Persistent spin waves above the Néel temperature in $YMnO_3$

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We have investigated the spin excitations in the frustrated triangular antiferromagnet $YMnO_3$ by inelastic neutron scattering in the ordered magnetic state and also above the Néel temperature. In the ordered phase, we observe broad spin excitations, which can be described by an anisotropic Heisenberg model. Above T_N, well defined signatures of persistent spin waves were found evidenced by inelastic peaks in energy spectra. Neatly separated excitations from the quasielastic fluctuations have been identified in this two-dimensional magnetic system. The clear observation might be related to the geometrical frustrated and short ranged ordered nature of this triangular antiferromagnet.

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Much of condensed matter physics can be learned from their elementary collective excitations, such as phonons and spin waves.[1] These quasiparticles are described by their momentum and energy, are well defined in the ordered state, and can often survive a multitude of oscillations. In the disordered state, e.g., liquids, it has been recognized since long now that acousticlike phonons, although often heavily damped, are part of the spectrum, in particular, for liquid metals.[2] Much more confusing is the situation with collective magnetic excitations in the paramagnetic state. In particular, over three decades, the nature of the magnetic excitations in itinerant ferromagnetic metals above the Curie temperature has been a matter of heated controversy. Neutron scattering experiments on iron by Collins *et al.*[3] suggested that spin waves were normalized to zero above T_C. Later, Lynn[4] reported persistent spin waves in Fe and Ni up to the highest measured temperature $1.4T_C$ and no further renormalization of the dispersion was observed above T_C. This result was, however, questioned by Shirane and co-workers.[5] One of the reasons for the debate was and is a lack of convincing energy spectra, which show an inelastic excitation nicely separated from the quasielastic intensity. Recently, Tao *et al.*[6] have used computer simulations to elucidate this longstanding controversy. Spin dynamics simulations of the dynamic structure factor of a Heisenberg model of Fe with first principles interactions reveal that well defined peaks persist above T_C. At large wave vectors, these peaks have been attributed to propagating spin waves and at small wave vectors only overdamped spin waves have been observed. Common ground on the controversy can be found accepting that short range order may exist above T_C and on these length scales spin waves are supported. In contrast to these itinerant systems, inelastic peaks have been reported for the three-dimensional ideal Heisenberg model EuS.[7] Assuming that short range correlations are a necessity, a natural system to look for would be materials with anisotropic interactions such as low-dimensional magnets. Even though long range order is erased above the transition temperature, within the strong interaction paths short range order could persist and support collective excitations. In fact, Skalyo *et al.*[8] reported spin waves of the zone-boundary mode in the linear chain antiferromagnet $CsMnCl_3$ with energy scans up to $2T_N$ and related this result to intrachain correlations. In the planar antiferromagnet K_2NiF_4, Skalyo *et al.*[9] and Birgeneau *et al.*[10] observed magnons over most of the Brillouin zone to persist well above T_N, but only constant-Q scans have been shown. Recently, we demonstrated that a spin wavelike dispersion persists above T_C in the two-dimensional bilayer manganite $La_{1.8}Sr_{1.2}Mn_2O_7$.[11] Once more, the results have been obtained by constant energy cuts and constant-Q cuts only deliver a monotonously falling spectrum. In this Brief Report, we provide direct evidence from energy spectra for persistent spin waves above the ordering temperature T_N in the quasi-two-dimensional antiferromagnet $YMnO_3$ applying mapping techniques for three-axis spectrometer.

