Orbital domain dynamics in a doped manganite

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Abstract. We explore a number of novel effects near the orbital-order phase transition in a half-doped manganite, Pr$_{0.5}$Ca$_{0.5}$MnO$_3$. To probe the unusual short-range orbital order in this system, we have performed coherent soft x-ray resonant scattering measurements in a Bragg geometry to measure dynamics. Near the transition temperature, we observe a small fluctuating component in the scattered signal that is correlated with three effects: a rapidly decreasing total signal and orbital domain size, as well as an abrupt onset of a broad background intensity that we attribute to the thermal production of correlated polarons. Our speckle results suggest that the transition is characterized by a competition between a pinned orbital domain topology that remains static and mobile domain boundaries that exhibit slow, temporal fluctuations.
1. Introduction

In transition metal oxides, such as cuprates, nickelates and cobaltates, spin and charge order are intimately coupled. Neutron scattering studies that can compare how correlations in spin and charge order grow with decreasing temperature provide important input for the theoretical models of these materials. For example, a comparison of spin and charge correlations (‘stripe phases’) in the copper-oxide plane of doped cuprates [1] suggest that the correlations are coupled, while in similarly structured cobaltates the spin and charge correlations develop independently, forming a glass-like state [2].

In the half-doped manganites, three-ordered phases—charge, magnetic and orbital—are coupled. In a useful, but oversimplified picture of these materials [3], there is one excess hole per pair of Mn sites. Below $T_{co}$ (or $T_{oo}$), these holes order into a checkerboard configuration [3] that creates a sub-lattice of Mn$^{3+}$ sites each with one electron in the $e_g$ degenerate 3d level (figure 1(A)). A cooperative Jahn–Teller (JT) distortion of oxygen anions surrounding the Mn$^{3+}$ sites lifts this degeneracy and sets the preference of the local orientation of the $e_g$ orbitals. Experimentally, charge and orbital order occur at the same temperature [4] ($T_{co} = T_{oo}$), while magnetic order occurs at or below [5, 6] the charge- and orbital-order transitions.

In the simplest—and longest held—picture [3] of half-doped manganites (see [7] and references therein), orbital and charge order [8]–[10] are seen as precursors to the magnetic ground state [3]. Recent resonant soft x-ray scattering measurements challenge this conventional picture of OO in half-doped manganites. In particular, it was found that the average correlation length of the orbital domains is $\sim$300 Å, about half the size of a typical magnetic domain [11, 12] ($\sim$700 Å) and much less than the correlation length associated with charge order [4] ($>2000$ Å). It remains unclear how to interpret these varying levels of mesoscale order between degrees of freedom that, in the simple model discussed above, are considered to be closely coupled. It should be noted that there have been some recent developments in the ordering models of the manganites [13], but because of the vectorial nature of the order parameter associated with orbital order, mean field theory is not capable of addressing the complex orbital phases. Strain fields [14], doping heterogeneity [15] and intrinsic structural defects [16] could all play a critical role in determining the short correlation length, in particular, of the orbital ordering.

One possible way to shed light on this transition and the role of these various factors is to study how the orbital correlations evolve dynamically as a function of temperature. To address this question, we use a coherent resonant soft x-ray scattering technique that is sensitive to temporal fluctuations in the orbitally ordered domain state in the half-doped manganite Pr$_{0.5}$Ca$_{0.5}$MnO$_3$ (PCMO). At temperatures 1–2 K below the OO phase transition, we find that the
Figure 1. Coherent scattering experimental set-up. The scattering geometry is set to measure the orbital Bragg condition at \( \mathbf{Q} = (0, 1/2, 0) \) in PCMO. Inset A schematically depicts the atoms of the Mn sub-lattice, the only sites probed with resonant scattering. The CE-type orbital-ordering of the \( e_g \) electron of Mn is shown for a single domain, illustrated in the \( a-b \)-plane. Coherent interference between scattered waves from the domain structure of the sample results in a ‘speckle’ pattern. Inset B shows a close-up view of one small region of the speckle image taken with a CCD camera, demonstrating the well-developed speckle with a fringe contrast of about \( \sim 80\% \).

