Novel Longitudinal Mode in the Coupled Quantum Chain Compound $KCuF_3$

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Inelastic neutron scattering measurements are reported that show a new longitudinal mode in the antiferromagnetically ordered phase of the spin-1/2 quasi-one-dimensional antiferromagnet $KCuF_3$. This mode signals the crossover from one-dimensional to three-dimensional behavior and indicates a reduction in the ordered spin moment of a spin-1/2 antiferromagnet. The measurements are compared with recent quantum field theory calculations and are found to be in excellent agreement. A feature of the data not predicted by theory is a damping of the mode by decay processes to the transverse spin-wave branches.

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Developing a comprehensive understanding of nonlinear many-body quantum phenomena is a major objective of condensed matter physics. Low-dimensional spin systems remain at the forefront of research because large zero-point fluctuations and nonlinearity inherent in the spin commutation relations create a wealth of exotic quantum phases with unusual dynamics. A model of central importance is the spin-1/2 ($S = 1/2$) Heisenberg antiferromagnetic chain (HAFC) defined by the simple Hamiltonian

$$\mathcal{H}_{1\mathrm{D}} = J \sum_i \vec{S}_i \cdot \vec{S}_{i+1}, \qquad (1)$$

where i is the site index and J is the antiferromagnetic exchange constant. The ground state of the HAFC is a spin singlet, and for half-odd-integer values of S the natural excitations are free spinons with a spin value of $S = 1/2$, rather than the $S = 1$ spin waves found in conventional magnets. Spinons are restricted to creation in pairs and obey fractional statistics that are neither Bose-Einstein nor Fermi-Dirac [1,2]. The spinon picture has been confirmed in some detail by measurements of the triplet excitation continuum in $KCuF_3$ [3], and evidence has also been seen in a number of other materials [4]. Thus the dynamics of an isolated HAFC are relatively well understood theoretically and experimentally.

Nevertheless, a crucial gap remains in our understanding of real systems containing embedded HAFCs with interchain interactions leading to three-dimensional (3D) antiferromagnetic (AF) order. The long wavelength excitations are Goldstone modes, so near the antiferromagnetic zone center (AFZC) one expects well-defined transverse spin waves obeying a linear dispersion relation. In contrast, at energies high compared to the ordering temperature 1D quantum fluctuations persist. Exactly how the classical dynamics characteristic of the ordered state evolve into the high-energy quantum fluctuations is an open question. In light of this, the proposal [5,6] that a novel longitudinal mode accompanies the crossover from 1D to 3D physics upon ordering in $S = 1/2$ quasi-1D compounds is of great importance. In this Letter we present a neutron scattering study of such a crossover in $KCuF_3$ and confirm the existence of this mode.

The magnetic properties of $KCuF_3$ are well characterized: It has a nearly tetragonal crystal structure with lattice parameters $a = b = 4.126$ Å, and $c = 3.914$ Å (at $T = 10$ K). The magnetic Cu^{++} ions have $S = 1/2$ and these are coupled by a strong antiferromagnetic superexchange interaction ($J \approx 53.5 \times 2/\pi$ meV [7]) in the c direction. In contrast superexchange in the a and b directions is weakly ferromagnetic, $J' \approx -10^{-2}J$. The interchain interactions induce magnetic ordering at $T_N = 39$ K. The spins are confined to the ab plane, with antiferromagnetic alignment along the chains (c direction) and ferromagnetic alignment between the chains. The ordered moment-per-spin $m_0 = |\langle S^z \rangle|$ (where z is the ordering direction) is measured to be about 0.27 [8] for temperatures $T \ll T_N$, indicating considerable reduction from the saturation value of 1/2 by zero-point fluctuations.

