

Origin of correlated diffuse scattering in the hexagonal manganites

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We use a combination of first-principles density-functional calculations and spin-dynamics simulations to explain the unusual diffuse inelastic neutron scattering in the hexagonal multiferroic yttrium manganite, YMnO_3 . Using symmetry considerations, we construct a model spin Hamiltonian with parameters derived from our density-functional calculations and show that it captures the measured behavior. We then show that the observed directionality in the structured diffuse scattering in momentum space is a hallmark of the triangular geometry, and that its persistence across a wide range of temperatures, both above and below the Néel temperature T_N , is a result of the strong magnetic frustration. We predict that this diffuse scattering exists in a yet-to-be-observed modulated continuum of energies, that its associated spin excitations have distinct in-plane and out-of-plane character, and that vestiges of the magnetic frustration persist into the sub- T_N state. Finally, we show that visualizing the magnetic order in terms of composite trimer magnetoelectric monopoles and toroidal moments, rather than individual spins, provides insight into the real-space fluctuations, revealing clusters of emerging order in the paramagnetic state, as well as collective short-range excitations in the ordered antiferromagnetic phase. Our understanding of this directional diffuse scattering in such a wide temperature range, both below and above T_N , provides new insight into the magnetic phase transitions in classical frustrated systems.

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It has been generally accepted that magnetic order follows the description of Landau phase transitions as magnetic fluctuations overcome the thermal excitations. However, the disordered paramagnetic state expected above, in temperature, the magnetic phase transition is sometimes not achieved until significantly higher temperatures. Instead, short-ranged diffuse-scattering signatures can be observed. Diffuse scattering, a measure of local correlation, is ubiquitous in finite-temperature inelastic neutron scattering (INS) [1,2], and has distinctive features in frustrated materials [3–5]. Frustration occurs in a multitude of systems, and can arise from competing polar phases [6], structures [3,7], or magnetic interactions [8]. These give rise to a plethora of ground states as well as excited metastable states [9,10] which don't have long-range order. In magnetic systems, the associated fluctuations, which can be classical or quantum [11], can suppress the long-range ordering temperature—sometimes fully [12]—resulting in local correlations that cannot be described within a mean-field description [13] and are not fully understood. Magnetic systems are particularly good models for studying

frustrated phenomena, owing to the large variety and availability of magnetic materials [14], and can show unusual properties such as highly tunable superconductivity [15,16], magnetic fragmentation [17], or exotic spin-orbit tunability [18]. Diffuse scattering can provide useful insight into the magnetic structure [2] as well as the nature of magnetic phase transitions [19]. In particular, many frustrated magnetic systems show diffuse scattering confined to specific directions in reciprocal space [8,11,17,20], revealing a particular momentum dependence of the associated short-range correlations.

The hexagonal manganites, of which h-YMnO_3 is the prototype, provide a natural playground for studying magnetic frustration, since they contain triangular ab planes of antiferromagnetically coupled Mn ions that are only weakly interacting along the c direction [21–24]. The long-range complex noncollinear ground state gives rise to the well-studied magnons [23,25–28]. INS has revealed unconventional scattering signals in this system, such as low-energy sub- T_N quasielastic continua [22], inelastic peaks above T_N [29], and diffuse scattering well above and below T_N [30], as well as signatures of magnon decay, such as roton-like minima, flat modes, and line-width broadening [23,27]. These behaviors have been variously attributed to closely related phenomena such as cooperative paramagnetism [22], Kosterlitz-Thouless phases [22], and classical spin liquids [21], or attributed to quasiparticle breakdown [23,27]. In this work, we focus in particular on the directional “rodlike” diffuse-magnetic excitations, persisting across T_N in an unusually large temperature regime and coexisting with magnon

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TABLE I. Parameters extracted from density-functional theory (DFT) in units of meV.