average domain size decreases markedly with increasing temperature. Surprisingly, at constant temperature in this narrow range, the domain structure remains largely static but is accompanied by small-amplitude temporal fluctuations on the timescale of minutes. We suggest that this disorder may determine the short-range order as well as the overall magnitude of the domain-wall fluctuations. Our results indicate that studies in the time-domain are essential for a more complete portrait of the complex mesoscale physics of strongly correlated electron systems.

2. Experimental procedures

While most neutron and x-ray experiments measure the atomic shifts associated with the JT-distortion, it is the OO electronic wavefunctions that determine the sign and magnitude of the magnetic interaction between neighboring Mn sites [3, 8]. To increase the sensitivity and selectivity of an x-ray diffraction measurement to the ordering of electronic orbitals, the x-ray energy can be tuned near the Mn L\(_{\text{III}}\) absorption edge (\( \sim 650 \) eV), where the photon causes virtual electronic excitations between the Mn 2p core and 3d valence electronic levels [17]. In this case, the Mn form factor depends sensitively on the local electronic structure of the
3d levels and provides scattering contrast between Mn sites with distinct orbital occupancies and orientations. The L-edge resonant enhancement allows the orbital correlations to be probed directly as they develop near the OO transition temperature \([12, 18]\). In comparison, previous hard x-ray studies near the Mn K-edge have been demonstrated, both theoretically \([19]\) as well as experimentally \([20]\), to provide only indirect sensitivity to orbital ordering through the JT-distortion.

We combine this L-edge resonant enhancement with sufficient transverse coherence to enable measurement of temporal fluctuations of the orbital domain boundaries. Coherent illumination provides a more detailed measure of the orbital-ordered domains themselves and allows us to begin to address the issues surrounding the spin, charge and orbital correlation lengths discussed above. Our experiments were performed on Beamline 12.0.2.2 of the ALS at the Lawrence Berkeley National Laboratory with an energy band width of \(\sim 1\) eV. The partially coherent x-ray source is prepared by focusing the beam exiting our monochromator onto a \(\sim 10\) \(\mu m\) pinhole located 3 mm in front of the sample (figure 1). The transverse coherence length of the beam at the sample is \(\sim 5\) \(\mu m\), which means \(\sim 10^5\) orbital domains are coherently illuminated. With coherent illumination, constructive and deconstructive interference occurs between x-rays diffracted from different orbital-ordered domains, and the resultant scattering exhibits a characteristic speckle pattern \([21]\) (figure 1(B)). We exploit the fact that changes in the real space domain configuration will be reflected in changes in this speckle pattern to monitor orbital domain dynamics as a function of temperature, the x-ray analog of dynamic laser light scattering.

Similar dynamical measurements were attempted previously at the Mn K-edge (6.55 keV), though the signal rates were very low and definitive conclusions about OO domain fluctuations could not be drawn in that study \([22]\). Since coherent flux scales with the square of the wavelength, comparative experiments give much higher counting rates in the soft x-ray regime \([23]\). Rates such as these allow us to make more precise statements concerning the thermal fluctuation of the orbital domain structure. Here the coherent flux is in the range of a \(2-5 \times 10^{11}\) photons s\(^{-1}\), two orders of magnitude larger than that available at higher energies. The combination of higher scattering rates with the direct sensitivity to orbital order provided by L-edges is of great utility in studying complex phase phenomena in transition metal oxides.