The one-dimensional dynamics of $KCuF_3$ have been studied extensively. Neutron scattering measurements, made above T_N where 1D effects dominate, showed that the energy (ω) and wave vector ($Q = [q_a, q_b, q_c]$) dependence of the spin correlation functions are in good agreement with the spinon model [3] as expressed by the ansatz proposed by Müller *et al.* [9]. Figure 1(a) illustrates the dynamical correlation functions of the Müller ansatz for the energies of interest around the AFZC located at $[0, 0, -3/2]$ in $KCuF_3$; scattering is expected within a V-shaped region centered at $q_c = -3/2$. In the three-dimensionally ordered phase below T_N the spin correlations at energies above ~27 meV were not noticeably affected by the ordering. However, below this energy well-defined spin-wave modes were found but with additional scattering lying between them for energies $\omega \gtrsim 12$ meV. Conventional 3D spin-wave theory (SWT) with the inclusion of two-magnon terms could qualitatively explain the observations at low energies [10] but not the continuum at higher energies. Figure 1(b) shows the scattering calculated from SWT [10] near $[0, 0, -3/2]$. The magnon branches are well-defined transverse modes, whereas the two-magnon signal is longitudinal and forms a broad

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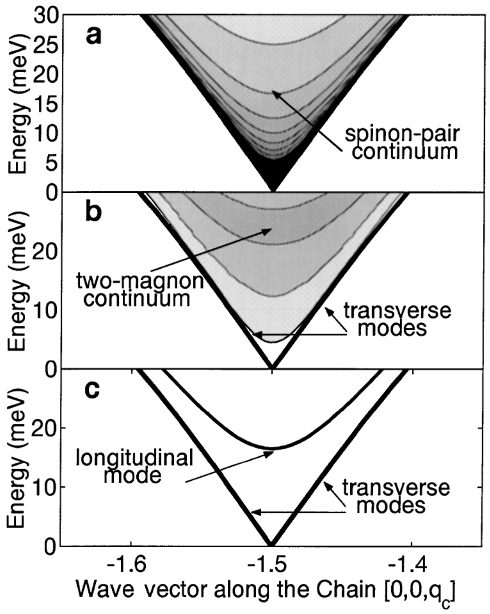

FIG. 1. Three theories of the magnetic correlations in $KCuF_3$ plotted as functions of wave vector and energy. (a) shows the two-spinon continuum given by the Müller ansatz [9], where the intensity of the scattering is indicated by the shading of the contours. (b) shows the scattering from SWT which gives both transverse modes (thick black lines) and a two-magnon continuum (intensity indicated by the shading) [10]. (c) shows the longitudinal mode predicted by field theory [6].

continuum with a maximum at 23.5 meV and a full width at half maximum (FWHM) of 24.0 meV. At this wave vector unpolarized neutron scattering measures half of the transverse cross section plus the full longitudinal cross section.

Recently, the $T = 0$ dynamics of coupled $S = 1/2$ HAFCs have been approached theoretically by considering the solution of the isolated chain in the continuum limit, treating the interchain interactions as a staggered field and applying the random phase approximation (RPA) [5,6]. The predicted spectrum near the AFZC consists of a doubly degenerate, well-defined, gapless transverse spin-wave mode, and a well-defined longitudinal mode with an energy gap, Δ_L, proportional to m_0^2. The longitudinal mode contributes to the dynamics only when the ordered moment is suppressed by zero-point fluctuations. Figure 1(c) shows these theoretical predictions: The longitudinal mode lies between the dispersion branches of the transverse modes and has a gap of $\sim$17 meV at the AFZC. In addition Essler *et al.* predict continuum scattering starting at 22 meV and extending upwards in energy [6]. The scan coverage and resolution in previous measurements [10] was insufficient to differentiate between the RPA and SWT so new experiments probing the low-energy sector are necessary to determine the existence of the longitudinal mode.

