Ba$_8$MnNb$_6$O$_{24}$: A model two-dimensional spin-$\frac{5}{2}$ triangular lattice antiferromagnet

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(Received 24 January 2019; revised manuscript received 28 March 2019; published 28 May 2019)

We successfully synthesized and characterized the triangular lattice antiferromagnet Ba$_8$MnNb$_6$O$_{24}$, which comprises equilateral spin-$\frac{5}{2}$ Mn$^{2+}$ triangular layers separated by six nonmagnetic Nb$^{5+}$ layers. The detailed susceptibility, specific heat, elastic and inelastic neutron scattering measurements, and spin-wave theory simulation on this system reveal that it has a 120$^\circ$ ordering ground state below $T_N = 1.45$ K with in-plane nearest-neighbor exchange interaction $\approx 0.11$ meV. While the large separation 18.9 Å between magnetic layers makes the interlayer exchange interaction virtually zero, our results suggest that a weak easy-plane anisotropy is the driving force for the $k_{\parallel} = (1/3, 1/3, 0)$ magnetic ordering. The magnetic properties of Ba$_8$MnNb$_6$O$_{24}$, along with its classical excitation spectra, contrast with the related triple perovskite Ba$_3$MnNb$_2$O$_9$, which shows easy-axis anisotropy, and the isostructural compound Ba$_8$CoNb$_6$O$_{24}$, in which the effective spin-$\frac{1}{2}$ Co$^{2+}$ spins do not order down to 60 mK and in which the spin dynamics shows signs of strong quantum effects.

DOI: 10.1103/PhysRevMaterials.3.054412

I. INTRODUCTION

Truly two-dimensional (2D) lattices of interacting spins, including triangular, honeycomb, and kagome antiferromagnets, are of central interest to stabilize, explore, and understand exotic quantum states and their excitations [1–22]. However, the experimental realization of an ideal 2D magnetic system embedded in a bulk crystal is very difficult, since undesired factors such as lattice distortions, interplane interactions, and anisotropies are often present and transform the system of interest into, at best, a quasi-2D environment.

The design and synthesis of bulk materials with an ideal 2D magnetic lattice has been a challenge in the materials science community. Here, our strategy to achieve two-dimensionality is to insert nonmagnetic buffer layers between magnetic layers in a three-dimensional crystal structure. This idea has been applied to the perovskite structure (ABO$_3$) comprised of stacked triangular layers of B ions to yield triple perovskite structure (A$_3$B$^+$B$^-$O$_9$). By having triangular layers of magnetic B$^+$ ions separated by two triangular layers of nonmagnetic B ions, the interlayer distance gets increased and the interlayer interactions weakened. As a result, A$_3$B$_2$O$_9$ realizes ideal quasi-2D magnets for exploring exotic magnetic properties. Examples of such a triple perovskite structure are Ba$_3$B$^+$Nb$_2$O$_9$ and Ba$_3$B$^+$Sb$_2$O$_9$ (B$^{2+} = $ Co$^{2+}$, Ni$^{2+}$, and Mn$^{2+}$ with spin number 1/2, 1, and 5/2, respectively) [14,15,20–25].