The PCMO sample used in these studies exhibits two transitions, a simultaneous orbital- and charge-ordering transition near \(T_{oo} \sim 235\) K and an antiferromagnetic transition at \(T_N \sim 170\) K \([5]\). It is a twinned single crystal and is grown by the floating zone method, described elsewhere \([6]\). Using an orthorhombic lattice unit cell, the only orbital Bragg reflection on the Ewald sphere accessible at the Mn L-edge wavelength appears at \((0 1/2 0)\), giving a 2\(\theta\) scattering angle of \(\sim 124^\circ\). The surface of our crystal is misaligned from the \((0 1 0)\) plane by several degrees to move light diffusely scattered from the surface away from the Bragg scattered x-rays. The superlattice peak measured at \(G = (0 1/2 0)\) is then due solely to the resonant scattered signal from the modulation of the orbital wavefunctions of manganese. The experimental system allows control of sample temperature with 0.01 K precision. Images are collected with a directly illuminated \(2048 \times 2048\) pixel, thermoelectrically cooled charge-coupled device (CCD) detector.

3. Results and discussion

Images taken with the CCD of the orbital Bragg peak are shown in figures 2(A) and (C) and were collected at \(T = 200\) and 232 K, well below, and near \(T_{oo}\), respectively. These were collected
Figure 2. Time evolution of speckle. (A) Speckle pattern taken at the orbital Bragg peak deep within the orbital ordering phase at $T = 205$ K. The exposure time for this image was 10 s. (B) Time evolution of the speckle pattern in (A) for a vertical slice (yellow line) through the center of the image. This figure represents the evolution of a one-dimensional (1D) scan through the image like that shown in (A) of a 100-image set, taken over a 20 min period. The speckle structure is largely static as evidenced by the appearance of straight horizontal lines. Panels (C) and (D) are the same as (A) and (B) for $T = 232$ K, near the OO phase transition.

using 10 s exposures, indicating the high signal available in these studies and confirming the possibility of measuring slow dynamics of the orbital domains. The images exhibit two primary types of structure: a broad scattering envelope, related to the average OO domain properties, and high frequency speckle, related to the specific illuminated OO domain pattern. As discussed further below, we have systematically analyzed many images like these as a function of temperature and time to illuminate the close relationship between these average and specific domain properties.

3.1. Average orbital domain properties

We quantified the evolution of the scattering envelope as a function of temperature by fitting the images to elliptical Lorentzian functions. We found, however, that including a constant, isotropic
background was required to obtain a good fit, particularly near $T_{oo}$. Such fits essentially ignore the speckle structure and can be compared to previous x-ray or neutron scattering studies that were performed using poor transverse coherence. Our results are summarized in figure 3. To illuminate the development of the order parameter over a large temperature range, panel (A) plots both the intensities of the OO Bragg peak and of the background for a 100 K window. As has been observed previously [24], the former clearly delineates thermal destruction of the OO phase as $T_{oo} \sim 235$ K is approached from below, as evidenced by the disappearance of the Lorentzian weight. One additional piece of the puzzle is the background: it slowly appears near 200 K, increases suddenly just as the order parameter starts to decay rapidly in a narrow range 1–2 K below $T_{oo}$, and then saturates in the paramagnetic state. Based on previous studies using hard x-rays [25], though different in overall behavior, we attribute this background to the presence of correlated polarons which would produce an unstructured, diffuse scattered signal due to the inability to resolve lattice-sized structures at soft x-ray wavelengths. For instance, the background signal is observed to persist well above 300 K, consistent with earlier studies [25].

In addition to the changes in Lorentzian amplitude and background discussed above, figures 2(A) and (B) clearly indicate that the Lorentzian width is larger for $T \sim T_{oo}$ than for $T \ll T_{oo}$. After correcting for our scattering geometry, the ellipticity extracted from our fits is consistent with isotropic OO domains. The temperature dependence of this isotropic Lorentzian width is presented in the panel (B) of figure 3. Well below the transition, our measured width corresponds to an average orbital domain size of 300 Å. This is consistent with recent measurements on PCMO samples at different doping [12] as well as with several other cubic manganites [18]. As discussed in the introduction, the source of this short-range orbital order is not well understood. More importantly, we observe a dramatic broadening of the scattering envelope 1–2 K below $T_{oo}$. This broadening indicates a decrease in the average size of the orbital domains over the same narrow temperature range where the order parameter rapidly diminishes and the polaron background increases. The set of points in figure 3(B) indicates that the rapid change in Lorentzian amplitude, the onset and saturation of the polaron background, and the rapid decrease in orbital domain correlation length 1–2 K below $T_{oo}$ are all accompanied by an onset of slow dynamics of the orbital domains.