	J_1^{ab}	J_2^{ab}	J_1^z	J_2^z	A^{zz}	$A^{x^*x^*}$	$ \vec{D} $
DFT [33]	7.779	4.874	-0.015	-0.003	0.530	-0.117	0.239

excitations. Such rodlike diffuse scattering [21] has also been observed in other triangular frustrated systems [31,32] and its origin is not yet understood.

Within each h-YMnO₃ *ab* plane, the S=2 Mn³⁺ lattice distinguishes itself from a perfect triangular frustrated lattice (TFL) through a trimerization which displaces the Mn ions away from their ideal 1/3 positions [33]. MnO₅ bipyramids then tilt towards their trimerization center, and layers of yttrium sites, which separate the magnetic planes, buckle [34]. The outcome of this particular crystal chemistry is a hierarchy of magnetic interactions [24,33,35] which is described by the following Hamiltonian:

$$\begin{aligned} \mathcal{H} = & J_1^{ab} \sum_{\substack{\langle i,j \rangle \\ 1^{\text{st}} \text{ n.n.}}} \hat{e}_i \cdot \hat{e}_j + J_2^{ab} \sum_{\substack{\langle i,j \rangle \\ 2^{\text{nd}} \text{ n.n.}}} \hat{e}_i \cdot \hat{e}_j \\ & + J_1^z \sum_{\substack{\langle i,j \rangle \\ 3^{\text{rd}} \text{ n.n.}}} \hat{e}_i \cdot \hat{e}_j + J_2^z \sum_{\substack{\langle i,j \rangle \\ 4^{\text{th}} \text{ n.n.}}} \hat{e}_i \cdot \hat{e}_j \\ & + \sum_i A^{zz} (e_{i,z})^2 + \sum_i A^{x^*x^*} (e_{i,x^*})^2. \end{aligned} \quad (1)$$

The parameters, which we obtained from Ref. [33] following the method described in Ref. [36], are listed in Table I. Here, $\hat{e}_i$ are the unit-vector magnetic moments. The first and second lines list the symmetric exchange interactions, with the

in-plane dominant first- and second-nearest neighbor AFM interactions, J_1^{ab} and J_2^{ab} , respectively, and the weak interplanar ferromagnetic interactions, J_1^z and J_2^z [see Fig. 1(a)]. The third line describes the single-ion anisotropy (SIA), which we compute to have a hard axis along *c*, described by A^{zz} and a weaker in-plane anisotropy, where x^* denotes the local in-plane easy-axis direction, which lies along the vector connecting the magnetic site to its trimerization center [see Fig. 1(a)]. This minimal Hamiltonian model yields the correct ground state. In the Supplemental Materials [37] we show that adding the Dzyaloshinskii-Moriya interaction—which we compute to have magnitude $|\vec{D}| = 0.239$ meV (Table I)—does not influence the correlated diffuse scattering (Fig. S1).

We simulate neutron-scattering results from Ref. [21] using a combined first-principles and spin-dynamics approach and clarify the nature of the short-range correlations, as well as their impact on the low-temperature magnetism in h-YMnO₃. We use the parameters from Table I in our Hamiltonian in Eq. (1) to calculate the spin dynamics using the UppASD code [38–41]. We calculate $T_N = 52$ K, which is in reasonable agreement with the experimentally measured $T_N^{\text{exp.}} \simeq 70$ K [42]. We note that while density-functional theory (DFT) over estimates the interaction parameters, their relative strengths are comparable to those fitted to INS data (which we list in Table S1 in the SM): $|J_1^z|/J_1^{ab} \simeq 0.002$ and $A^{zz}/J_1^{ab} \simeq 0.09$ [33], where J^z and J^{ab} are the average intra- and interplanar exchanges, respectively. We compute energy dispersions by converting spin-spin correlations into the usual dynamical structure factor $S(\vec{q}, \omega)$, as implemented in UppASD, using this quantity to compare our theoretical model predictions with experimental scattering data [43]. We create $50 \times 50 \times 10$ supercells, and cool our system from 400 K down to 2 K, in steps of 10 K from 400 K to 100 K and

