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FOR MORE INFORMATION

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Orbital and Spin Correlations in a Manganite Probed with Soft X-ray Resonant Diffraction

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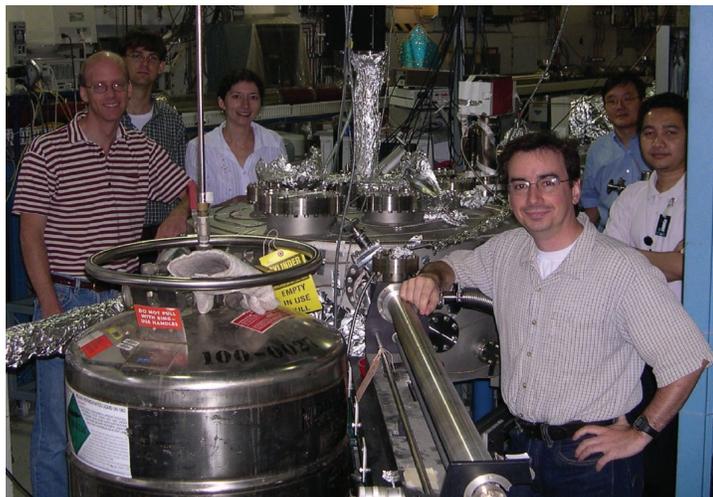
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Soft x-ray resonant diffraction was used to directly probe spin and orbital correlations in a near-half doped manganite. The diffraction was performed at the manganese (Mn) L_{II} and L_{III} absorption edges, providing a sensitive spectroscopy of the Mn 3d states in the spin and orbitally ordered phases. These measurements suggest that the established "checkerboard" model for charge ordering of the Mn^{3+} and Mn^{4+} ions is too simplistic, and reveal a surprising discrepancy between the orbital and magnetic correlation lengths.

In doped manganites of the form $RE_{1-x}A_xMnO_3$ (where RE is a rare earth and A is a divalent element), the magnetic coupling between Mn sites depends on the overlap of the Mn 3d electron orbitals. Therefore, a complex behavior arises in these materials when the direction of the highest occupied Mn 3d orbital is itself a degree of freedom. For example, the Mn 3d orbitals can undergo long-range order, usually in association with cooperative distortions of the oxygen octahedra that surround the Mn sites. Understanding how the orbital physics drives the overall ground state necessitates a direct probe of both orbital and magnetic order.

orbital ground state is shown in **Figure 1**. The incident energy at a magnetic, $(\frac{1}{2} 0 0)$, or orbital, $(0 \frac{1}{2} 0)$, Bragg peak was tuned through the Mn L_{II} and L_{III} atomic absorption edges (~ 650 eV). At

strength of the $2p \rightarrow 3d$ resonance at an Mn site depends on the local charge distribution of the occupied 3d orbitals. This leads to a large contrast between the resonant scattering factors on sites 1 and 2 (**Figure 1**) and a large enhancement of the orbital $(0 \frac{1}{2} 0)$ Bragg peak. The resonance matrix element also depends on the direction of the spin in the 3d levels, resulting in magnetic resonant scattering at the antiferromagnetic Bragg peak, $(\frac{1}{2} 0 0)$. The magnetic enhancement at the L-edges is truly enormous – off resonance, the magnetic scattering is too weak to be observed in these materials – and thus provides a unique opportunity to directly compare the orbital and magnetic correlations in a manganite.



The experimental team. From left, John Hill, Peter Abbamonte, Jessica Thomas, Young-June Kim, Andriwo Rusydi, and Stephane Grenier.