### 3.2. Orbital domain fluctuations

At a fixed temperature, a sequence of speckle patterns of the orbital diffraction peak separated by equal increments of time was recorded. The images shown in figures 2(A) and (C) are actually the first images in such movies, each consisting of 100 speckle patterns collected with a 10 s exposure, for a total measurement time of 20 min. Any large-scale domain wall motion would necessarily lead to changes in the speckle pattern that would be apparent in the time scans. The evolution of the speckle intensity as a function of time is shown for a vertical line of pixels on the CCD through the middle of the Bragg peak. This is presented for figures 2(A) and (C), as an image plot in figures 2(B) and (D), respectively. Straight horizontal lines are apparent in both time scans, indicating that the domains are largely static on the timescale of the data set. Surprisingly, this is true even at $T = 232$ K, which is within the narrow range where the dramatic changes in the scattering envelope discussed above occur. A more visual indication of the lack of strong fluctuations is offered by movies 1(A) and (B), which present a section of the images associated with figures 2(A) and (C), respectively. The constancy of the speckles is more obvious in movie 1(A) than in movie 1(B), as it is for one-dimension (1D) in the time scans for figure 2(B) versus figure 2(D). Partly this is due to the lower signal and consequent lower
Figure 3. Near-transition behavior. (A) The integrated intensity for the orbital ordering mechanism shown in red is plotted in units of $10^6$ average photons collected per second. The error bars are well below the size of the data points shown. The green plot shows the contribution of the total intensity due to a constant background. The increase of this contribution, which we believe to be due to polarons, occurs in the same temperature region of the transition as the order parameter decay. (B) The orbital peak width or average inverse correlation length is shown in yellow as a function of temperature, in the expanded region highlighted in the panel A. The average domain volume starts to decrease as the temperature approaches the transition from below, manifesting itself as an increase in the Bragg peak width. Well below the transition, the orbital domain size is of the order of 300 Å. The purple points show the correlation coefficient, shown in the maps in figure 4, averaged over long times at a given temperature. The point where the fluctuations ‘turn on’ is in a narrow window right near the orbital ordering phase transition.

signal-to-noise ratio available at the higher temperature, but the result at 232 K nonetheless indicates that the orbital structure remains largely static.

To measure the dynamics more thoroughly, we systematically raised the sample temperature in one degree increments through the transition, waiting for 1 h for the system to stabilize at each point before collecting a movie. We make measurements with temperature always increasing to maximize the thermal stability of our apparatus and to minimize the impact...
Figure 4. Cross-correlation coefficient maps. Each frame represents the average of correlations for three separate maps for a given temperature. Time-dependence is calculated by averaging over all pairs of images separated by delay time $\tau$. The quantity $q$ was calculated by computing all three components of the wavevector transfer $q = Q - G$ and taking the magnitude, where the wavevector magnitude $|k|$ is $2\pi/\lambda$, $Q$ is given by $k_f - k_i$, and $G$ is the orbital ordering reciprocal lattice vector. The $q$-resolution shown in the map is 0.01 nm$^{-1}$ per pixel. Even for the $q$ and $\tau$ dependence shown above, the absolute changes are small.