The hexagonal manganites $RMnO_3$ (R=Ho,Er,Tm,Yb, Lu,Y,Sc) have attracted considerable interest due to their multiferroic properties, i.e., they are both ferroelectric and antiferromagnetic (e.g., see for recent reviews[12]) and the corresponding order parameters couple to each other although the ferroelectric transition temperature is as high as $T_c \sim 900$ K, whereas the Néel temperature is $T_N \sim 70$ K.[13] $YMnO_3$ crystallizes[14] in the hexagonal space group $P6_3cm$ with lattice parameters $a=6.1553\pm0.0003$ and $c=11.4026\pm0.0008$ Å. $YMnO_3$ orders antiferromagnetically below $T_N \approx 70$ K with the spin moments lying in the ab plane. The magnetic structure of $YMnO_3$ has recently been measured by three dimensional neutron polarimetry.[15] This investigation unveiled that the magnetic space group of the antiferromagnetic phase of $YMnO_3$ is $P6_3'$. Due to the quasi-two-dimensional triangular lattice, the magnetic system is geometrically frustrated, which is evidenced[16] by the large ratio of Weiss-to-Néel temperatures, $\frac{\Theta}{T_N} \sim 10$. Diffuse magnetic scattering demonstrated the quasi-two-dimensional character of $YMnO_3$ by streaks of intensity parallel to c^*.[17] This behavior persisted up to $2T_N$ and indicates short range ordering far above the ordering temperature. Spin dynamics of $YMnO_3$ at low temperatures has been, studied by inelastic neutron scattering on a polycrystalline sample[18] and a single crystal.[19] The spin wave dispersion was described by an anisotropic two-dimensional (2D) Heisenberg model. The interplane interaction turned out to be two orders of magnitude smaller than the inplane interaction and $YMnO_3$ can be assumed as a good realization of a 2D triangular lattice antiferromagnet. The measurements from the polycrystalline sample provided evidence for short range order and a spin liquid state above T_N;[18] Roessli *et al.* investigated the spin

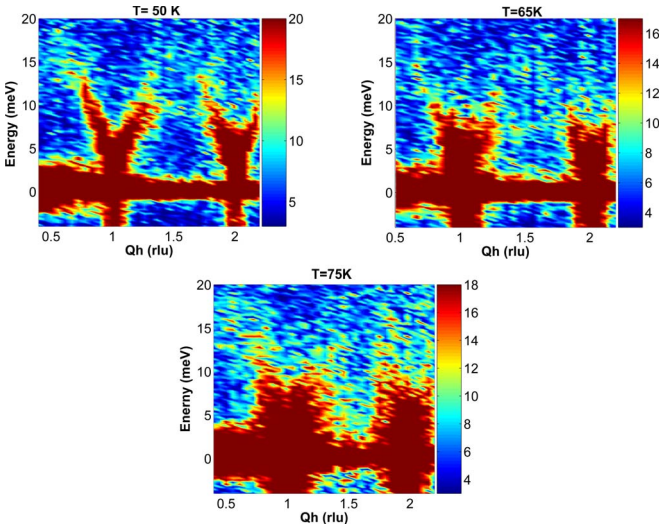

FIG. 1. (Color online) Maps of the spin excitations at $T=50$, 65, and 75 K in the (Q,ω) space. At 75 K at small Q_h values still an excitation branch is visible separated from the quasielastic intensity.

fluctuations around T_N of a $YMnO_3$ single crystal with polarized neutrons and revealed an inelastic peak at the zone center just above the ordering temperature related to nondispersive out of plane spin fluctuations.[20] Therefore, all ingredients, such as low dimensionality and short range order, are given for a possible finding of persistent spin waves in $YMnO_3$.

We mapped the spin dynamical response in the (Q,ω) space on the three-axis spectrometer IN3 equipped with the multianalyzer detector. The instrument IN3 is located in the neutron guide hall of the Institut Laue-Langevin in Grenoble and has negligible background. A $YMnO_3$ single crystal of size $4\times5\times4$ mm^3 has been used for the investigations. The multianalyzer technique is particularly useful for low-dimensional systems, such as $YMnO_3$.[21] All 45 copper ana-