We used resonant x-ray diffraction to directly probe the magnetic and orbital order in the near half-doped manganite $Pr_{0.6}Ca_{0.4}MnO_3$, for which the proposed spin, charge, and

the L-edges, core Mn $p_{1/2}$ and $p_{3/2}$ electrons are resonantly excited into unoccupied 3d levels, which enhances the Mn sites' contribution to the diffracted intensity. The

Figure 2(a) shows the orbital and magnetic resonant line shapes (energy scans at fixed Q). The features

in the line shapes are the different excited states in the $3d$ band, which are probed with increasing incident energy. The 3 eV shift in spectral weight and the large difference in intensity between the orbital and magnetic spectra suggests that

the checkerboard charge-ordered picture in **Figure 1** is too simple [see publication]. Furthermore, longitudinal scans through the orbital and magnetic Bragg peaks show that the orbital peak is approximately two times wider than

the magnetic peak (**Figure 2(b)**). This suggests that the orbital correlations are shorter ranged than the magnetic correlations, a result that appears at odds with orbitally driven magnetic order.

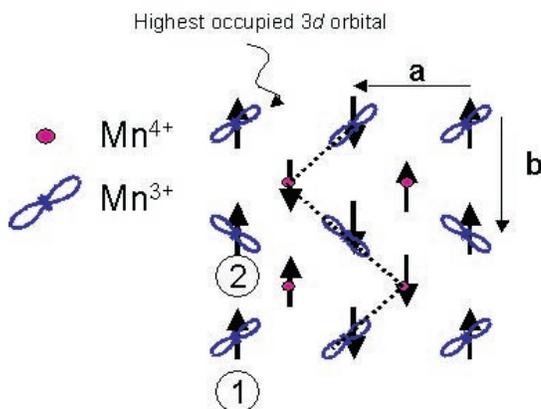


Figure 1. Schematic of the ground state exhibiting charge-orbital-magnetic order in $\text{Pr}_{0.6}\text{Ca}_{0.4}\text{MnO}_3$, showing only the Mn sites in the Mn-O plane. In $\text{Pr}_{0.6}\text{Ca}_{0.4}\text{MnO}_3$, the charge order is modeled as a checkerboard of Mn^{3+} and Mn^{4+} ions and only the Mn^{3+} ions have the orbital degree of freedom. (The symmetry of the highest occupied electron orbital is indicated by a blue lobe). The orbital zig-zag chains couple the Mn sites ferromagnetically along the chain and antiferromagnetically between the chains. The orbital order doubles the unit cell along **b**, leading to $(0 \frac{1}{2} 0)$ type reflections, while the spin order doubles the unit cell along **a**, leading to $(\frac{1}{2} 0 0)$ reflections.

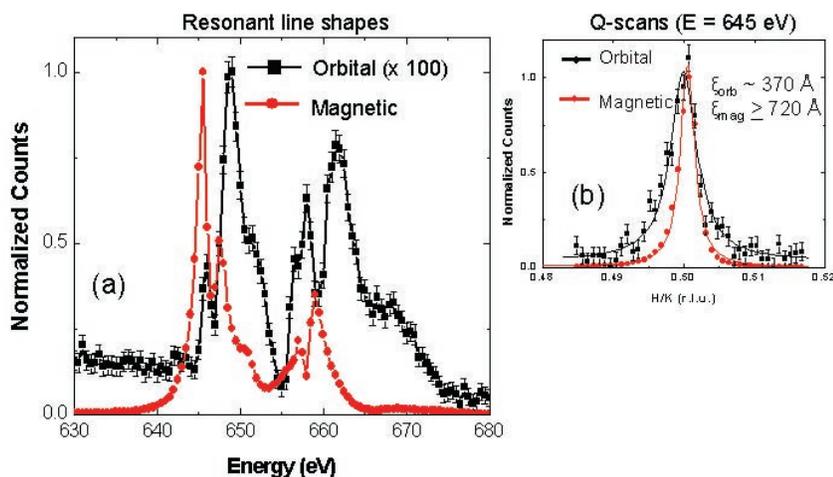


Figure 2. (a) Resonant diffraction line shapes at fixed \mathbf{Q} at the orbital $(0 \frac{1}{2} 0)$ position (black curve) and the magnetic $(\frac{1}{2} 0 0)$ (red curve). Note that the magnetic resonant line shape is peaked ~ 3 eV below the orbital line shape. (b) Longitudinal \mathbf{Q} scans through the magnetic and orbital Bragg peaks, at the same energy.