

Microscopic nature of orbital disorder in LaMnO_3

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We present a revised atomistic picture of the order-disorder transition in the archetypal orbital-ordered perovskite material, LaMnO_3 . Our study uses machine-learning-driven molecular-dynamics simulations which describe the temperature evolution of pair distribution functions in close agreement with experiments. We find the orbital-disordered phase in LaMnO_3 to comprise a mixture of differing structural distortions with and without inversion symmetry, implying a mixture of different orbital arrangements. These distortions are highly dynamic with an estimated lifetime of ~ 40 fs at 1000 K, and their fluctuations converge with the timescales of conventional thermal motion in the high- T phase—indicating that the electronic instability responsible for static Jahn-Teller distortions at low temperature instead drives phonon anharmonicity at high temperatures. Beyond LaMnO_3 , our work opens an avenue for studying a wider range of correlated materials.

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I. INTRODUCTION

Orbital correlations can play a key role in determining electronic and magnetic properties of materials. In many transition-metal compounds, the occupation of d -orbitals and their orientations control overlap, which influences electronic transport and magnetic coupling. These orbital arrangements can be crystallographically ordered or disordered, and the balance between the two extremes can be critical to emergent phenomena such as superconductivity [1,2], colossal magnetoresistance [3–5], charge-orbital ordering [6], and ferroelectricity [7]. Therefore, understanding orbital-ordered and -disordered states in crystalline solids is an important research objective.

While the long-range ordered structural distortions induced by orbital ordering can be directly deduced from bulk diffraction studies [8,9], it is much more difficult to describe orbital-disordered states, both experimentally and computationally. In general, three pictures are usually considered for orbital-disordered phases. The first is that of an orbital liquid or glass, where local orbital correlations resemble those of a low-temperature ordered state, but correlated only over finite lengthscales (e.g., domains) [10,11]; the second is a correlated-disorder model, in which the disordered state supports orbital correlations not found directly in the related ordered state [12]; and the third involves suppression of structural distortions through a displacive transition [13–15]. Differentiating these descriptions requires probes that are sen-

sitive to the local distortions in the orbital-disordered phase and their corresponding dynamics.

The archetypal orbital-ordered system is LaMnO_3 [10], which contains Mn^{3+} with a $(t_{2g})^3(e_g)^1$ high-spin configuration. Under ambient conditions, the compound adopts a perovskite structure in which each $[\text{MnO}_6]$ octahedron is Jahn-Teller (JT) distorted to have two long, two medium, and two short Mn–O bonds. The distortions order such that two opposing long Mn–O bonds alternate orientations between two orthogonal directions, in a way that repeats along a third direction (“C-type” ordering) [16]. This anisotropy is evident in the orthorhombic strain of LaMnO_3 . Above 750 K, the lattice parameters collapse to yield a pseudocubic phase in which the JT distortion is apparently quenched [9]. However, local-structure probes show that the asymmetry in Mn–O bond-length distributions remains after the transition, which was interpreted as there being no change in the Mn–O bond lengths with increasing T . The implication is that the JT distortions persist, but now in a crystallographically disordered manner which, on average, gives the observed pseudocubic phase [11,17].

Experiments have yielded conflicting descriptions of this order-disorder transition. Resistivity and thermoelectric measurements of the orbital-disordered phase of LaMnO_3 led to an interpretation involving dynamic cooperative JT distortions, in which the C-type order fluctuates above 750 K [10]. Additionally, a model containing nanoregions of static C-type order was proposed based on neutron pair distribution function (PDF) analysis [11]. In contrast, a three-state Potts (“3SP-type”) model, which allows the two opposing long Mn–O bonds to align along all three principal axes, was found to reproduce the behavior of LaMnO_3 in resonant x-ray scattering [12,18]. Furthermore, reverse Monte Carlo (RMC) fits to PDFs calculated from x-ray and neutron diffraction experiments yielded a model with the long bonds in the JT-distorted octahedra aligning perpendicularly to each other (“L-type” ordering) [19]. Notably, the PDFs showed a

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discontinuous change at 750 K, indicating that the high- T phase has a different local structure to the ordered one. However, the conclusions were not definitive as the differences between fits to high- T PDF data using C-type, 3SP-type, and L-type models were not meaningful in the presence of strong thermal fluctuations [19].

