The self-organization of strongly interacting electrons into superlattice structures underlies the properties of many quantum materials. How these electrons arrange within the superlattice dictates what symmetries are broken and what ground states are stabilized. Here we show that cryogenic scanning transmission electron microscopy (cryo-STEM) enables direct mapping of local symmetries and order at the intra-unit-cell level in the model charge-ordered system Nd$_{1/2}$Sr$_{1/2}$MnO$_3$. In addition to imaging the prototypical site-centered charge order, we discover the nanoscale coexistence of an exotic intermediate state which mixes site and bond order and breaks inversion symmetry. We further show that nonlinear coupling of distinct lattice modes controls the selection between competing ground states. The results demonstrate the importance of lattice coupling for understanding and manipulating the character of electronic self-organization and that cryo-STEM can reveal local order in strongly correlated systems at the atomic scale.
Strong interactions between electrons and the atomic lattice often lead to their self-organization into ordered spatial patterns. One well-known example is charge ordering, the spatial modulation of the electronic charge density which forms superlattices and governs the properties of many exotic materials, from oxides to transition-metal chalcogenides to charge-transfer salts. In general, charge ordering is studied at scales larger than the superlattice (inter-unit-cell structure), with a focus on the average periodicity of the charge modulation or the degree of long-range order. On the other hand, the microscopic arrangement at sub-unit-cell length scales (intra-unit-cell structure), such as whether the electrons reside on the atomic sites or bonds, is significantly more challenging to measure but dictates the symmetry of the system in addition to the mechanism underlying electronic order.

Perovskite 3d transition-metal oxides are a class of materials in which charge order plays an especially important role, influencing antiferromagnetic order, high-temperature superconductivity, and colossal magnetoresistance. The strong hybridization between the transition metal (site) and oxygen (bond) in these systems contributes to their rich, intricate electronic structure landscape. In cuprates, for instance, recent bulk measurements suggest that the charge-ordered phase may contain a site-centered modulation in addition to the putative bond-centered d-wave modulation. A similarly complex and fundamental debate concerns the half-doped manganites, an ideal playground for exploring coupled charge and orbital orders and phase competition.

The material we focus on is a Nd$_{1/2}$Sr$_{1/2}$MnO$_3$ thin film which was grown using pulsed laser deposition on a (110)-oriented SrTiO$_3$ substrate. The particular substrate orientation, which imparts in-plane anisotropy on the film, reproduces the electronic and structural transitions found in the bulk counterpart including the charge-order transition. The temperature dependence of the magnetization in this film matches that of previously reported epitaxial films. Figure 1e shows a projection HAADF-STEM image of the crystal along the [010] orientation (in the Pnma space group setting) below the charge-order transition temperature ($T_N \sim 150$ K). The atomically resolved Nd/Sr columns appear brighter than the Mn columns because the contrast in HAADF scales strongly with the atomic number. Near the crystalline peaks in the Fourier transform amplitude (Fig. 1f), superlattice peaks (blue arrows) appear at low temperatures, indicating the formation of a modulated structure. These are located at $Q^{CO} = (1/2, 0)$ as expected for charge ordering at half doping. The modulation is unidirectional, however, some regions of the sample exhibit bi-directional modulations. These might arise from the coexistence of small charge-order domains (either in-plane or out-of-plane).

Results

The material we focus on is a Nd$_{1/2}$Sr$_{1/2}$MnO$_3$ thin film which was grown using pulsed laser deposition on a (110)-oriented SrTiO$_3$ substrate. The particular substrate orientation, which imparts in-plane anisotropy on the film, reproduces the electronic and structural transitions found in the bulk counterpart including the charge-order transition. The temperature dependence of the magnetization in this film matches that of previously reported epitaxial films. Figure 1e shows a projection HAADF-STEM image of the crystal along the [010] orientation (in the Pnma space group setting) below the charge-order transition temperature ($T_N \sim 150$ K). The atomically resolved Nd/Sr columns appear brighter than the Mn columns because the contrast in HAADF scales strongly with the atomic number. Near the crystalline peaks in the Fourier transform amplitude (Fig. 1f), superlattice peaks (blue arrows) appear at low temperatures, indicating the formation of a modulated structure. These are located at $Q^{CO} = (1/2, 0)$ as expected for charge ordering at half doping. The modulation is unidirectional, however, some regions of the sample exhibit bi-directional modulations. These might arise from the coexistence of small charge-order domains (either in-plane or out-of-plane).
within a single crystal twin or from the presence of crystalline twins that establish the direction of the charge-order wavevector. By mapping both the charge order and the crystalline order parameters, we find that the orthogonal charge-order domains are coupled to crystalline twins in the sample (Supplementary Fig. 3).