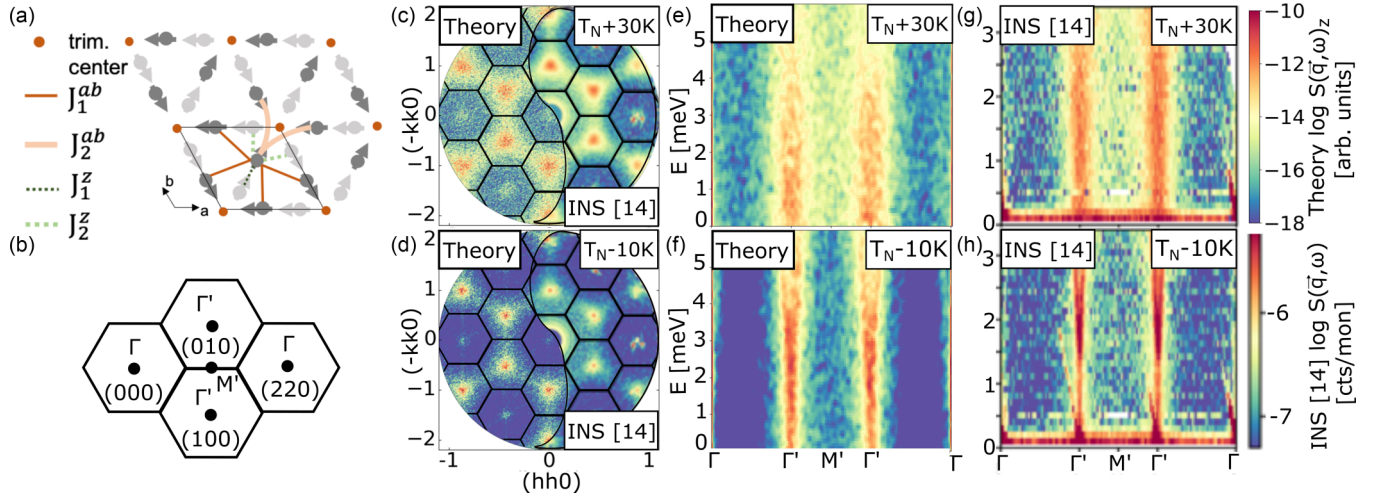


FIG. 1. (a) Magnetic ground state of h-YMnO₃ and symmetric exchanges of one representative spin. Lighter and darker colored arrows represent Mn spins belonging to consecutive *ab* planes along *c* and black lines indicate the boundaries of the six Mn-atom magnetic unit cells. Red dots indicate trimerization centers (trim. center). (b) Studied points in reciprocal space. Black lines indicate the reciprocal lattice and the magnetic and structural Bragg peaks occur at Γ' and Γ , respectively. (c)–(h) Color maps of the dynamical structure factor $S(\vec{q}, \omega)$ 30 K above (top panels) and 10 K below (bottom panels) T_N , computed using spin-dynamics simulations (Theory) and compared with INS data (INS [21]) adapted with permission from Fig. 1 from Ref. [21] (copyrighted by the American Physical Society). (c)–(d) Constant $\hbar\omega = 1.04 \pm 0.07$ meV cuts. (e)–(f) Calculated low-energy spectrum along the Γ - Γ' - Γ'' - Γ''' - Γ path, sketched in (b). (g)–(h) INS-measured low-energy spectrum along Γ - Γ' - Γ'' - Γ''' - Γ from Ref. [21].