The orbital order-disorder transition in LaMnO_3 is similarly difficult to interpret using computations. First-principles studies indicated that the transition is multifaceted, involving a combination of octahedral rotations, antipolar distortions of the A-site cations (La^{3+}) and, importantly, a compressive tetragonal shear strain on the lattice stabilizing the cooperative JT distortions in the low- T antiferromagnetic phase [20]. Comparing the computed energies of different magnetic orderings in the high- T paramagnetic phase shows that antiferromagnetic (AFM) ordering yields a stronger stabilization of the JT distortion modes than ferromagnetic ordering. This stabilization results in an improved prediction of the transition temperature, implying that spin symmetry breaking, and thus dynamics, affect the transition. While these simulations have allowed the study of distortion modes and magnetic ordering in LaMnO_3 , they have been limited to relatively small system sizes.

Here, we use large-scale, machine-learning-driven molecular-dynamics (MD) simulations to re-examine the microscopic nature of orbital order and disorder in LaMnO_3 . We first validate our simulations by computing the temperature evolution of average structural parameters and PDFs, comparing to experimental data. We then analyze the evolution of JT distortions with temperature, demonstrating that the high- T phase of LaMnO_3 actually adopts a mixture of the orbital orderings from the 3SP-type and L-type models. Finally, by computing the time-dependent evolution of the amplitudes of JT distortions, we show that these distortions become dynamic with increasing temperature and that their lifetimes shorten to the same timescale as thermal fluctuations at 1000 K. Hence, the apparent persistence of JT distortions in orbital-disordered LaMnO_3 is really a signature of JT-driven bond anharmonicity.

II. METHODS

Our MD simulations are driven by a machine-learned interatomic potential (MLIP). Recent advances in on-the-fly-accelerated first-principles MD [21–23] provide a route for the efficient generation of training data for MLIPs [24–26]. We here parametrized an MLIP model using the NequIP architecture [27,28] together with an existing dataset of 1384 LaMnO_3 structures from Ref. [26], generated from a 100 $\rightarrow$ 1100 K heating run of a $2 \times 2 \times 2$ cubic unit cell with initial AFM ordering [26]. As discussed in Ref. [26], the initial magnetic order is lost at high- T during the AIMD simulation, which leads to the sampling of magnetically disordered phases.

The MLIP model shows a test-set root mean square error of 9.6 meV/atom for energies and 65 meV/Å for force components. It also predicts the atomic volumes of representative low- T and high- T configurations in good agreement with DFT (see equations of state in Ref. [29]).

For MD simulations, we used the isothermal-isobaric (NPT) ensemble with a Nosé-Hoover thermostat and barostat

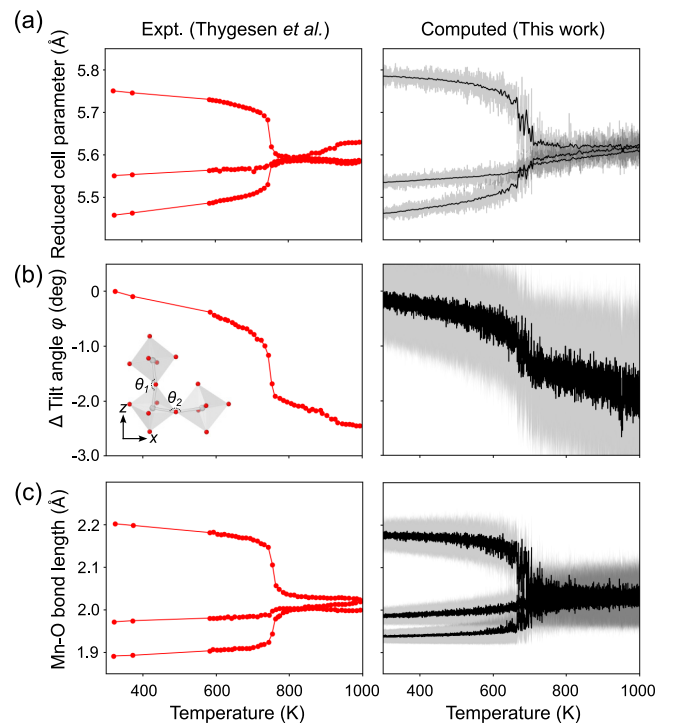


FIG. 1. Change in average structure of LaMnO_3 with temperature. Experimental (red) and simulated (black) data are shown. (a) Reduced lattice parameters. (b) Change in tilting angle, ϕ , of octahedra along the (111) axis. (c) Mn–O bond lengths showing two long, two medium, and two short bonds and their evolution. Experimental data taken from Ref. [19].

as implemented in LAMMPS [30]; specific details are given in the context of each simulation. Neutron and x-ray PDFs were predicted using DebyeCalculator [31] using reciprocal-space ranges, real-space resolutions, and damping parameters from Ref. [19].

