of the Free Electron Laser (FEL) [3].

Reconstruction of the 3D reciprocal space volume is aided by using a pixelated detector. Each pixel of the detector corresponds to a distinct position in reciprocal space (H K L), providing a means for parallel acquisition of many (H K L) points at once. Transformation of each image into reciprocal space is achieved by applying the appropriate UB matrix [4] to the scattering geometry. Through a combined motion of the sample angle, q and detector angle, 2q, an ensemble of images is acquired as the camera is moved through the scattering volume. The full 3D reciprocal space representation of the scattering volume is then reconstructed from these images.

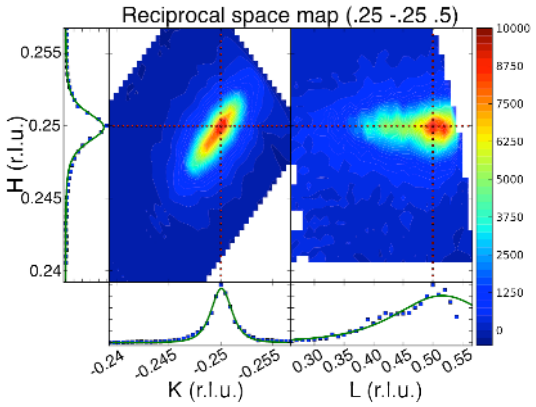

Fig. 1. Reciprocal space map of the antiferromagnetic reflection. Cuts through the scattering volume show elongation along the L direction due to poor correlations. Asymmetry within the HK plane is due to finite x-ray penetration depth.

The outcome of this transformation is shown in figure 1, in which slices through the reconstructed 3D volume are presented for the case of no laser excitation. Data along the L direction are limited by the maximum detector angle of 150° and thus a measurement of just over half the scattering volume is possible in this direction. Line cuts through the maximum in the scattering volume along the three principal directions in reciprocal space and corresponding squared Lorentzian fits are also shown. Extracted correlation lengths are $x_a = x_b = 540 \pm 10$ Å and $x_c = 20 \pm 5$ Å, in agreement with published results [5].

The temporal evolution of the magnetic scattering is shown in figure 2a, where we display normalized line cuts along (00L) at time delays before and after excitation by the laser. We see a transient shift to lower L values in the peak position 1.3 ps after excitation, which subsequently

recovers its original position once the sample has entered the metastable state, measured at 9 ps. Shown in figure 2b is the evolution of the 'order parameter', which we take as the scattering intensity of the peak of the scattering volume with vertical lines showing the temporal positions for which the 3D volumes are reconstructed. Importantly, the transient shift is apparent at the earliest time delays, on timescales we attribute to electronic processes and suggestive of electronic correlation effects.

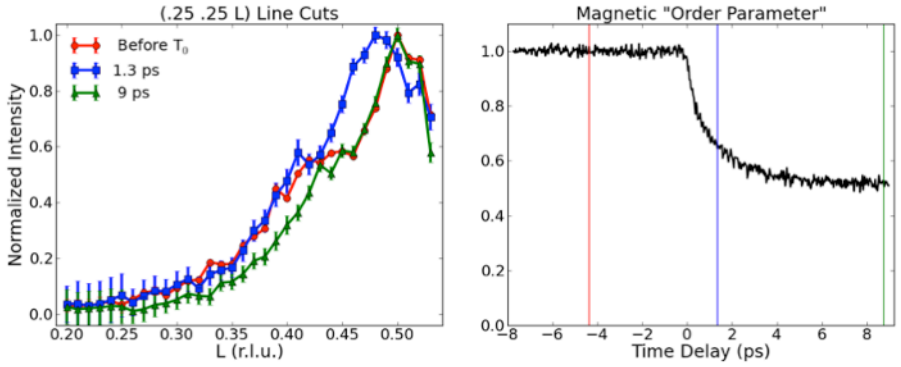

Fig. 2. a) Line cuts through the scattering volume for time delays before and after laser excitation. A transient shift to lower L values is seen at 1.3 ps which recovers when the sample is in the metastable state (9 ps). b) Magnetic 'order parameter' as measured by the time dependent intensity at the peak of the scattering volume.

The observed dynamics suggest a photo-induced incommensurate magnetic order at the earliest time delays, followed by at recovery and narrowing of the scattering lineshape along (000L). Experiments are underway to further clarify the importance of transient incommensurability in photo-induced transitions in strongly correlated oxides.

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