New Nonlinear Modes in Planar Antiferromagnets: A Study of Polarized-Neutron, Inelastic Scattering of $(CD_3)_4NMnCl_3$ in a Transverse Field

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Neutron inelastic scattering combined with neutron-polarization analysis has been used to observe new, nonlinear excitations in the planar antiferromagnet $(CD_3)_4NMnCl_3$ (TMMC) at low temperatures and in the presence of a large, transverse magnetic field. The new excitations result from a field-induced nonlinear coupling of in-plane and out-of-plane magnons.

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In recent years, fascinating dynamical effects associated with nonlinearity have been predicted for a variety of physical systems. Most often these effects have been observed in systems such as plasmas, fluids, and optic fibers[1] but a few examples of novel nonlinear phenomena have also been reported in condensed matter. For example, studies of topological solitons in quasi-one-dimensional (1D) magnetic systems have provided accurate tests of theories which describe the thermodynamics and dynamics of such quasiparticles.[2] In the present Letter we report the observation of new nonlinear excitations in the 1D antiferromagnet $(CD_3)_4NMnCl_3$ (TMMC).

A simple, linear theory for the excitations of a magnetic system predicts a fluctuation spectrum which includes both magnon and double-magnon (DM) excitations. Both of these excitations have been observed by neutron inelastic scattering as well-defined modes, which can be analyzed as a function of wave vector, **q**, and frequency, ω.[3] In the case of planar antiferromagnetic (AF) chains considered here, the spin-flop configuration (cf. inset A of Fig. 1) describes the equilibrium spin orientations at low temperatures when a large magnetic field, **H**, is applied perpendicular to the chain axis, z. If **H** is parallel to x, the spins are directed alternately along $+y$ and $-y$. In such a system, both magnons and double magnons are manifestations of small angular oscillations of the spins in the x-y and y-z planes. These oscillations, which correspond to small changes of the in-plane and out-of-plane angles ϕ and θ defined in Fig. 1, inset A, give rise to spin fluctuations which are predominantly in the x and z directions, but with a small component along y. The x and z fluctuations, deemed in-plane (IP) and out-of-plane (OP), respectively, are *perpendicular* to the equilibrium spin direction and correspond to magnon excitations. Conventional DM modes correspond to the small changes which the θ and ϕ oscillations induce in the spin component *parallel* to the equilibrium spin direction, y.

Our experiments demonstrate that the predictions of the linear theory are incomplete: There are at least *five* well-defined modes at low temperature rather than the four predicted. We shall demonstrate that the additional mode, which is associated with spin fluctuations parallel to the applied field, is a new type of DM excitation which results from nonlinear coupling of the IP and OP magnon fluctuations. This explanation predicts a sixth excitation, also a new DM mode, whose intensity is strongly dependent on temperature. We have searched for, and found, evidence for this sixth mode. Our general conclusion is that nonlinear effects, which are usually important only for large-amplitude fluctuations, may modify drastically the spectrum of small-amplitude motions predicted by a linear theory.

TMMC is a good practical realization of a one-dimensional planar antiferromagnet. Such systems have been studied extensively for manifestations of nonlinear spin dynamics. In particular, soliton excitations which correspond to a π rotation of spins about the chain axis have been characterized in TMMC.[2] Nonlinear interactions between single-magnon and conventional DM modes have also been observed.[4] In TMMC, an interaction of this sort occurs between the IP single-magnon mode and a DM mode over the limited range of applied field (2.5 to 4.5 T) where the two excitation branches cross. The theory advanced in this case[5] predicts that the conventional DM modes can manifest an out-of-plane (z in Fig. 1, inset A) polarization in addition to the conventional y polarization as a result of the coupling to IP modes. This additional component arises because the equilibrium spin configuration is not quite the spin-flop state depicted in Fig. 1, inset A: There is a slight canting of the spins in the direction of the applied field. The nonlinear coupling of the IP magnon and a DM mode does not change the qualitative predictions of the linear theory, however. There are still four modes which should be observed as finite-energy peaks in the spectra of magnetically scattered neutrons.

Our experiments were carried out with the thermal-

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beam, three-axis spectrometer, IN20, at the Institut Laue-Langevin in Grenoble, France. This instrument is equipped with vertically focusing, Heusler-alloy monochromator and analyzer crystals which provide intense beams of polarized neutrons, a feature which allows an unambiguous discrimination between spin fluctuations perpendicular and parallel to the field applied to the sample. For magnetic scattering processes during which the neutron spin is unchanged [non-spin-flip (nsf) processes] only magnetic fluctuations of the sample which are parallel to the applied-field direction are detected. Magnetization fluctuations in the plane perpendicular to **H** give rise to spin-flip (sf) scattering of neutrons. Our measurements were made with a 7 g, ^{37}Cl-enriched single crystal of TMMC, mounted in an asymmetric split-pair superconducting magnet which provided a field up to 10 T perpendicular to the y-z scattering plane of the spectrometer (cf. Fig. 1, inset A). All measurements were made using neutrons of incident energy 14.7 meV with a graphite filter to suppress second-order contamination. Beam collimations were 30′-30′-30′-40′ full width at half maximum.