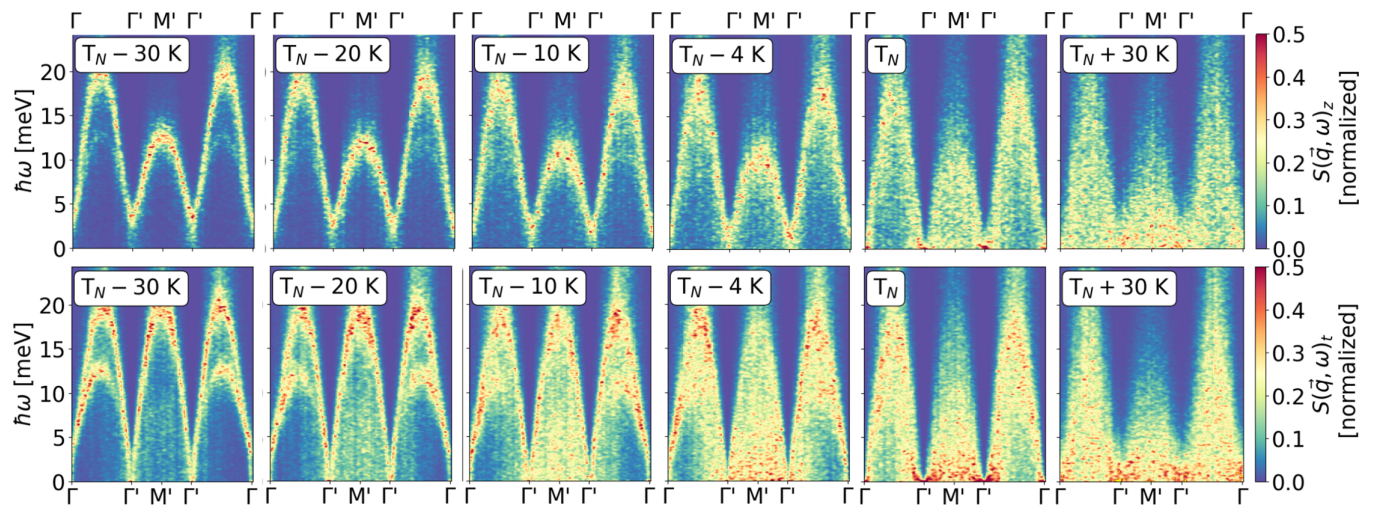


FIG. 2. Calculated temperature dependence of the out-of-plane z (top panel) and transverse t (bottom panel) components of the dynamical structure factor, $S(\vec{q}, \omega)_z$ and $S(\vec{q}, \omega)_t$, respectively, along the Γ - Γ' - M' - Γ' - Γ path.

then in 2 K steps from 100 K to 2 K, evolving the system at each temperature for 60 000 femtosecond time steps; chosen to match the energy resolution to the experimental value of 0.07 meV in Ref. [21]. After initializing the spin directions randomly and cooling the system down to $T_N - 50$ K, the system stabilizes in its ground state configuration [shown in Fig. 1(a)], indicating that the Hamiltonian of Eq. (1) with the parameters of Table I correctly captures the experimentally observed ground state [42].

We now present our calculated dynamical structure factors and compare them to the INS measurements from Ref. [21] in Figs. 1(c)–1(h). Since Ref. [21] only measures the component of the spins perpendicular to the momentum transfer, we first plot our calculated $S(\vec{q}, \omega)_z$ in the $(hk0)$ plane in Fig. 1 at 30 K above and 10 K below our calculated T_N . Figures 1(c) and 1(d) are in-plane $(hk0)$ cuts at constant $\hbar\omega = 1.04 \pm 0.07$ meV. Our simulations reproduce the rodlike diffuse scattering observed in Ref. [21] well above and below T_N , connecting neighboring magnetic Bragg peaks (Γ') along the $(h00)$, $(0k0)$, and $(hh0)$ directions. We reproduce other characteristics of the experimental data, namely the correct energy scale as well as the narrowing of the diffuse rod width in $\vec{q}$ space and the decrease in diffuse scattering around the Γ' points with decreasing temperature. In Figs. 1(e) and 1(f), we plot the calculated frequency dependence of $S(\vec{q}, \omega)_z$ along the Γ - Γ' - M - Γ' - Γ path beside the measured spectra in Figs. 1(g) and 1(h). Our simulations recreate the measured sheet of diffuse scattering along the Γ' - M - Γ' rod path, as well as its attenuation with decreasing temperature. Below T_N , a Γ' -gapped excitation appears at the bottom of the parabolic spin wave at around $\Delta = 2$ meV (we fit a parabola to the low-energy spectrum in Fig. S2 in the SM). The amplitude of this excitation gap is in reasonable agreement with the experimentally measured $\Delta \simeq 1.7$ meV in Refs. [21,44].