Among these materials, Ba$_8$CoSb$_2$O$_9$ is arguably the most interesting one stabilizing a one-third magnetization plateau as well as carrying anomalous zero-field magnetic excitations [14–19,26,27]. Proceeding from the triple perovskite structure, and with the strategy outlined above, we previously modified and expanded Ba$_3$CoSb$_2$O$_9$ to Ba$_8$CoNb$_6$O$_{24}$ with six layers of nonmagnetic Nb a vacant layer between each layer of magnetic Co$^{2+}$ ions [28,29]. In Ba$_8$CoNb$_6$O$_{24}$, the intraplane Co-Co distance is 5.86 Å and the interplane Co-Co distance is 7.23 Å, which yields an interlayer exchange interaction (J) around 5% the strength of the intralayer exchange interaction (J) [14–19,26,27]. Also present in Ba$_8$CoSb$_2$O$_9$ is a small easy-plane XXZ anisotropy (the ratio between the longitudinal and transverse exchange interactions is $\Delta \approx 0.9$). In contrast, Ba$_8$CoNb$_6$O$_{24}$ has a similar intralayer Co-Co distance of 5.79 Å, while the interlayer distances expand to 18.90 Å. This reduces the interplane interaction below detectable limits of susceptibility and specific-heat measurements and also removes any resolvable anisotropy, producing a virtually ideal 2D magnetic lattice with no ordering down to 60 mK, as our recent studies show. Moreover, its inelastic neutron scattering spectrum reveals a high-energy continuum also known as two-magnon scattering, which reflects the reduction of the ordered moment by quantum fluctuations. Therefore, Ba$_8$CoNb$_6$O$_{24}$ is a rare example of a spin-$\frac{1}{2}$ triangular-lattice Heisenberg antiferromagnet in the 2D limit [28,29].
The successful reduction of $J'$ and anisotropy in Ba₈CoNb₆O₂₄ calls for engineering a similar material with classical spins. In this paper, we report the synthesis and characterization of Ba₈MnNb₆O₂₄, which is isomorphous to Ba₈CoNb₆O₂₄, but with a Mn²⁺ (spin-$\frac{5}{2}$) 2D triangular lattice. The detailed dc, ac susceptibility, specific heat, elastic, and inelastic neutron scattering measurements on this system reveal that it has a 120° ordering ground state below $T_N = 1.45$ K. The linear spin-wave simulation along with the inelastic neutron scattering (INS) spectra extract the antiferromagnetic nearest-neighbor exchange $J = 0.11$ meV. Unlike the related triple perovskite Ba₃MnNb₂O₉ that shows the easy-axis anisotropy [24], we suggest Ba₈MnNb₆O₂₄ could have a rather weak easy-plane anisotropy.

Our paper is organized as follows. Section II presents our experimental and theoretical methods. Section III presents the results of our thermomagnetic, diffraction, and inelastic neutron scattering characterizations of powder samples of Ba₈MnNb₆O₂₄. Section IV discusses and interprets our experimental results. Section V serves as a conclusion.

### II. EXPERIMENT

Our polycrystalline sample of Ba₈MnNb₆O₂₄ was prepared by solid-state reaction. Stoichiometric amounts of BaCO₃, MnO, and Nb₂O₅ were mixed in agate mortars, compressed into pellets, and annealed for 20 h at temperatures of 1525 and 1600 °C under Ar atmosphere with intermediate mixing. High-resolution neutron powder diffraction (NPD) measurements were performed by a neutron powder diffractometer, HB2A, at the High Flux Isotope Reactor (HFIR), Oak Ridge National Laboratory (ORNL), Oak Ridge, TN. Around 3 g of powder was loaded in an Al-cylinder can and mounted in a close-cycled refrigerator. We used a neutron wavelength of $\lambda = 1.5405$ Å with a collimation of 12′-open-6′. The NPD patterns were analyzed by the Rietveld refinement program FULLPROF [30].

#### TABLE I. Structural parameters for Ba₈MnNb₆O₂₄ at 10 K (space group $P-3m1$) determined from refined NPD measurements.