The exact intra-unit-cell arrangement within charge-order superlattices is key to understanding their microscopic origins and interactions with other electronic phases. In the majority of theoretical treatments of charge-ordered phases, the ground state is discussed in terms of pure electronic degrees of freedom. Site-centered charge order in half-doped manganites, for instance, is described as the alternation of Mn$^{4+}$ and Mn$^{3+}$ species$^{18,19}$. However, the degree of charge disproportionation is much smaller and is better described by Mn valences of $3.5 \pm \delta$ ($\text{Mn}^{4+}$-like) and $3.5 - \delta$ ($\text{Mn}^{3+}$-like) with $\delta \ll 0.2$. Measurements of the charge modulation in manganites, whether using resonant X-ray scattering or electron energy loss spectroscopy, have been complicated by the small amplitude of the valence modulation and by the presence of strong lattice distortions. In fact, the crystal structure in the charge-ordered state undergoes a variety of complex atomic displacements, such as Jahn–Teller and breathing distortions, which alter the bonding network and hence the electronic configuration. Therefore, emergent charge and orbital textures are closely linked to the pattern and symmetry of said displacements. For site-centered order, bulk X-ray or neutron structural data suggest that the crystal adopts $P2_1/m$ space group symmetry with bond distortions consistent with charge localization on the Mn sites. The experimental report for the bond-centered model, on the other hand, found a different space group symmetry ($Pmnm$) and a distinct displacement pattern associated with it. The ability to probe intra-unit-cell lattice distortions can therefore determine or even reveal novel ground states.

To firmly connect patterns of atomic displacements to the reported crystal symmetries and hence to the models of electronic order, we first explore the possible distorted structures that emerge from the high-symmetry phase (space group $Pnma$) using group theory. Any distorted structure must double the unit cell, therefore we require atomic displacements whose wavevector is $q = (1/2, 0, 0)$. The relevant distortion consistent with this requirement is the $X1$ displacement mode, or irreducible representation (irrep), which affects two inequivalent Mn sublattices in the high-symmetry unit cell (see Supplementary Information §4). In other words, the X1 irrep is two-dimensional with the first dimension corresponding to the first Mn sublattice (Mn-1 in blue) and the second dimension to the second Mn sublattice (Mn-2 in red), as shown in Fig. 2a. If X1 displacements, which consist of a complex set of atomic distortions including the transverse Mn displacements shown in Fig. 2, affect only the first sublattice, the resulting crystal structure has $P2_1/m$ symmetry, which is consistent with site-centered order (Fig. 2b, c). If they occur in both sublattices and with equal magnitude (Fig. 2b, d), the resulting crystal structure has $Pnnm$ symmetry, which matches that of bond-centered order. The third structure with $Pm$ symmetry can be obtained by having displacements on both sublattices but with different magnitudes (Fig. 2b, e). Such an intermediate state combines aspects of both site and bond order, however, experimental refinements of atomic positions in this symmetry have not been performed so far. The group theory analysis thus shows that the pattern of Mn displacements uniquely defines the character of the charge-ordered phase, without resorting to measurements of the electronic charge.

We now pinpoint the underlying ordering model in Nd$_{1/2}$Sr$_{1/2}$MnO$_3$ by mapping the picoscale lattice degrees of freedom using HAADF-STEM at low temperature. A reference image lacking the magnetic phase provides the geometric parameters, we report for the bond-centered model, on the other hand, found a different space group symmetry ($Pmnm$) and a distinct displacement pattern associated with it. The ability to probe intra-unit-cell lattice distortions can therefore determine or even reveal novel ground states.

Remarkably, we discover that within the same sample another region has a distinct displacement pattern: prominent Mn displacements occur in both Mn sublattices (Fig. 3b). Importantly, the displacements have different amplitudes on each sublattice, unlike the predicted pattern for purely bond-centered order. By comparing to the group theory analysis of displacement patterns, the observed structure is consistent with a state which is

Fig. 2 The symmetry of distorted structures. a High-symmetry unit cell with two Mn sublattices. b Group-subgroup relations for displacements with the wavevector $q = (1/2, 0, 0)$ and irreducible representation $X1$. The $X1$ order parameter is two-dimensional with each dimension corresponding to a Mn sublattice. c-e Displacement patterns for the site-centered ($s_1, s_2 = 0$), bond-centered ($s_1, s_2 = s_3$), and intermediate ($s_1, s_2 \neq s_3$) phase, respectively.
intermediate between pure site and bond order (Pm structure). Figure 3d shows the aggregated Mn displacements and their sinusoidal envelope. The intermediate structure is readily apparent since the maximum of the sinusoidal function is not centered on Mn sites nor exactly at the middle. The phase is $\phi = 0.35(1) \pi$ and the mean displacement amplitudes on the first and second sublattices are 2.9(9) pm and 5.8(7) pm, respectively.