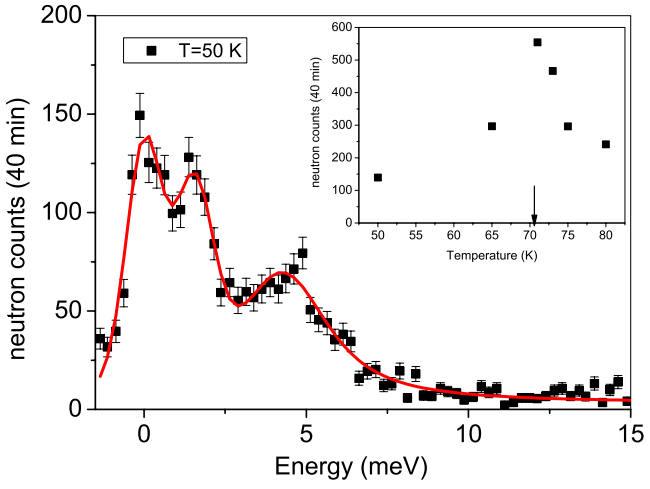

FIG. 2. (Color online) A cut at $Q_h=1.0$ of the 50 K data is depicted with a fit using three Lorentzians. The inset shows the temperature evolution of the elastic intensity at the momentum transfer $Q_h=(1.0,0,-0.5)$.

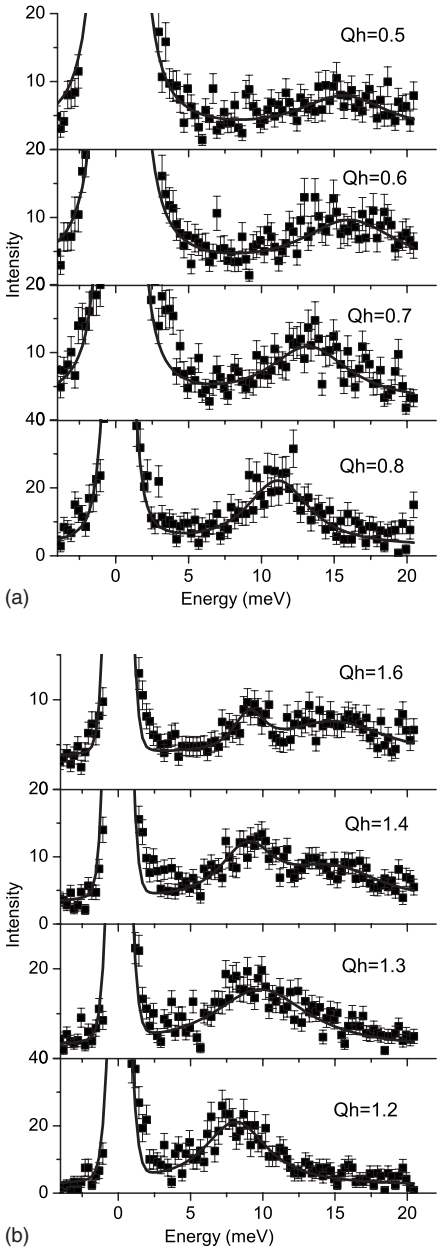

FIG. 3. Constant-Q_h scans at $T=50$ K with fit functions. At small Q_h, one excitation can be identified. At $Q_h\approx1.5$, two excitations are visible.

lyzers were aligned to the same final energy $E_f=31$ meV. The energy resolution was 1.2 meV and the Q resolution was about ±0.03 Å^{-1} for each channel. The crystal was aligned with the $\mathbf{a}^*$ and $\mathbf{c}^*$ axis in the scattering plane and the $\mathbf{k}_i$ vector parallel to $\mathbf{c}^*$. In this setup, the different analyzers cover a range of different Q_h vectors and by changing the incoming energy they remain on their particular Q_h value. Concomitant with the change in energy is a change in Q_l. Because the dispersion of this low-dimensional material does not change appreciable in this direction, all analyzers together scan the whole dispersion simultaneously, a decisive advantage with this technique. The crystal was mounted in an orange cryostat with a temperature stability of ±0.5 K during the experiment. Up to seven scans have been per-