<table>
<thead>
<tr>
<th>Refinement</th>
<th>Atom</th>
<th>Site</th>
<th>$x$</th>
<th>$y$</th>
<th>$z$</th>
<th>Occupancy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ba₈MnNb₆O₂₄</td>
<td>Ba1</td>
<td>2c</td>
<td>0</td>
<td>0</td>
<td>0.18732(84)</td>
<td>0.16666</td>
</tr>
<tr>
<td></td>
<td>Ba2</td>
<td>2c</td>
<td>1/3</td>
<td>2/3</td>
<td>0.05867(120)</td>
<td>0.16666</td>
</tr>
<tr>
<td></td>
<td>Ba3</td>
<td>2c</td>
<td>1/3</td>
<td>2/3</td>
<td>0.44997(89)</td>
<td>0.16666</td>
</tr>
<tr>
<td>Bragg $R$-factor = 7.28</td>
<td>Ba4</td>
<td>2c</td>
<td>1/3</td>
<td>2/3</td>
<td>0.68171(92)</td>
<td>0.16666</td>
</tr>
<tr>
<td></td>
<td>Mn</td>
<td>1a</td>
<td>0</td>
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<td>0</td>
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</tr>
<tr>
<td></td>
<td>Nb1</td>
<td>2c</td>
<td>0</td>
<td>0</td>
<td>0.38767(61)</td>
<td>0.16667</td>
</tr>
<tr>
<td></td>
<td>Nb2</td>
<td>2d</td>
<td>1/3</td>
<td>2/3</td>
<td>0.25790(82)</td>
<td>0.16667</td>
</tr>
<tr>
<td></td>
<td>Nb3</td>
<td>2d</td>
<td>1/3</td>
<td>2/3</td>
<td>0.87545(74)</td>
<td>0.16667</td>
</tr>
<tr>
<td></td>
<td>O1</td>
<td>6i</td>
<td>0.17214(116)</td>
<td>0.82786(116)</td>
<td>0.30799(46)</td>
<td>0.50</td>
</tr>
<tr>
<td></td>
<td>O2</td>
<td>6i</td>
<td>0.16375(92)</td>
<td>0.83615(92)</td>
<td>0.57010(49)</td>
<td>0.50</td>
</tr>
<tr>
<td></td>
<td>O3</td>
<td>6i</td>
<td>0.16731(117)</td>
<td>0.83259(117)</td>
<td>0.93445(58)</td>
<td>0.50</td>
</tr>
<tr>
<td></td>
<td>O4</td>
<td>6i</td>
<td>0.49493(118)</td>
<td>0.50497(118)</td>
<td>0.18831(31)</td>
<td>0.50</td>
</tr>
</tbody>
</table>

**Space group** $P-3m1$

**Lattice parameters (Å)**

$a = b = 5.80701(6)$, $c = 18.94654(29)$

**Overall B-factor** = 0.156 (Å$^2$)

**Magnetic space group** $P-1$

**Momentum** $4.05(35) \mu_B$
FIG. 2. (a) Stacked layer structure of Ba₈MnNb₆O₂₄. (b) Unit cell of Ba₈MnNb₆O₂₄ and spin structure of Mn²⁺ ions at zero field. (c) Triangular lattice composed of Mn²⁺ ions in the ab plane.

The ac susceptibility measurements were taken using the conventional mutual inductance technique with a homemade setup [31]. The specific-heat data were obtained using a commercial physical property measurement system (PPMS, Quantum Design).

III. RESULTS

A. Lattice structure

Figure 1 shows the NPD pattern of Ba₈MnNb₆O₂₄ measured at T = 10 K with wavelength λ = 1.5405 Å. A Rietveld refinement yields a pure phase of the space group P-3m1 with the lattice parameters of a = 5.8070(1) Å and c = 18.9465(3) Å. The details of the structural parameters are listed in Table I and confirm that Ba₈MnNb₆O₂₄ is isostructural to Ba₈CoNb₆O₂₄. No antisite disorder between Mn²⁺ and Nb⁵⁺ ions was observed, while ∼3% Ba₃MnNb₂O₉ had been identified as the impurity. A crystallographic unit-cell [Fig. 2(a)] contains a vacant layer and six layers of nonmagnetic NbO₆ octahedral separating triangular layers of Mn²⁺ ions. While the intralayer Mn-Mn distance is around 5.81 Å, the interlayer Mn-Mn distance is as large as 18.91 Å [Fig. 2(b)]. This remarkable structure is expected to guarantee that the interlayer interaction of Ba₈MnNb₆O₂₄ is approaching the zero limit.