We now determine which terms in our Hamiltonian explain the behavior of Fig. 1 by setting individual parameters to zero, and comparing the resulting calculated inelastic magnetic spectra. We find that diffuse rods persist even for a simple model (see Fig. S2 in SM) in which we only include the first-nearest neighbor AFM symmetric exchanges. This

shows that strong geometric frustration arising from AFM-coupled classical spins on a triangular network causes the correlated rodlike diffuse scattering. Additionally, we find that the low-temperature energy dispersion is affected by A^z , with the 2 meV Γ' -gapped excitations [shown in Fig. 1(f)] disappearing when we turn off the hard axis SIA (see Fig S3 in the SM).

Reference [21] models scattering arising from clusters of coherently oscillating spins and since their Hamiltonian is, by definition, isotropic, it cannot describe the $\vec{q}$ space directionality of the short-range correlations. In our simulations, pairwise interactions on a triangular lattice allow for the formation of temperature-dependent clusters of correlated spins. As a result, anisotropic short-ranged correlated fluctuations persist above and below T_N . Note also that the model captures the temperature dependence of the short-range correlations, which is different from that of the long-range order and of conventional thermal diffuse scattering. We illustrate the coexistence and distinct temperature dependencies of the spin wave and the short-range orders in Fig. 3(a), where we compare the temperature evolution of $S(\Gamma', \omega)$, $S(M', \omega)$, and $S((0.1, 0.1, 0), \omega)$, integrating over ω from the elastic line up until 5 meV. We calculate, as expected, that scattering at the Bragg peak has a characteristic sharp increase at T_N , but that the correlated diffuse scattering evolves independently, and deviates from the behavior of the thermal diffuse scattering at $\simeq T_N + 200$ K.

In the following, we analyze the high-frequency and spin-component dependence of the rodlike scattering, and predict and explain characteristics of the diffuse signal that have not yet been measured experimentally. We plot our calculated $S(\vec{q}, \omega)_z$ and $S(\vec{q}, \omega)_t$ on the top and bottom panel of Fig. 2, respectively.

We begin by pointing out the calculated low-temperature magnon branches in the top left plot of Fig. 2 for $S(\vec{q}, \omega)_z$, where we normalize along each $\vec{q}$ point to showcase the fainter higher energy and diffuse scattering (note that our energy dispersions match earlier nonlinear spin-wave theory calculations [25,45]). We distinguish between spectra along paths linking two magnetic Bragg peaks, along the Γ' - M' - Γ'

