

Nanoscale distortions and ground state selection in geometrically frustrated magnets

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Geometrically frustrated magnets, such as triangular networks of antiferromagnetically coupled spins, can display incredibly rich physical properties that may have potential applications in quantum information science and other technologies. Determining if and how magnetic order emerges from competing magnetic tendencies is an important objective in this field. Here, we discuss the Jahn-Teller active triangular AMnO₂ (A= Na, Cu; Fig. 1) antiferromagnets [1] to highlight that the degree of frustration, mediated by residual disorder, contributes to the rather differing pathways towards a single, stable magnetic ground state, albeit with varying ordering temperatures.

For these insulating sister compounds, complementary high-resolution synchrotron XRD, local-probe muon-spin relaxation (μ^+ SR) studies, corroborate that the layered NaMnO₂ adopts a remarkable magnetostructurally inhomogeneous ground state. [2] In view of this peculiarity, we employ powerful neutron total scattering and magnetic pair distribution function (PDF) analysis to uncover that although CuMnO₂ undergoes a conventional symmetry-lowering (monoclinic to triclinic) lattice distortion driven by Néel order, in the Na-derivative an enhanced, short-range triclinic distortion (Fig. 2) lifts the degeneracy of the isosceles triangular network on the nanoscale, thereby enabling long-range magnetism to develop with enhanced magnetic correlations above the transition. [3] More generally, the work illuminates the cooperative intertwining of the local atomic and magnetic structures that permits ground state selection when spatial inhomogeneity meets geometrical frustration, a mechanism that may also be operative in other frustrated materials with electronically active transition metal cations.

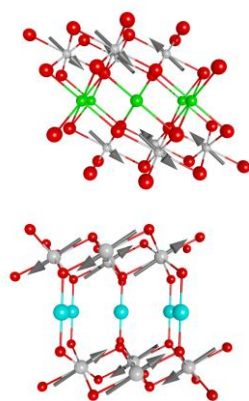


Figure 1. Atomic and magnetic structures of NaMnO₂ (top), and CuMnO₂ (bottom).

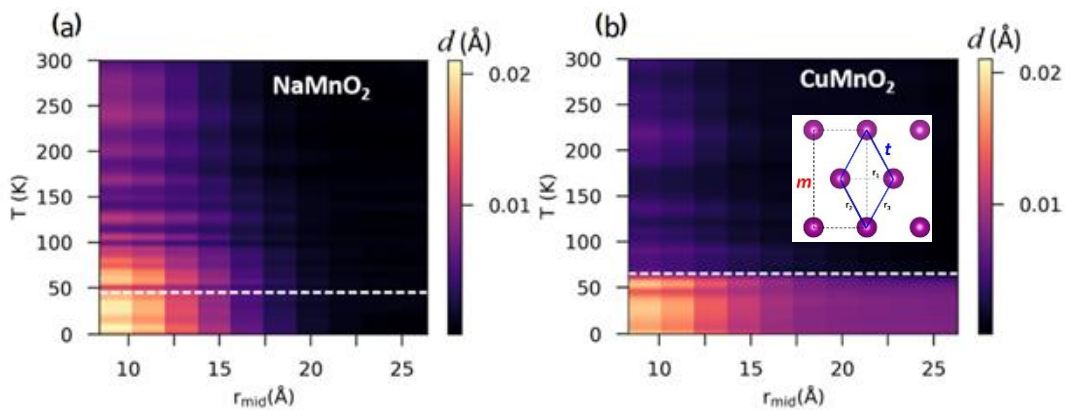


Figure 2. Color map of the strength of triclinic distortion, d ($=r_2-r_3$; see inset to panel (b)), as a function of temperature in AMnO₂ (A= Na, Cu); the horizontal dashed lines mark the Néel order, at 45 (a) and 65 K (b). Inset: schematic of basal plane Mn³⁺ sublattice, and its monoclinic to triclinic distortion.

[1] Giot, M., Chapon, L. C., Androulakis, J., Green, M. A., Radaelli, P. G., & Lappas, A. (2007). *Phys. Rev. Lett.* **99**, 247211.

[2] Zorko, A., Kokalj, J., Komelj, M., Adamopoulos, O., Luetkens, H., Arcon, D., & Lappas, A. (2015). *Sci. Rep.* **5**, 9272.

[3] Frandsen, B. A., Bozin, E. S., Aza, E., Martinez, A. F., Feyngenson, M., Page, K., & Lappas, A. (2020). *Phys. Rev. B* **101**, 024423.

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