B. dc and ac susceptibility

Figure 3(a) reports the temperature dependence of the magnetic dc susceptibility χ of Ba₈MnNb₆O₂₄. No sign of magnetic ordering is observed down to 1.8 K, and a Curie-Weiss fit of the inverse dc susceptibility in the range of temperatures 100–350 K yields μₑff = 6.04 μₜ and θ_W = −10.7 K. The negative Weiss constant indicates overall antiferromagnetic exchange interactions, while the obtained effective magnetic moment agrees well with the value 5.93μₜ expected for spin-only S = 5/2 magnetic moments [32]. Hereafter, we will consider the Hamiltonian of a triangular Heisenberg antiferromagnet,

\[ \mathcal{H} = J \sum_{\langle i,j \rangle} \left( S_i^x S_j^x + S_i^y S_j^y + \Delta S_i^z S_j^z \right) \]  

where \( \langle i, j \rangle \) indicates the nearest neighbors and \( \Delta \) indicates the potential easy-plane anisotropy which is not considered until Sec. III D. Considering z = 6 nearest-neighbour magnetic moments coupled with the Heisenberg exchange interaction \( J \), mean-field theory yields \( \theta_W = -J/S(S+1)/6k_B \), which corresponds to \( J/k_B = -4/3S/\theta_W = 1.22 \) K for Ba₈MnNb₆O₂₄. The isothermal dc magnetization taken at T = 0.6 K is shown in Fig. 3(b) and yields a moment of 5.5μₜ above the saturation magnetic field \( \mu_0 H_s \approx 22.5 \) T. The value of the saturated moment corresponds to a powder-averaged gyromagnetic ratio \( g = 2.1 \) for Mn²⁺ ion assuming \( S = 5/2 \). Therefore, the exchange interaction, \( J = \mu_0 S^2 \theta_W^{-1} \approx 1.41 \) K, from the saturated field agreed with the temperature-dependence measurement of magnetization. The field derivative of the magnetization shows no obvious valley or peak that would indicate possible spin-state transitions.

The ac susceptibility, \( \chi_{ac} \), measured down to 0.3 K at a frequency of 347 Hz was used to probe lower-temperature
magnetic properties of Ba$_8$MnNb$_6$O$_{24}$, as reported in Fig. 4. At zero dc field, $\chi_{ac}$ shows a sharp peak at 1.45 K, which indicates a transition to a long-range magnetic order. Upon application of a small dc magnetic field of 0.1 T, the peak broadens with a possible shoulder around 2.3 K. The susceptibility was fitted by two Gaussian peaks to determine the transitions of $T_{N1}$ and $T_{N2}$, Fig. 4(a) (solid lines). A possible scenario to explain this behavior is that the single magnetic transition at zero dc field evolves into two transitions upon increasing the dc field, with $T_{N1}$ referring to the low-temperature transition and $T_{N2}$ the high-temperature one. With increasing dc field, Fig. 4(a), $T_{N2}$ shifts to higher temperatures while $T_{N1}$ shifts to lower temperatures and ultimately reaches below 0.3 K at 3.0 T. With even larger dc fields ($H > 3.0$ T), $T_{N2}$ changes behavior from a maximum above 3.5 K around 4.0 T and shifts to lower temperatures with increasing dc field. Using the saturation field obtained from the dc magnetization and the $T_{N}$‘s obtained from the ac susceptibility data, we draw a magnetic phase diagram, Fig. 5(a), for Ba$_8$MnNb$_6$O$_{24}$ and compare it to recent results for Ba$_3$MnNb$_2$O$_9$ [24]. A more detailed discussion of this phase diagram will be presented below.