III. RESULTS AND DISCUSSION

To validate the MLIP model, we computed key structural parameters and compared their change with temperature to the experimental data of Ref. [19] (Fig. 1). A $6 \times 6 \times 6$ expansion of the cubic unit cell was used for a heating simulation from 200 to 1200 K at a rate of 1 K/ps at constant atmospheric pressure. Reduced cell parameters were calculated using equations outlined in Ref. [26], and the tilting angle, ϕ , along the (111) axis was calculated using the O–O–O angles θ_1 and θ_2 according to the methods in Refs. [32] and [33]. The average structural parameters were obtained from a trajectory with a time averaging of 0.1 ps. For the lattice parameters, a moving average was calculated and overlaid on the time-averaged values. The tilting angles and bond lengths were averaged over all 256 octahedra within the supercell. Overall, Fig. 1 shows excellent agreement between simulated and experimental data: in particular, the orthorhombic/pseudocubic transition is replicated, as in Ref. [26]. The temperature at which this transition occurs is underestimated by ~ 60 K, which may be due to the underlying DFT level [20,26]. With increasing T , the lattice parameters collapse to

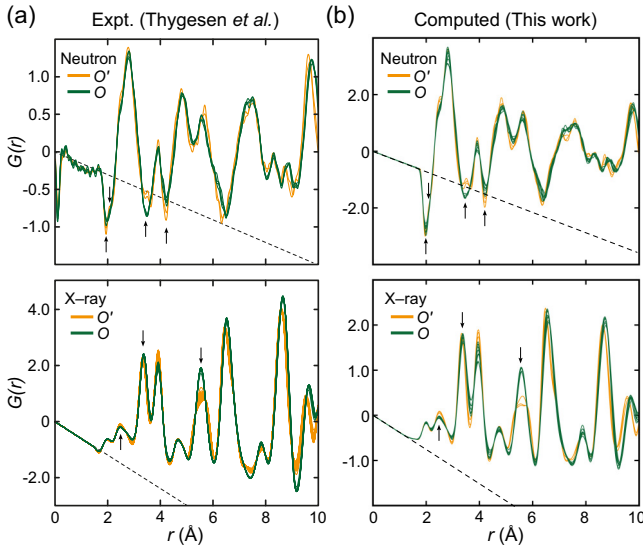


FIG. 2. Pair distribution functions for LaMnO_3 and their evolution with temperature. Experimental (a) and computed (b) neutron (top) and x-ray (bottom) PDFs for the temperature range 500–1000 K are shown. Baselines are shown using dotted lines; features of particular interest are marked with arrows. As in Ref. [19], O' (orange) denotes the orbital-ordered low- T and O (green) denotes the orbital-disordered high- T phase. Panel (a) adapted from Ref. [19].

similar values accompanied by a decrease in shear strain [29], the octahedral tilting decreases, and the JT distortions appear to be suppressed. At ~ 1200 K, the tilting pattern changes to $a^-a^-a^-$, consistent with a transition to the rhombohedral phase as observed experimentally [29].

Next, we calculated averaged PDFs from structures sampled from 10 ps MD runs. Figure 2 shows that the calculated PDFs agree remarkably with experimental data from Ref. [19], indicating that the simulation captures the change in local structure within the temperature range. Notably, it replicates the change in peaks corresponding to the Mn–O bonds, from two split peaks to a broad single peak at ~ 2 Å. While the changes in lattice parameters, volume, and Mn–O bond lengths have been previously simulated [12,20,26,34,35], our study replicates the experimental PDFs for LaMnO_3 —and their evolution across the order/disorder transition— from first principles.

Overall, Figs. 1 and 2 show that the temperature dependence of both average and local structure in LaMnO_3 is well described by our simulations. It is perhaps surprising that the MLIP model is able to replicate structural changes through the transition in such close agreement with experiments, even though the underlying DFT approach, LDA + U , is heavily scrutinized for such correlated electron systems [36,37]. This observation justifies now using the atomistic configurations to explore the nature of orbital order and disorder in LaMnO_3 . Figure 3 shows the orientations of the respective two longest Mn–O bonds in each $[\text{MnO}_6]$ octahedron with temperature, as well as snapshots of slices in the xy plane. The MD trajectory has been time averaged with a window of 0.1 ps to remove thermal fluctuations. We found that larger time-averaging windows resulted in the bond lengths averaging out in the