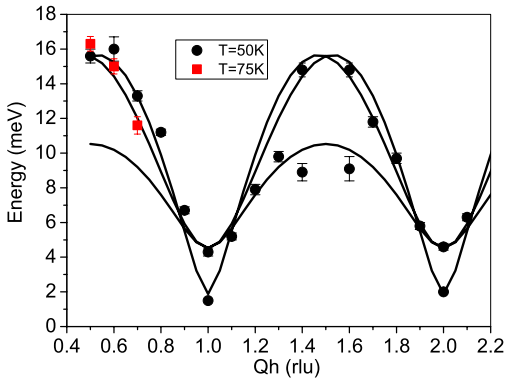

FIG. 4. (Color online) Dispersion relation derived from the 50 K data. The lines show the dispersion calculated from a nearest-neighbor Heisenberg model with two anisotropies. The squares denote the fit results from the 75 K data.

formed in steps of 0.25 meV with a total counting time of about 40 min per energy point. Figure 1 shows such mapping at $T=50$, 65, and 75 K. The large array of analyzers allowed covering of nearly two Brillouin zones at once. At $T=50$ K, the relatively sharp excitations in the map suggest propagating spin waves. At $T=65$ K, which is about 5 K below $T_N \approx 70$ K, the excitations become already much broader and one mode shifts to lower energies as one can see for the lower excitation branch near $Q_h=1.5$. At the highest temperature $T=75$ K, the overall picture looks diffusivelike. However, at the smallest Q vectors $Q_h \approx 0.5$, one can still identify a separated excitation branch from the elastic line, even though strongly broadened. We obtained constant-Q_h cuts from these maps by searching within the measured $(Q\ \omega)$ grid. For the constant-Q_h scans, single analyzer channels have been used where the dispersion is steep and at the zone boundaries $(Q_h=0.5,0.6,1.4,1.6)$ three or four channels have been averaged. The constant-Q spectra have been fitted by a model function of a sum of Lorentzians convoluted with the Gaussian resolution function. Figure 2 shows a cut at $Q_h=1.0$ for $T=50$ K with the fitted model function depicted as a line. The energy resolution is just good enough to identify two gap excitations due to anisotropies. The derived values from the fit, $\omega_1=1.5$ meV and $\omega_2=4.3$ meV, are in accord with depicted data from Sato *et al.*[19] interpolating their different temperatures and keeping in mind the slightly different Q_l. In the inset, the evolution with temperature of the elastic intensity $I(Q=(1.0,0,-0.5),\omega=0)$ at this momentum transfer value is provided. The arrow indicates $T_N=71$ K determined previously[22] from the same single crystal. From the temperature dependence, it is evident that the 75 K measurement is well beyond the transition temperature. Figure 3 shows further constant-Q cuts including the model fit function demonstrating dispersing spin excitations. At the small Q values, only one excitation branch appears, probably due to a vanishing structure factor. At Q vectors around $Q_h=1.5$, two modes are visible. The higher energy branch is quite broad and it is not possible to differentiate further excitations. Except at the zone center, all excitations are much broader than the energy resolution. The extracted spin wave dispersion determined at $T=50$ K is shown in Fig. 4. In-