C. Specific heat

In Fig. 6(a) we show the specific heat, $C_p$, of Ba$_8$MnNb$_6$O$_{24}$ measured in magnetic fields of 0, 7, and 14 T. At zero field, $C_p$ shows a broad peak around 4 K but no significant feature matching the kink observed around 1.45 K in the temperature dependence of the ac susceptibility. Moreover, the data show a sharp increase below 200 mK, which we ascribed to the nuclear Schottky anomaly of the Mn ions, since the naturally abundant 55Mn has nuclear spin $\frac{5}{2}$. This can also explain why the upturn shifts to higher temperatures with higher magnetic fields, because the magnetic ordered moment increases with the magnetic field and hence opens up the nuclear spin levels through the hyperfine coupling, which leads to the transition at higher temperatures. With increasing fields, the broad peak is somewhat suppressed. We isolate the magnetic contribution to the specific heat, $(C_m)$, by
subtracting the specific heat of the isostructural nonmagnetic compound \( \text{Ba}_8\text{ZnTa}_6\text{O}_{24} \). This yields the magnetic entropy change calculated by integrating \( C_m/T \) with lattice subtracted starting at 200 mK.

D. Magnetic structure and excitations

In Fig. 7 we show the NPD pattern of \( \text{Ba}_8\text{MnNb}_6\text{O}_{24} \) measured at 0.3 K with wavelength \( \lambda = 2.4127 \text{ Å} \). The data reveal the presence of magnetic Bragg peaks that can be indexed by \( \mathbf{Q} = \mathbf{r} + \mathbf{k}_m \), where \( \mathbf{r} \) is a reciprocal-lattice vector and \( \mathbf{k}_m = (1/3, 1/3, 0) \). In the inset of Fig. 7, we highlight the difference between the 0.3 and 10 K data, which reveals the details of these magnetic Bragg peaks. The refined magnetic structure for the above propagation vector \( \mathbf{k}_m \) is shown in Fig. 1(c): it corresponds to a 120° spin-structure in the \( ab \) plane or in a plane containing the \( c \) axis (an easy-axis type 120° structure).

FIG. 7. Rietveld refinement of the neutron powder diffraction pattern measured at \( T = 0.3 \text{ K} \) with \( \lambda = 2.4127 \text{ Å} \) for \( \text{Ba}_8\text{MnNb}_6\text{O}_{24} \). Black circles are the measured intensity, the red line is the calculated intensity, and the blue line is the difference. Tick marks are the lattice Bragg peak and aluminum and magnetic Bragg peak positions, from top to bottom, respectively. Inset: Highlight of the difference between the 0.3 and 10 K neutron powder diffraction patterns to show the magnetic Bragg peak positions.

cannot reveal whether the spins lie in the \( ab \) plane or in a plane containing the \( c \) axis (an easy-axis type 120° structure).

Figure 8 shows the powder INS spectra of \( \text{Ba}_8\text{MnNb}_6\text{O}_{24} \) measured at 10, 4.0, 1.5, 0.4, and 0.05 K, respectively. The INS intensity as a function of momentum transfer \( E \) allows us to track the development of magnetic correlations upon lowering \( T \). At high temperatures such as 10 and 4 K, the INS intensity shows a clear short-range magnetic signal with a momentum dependence peaked at \( Q = 0.7 \text{ Å}^{-1} \). Upon decreasing temperature and approaching \( T_N \) (1.45 K), sharper ridges of intensity emerge from \( Q = 0.7 \text{ Å}^{-1} \) with less intense repetitions at 1.5 and 2.0 Å\(^{-1} \), in perfect agreement with the powder-diffraction results. The energy dependence of the main signal reveals gapless excitations extending up to 1.0 meV. These features change very little below \( T_N \) from 0.4 to 0.05 K. This further confirms the occurrence of a well-correlated magnetic state below 1.0 K, consistent with long-range magnetic order.