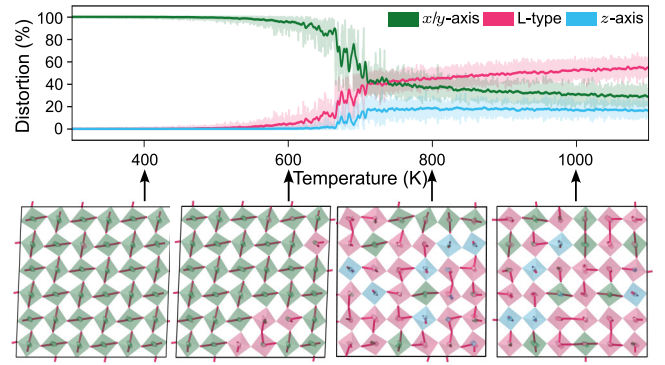


FIG. 3. Evolution of long Mn–O bond orientations with temperature. We plot the percentage of parallel long bonds aligning with the x or y axis (green) and z axis (cyan) as well as L-type (magenta). Slices in the xy plane highlighting the two longest bonds are shown at relevant temperatures [38].

high- T phase due to the constant fluctuation of long-bond orientations [29]. As temperature rises, the system undergoes a drastic change, losing the C-type order and adopting a crystallographically disordered configuration. Unexpectedly, the high- T phase consists of $\sim 50\%$ of L-type distortions and the remaining distortions having opposing long bonds aligning along all three axes in equal proportions. This result is consistent with the meaningful change in local structure observed in experimental PDFs [19], but is a markedly different picture than previously considered.

We computed phonon dispersion curves and power spectra from velocity autocorrelation functions (VACFs) obtained from MD trajectories (Fig. 4), in order to validate the predictions of dynamical properties and to probe the changes in the phonon density of states with temperature. At each temperature, supercells containing 13 720 atoms supercells were thermalized with NPT MD for 20 ps and the cell parameter for the last snapshot of the trajectory was scaled to the average value from the last 50 steps of the trajectory. The resultant supercell was thermalized with NVT MD for 5 ps, followed by a 25 ps NVE MD run, to obtain dynamics unaffected by thermostats and barostats. The timestep of each run was 1 fs and snapshots were dumped from the trajectory every 3 fs. Neutron-weighted longitudinal and transverse correlations were calculated using dynasor [39,40]. Per-element VACFs were calculated from the trajectories and Fourier transformed to obtain per-element power spectra. The per-element power spectra were then neutron-weighted using factors from Ref. [41], summed up and normalized. Convergence of the spectra was tested by comparing to the power spectra for a longer 40 ps NVE trajectory, which showed minimal changes [29]. The energies were scaled by a factor of 1.12 to compare to the DFT phonon density of states from Ref. [41]. It is not uncommon to use constant scaling factors when calculating vibrational frequencies with DFT, as they have a systematic shift corresponding to the functional used [42]. Similarly, some MLIPs were shown to have a systematic shift in energies for vibrational modes w.r.t. DFT [43]. With the slight scaling, the power spectra are in qualitative agreement with the DFT results [Fig. 4(b)]. The phonon density

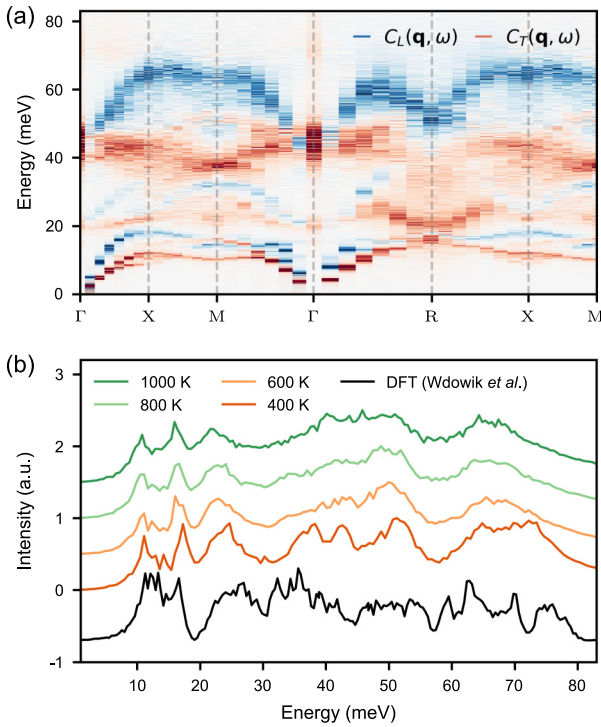


FIG. 4. Dynamic properties of LaMnO_3 computed using a 13 720-atom supercell. (a) Scaled neutron-weighted phonon dispersion curves with longitudinal (blue) and transverse (red) correlations at 800 K. (b) Scaled neutron-weighted power spectra below (orange, red) and above (lime, green) the transition temperature, as well as a DFT phonon density of states (black) are shown. The DFT data were taken from Ref. [41].