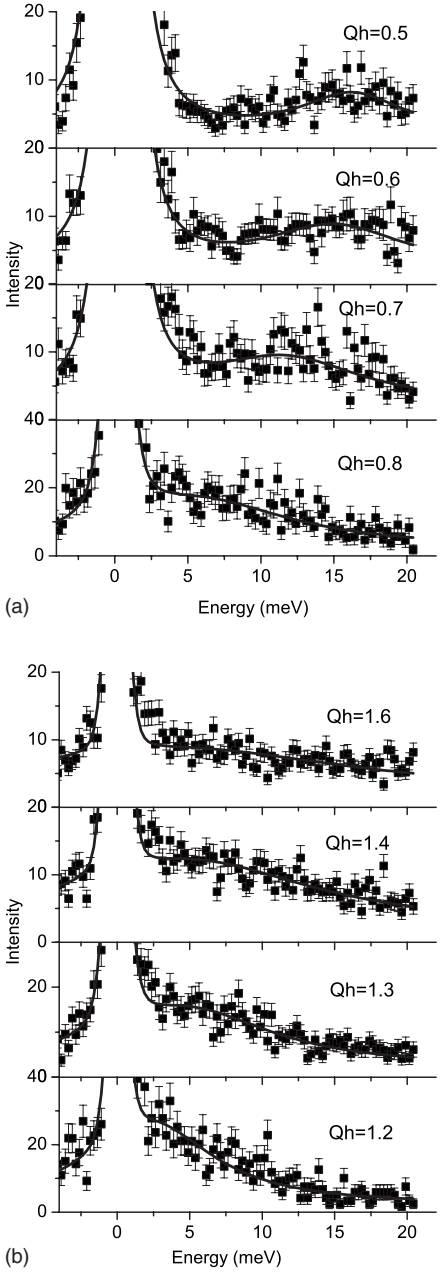

FIG. 5. Constant-Q_h scans at 75 K. Included are fits with a two Lorentzian model.

cluded is the dispersion calculated from an anisotropic nearest-neighbor Heisenberg model, similar to the one used to describe $HoMnO_3$.[23] The dispersion of the three modes i, now including two anisotropies, is given then by[22]

$$\hbar\omega_i = 3JS\sqrt{(1+4z_i+2D_z/3J)(1-2z_i+2D_y/3J)} \qquad (1)$$

with $S=2$ and the anisotropies D_z and D_y. The z_i are defined by lattice Fourier sums identical to Ref. 23. The antiferromagnetic nearest-neighbor interaction in the ab plane J is $J=2.4$ meV and the anisotropies are $D_z=0.24$ meV and $D_y=0.02$ meV. The interaction J is in good agreement with that determined at low temperature by conventional three-axis

measurements, but the anisotropies are reduced with increasing temperature.[22]

Now, we consider the excitation spectra above T_N. Figure 5 shows the constant-Q_h cuts for $T=75$ K. For Q vectors beyond $Q_h=1.0$, no distinct inelastic peak can be identified and the spectra look diffusive. Surprisingly for smaller Q vectors, inelastic peaks still persist at $T=75$ K, which is 5 K above T_N. For three Q vectors, the fit with the model delivers excitation frequencies, which are larger than the width. At $Q_h=0.8$, these numbers change to an overdamped excitation, easily visible from the spectrum. The three excitation energies from the fit are included in Fig. 4 and show only small changes to the values below T_N. However, the widths of the excitations have increased considerably. The shortened lifetime combined with the appearance of two excitation branches is probably the reason why beyond $Q_h>1.0$, no distinct excitation can be identified. Note that constant energy scans would provide a nice dispersion over a large range of Q vectors. To our best knowledge, these are the first experimental evidences of clearly visible inelastic peaks above the ordering temperature in a constant-Q scan in a 2D system. This fact implies the existence of persistent spin waves above the ordering temperature in $YMnO_3$. In this two-dimensional magnetic system, the spins in the magnetic planes have short range ordering above T_N and therefore could sustain propagating, albeit damped spin waves. The large Curie-Weiss temperature [$\theta=705$ K (Ref. 16)] indicates a strong spin-spin interaction inside the planes, but geometrical frustration prevents long range ordering at higher temperatures. The negligible renormalization of the excitation energies implies that the macroscopic transition temperature T_N does not influence the excitation frequency on short wavelengths. However, the induced disorder could be the reason for the shortened lifetime. We suppose that the combination of strong interaction and frustration, resulting in short range order above T_N, permits the propagation of spin waves above the transition temperature. The interplay of geometrical frustration and low dimensionality is a source for new aspects in spin dynamics—not only at low temperatures.

In conclusion, we showed inelastic spectra obtained by neutron scattering from the 2D triangular antiferromagnet $YMnO_3$ below and above T_N. Applying an alternative technique, we have been able to map out nearly two Brillouin zones simultaneously. Above the ordering temperature, clear evidence for spin waves was found by energy spectra from constant-Q scans. The relation of a strong interaction and frustration may be the key to understand our results. Further investigations on similar systems, simulations, and theoretical work are necessary to catch more than a glimpse of propagating spin excitations above T_N.

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