of thermal hysteresis in our sample. We then calculated momentum-resolved cross-correlation coefficients $\langle \rho(q, \tau, T) \rangle$ to quantify the fluctuations of orbital domains. The coefficient is normalized such that $\rho = 1$ for two perfectly correlated speckle patterns, and $\rho = 0$ for completely uncorrelated patterns. Similar computations were performed by Pierce et al [26] to probe return point memory in magnetic films. Here, however, we are interested in the change in the average coefficient as a function of $\tau$, the time between two images. The $q$-component is the magnitude of the vector $q = k_f - k_i - G$, where the $k$-vectors are the initial and final wavevectors of the photons (figure 1) and $G$ is the reciprocal lattice vector of the orbital ordered phase, $G = (0120)$ in an orthorhombic unit cell. Regions of approximately constant magnitude of $q$ form annuli centered on the orbital peak in our images. Correlating the speckle pattern within these annuli, the average $\rho(q, \tau)$ is computed using

$$\rho(q, \tau) = \frac{\sum_{t=1}^{n-\tau} A(q, t) \times A(q, t+\tau)}{(n-\tau) \times N(q, t, \tau)},$$

where $n$ is the number of images, and $\times$ represents a cross-correlation operation. $N(q, t, \tau)$ is a normalization constant that is a function of both the real time $t$, and image separation time $\tau$, given by:

$$N(q, t, \tau)^2 = A(q, t) \times A(q, t) \times A(q, t+\tau) \times A(q, t+\tau).$$
Movie 1. Static structure with small fluctuations, available from stacks.iop.org/NJP/10/053023/mmedia. The movies shown in panels (A) and (B) correspond to the same small region of the speckle patterns in figures 2(A) and (C), respectively. Movie (A) was collected at $T = 205$ K, well below $T_{oo}$, and shows static structure over its entire duration of 1200 s. Movie (B) was collected at $T = 232$ K, very close to $T_{oo}$, shows mostly static structure as well though the contribution from statistical noise is higher due to the lower signal available. The movies are both 100 frames and complement the visual 1D time scans shown in figures 2(B) and (D).

The correlation map $\rho(q, \tau, T)$ averages over all pairs of images separated by time $\lambda$ within a 100-image set for a given temperature $T$. Multiple image sets per temperature are then averaged to obtain $\langle \rho(q, \tau, T) \rangle$.

A sampling of the correlation map results is presented in figure 4. For temperatures less than $T = 231$ K, $\langle \rho(q, \tau, T) \rangle$ is very close to one for all delay times and wavevectors measured, confirming that the low-$T$ speckle patterns are in fact static and that the experimental set-up and the analysis procedure is stable enough for the detailed measurements outlined here. The correlation maps for $T = 232$ and 233 K show small, but measurable deviations from unity. In addition, the contours show a downward slope at increasing $q$ for $T = 232$ K. This $q$-dependence is consistent with smaller length scales fluctuating faster, and decorrelating sooner, as might be expected. Though it seems that the fluctuations are longer in the map at $T = 233$ K, we believe this is due to the fact that the window where these dynamics can be measured is extremely narrow. As such, at 233 K, the fluctuations have become faster than the exposure time of each CCD image in the experiment, and we are unable to resolve the short time behavior with accuracy. Two key points arise from this analysis. Firstly, only a small fraction of the domains structure is fluctuating, even right in the vicinity of the transition, as evidenced by the small deviation of $\langle \rho(q, \tau, T) \rangle$ from unity. Secondly, this small fluctuating fraction comprises real material physics with slow, measurable dynamics near the transition. The average magnitude of this deviation of $\langle \rho(q, \tau, T) \rangle$ from unity is plotted for more temperatures in figure 3(B). The dramatic onset of this small fluctuating component in our speckle patterns evidently occurs over the same narrow temperature range where we observed changes in the scattering envelope.
4. Conclusions