shows no significant change through the transition except for a slight broadening and decrease in intensity of the high-energy features associated with the oxygen sublattice. This trend is in good agreement with experimental phonon density of states data obtained from inelastic neutron scattering [41], indicating the reliability of the dynamics obtained from the MLIP-driven MD simulations. Further analysis of the longitudinal correlations at specific q -vectors reveals oscillations that hint towards the anharmonic nature of the oxide anion vibrations [29].

To draw a comparison to other materials, a clear change in local structure is also apparent from PDFs for orbital order transitions in the layered nickelates, LiNiO_2 [14] and NaNiO_2 [15]: the Ni–O peaks broaden and merge with increasing temperature, similar to the Mn–O peaks in LaMnO_3 . However, the change in the Ni–O peaks was interpreted to be due to the JT distortions disappearing, in contrast to the canonical understanding of the behavior of LaMnO_3 . We can investigate this difference in interpretation by calculating the magnitude of the JT distortions of each octahedron through Van Vleck vibration modes, using VanVleckCalculator [44]. We focus on the E_g distortion space which is spanned by a planar rhombic Q_2 component (compression of two bonds and stretching of two bonds) and a tetragonal Q_3 component (compression of four bonds and stretching of two bonds). Figure 5(a) shows the evolution of the distributions of the octahedral distortions from snapshots of a $14 \times 14 \times 14$ supercell heated for 10 ps at each temperature. At 400 K, there are two high-intensity

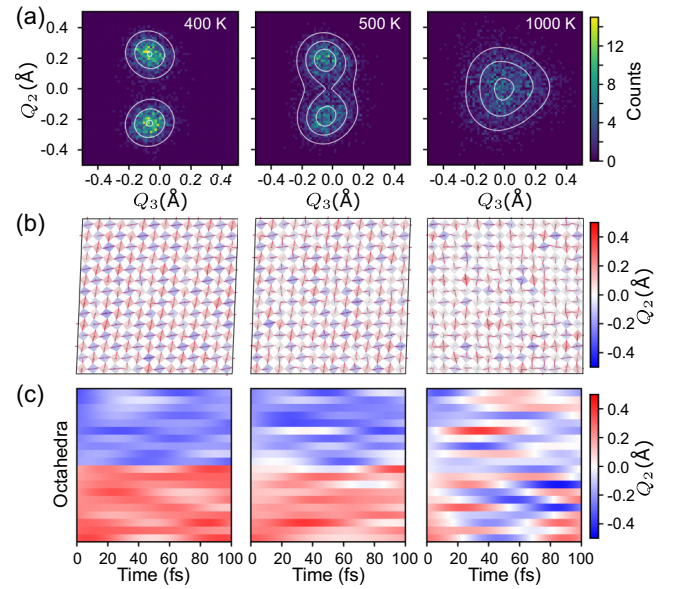


FIG. 5. Evolution of octahedral distortions in a 13 720-atom supercell with temperature and time. (a) Heatmaps with contour lines of the octahedral distortion mode components, Q_2 and Q_3 , at 400 K, 500 K, and 1000 K. (b) Slices along the xy plane of the supercells [38]. The amplitude of the Q_2 component for each octahedra and Mn–O bonds longer than 2.05 Å are colored. (c) Change in the amplitude of the Q_2 component with simulation time for 20 randomly selected octahedra, ordered by the initial amplitude, at each temperature.

regions showing that there are two major orientations of the JT distortions, as expected for C-type ordering. At 500 K, these regions are broader and closer together in distortion space. When slices of the 500 K configurations are visualized with color coding based on the amplitude of the Q_2 component [Fig. 5(b)], we see mostly C-type ordering, but a small proportion of the octahedra have near-zero Q_2 amplitudes. At 1000 K, the heatmap in Fig. 5(a) shows a clear centering around (0, 0), and the majority of the octahedra have reduced Q_2 or Q_3 amplitudes ($\sim 45\%$ within ± 0.05 Å). Slices of the 1000 K configurations show essentially complete melting of the original C-type order.

