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Emergent Phase Coherence of Stripe Order in Manganites Revealed with Cryogenic Scanning Transmission Electron Microscopy

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Low temperature phase diagrams of *strongly* correlated systems reveal complexity and competition between mismatched orders, best exemplified in manganites where the inhomogeneous coexistence of metallic domains with charge-ordered patches results in colossal magnetoresistance [1]. Charge order, or stripes, is a prevalent electronic instability where electrons form periodic patterns, breaking lattice symmetry and competing with other phases. Disorder is thought to govern the emergence of the low temperature striped phases, causing, for instance, a tendency for gradual crossover rather than sharp phase transitions [1]. Real space visualization of the evolution of stripe order promises a deeper understanding of the onset of ordered phases.

Dark-field transmission electron microscopy has allowed direct observation of stripes in manganites with nanometer resolution [2]. In those studies, contrast modulation is thought to originate from the choreographed expansion and contraction of oxygen octahedra due to charge localization and the Jahn-Teller effect. However, direct, atomic-scale mapping of stripes and their temperature dependence is still lacking. Here we demonstrate atomic-resolution high-angle annular dark field scanning transmission electron microscopy (HAADF STEM) at room and cryogenic temperatures (~93K), allowing vivid visualization of the evolution of stripes in $Bi_{1-x}(Sr,Ca)_xMnO_3$ (BSCMO), a model charge-ordered system with T_c ~300K. Room temperature and cryogenic HAADF imaging was performed in an aberration-corrected FEI Titan Themis operating at 300kV, using a 30mrad convergence angle and 68-340mrad collection angles. Due to reduced stage stability at cryogenic temperatures, we acquired stacks of 20-30 images using a 1-2µs/pixel dwell time and applied rigid-registration schemes to align and average frames.

Figure 1(a) shows a HAADF image of BSCMO at 93K, with the Bi/Sr/Ca (green) and Mn (red) columns clearly resolved, highlighting resolution and signal-to-noise ratio on par with room temperature imaging. The Fourier transform amplitude of the lattice image contains superlattice peaks decorating lattice Bragg peaks, reflecting the presence of a modulation with ~3 unit cell periodicity. We map the real space structure of the modulation that gives rise to the observed peaks. As shown in Fig. 1(c,d), stripes originate from picometer scale periodic lattice displacements of the cations. The order parameter (OP) is $\boldsymbol{\Delta}(\boldsymbol{r}) = \Re e\{\mathbf{A}(\boldsymbol{r})\exp[i\phi(\boldsymbol{r})]\exp[i\boldsymbol{q}.\boldsymbol{r}]\}$ where $\boldsymbol{q}$ is the wavevector, $\mathbf{A}(\boldsymbol{r})$ is the amplitude vector, and $\phi(\boldsymbol{r})$ is the phase.

We observe temperature-dependent stripe inhomogeneity, including shear deformations (Fig. 2). To visualize the defect-mediated evolution of stripes, we show in Fig. 3(a,b) coarse-grained $\phi(\boldsymbol{r})$ fields at 293K and 93K, respectively. The former displays significant phase inhomogeneity including 2π phase vortices (dislocations) and large phase gradients (shear). At low temperature, a more uniform, slowly varying phase configuration is observed, indicating emergent phase coherence well below T_c. In Fig. 3(c), we plot the correlations of the OP components, notably $\phi(\boldsymbol{r})$ and $|\mathbf{A}(\boldsymbol{r})|$, across temperatures. Amplitude correlations exhibit minimal decay at both temperatures, suggesting that amplitude variations have a negligible effect on stripe evolution. On the other hand, phase correlations decay rapidly at 293K, becoming uncorrelated after ~5nm. At 93K, the phase shows increased homogeneity with correlations remaining finite over accessible length scales (~10nm), supporting that emergent phase coherence governs the phenomenology of stripes. In addition to permitting vivid visualization of the emergence of electronic order, cryogenic STEM paves the way for accessing low temperature phases in correlated systems [3].