The first result discussed above can be seen from the fact that even at the longest times measured, $\langle \rho(q, \tau, T) \rangle$ does not decay by more than 10%. The system behaves as though it has frozen-in disorder that prevents any sizeable fluctuations in the orbital domains on the timescale of the experiment, even very close to $T_{\text{oo}}$. The short-range order in the orbital-ordered phase, then, is apparently related to this frozen-in disorder. Although x-rays at these energies only penetrate 500–1000 Å into the sample, we believe it is unlikely that this pinning is due to defects at the surface. In particular, we note that the size of the domains observed here, and the temperature dependence of the order parameter, are both comparable with the results of neutron and hard x-ray measurements, which probe the bulk of the material [4, 22, 27, 28]. In addition, short-range orbital order, with orbital domains of $\sim$300–500 Å, appears to be a general result from the more direct L-edge scattering studies on a variety of cubic manganite materials prepared in different laboratories [11, 12, 18]. These arguments suggest that the pinning of the domain walls is not surface related, but is intrinsic to the system. Given this, domain walls may be expected to be pinned at points where the cost in elastic or electronic energy for having neighboring OO wavefunctions out of phase is lowest. One candidate for such a location is the Ca-dopant ions and the strain fields they produce. This is at odds, however, with the fact that the sample is half-doped, meaning the average separation between dopants is much smaller than the orbital domains. If this is the responsible mechanism, then clearly the pinning is not very energetically relevant. It is possible nonetheless that this is a cumulative effect associated with a local statistical deviation from the average calcium concentration which produces a strain field that pins the domain walls. These regions of large variation from the average doping concentration could be abundant. Another issue that may be related to the static structure is the level of hole doping. With an equal number of divalent and trivalent sites, the ordering is thought to be more robust with respect to perturbations, and hence, to equilibrium domain dynamics.

We now turn to the small-amplitude fluctuating component that emerges only near the phase transition (figure 4). The dynamics are apparently related to limited domain wall motion and exhibit fluctuations that occur on the order of hundreds of seconds. These long timescales are slower than any measured mechanism that we know of at this time. These results provide a clue that understanding the orbital ordering transition in the manganites may be through mesoscopic, rather than microscopic physics. Furthermore, not only are the dynamics slow, but they are also associated with domains that occur on large length scales, about 50 lattice spacings.

An interpretation that is consistent with our results is a picture in which, at a given temperature, the bulk of a domain volume is static. The domain walls are pinned except for temperatures very close to the transition where the domain walls are able to execute small-amplitude motion. Such behavior contrasts, for example, with the domain wall dynamics associated with charge density waves in the simple antiferromagnet Cr [29]. In chromium, the domains are a few microns in size and fluctuate on a length scale comparable to the domain size itself, resulting in speckle patterns that become completely uncorrelated at long times [29]. Thus, it appears that in contrast with these systems, the manganites possess a source of quenched disorder which couples to the OO-order parameter. This coupling pins the domain walls, and though critical fluctuations are absent in this regime, may demonstrate that the manganites are more analogous to recent observations in the displacive, SrTiO$_3$ antiferrodistortive transition [30], a second-order structural phase transition. However, the onset
of the unusual, slow orbital domain wall motion is a completely novel observation, unlike either of these systems.

In the manganites and other complex oxides, the coupling of multiple degrees of freedom—charge, spin and lattice—has mostly been considered at the microscopic level. Here, we argue that a thorough understanding of the mesoscopic physics [31] is also needed to shed light on the governing mechanisms of the macroscopic properties. Indeed, the interactions between order parameters are complex and involve a range of length scales. Our results show that a great deal of information can be collected by probing these systems in the time-domain. In particular, we have shown a number of new results regarding the orbital transition, including that short-range orbital correlations are largely static with only small-magnitude, domain wall fluctuations that become manifest only as the transition is approached. This highlights the importance of quenched disorder in this system for determining its ground state properties. It is also interesting to note that the short-range orbital order is not universally observed in doped manganite single crystals. For instance, in some layered manganite systems, the domain size has been shown to be much larger [32]. It seems intuitive to assume that these longer-ranged ordered systems may lack appreciable quenched disorder, giving a larger probability that motion associated with domain walls will be more considerable. Finally, if the domain wall dynamic measurements reported here can be applied to other complex oxides, such as the dynamic stripe correlations in the high-temperature superconductors [33], then many forms of time-dependent heterogeneity may be readily probed, helping to reveal a new understanding of the interconnection between mesoscopic physics and complex order parameters.

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