Neutron spectra for both nsf and sf scattering are

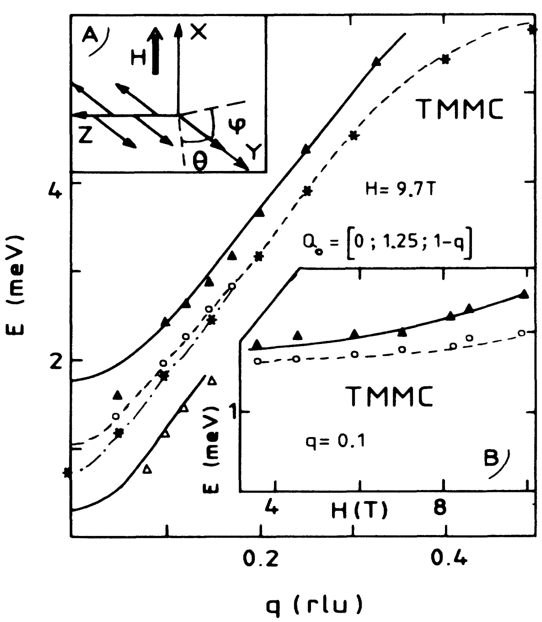

FIG. 1. Dispersion relations for the new DM modes in TMMC. While the high-frequency modes (▲) give unambiguous peaks in the neutron spectra, the frequencies of the low-frequency modes (△) can only be obtained from detailed fits to the data or by a maximum-entropy analysis. The IP (○) and OP (✱) single-magnon modes are also shown in the figure. As explained in the text, the full lines represent the Van Hove singularities associated with the fluctuation spectrum $S^x(q,\omega)$. The dashed lines represent fits to the single-magnon data. Inset A: Definition of the spin direction with respect to the chain and field axes, and of the oscillation angles φ and θ. Inset B: Field dependence of the new high-frequency DM modes.

shown in Figs. 2(a) and 2(b). These data correspond to measurements for scattering vectors [in reciprocal-lattice units[2] (rlu)] of $\mathbf{Q}_0=(0,1.25,1-q)$, at a temperature of 1.4 K and an applied field of 9.7 T. The most intense peaks in Figs. 2(a) and 2(b) are the IP and OP single-magnon modes, whose widths are resolution limited (0.2 meV full width). Three peaks are observed in addition to the two magnons: two in the sf spectrum and one in the nsf spectrum. The spectrum shown in Fig. 2(b) is qualitatively consistent with traditional theory, in that there are two peaks, at ≈ 2.2 and ≈ 2.8 meV, which may be identified as conventional, y-polarized DM modes. On the other hand, the weak x-polarized mode at ≈ 2.4 meV in the nsf spectrum [Fig. 2(a)] is *not* explained by existing theories. The 2.4-meV peak in Fig. 2(a) is one of the new DM modes which we describe and explain in this Letter. Since the modes in both the sf and nsf channels tend to overlap as q increases [cf. Fig. 2(c)], it is not clear *a priori* that the spectra should be described in terms of separate modes for $q \geq 0.2$. Nevertheless, we have chosen to fit each measured spectrum at 1.4 K by a sum of Gaussian peaks whose positions allow us to represent the data by the three upper dispersion curves shown in Fig. 1. For clarity, the two conventional DM modes, observed in the sf channel, have been omitted from this figure.

The theory presented briefly below[6] shows that, when an intense transverse field is applied to a planar system, there is a nonlinear coupling between IP and OP fluctuations. While a coupling of this sort was known for soliton excitations,[7] it has not been considered before in the