To model the dynamic magnetic correlations in the ordered state, we resort to linear spin-wave theory at zero temperature [33]. We use a canonical Heisenberg Hamiltonian [Eq. (1)] with the 120° magnetic structure as the ground state. Due to the powder averaging effect, information about possible exchange anisotropies, which we expect to be relatively small, cannot be accurately extracted from the INS data and therefore are not considered here. The best match between the experimental data measured at \( T = 0.05 \text{ K} \) and the simulation, Fig. 8(f), is achieved with the nearest-neighbor exchange interaction \( J = 0.11 \text{ meV} \) (or 1.28 K). This value is consistent with the \( J = 1.22 \text{ K} \) calculated from the Curie-Weiss temperature. As shown in Figs. 8(e) and 8(f), the calculated spectrum reproduces the main features of the experimental data, such
as the positions of the zone centers and the bandwidth of the magnetic excitations. A more detailed illustration of the good match between experiment and our model is evident in $E$-integrated [Fig. 9(a)] and $Q$-integrated [Fig. 9(b)] cuts. We may also estimate the upper bound of the easy-plane anisotropy $\Delta$ from the INS data. Although such anisotropy does not gap the entire spin-wave dispersion relation in the Brillouin zone, there will be major intensity shifted up in the calculated spin dynamical structure factor at the ordering wave vector. This shift is essentially the energy gap of the out-of-plane mode. Within the linear spin-wave theory, the gap $\Delta \varepsilon$ is proportional to

$$\Delta \varepsilon = 3JS\sqrt{3(1-\Delta)/2}.$$  

(2)

Meanwhile, the inelastic neutron scattering data we obtained have an incoherent elastic line that extends to roughly 0.2 meV. Therefore, we cannot resolve any potential gap if it is smaller than that. Now we just plug the values $J = 0.11$ meV, $S = 5/2$ into the above equation. We can obtain that $\Delta > 0.96$.

IV. DISCUSSION

A noteworthy feature of $\text{Ba}_8\text{MnNb}_6\text{O}_{24}$ is that the heat-capacity $C_p$ shows no clear sign of long-range magnetic ordering but a broad peak around 4 K. Previous quantum Monte Carlo studies on quasi-2D antiferromagnetic Heisenberg models have shown that the onset of long-range magnetic ordering is accompanied with a sharp peak in $C_p$ even for interlayer exchange interactions as small as $J'/J = 2 \times 10^{-4}$; see Ref. [34]. Upon further decreasing the interlayer coupling, the sharp peak disappears and only a broad peak remains.

Therefore, we believe the sole broad peak in $C_p$ hints at the almost ideal two-dimensional nature of magnetism in $\text{Ba}_8\text{MnNb}_6\text{O}_{24}$. At the same time, the broad peak indicates that the short-ranged spin correlations have already developed at temperatures higher than $T_N$. This is consistent with the INS observation, which shows that broad magnetic signals have already developed at as high as 10 K.

In spite of its almost ideal two-dimensional magnetism, $\text{Ba}_8\text{MnNb}_6\text{O}_{24}$ still appears to order at $T_N = 1.45$ K with a $120^\circ$ ordering structure as confirmed by the ac susceptibility and neutron diffraction experiments. Since $T_N$ increases logarithmically in the interlayer interaction or in the exchange anisotropy [35–41], we conjecture that the magnetic transition of $\text{Ba}_8\text{MnNb}_6\text{O}_{24}$ is most likely driven by an easy-plane anisotropy.

It is instructive to compare the magnetic properties of $\text{Ba}_8\text{MnNb}_6\text{O}_{24}$ with those of the related quasi-2D compound $\text{Ba}_3\text{MnNb}_2\text{O}_9$. For this purpose, the magnetic phase diagram for $\text{Ba}_3\text{MnNb}_2\text{O}_9$ is reproduced in Fig. 5(b). The major differences between these two phase diagrams are as follows. First, a two-step transition at $T_{N1} = 3.4$ K and $T_{N2} = 3.0$ K occurs in $\text{Ba}_3\text{MnNb}_2\text{O}_9$, which indicates its easy-axis anisotropy. This is not only the normal behavior for Mn$^{2+}$ ions on octahedral sites as for $\text{Rb}_2\text{Mn(MoO}_4)_3$ [42], but also for the distorted triangular lattice in $\text{A}_3\text{NiNb}_2\text{O}_9$ [43] and $\text{NaCrO}_2$ (NiGa$_2$S$_4$) [44]. For some special case, the TLAF could exhibit easy-plane anisotropy. For example, the recently studied TLAF $\text{Ba}_2\text{La}_2\text{MnW}_2\text{O}_{12}$ shows a single-step transition, in which the competition between the antiferromagnetic Mn-O-O-Mn and ferromagnetic Mn-O-W-O-Mn could be the reason for the easy-plane anisotropy. In contrast, $\text{Ba}_8\text{MnNb}_6\text{O}_{24}$, potentially with a weak easy-plane anisotropy with transferring from one
magnetic transition at 0 T to two transitions at very low field (0.1 T), sits closer to the limit of Heisenberg spins. Second, the magnetic phase diagram for Ba$_3$MnNb$_2$O$_9$ evolves from the zero-field $120^\circ$ ordering to canted $120^\circ$ ordering, UUD phase, and then oblique phase with increasing applied field, while there is no indication for the existence of the UUD phase in Ba$_8$MnNb$_6$O$_{24}$. Whether this disappearance of UUD phase is intrinsically related to dimensional reduction in Ba$_8$MnNb$_6$O$_{24}$ or is extrinsic due to the polycrystalline sample nature (in which the powder average effect smears the phase boundaries) needs to be clarified by more studies on putative single crystalline samples in the future.