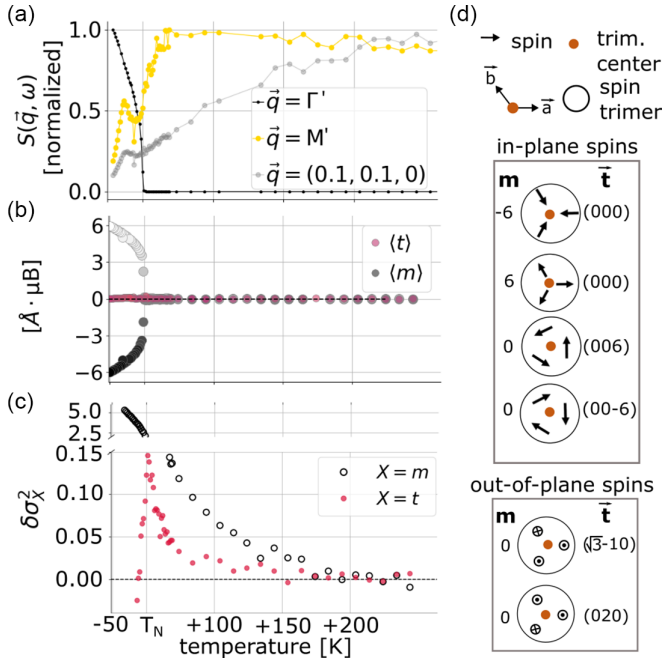


FIG. 3. (a) Three different scattering signals: Bragg peak at Γ' (black), thermal diffuse scattering at a representative $\vec{q} = (0.1, 0.1, 0)$ point (gray), and correlated diffuse scattering at M' (yellow) as a function of temperature, integrated from the elastic line up to 5 meV. (b) Temperature evolution of the monopolar order parameter, averaged over layers converging to a positive $m = +6$ (white) and negative $m = -6$ (black) ground state, as well as of the average of the norm of the toroid $\langle t \rangle$. (c) Temperature evolution of $\delta\sigma_m^2$ and $\delta\sigma_t^2$; the $T \rightarrow \infty$ asymptote is indicated by the black dotted line. (d) Examples of magnetoelectric multipoles. The fractional coordinates of the Mn sites within each trimer of spins are, clockwise from the top left spin, $(\frac{1}{3} + \delta)(-\vec{a} + \vec{b})$, $(\frac{1}{3} + \delta)\vec{a}$, and $(\frac{1}{3} + \delta)(-\vec{a} - \vec{b})$, where δ is the small Mn off-centering.

rod directions, and paths along Γ - Γ' ($\equiv \Gamma'$ - Γ) that join a structural and a magnetic Bragg peak. Along Γ - Γ' , we see a low- and a high-energy magnon branch, peaking at around 13 and 20 meV, respectively, and only the former visibly appears along Γ' - M' - Γ' .

Next, we focus on the temperature dependence of the $S(\vec{q}, \omega)_z$ signal, shown on the top panel of Fig. 2, at five different temperatures. The first notable features are the tails of diffuse scattering above the previously described magnon branches. Most of the scattering, however, occurs below the magnon branches: in particular along Γ' - M' - Γ' , a remarkable continuum of excitations, bounded at high frequency by the magnon branch, starts to appear at $T_N - 4$ K, and persists up to $T_N + 150$ K (see higher-temperature calculations in Figs. S4 and S5 in the SM). Fluctuations at frequencies up to that of the magnon-branch energy therefore dominate the diffuse spectrum. Excitation continua also occur along Γ - Γ' , but have, in addition to an upper bound, a lower bound below T_N , set by the lower magnon branch.

Next, we calculate the as-yet-unmeasured transverse component $S(\vec{q}, \omega)_t$, shown in the bottom panel of Fig. 2, to compare with $S(\vec{q}, \omega)_z$ presented above. Comparing the top and bottom panels of Fig. 2, we see no significant differences

between the in-plane and out-of-plane dynamical structure factors above T_N (see the $T_N + 30$ K plot in Fig. 2, as well as the higher temperature plots in Figs. S3 and S4 in the SM). Notable differences occur below T_N however, where we see that the bounded $S(\vec{q}, \omega)_t$ excitation continuum is more clearly defined than that of $S(\vec{q}, \omega)_z$ along the whole Γ - Γ' - M' - Γ' - Γ path and that, along Γ' - M' - Γ' , the upper-magnon branch [as opposed to the lower-magnon branch for $S(\vec{q}, \omega)_z$] now modulates the higher-energy scattering. These bounded regions of continuous excitations are a classical analog to the multipinon continua observed in low-dimensional magnetically frustrated quantum systems [46–49] and have, to our knowledge, not previously been predicted.