The unimodal distribution of octahedral distortion types we observe is inconsistent with previous interpretations based on the 3SP model [35], for which the (Q_2, Q_3) heatmap would show three maxima at high temperature, corresponding to JT distortions aligned along the three principal axes. Hence, our results indicate that the high- T phase of LaMnO_3 involves suppression of the conventional JT distortion of $[\text{MnO}_6]$ octahedra on long timescales. The broadening and merging of the Mn–O peaks in the PDFs is probably a signature of this local-structure transition. Indeed, this interpretation has much in common with that of JT suppression in LiNiO_2 [14] and NaNiO_2 [15].

Next, we ask whether the octahedral distortions in the high- T phase are static, as had been inferred from experimental studies [11,45]. We qualitatively analyzed JT mode lifetimes by tracking the amplitude of the Q_2 component of each octahedron over time. Figure 5(c) compares the lifetimes

at different temperatures for 20 randomly selected octahedra. Each horizontal line in the plot represents the evolution of the Q_2 component of one octahedron in the last 100 fs from 10 ps runs at constant T . While essentially static at 400 K, the lifetimes shorten and the distortions start to fluctuate between different directions at 500 K [red $\leftrightarrow$ blue in Fig. 5(c)]. At 1000 K, the distortions are constantly swapping orientations with a lifetime of ~ 40 fs [29], showing that distortions in the high- T phase are highly dynamic. Indeed, projecting velocities onto the cooperative JT distortion phonon mode, calculating the autocorrelation functions, and fitting it using a damped harmonic oscillator leads to a phonon lifetime of 42 fs [29]. This behavior is reminiscent of JT-driven fluctuations in paraelectric BaTiO₃, in which asymmetric broadening of the Ti–O PDF peaks at high temperatures is associated with anharmonicity of Ti-off-centering dynamics [7,46]. Similar interplay of JT instabilities and strongly anharmonic lattice dynamics is found in the thermoelectrics PbTe and SnSe [13,47–49]. For PbTe, single-crystal inelastic neutron scattering showed that the electronic instability causes phonon anharmonicity resulting in dynamic dipoles that give asymmetric PDF peaks with increasing temperature [50]. The dynamic nature of the octahedral distortions in LaMnO₃ might be confirmed experimentally by the same approach. For comparison with future studies, simulated single-crystal diffuse scattering patterns from the configurations with 13 720 atoms are provided online [29].

While the present work was prepared for revised submission, Ref. [51] was published, which also employs MLIP-based MD to study the phase transition in LaMnO₃. As in Ref. [26], the training dataset is generated using on-the-fly learning, and production simulations are performed on 1080-atom supercells. The results reported in Ref. [51] are in close agreement with those presented here, including the evolution of lattice parameters, Mn–O bond lengths, and the Q_2 and Q_3 octahedral distortion mode components across the transition. Additionally, Ref. [51] analyzes the temperature dependence of the $A_g(1)$ phonon mode by projecting oxygen atom velocities onto the corresponding eigenvector, and describes pronounced phonon softening indicative of strong anharmonicity. Importantly, Ref. [51] frames the transition in terms of an order-disorder picture, whereas our

analysis instead interprets the high- T phase in terms of JT-driven anharmonicity, emphasizing the coexistence of distinct local distortion motifs and rapid fluctuations. We note that the interpretation of the structural and dynamic signatures differ between the studies. However, this distinction does not alter our central conclusion that, at high temperatures, LaMnO₃ is best described as a dynamic, strongly anharmonic state rather than in a static order/disorder picture.

IV. CONCLUSIONS

Our key result has been to show, by detailed analysis of local atomic environments, that the conventional JT distortion found in octahedral Mn³⁺ perovskites is indeed actually quenched in the orbital-disordered phase of LaMnO₃. The distortions that are present at high temperatures are more varied and fluctuate on a timescale that is similar to that of typical thermal motion. Hence, the orbital arrangements in LaMnO₃ are not well described by a conventional order/disorder picture, but instead involve a transition from static correlated JT to dynamic JT-driven anharmonicity. Because CMR emerges from this “orbital-disorder” phase on doping, our analysis suggests that MLIP-driven MD simulations may offer insight into the microscopic nature of orbital disorder within the CMR manganites.

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DATA AVAILABILITY

Data supporting this work are openly available in Ref. [29].

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