Finally, we compare Ba$_8$MnNb$_6$O$_{24}$ with Ba$_8$CoNb$_6$O$_{24}$, where the main difference lies in the spin quantum number. With $S = 1/2$ magnetic moments, Ba$_8$CoNb$_6$O$_{24}$ is subject to stronger quantum fluctuations and thus exhibits no long-range magnetic order down to $T = 60$ mK and calls for $1/S$ spin-wave theory to model its magnetic excitations. Meanwhile, Ba$_8$MnNb$_6$O$_{24}$, with $S = 5/2$, is essentially in the classical limit and the resulting spin dynamics are well described by linear spin-wave theory without considering any form of magnon-magnon interactions. On top of quantum effects, thermal fluctuations should play an influential role in both systems. Therefore, Ba$_8$MnNb$_6$O$_{24}$, free from strong quantum fluctuations, may serve as a good candidate to investigate the role of thermal fluctuations.

V. CONCLUSION

We presented a detailed experimental study of the triangular lattice antiferromagnet Ba$_8$MnNb$_6$O$_{24}$ with $S = 5/2$ Mn$^{2+}$ ions forming equilateral triangular lattices. Our results reveal that despite the almost ideally 2D nature of the magnetism and the likely vanishing interlayer interaction, long-range magnetic order develops at $T_N = 1.45$ K. Specific-heat measurements along with an inelastic neutron scattering study show that short-ranged spin correlations are well formed at a temperature as high as $T = 10$ K. Linear spin-wave theory simulations yield a nearest-neighbor interaction around 0.11 meV, in good agreement with an estimate from magnetic susceptibility measurements. By comparing with the related triple-perovskite Ba$_4$MnNb$_2$O$_8$ and the isostructural Ba$_8$CoNb$_6$O$_{24}$ with effective spin-$1/2$ Co$^{2+}$ ions, we elucidated the subtle role played by the quantum spin number and putative weak anisotropies to produce long-range magnetic ordering in the 2D triangular lattice Heisenberg antiferromagnet Ba$_8$MnNb$_6$O$_{24}$.

ACKNOWLEDGMENTS

J.M. gratefully acknowledges the support from the Chinese Spallation Neutron Source (CSNS) User Special Grant, the NSF China (11774223), and the Ministry of Science and Technology of China (2016YFA0300500). R.R., Q.H., and H.D.Z. gratefully acknowledge the support from NSF-DMR through Award No. DMR-1350002. The work at Georgia Tech (L.C. and M.M.) was supported by the National Science Foundation through Grant No. NSF-DMR-1750186. This research used resources at the High Flux Isotope Reactor, a DOE Office of Science User Facility operated by the Oak Ridge National Laboratory. A portion of this work was performed at the NHMFL, which is supported by National Science Foundation Cooperative Agreement No. DMR-1157490 and the State of Florida. E.S.C. and M.L. acknowledge the support from NSF-DMR-1309146.