Again, we rationalize the spectra by setting parameters in our Hamiltonian to zero and recalculating $S(\vec{q}, \omega)_{t,z}$. Our main finding is that the hard axis SIA plays a defining role in distinguishing between the $S(\vec{q}, \omega)_t$ and $S(\vec{q}, \omega)_z$ spectral weight distributions in the low-temperature diffuse spectrum. When $A^{zz} = 0$, there is no preference for the spins to lie in the ab plane, and both $S(\vec{q}, \omega)_t$ and $S(\vec{q}, \omega)_z$ carry both the upper and lower magnon branches along Γ' - M' - Γ' (Fig. S6 in the SM). When the spins adopt the in-plane configuration favored by the SIA, in-plane and out-of-plane fluctuations become inequivalent. Since short-range out-of-plane fluctuations can be accommodated without breaking the local 120° nearest-neighbor antiferromagnetic configuration, they contribute to the low-energy mode that emerges in the out-of-plane spectrum below T_N along the Γ' - M' - Γ' rod path, seen on the top panel of Fig. 2. On the other hand, in-plane fluctuations break local 120° nearest-neighbor order and so occur at higher frequency, with dominant spectral weight around the upper magnon branch (lower panel of Fig. 2).

Summarizing our understanding of our simulated spectra so far, from at least 150 K above T_N down to T_N , the SIA is far smaller than the thermal energy: rodlike diffuse scattering originates from first-nearest-neighbor AFM interactions between all three x, y , and z spin components, and we can neglect the SIA and the weak interplane coupling. On further cooling through T_N , we observe the effect of the SIA which breaks the local spin symmetry and the in-plane and out-of-plane correlations start to differ. We further demonstrate the generality of our model by recalculating the $S(\vec{q}, \omega)$ spectra for a LuMnO_3 Hamiltonian (Table S2) in Figs. S8 and S9, reproducing the correlated rodlike diffuse scattering, its upper-bounded energy continuum and its distinct in-plane and out-of-plane characteristics.

Finally, we visualize the correlations in real space. Since triangular arrangements of spins dominate the spectrum in the Néel phase, we find it useful to visualize excitations away from the ground state by associating two multipolar quantities, the local moment magnetoelectric monopole m and toroidal moments $\vec{t}$ [50] of the trimer, with each trimer of spins belonging to the same trimerization center (hereafter, we refer to these as a monopole and toroid, respectively):

$$m = \sum_i \vec{r}_i \cdot \hat{e}_i \quad \text{and} \quad \vec{t} = \sum_i \vec{r}_i \times \hat{e}_i. \quad (2)$$

Here the sum is over the three spin sites, $\vec{r}_i$ is the position of spin i in fractional lattice coordinates, relative to the trimer center, and $\hat{e}_i$ is the unit vector describing the direction of

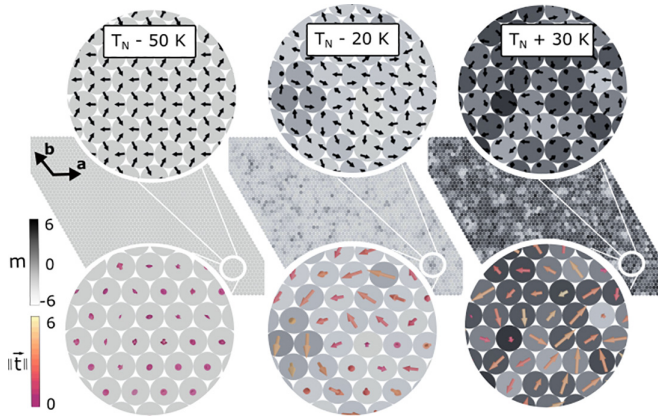


FIG. 4. Spin configurations (top insets) and their associated trimer monopoles (gray-scale disks) and toroids (colored vectors in the bottom insets) at three different temperatures.

the spin. The h -YMnO₃ ground state [shown in Fig. 1(a)] is described by an antiferromonopolar order, corresponding to $m = \pm 6.1 \simeq \pm 6$ alternating along c monopoles and $\vec{t} = (000)$ [see Fig. 3(b)]; changes in these values capture fluctuations away from the ground-state antiferromonopolar order.