Our measurements were performed on the HB1 and HB3 triple-axis spectrometers located at the High Flux Isotope Reactor, Oak Ridge National Laboratory. PG(0,0,2) monochromator and analyzer crystals were used and the final energy was fixed at 13.5 meV with a PG filter placed after the sample to remove higher order contamination from the beam. Our sample of $KCuF_3$ was a high quality single crystal with a mosaic of $10'$, mass of 6.86 g, and volume of 1 cm^3. It was mounted in a variable flow cryostat which provided a base temperature of 2 K and temperature control to within $\pm$0.1 K. The crystal was aligned with the $\mathbf{a}^\star$ and $\mathbf{c}^\star$ reciprocal lattice vectors in the scattering plane and most measurements took place around the $[0, 0, -3/2]$ AFZC. This point was found to be the best compromise between freedom from phonon background sharpness of resolution, and maximization of longitudinal magnetic intensity. Using the collimation $48'$-$40'$-$40'$-$240'$ the transverse modes had a FWHM of 0.057 Å^{-1} (0.03525 r.l.u.) and the energy resolution was 1.3 meV FWHM at 16 meV energy transfer.

Several constant-energy and constant-Q scans were made over the energy range 7 to 27 meV at wave vectors with various values of h and l and at a variety of temperatures above and below T_N. Figure 2(a) shows two constant-Q scans made at the AFZC $[0, 0, -3/2]$. The filled circles are data measured at $T \ll T_N$ in the temperature range 2 to 10 K; while the open circles were measured at $T = 200$ K. The temperature dependence of the peaks at 19.5 and 25 meV, along with measurements in other Brillouin zones, show that they are phonons. At low temperatures, a broad peak around 16 meV is clearly visible in constant-Q scans with $h = 0$, where the longitudinal contribution to the magnetic scattering is maximized. In other zones the mode is either much weaker or masked by phonons. At 200 K the magnetic signal at low energies is reduced to an insignificant level and the observed scattering, adjusted for phonon thermal population factors, is representative of the nonmagnetic background. At energies larger than $k_B T$, however, one still expects the magnetic continuum scattering to have some weight.

Figure 2(b) shows the $T \leq 11$ K data with the smoothed nonmagnetic background subtracted. The peak at 25 meV is slightly undersubtracted, possibly indicating the presence of residual magnetic scattering at 200 K, alternatively this could be due to anharmonic effects which are not taken into account in the phonon subtraction. Two features dominate the magnetic scattering: a large signal at low energies and a broad peak centered at 16 meV. The low-energy scattering comes from the capture of the transverse modes by the resolution function. The broad peak lies at the anticipated longitudinal mode energy. It has been characterized by fitting it using four different models of $S(q, \omega)$ convolved with the experimental resolution. These correspond to two different line profiles (Gaussian or Lorentzian) and a structure factor characteristic of either a local mode or a sinusoidally dispersive mode [6] with a $1/\omega$ intensity variation. The observed scattering is characterized better

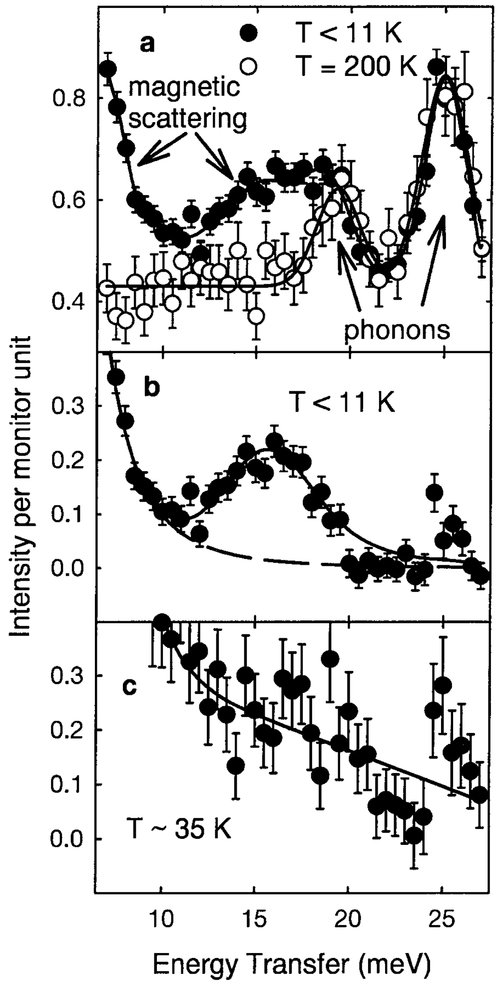

FIG. 2. Constant-Q scans at $[0, 0, -3/2]$. (a) The scattering at $T < 11$ K (closed circles) compared to the scattering at 200 K (open circles), both magnetic and phonon signals are observed. The lines are guides to the eye. (b) The magnetic scattering at $T < 11$ K with the phonon background subtracted off. The solid line is a representative fit to a dispersive mode with a Gaussian profile as described in the text. The dashed line is a guide to the eye showing the tail of the transverse scattering. (c) Magnetic scattering measured in the temperature range 30–40 K. The line is a guide to the eye.