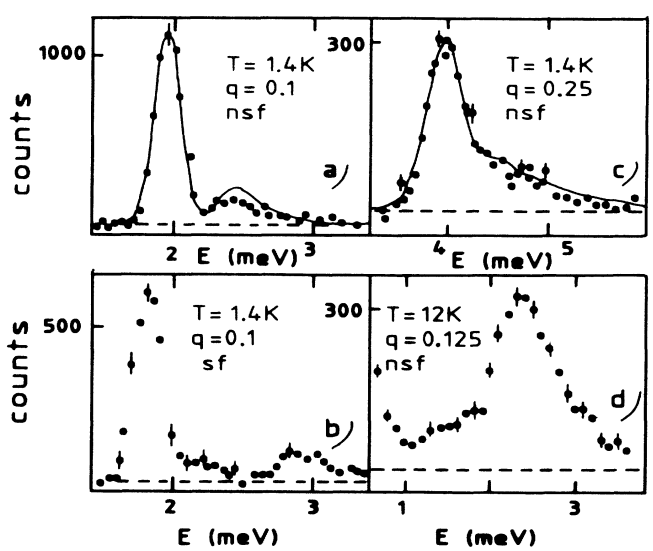

FIG. 2. Spin-flip (sf) and non-spin-flip (nsf) spectra observed in TMMC for $H=9.7$ T. Data have been corrected for the finite polarization of the neutron beams using the observed flip ratio of 18. The full lines are theoretical curves (see text) while the dashed lines represent the background. For each data point the counting time was approximately 20 min.

context of small-amplitude spin fluctuations. As a result of the coupling, the general oscillatory solutions for the spin system include extra DM modes whose frequencies are the sum and difference of IP and OP single-magnon frequencies. The mode whose frequency is the *sum* of IP and OP magnon frequencies is x polarized and is observable at low temperatures. It corresponds to the 2.4-meV mode in Fig. 2(a). The nonlinear coupling also produces a new DM mode whose frequency is the *difference* of the IP and OP magnon frequencies. Although this DM mode is also x polarized, its intensity is vanishingly small at low temperatures and it may only be observed as the sample is heated. An attempt has been made to identify this mode in a second experiment performed at 12 K. An example of the data obtained is shown in Fig. 2(d). These data have been treated by maximum-entropy reconstruction,[8] by fitting multiple peaks, and by direct subtraction of "background" measured at low temperature. For each method, extra intensity is found on the low-energy side of the IP magnon. The dispersion of this feature is shown by the open triangles in Fig. 1.

The magnetic properties of TMMC are well described by the Hamiltonian

$$\mathcal{H}=\sum_n 2J\mathbf{S}_n\cdot\mathbf{S}_{n+1}+2D(S_n^z)^2-g\mu_B HS_n^x\,, \tag{1}$$

with $J=6.8$ K, $D=0.16$ K, $g=2$, and $S=\frac{5}{2}$. The spin dynamics can be described by the equations of motion for the IP and OP angles, ϕ and θ defined in Fig. 1, inset A. For simplicity, the ground state of the spins is assumed to be $\theta_0=0$ and $\phi_0=0$.[7] This approximation is reasonable because the spin canting caused by the applied field does not give rise to new modes[5] and because we do not expect the qualitative predictions of our nonlinear theory to be modified by the inclusion of canting. In the continuum approximation, we may use Eqs. (3a) and (3b) of Wysin, Bishop, and Oitmaa[7] to obtain, in the small-angle limit $\theta\rightarrow\epsilon\theta$ and $\phi\rightarrow\epsilon\phi$ $(\epsilon\ll 1)$,

$$L\phi=-2\epsilon^2\Omega_\beta^2\phi^3/3+2\epsilon^2(C_0^2\phi_z\theta_z-\phi_t\theta_t)+2\epsilon\Omega_\beta\theta_t\phi+o(\epsilon^3)\,, \tag{2}$$

with $\Omega_\beta=g\mu_B H$, $\Omega_a=2S\sqrt{8}DJ$, $C_0=4JS$, $L+\Omega_\beta^2=C_0^2\delta^2/\delta z^2-\delta^2/\delta t^2=L'+\Omega_a^2$, and subscripts t and z implying differentiation with respect to t or z. A similar equation pertains for $L'\theta$. To lowest order, $L\phi=L'\theta=0$, and the solutions are

$$\phi_1=B\exp(i\psi_\beta)+\text{c.c. and } \theta_1=A\exp(i\psi_a)+\text{c.c.}\,, \tag{3}$$

with $\psi_\beta=kz+\omega_\beta t$ and $\psi_a=k'z+\omega_a t$, and where $\omega_\beta=(\Omega_\beta^2+C_0^2k^2)^{1/2}$ and $\omega_a=(\Omega_a^2+C_0^2k'^2)^{1/2}$ are the dispersion relations in the long-wavelength limit. Equations (3) are the IP and OP single-magnon oscillations. To obtain higher-order solutions,[9] the term proportional to ϵ in Eq. (2) is retained. This term introduces a coupling between the lowest-order solutions ϕ_1 and θ_1 and is responsible for the appearance of the new DM modes.