In Fig. 3(b), as a function of temperature, we plot the expectation value of m , $\langle m \rangle$, for layers which reach a $+6$ (white) or -6 (black) monopolar ground state at low temperature, as well as of the norm of the system's average $\vec{t}$ vector, $\langle t \rangle$. At 0 K, $\langle m \rangle$ and $\langle t \rangle$ converge to their ground-state values, ± 6 and 0, respectively; we illustrate a near-perfect $m = -6$ monopolar arrangement of spins in a single ab layer at $T_N - 50$ K in the left panel of Fig. 4(a). Collective excitations around this ground state give rise to the low-temperature $S(q, \omega)_{t,z}$ magnon branches in Fig. 2. As the system approaches T_N from below, short-range fluctuations away from the monopolar ground state are superimposed on the long-range order: we see the expected decrease in magnitude in Fig. 3(c) in both $\langle m \rangle$ branches. The real-space behavior in this temperature regime is illustrated by the multipolar projections at $T_N - 20$ K in the middle panel of Fig. 4, where we observe perturbations of the order in the monopolar projection, and a corresponding short-range swirl-like pattern in the $\vec{t}$ inset, reminiscent of the predicted vortex configurations [22,51,52] in this compound. We highlight the surprisingly large out-of-plane collective excitations, captured by the t_x and t_y components, seen in the $\vec{t}$ arrows in the ab plane. We calculate the variance of m and of t (σ_m^2 and σ_t^2) as a function of temperature and

plot in Fig. 3(c) the fractional deviation from their respective $T \rightarrow \infty$ asymptotic values ($\delta\sigma_X^2$) following

$$\delta\sigma_{X=m/t}^2 = \frac{\sigma_X^2 - \sigma_{X,T \rightarrow \infty}^2}{\sigma_{X,T \rightarrow \infty}^2}. \quad (3)$$

We use $\delta\sigma_X^2$ as our real-space metric for the degree of local correlation (in analogy with the reciprocal space correlated diffuse scattering). We calculate that, as the temperature increases above T_N , σ_m^2 and σ_t^2 converge towards their high-temperature paramagnetic asymptotes at $\simeq T_N + 200$ K, consistent with the temperature dependence of $S(\vec{q}, M')$ in Fig. 3(a). We show the spin configuration at $T_N + 30$ K in the right panel of Fig. 4, where nanoscale regions of similar monopolar value indicate that spins still have a preferred local 120° nearest-neighbor configuration. We observe a decrease in correlation between the toroids, compared to its value below T_N , consistent with the increase in dispersion width with increasing temperature in Figs. 1(c)–1(d).

In summary, we have reproduced structured diffuse INS from first principles, capturing its $\vec{q}$ -space directionality and frequency dependence using a combination of a DFT-extracted magnetic Hamiltonian and spin dynamics. New features of this diffuse scattering are also predicted, namely its transverse character, the strong nonlinearity of its associated spin-spin correlations, as well as its bounded excitation continuum. We have provided intuition on the spin excitations responsible for the scattering through a trimer multipolar formalism, which facilitates visualization of the short-range correlations beyond the 120° spin arrangement, highlighting the surprisingly large out-of-plane-correlated spatial fluctuations in the Néel state. We have also shown that the planar magnetoelectric monopole $\langle m \rangle$ is a good order parameter for this system. Our work motivates experimental measurements of the continuous scattering, t - and z -component resolved structure factors and diffuse-rod reciprocal-space direction in h -YMnO₃. We hope that it also inspires theoretical characterization of such structured diffuse scattering in other systems.

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- Additionally, the document contains parameter tables derived from density functional theory and calculated dynamical structure factors for LuMnO_3 .
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