by a Gaussian in both cases. The fitted peak position, Δ_L, is 15.8 $\pm$ 0.1 meV (localized) or 14.9 $\pm$ 0.1 meV (dispersive). The difference reflects the effect of resolution in each case. The observed Δ_L is similar to the theoretical value of 17.4 meV [6] but quite different from the position of the two-magnon maximum calculated from SWT which should appear at 23.5 meV. The mode is intrinsically broadened with FWHM 5.0 $\pm$ 0.4 (Gaussian fits) or 4.2 $\pm$ 0.4 (Lorentzian fits). This suggests that the lifetime may be shortened by decaying into spin waves. It should be noted that this feature is still much sharper than two-magnon scattering predicted by SWT.

The longitudinal mode associated with ordering in the quasi-1D, spin-1/2 HAFC is not expected to exist above the transition temperature. The scan made at 200 K [Fig. 2(a)] shows that the observed mode is not present at temperatures $T \gg T_N$. This measurement was repeated close to T_N over the range 30 to 40 K and is displayed in Fig. 2(c). It shows increased scattering at low energies compared to the $T \ll T_N$ data, with the region between the transverse and longitudinal modes filled in. The longitudinal mode is indistinguishable from the smooth continuum scattering although its presence cannot be entirely ruled out by the statistics. Previous experiments [3] have shown that the scattering above T_N is consistent with the free spinon continuum expected for an ideal 1D chain. As the AF order in the low-temperature phase is reduced with increasing temperture towards T_N one expects the spinon continuum to fill in at lower energies.

It is important to eliminate the possibility that the feature at 16 meV is the signal from transverse branches that has been distorted by the resolution function to give the appearance of a mode. Figure 3 shows constant-energy scans at 10 meV (a) and 16 meV (b). Each scan shows two peaks which come from the transverse branches, and the solid lines are fits of the transverse mode dispersion [Fig. 1(c)] convolved with the instrumental resolution, where the only fitted parameter is the amplitude. At 10 meV the fit is remarkably good and shows the accuracy with which the resolution is known. At 16 meV, where the longitudinal mode is seen in the constant-Q scan of Fig. 2(b), the profile can no longer be fitted by the transverse modes alone. The extra scattering occurring in between the peaks demonstrates that the feature at 16 meV cannot originate from the transverse modes.

A series of constant-Q scans was performed at 10 K to map out the scattering as a function of energy and wave vector. The phonon at 19.5 meV [Fig. 2(a)] was modeled and subtracted using additional scans at 200 K and the resulting data are displayed as a contour plot in Fig. 4(a). The transverse modes form the red V-shaped rods dispersing from $[0, 0, -3/2]$ and the longitudinal mode is the yellow band lying between the transverse branches. Figure 4(b) shows a simulation of the predicted

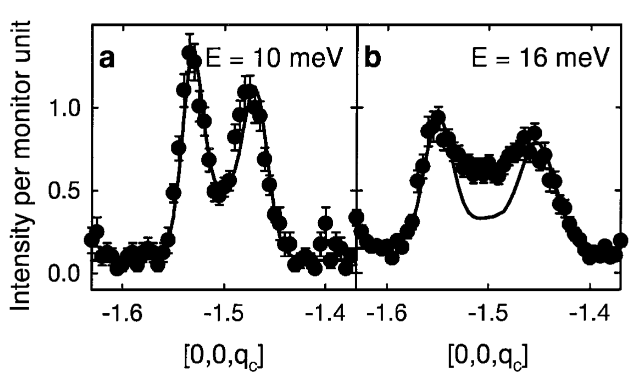

FIG. 3. Constant-energy scans measured at 10 K for the energies (a) 10 meV and (b) 16 meV. The solid line is a fit of the transverse modes convolved with the resolution function.