Given that STEM is a projection imaging technique, we ruled out the possibility that the intermediate state merely reflects a projection of stacked site-centered states along the beam direction. This is achieved by analyzing both the contrast variations in the image and the patterns of displacements (see Supplementary Information section 3 for more details). Cryo-STEM mapping of lattice displacements, therefore, provides direct evidence for intermediate charge order in manganites, an observation with implications for other oxides in which the exact character of charge order remains unresolved.

An intriguing consequence of the overlap of the site- and bond-centered order is that additional crystal symmetries may be broken. In oxides or charge-transfer salts, intermediate order is predicted to break inversion symmetry due to the formation of uncompensated dipoles, motivating proposals for unconventional ferroelectricity emerging from electronic order. To determine whether the intermediate phase is indeed noncentrosymmetric, we compute local complex-valued Fourier transforms of the site-centered and intermediate regions. The imaginary part, $\text{Im}[F(q)]$, shows clear differences between the $\pm q$ peaks, an indication that the latter phase breaks inversion symmetry.

Figure 3 Nanoscale coexistence of site order and noncentrosymmetric intermediate order. a Map of periodic lattice displacements of manganese atomic columns ($\Delta_{\text{Mn}}$). The area of the arrows scales with the amplitude of the distortion and the color represents the angle relative to the wavevector direction. In this case, the colors (blue/yellow) indicate a transverse polarization of the displacements ($\pm 90^\circ$). The displacement pattern within the sub-region is consistent with site-centered order; one Mn sublattice shows large displacement amplitude while the other shows much smaller displacements.

b Periodic lattice displacement map in another sub-region shows displacements on both sublattices. Further, the sublattice displacement amplitudes are not equal which indicates intermediate order. c Mn displacements (dots) and sinusoidal envelope function (line) in the site-centered phase. d Mn displacements (dots) and sinusoidal envelope function (line) in the intermediate phase. e, f Line cuts across the imaginary part of local Fourier transforms, $\text{Im}[F(q)]$, along the pseudocubic directions $a_{pc}$ and $c_{pc}$. The site-centered phase (green) maintains $\pm q$ symmetry whereas the intermediate phase (magenta) shows clear differences between the $\pm q$ peaks, an indication that the latter phase breaks inversion symmetry.

g Large field-of-view map of periodic Mn displacements shows a transition from site-centered order (left side) to intermediate order (right side). The scale bar corresponds to 2 nm.
is sensitive to the odd component of the atomic-resolution image and hence to the breaking of inversion symmetry. For a noncentrosymmetric structure, we, therefore, expect the crystalline Bragg peaks at 2q positions to be inequivalent. Figure 3e, f shows integrated line cuts through Im{N(q)} along the pseudocubic a\textsubscript{P} and c\textsubscript{P} directions, respectively. In the site-centered phase (green), the imaginary component of the Bragg peaks has the same amplitude at 2q coordinates for both pseudocubic directions, which indicates that inversion symmetry is maintained. In contrast, there is a significant difference in the intermediate case (magenta), confirming that inversion symmetry is broken in that region. The group theory analysis also tells a consistent story with the intermediate phase having a noncentrosymmetric (Pm) structure. If intermediate charge order can in principle be detected through the switching of bulk polarization, the finite conductivity and nanoscale spatial inhomogeneity of manganites have precluded such an approach. Atomic-scale cryo-STEM imaging, on the other hand, confirms the existence of intermediate charge ordering and the resultant breaking of inversion symmetry.

Another key insight from these local visualizations is that ground states with distinct symmetries may coexist within the same system. Both Fig. 3a, b are taken from the same field-of-view image (Fig. 3g), in which the site-centered phase (left side) transforms into the intermediate phase (right side) over a few unit cells. Within this field of view, strain variations are likely too small to govern the nature of the charge-order ground state (Supplementary Fig. 4a-c). Instead, a possible origin for the coexistence is the presence of quenched disorder which is known to lead to nanoscale phase coexistence in correlated oxides. In the case of the Nd\textsubscript{1/2}Sr\textsubscript{1/2}MnO\textsubscript{3} sample studied here, we detect local spatial inhomogeneity of manganites have precluded such an approach. Atomic-scale cryo-STEM imaging, on the other hand, confirms the existence of intermediate charge ordering and the resultant breaking of inversion symmetry.