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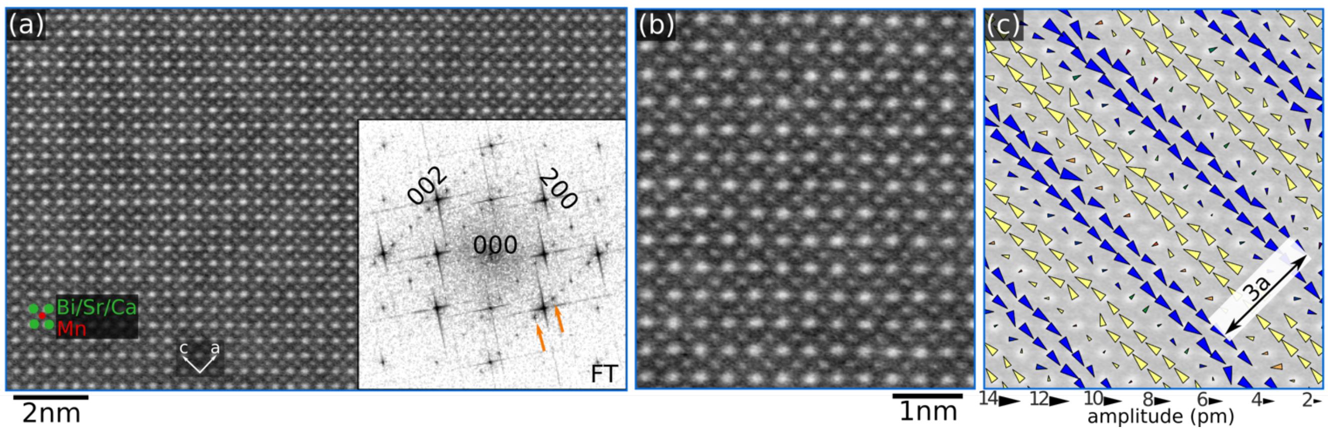

Figure 1. (a),(b) Cryogenic HAADF STEM images of BSCMO with Bi/Sr/Ca and Mn clearly resolved. The Fourier transform (FT) amplitude exhibits superlattice peaks (orange arrows) in addition to Bragg peaks. (c) Mapping picometer scale displacements (arrows) associated with superlattice peaks. The blue (yellow) arrows correspond to transverse displacements, oriented 90º (-90º) relative to $\hat{\boldsymbol{a}}$.

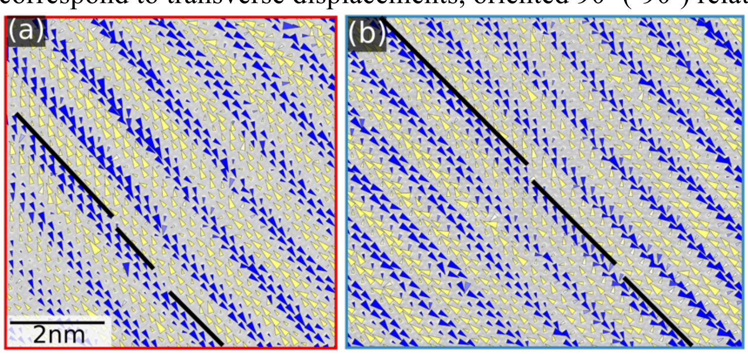

Figure 2. (a),(b) Mapping of shear deformations of stripes at 293K and 93K, respectively. The deformation appears milder and more extended at low temperature.

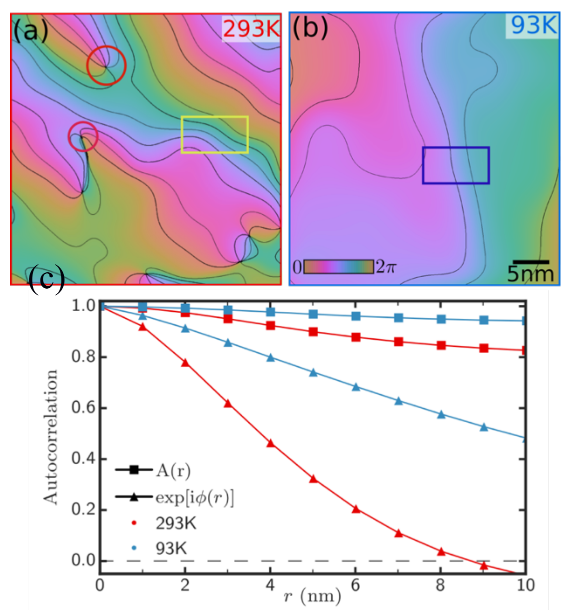

Figure 3. (a),(b) Phase configuration over a ~40nm field of view at 293K and 93K, respectively. Circles correspond to phase vortices (dislocations) and rectangles correspond to shear deformations. (c) Autocorrelations of the phase and amplitude fields at 293K and 93K.

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