For the second-order solution, ϕ_2, one obtains

$$\phi_2=i[ABF^+e^{i(\psi_\beta+\psi_a)}-AB^*F^-e^{i(\psi_\beta-\psi_a)}+\text{c.c.}]\,,$$

with

$$F^\pm=-\frac{2\epsilon\Omega_\beta\omega_a(k')}{[\omega_\beta(k)\pm\omega_a(k')]^2-C_0^2(k\pm k')^2-\Omega_\beta^2}$$

and amplitude functions, A and B, which may be calculated by retaining terms of order ϵ^2 in Eq. (2). The fluctuations observed in the nsf spectra are described by $S^x(z,t)\sim\langle\phi(z,t)\phi^*(0,0)\rangle$, where

$$\phi(z,t)\approx\sum_k\phi_1+\sum_{kk'}\phi_2+\cdots \tag{4}$$

accounts for all the possible oscillatory modes. The first term in Eq. (4) gives rise to the IP single-magnon mode:

$$S_{1M}^x(q,\omega)\approx(n_\beta+1)\delta(\omega-\omega_\beta)\,,$$

where $n_\beta=1/[\exp(\hbar\omega_\beta/kT)-1]$. The second term leads to new spectral contributions which are DM modes. Specifically,

$$S_{\rm DM}^x(q,\omega)\approx 4\Omega_\beta^2(S^++S^-)/[\omega^2-\omega_\beta^2(q)]^2\,, \tag{5}$$

with

$$S^\pm=\sum_k\omega_a^2(k)[n_\beta(q\mp k)+1][n_a(k)+\tfrac{1}{2}\pm\tfrac{1}{2}]\times\delta\{\omega-[\omega_\beta(q\mp k)\pm\omega_a(k)]\}\,. \tag{6}$$

At $T=1.4$ K, the only significant contribution to $S^x(q,\omega)$ is expected to arise from the S^+ term defined in Eq. (6). For the purposes of comparison with experimental data, the δ function in Eq. (6) may be replaced by a Gaussian whose width is equal to the spectrometer resolution. This simple procedure is reasonable because the widths of the observed single-magnon modes are resolution limited at 1.4 K and the calculated DM contribution is much broader than the instrumental resolution. For $q=0.1$ the result obtained after adding the IP magnon and the S^+ contributions to $S^x(q,\omega)$ is shown by the full line in Fig. 2(a). The theoretical curve, which has been scaled to reproduce the measured peak intensity of the IP magnon, provides an adequate description of the relative intensities of the IP magnon and the new DM mode, as well as the energy of the latter. At elevated temperatures, when the Bose factors in Eq. (6) become non-negligible, an additional, new DM mode which involves the difference of IP and OP magnon frequencies is predicted (S^-). We believe that this contribution has been observed at 12 K in data such as those displayed in Fig. 2(d): The low-energy DM mode contributes intensity between the IP magnon and the elastic, incoherent scattering.

For values of $q>0.2$, the simple perturbation theory described above breaks down,[10] giving rise to spurious divergences in the DM contributions to $S^x(q,\omega)$. Similar breakdown occurs at $T=12$ K when thermal fluctuations broaden the elementary excitations. In these cases, a more general treatment[6] allows a reasonable agreement to be obtained for any value of q. An example is given in Fig. 2(c) for $q=0.25$ and $T=1.4$ K.

The dispersion curves for the new DM modes can be deduced from the maxima in the fluctuation spectrum $S^x(q,\omega)$. These maxima are determined by the Van Hove singularities which characterize the magnon density of states in a one-dimensional spin system.[4] Thus, from the condition that $d[\omega_\alpha(q\pm k)\pm\omega_\beta(k)]/dk=0$, which yields the Van Hove singularities, the full lines in Fig. 1 may be obtained over the entire Brillouin zone by using the experimental values of ω_α and ω_β. These lines are in reasonable agreement with our data for both the dispersion curves and the field dependence shown in inset B.

The new DM modes which we have identified in TMMC are a striking example of the effect which nonlinearity may have on the spectrum of small-amplitude oscillations in anisotropic magnetic systems. When the theory introduced in this Letter is extended to order ϵ^3,[9] two coupled nonlinear Schrödinger equations, similar to those predicted for nonlinear dynamics in plasmas and optic fibers, are obtained. Yet again the ubiquitous nature of nonlinear dynamical effects is apparent.

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[10]The perturbation calculation and its result, Eq. (5), are valid far from resonance. For small values of q, S^+ is identically zero when ω is close to ω_β, so Eq. (5) can be used. For larger values of q or at finite temperatures a more sophisticated approach is required (Ref. 6).