We now consider the implications of the two coupling terms in Eq. (1). The first term stabilizes the breathing distortion and lowers the energy the most in the site-centered phase where the X1 displacements are (s\textsubscript{1} \neq 0, s\textsubscript{2} = 0). The second term lowers the energy most in the presence of the antipolar distortions and the bond-centered phase which has X1 displacements (s\textsubscript{1}, s\textsubscript{2} = s\textsubscript{1}). In both of these cases, where only one of the coupling terms is nonzero, we obtain a pure solution (site or bond). If both coupling terms are present (magenta) both s\textsubscript{1} and s\textsubscript{2} are finite but not equal indicating that intermediate order is favorable. The values for the coefficients in the Landau free energy and the Q\textsubscript{B}/Q\textsubscript{AP} amplitudes are listed in the Supplementary Information section 5. The amplitude of s\textsubscript{1} (in Å) is with respect to a 40-atom DFT supercell. c, d Atomic-scale maps of the \Gamma\textsuperscript{7} Mn displacements in the site-centered and intermediate region, respectively. The largest arrows correspond to 1 pm. The displacements are disordered in the former and coherent in the latter, in agreement with the Landau theory predicting that the \Gamma\textsuperscript{7} mode favors X1 displacements on both sublattices. Both scale bars correspond to 1 nm.

![Image](https://example.com/image.png)
in the X1 amplitude, indicating that site-centered order is favorable. When both coupling terms are present, the minimum of the free energy shifts away from \( s_1, 0 \) to \( s_1, s_2 \neq 0 \), thus stabilizing the intermediate phase through a nonlinear mechanism. A more complete picture of the energy dependence on the various coupling terms can be found in 2D plots of the energy surfaces (Supplementary Fig. 11). Based on this theory, a key prediction is that the antipolar \( \Gamma_2 \) displacements are absent (present) in the experimentally observed site-centered (intermediate) phase.

To test this prediction, we visualize the spatial interplay between the antipolar Mn displacements and the character of the charge-ordered ground state. Figure 4c, d show maps of the antipolar \( \Gamma_2 \) Mn displacements in the regions containing site and intermediate order, respectively. In the region with site-centered order, the \( \Gamma_2 \) mode is disordered, lacking any clear pattern of antipolar distortions. In the region with intermediate order, however, this mode is coherent and relatively strong, in agreement with the Landau theory prediction.

**Discussion**

Untangling the roles of electronic and structural mechanisms of charge ordering continues to be a major challenge. While initial predictions proposed purely electronic mechanisms behind the formation of the various charge-ordered states in manganites\(^4,6\), our observations show that lattice degrees of freedom play a more complex role than appreciated. For instance, textbook charge disproportionation on Mn sites is equivalent to the \( \Gamma_2^+ \) breathing distortion from both chemical and symmetry perspectives. The calculations herein, however, show that this charge disproportionation mode raises the energy of the system on its own and that it must couple to additional modes with large structural responses in order to materialize. Similarly, the emergence of the exotic intermediate phase relies on the softening of the \( \Gamma_2^+ \) displacements, further underscoring a critical role of the lattice.

These unusual lattice couplings not only challenge our microscopic understanding of the origin of charge ordering but also provide a mechanism for manipulating the character of electronic order and its associated electronic properties. In the case of the intermediate phase, an exciting theoretical prospect was to achieve ferroelectricity\(^21\). On the experimental front, however, the finite conductivity of manganites and the previously lacking evidence for the intermediate phase remained significant hurdles\(^23,25,26\). Our analysis provides evidence for broken inversion symmetry in the intermediate phase, which not only confirms previous theoretical proposals but also paves the way to achieving ferroelectricity if the polar state is stabilized throughout the sample. To achieve that, one approach inspired by our observations is to enhance the \( Q_{AP} \) amplitude which in turn would favor the polar intermediate phase over the centrosymmetric site-centered phase. Such manipulation could be achieved via elastic strain and rare-earth substitution which were found to modulate the amplitude of \( \Gamma_2^+ \) distortions in related theoretical calculations\(^41\).

Our study shows that cryo-STEM is an emerging methodology for probing complex electronic ordering phenomena through the important but often neglected lens of the lattice degrees of freedom. Using this approach, we discovered the coexistence of distinct charge-order configurations including a pure site-centered phase and a more exotic intermediate phase that breaks inversion symmetry. We also revealed that the stability of these different configurations depends on unique lattice couplings. Such a rich electronic and structural landscape should be relevant to other charge-ordered systems, including cuprates and nickelates where the exact microscopic arrangement associated with various electronic instabilities remains under intense scrutiny. Similar to the current study, microscopic insights may be achieved through direct, real space visualizations that can spatially disentangle different ordering models and characterize the intra-unit-cell structure and symmetry in detail.

**Data availability**

The data that support the findings of this work are available from PARADIM, a National Science Foundation Materials Innovation Platform (https://doi.org/10.34863/bg5n-4g68).

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Author contributions

D.L., Y.H., and H.H. synthesized the samples and performed magnetic characterization. D.J.B, I.E., M.J.Z., and L.F.K. acquired and analyzed the electron microscopy data. E.A.N. performed density functional theory calculations. E.A.N., I.E., and L.F.K. performed symmetry and theoretical analysis. I.E., E.A.N., D.J.B., and L.F.K. wrote the manuscript with input from all authors.

Competing interests

The authors declare no competing interests.

Additional information

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