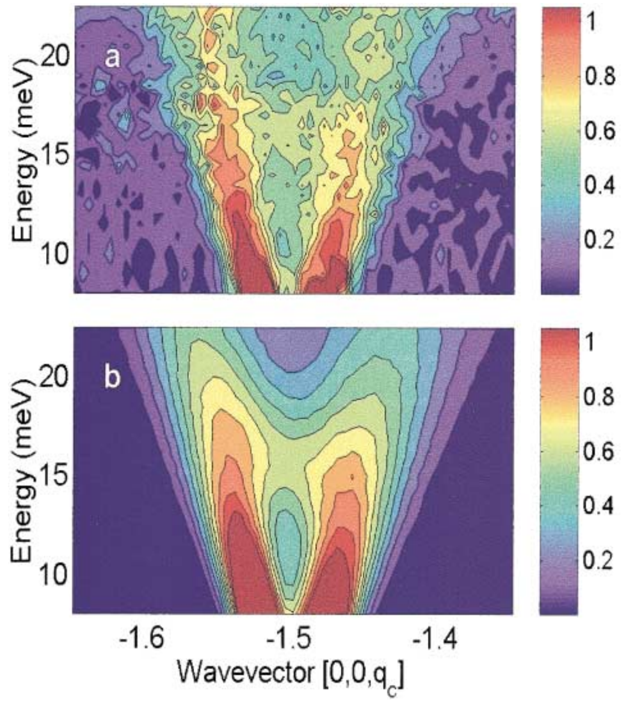

FIG. 4 (color). (a) Energy-wave vector contour map of the magnetic signal collected at 10 K; the colors indicate the relative scattering intensities. (b) Simulation of the magnetic signal over the same reciprocal space region using the theoretical dispersions for the transverse and longitudinal modes [6] convolved with the resolution function.

magnetic scattering over the same energy and wave vector region. The longitudinal mode was assumed to follow the theoretical dispersion [6], with a Gaussian profile of FWHM 4.95 meV and zone-center energy gap fixed at 14.9 meV as obtained from fitting. The transverse modes were given a resolution limited profile. The line shapes of the modes were normalized so that the integrated intensity of the transverse modes was 4 times greater than that of the longitudinal mode as predicted theoretically [6]. The calculation includes the thermal correction, the neutron polarization term, and the Cu^{++} magnetic form factor. The resemblance between Figs. 4(a) and 4(b) is striking and demonstrates not only the very real presence of the longitudinal mode, but also the accuracy with which the theories predict its intensity relative to the transverse modes. Finally, preliminary measurements have also been carried out using polarized neutrons. These further confirm the picture presented above, in particular, the longitudinal nature of the 16 meV mode. This data will be discussed in a forthcoming publication [11].

In summary our experiments have established the presence of a novel mode in $KCuF_3$, with an energy and intensity quantitatively consistent with the predictions of RPA theory for a longitudinal mode in coupled $S = 1/2$ chains. The mode was found to have a broadened linewidth suggesting an instability to decay to spin waves. Longitudinal modes should occur in other quasi-1D $S = 1/2$ systems that show significant zero-point reduction in their ordered spin moment. Interestingly, a similar longitudinal mode is also predicted when the spectrum of the $S = 1/2$ HAFC in a staggered magnetic field is calculated using pseudoboson dimer operators [12]. The dimer basis approach preserves most of the quantum fluctuations that are discarded by SWT and underscores the fact that the physical origin of the longitudinal mode is the zero-point fluctuations that suppress the ordered moment in the coupled chain system. We note that an isolated longitudinal mode is also present in the Haldane-gapped ($S = 1$) chain in the presence of a staggered field [13], but the physics there is different since this corresponds to a splitting of the well-defined gap mode.

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