A Novel Longitudinal Mode in the Coupled Quantum Chain

Compound KCuF$_3$.

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(March 24, 2022)

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 cross-over 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 results 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.
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 non-linearity 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

$$\mathcal{H}_{1D} = J \sum_i \vec{S}_i \cdot \vec{S}_{i+1}$$

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 of $S=1/2$, not $S=1$ spin-waves as in conventional magnets. Spinons are restricted to creation in pairs and obey fractional statistics that are neither Bose-Einstein nor Fermi-Dirac. 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 inter-chain 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. At energies high compared to the ordering temperature one-dimensional (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 [4]) in the $c$-direction. In contrast superexchange in the $a$ and $b$-directions is weakly ferromagnetic, $J' \approx -10^{-2}J$. The inter-chain 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 chains. The ordered moment-per-spin $m_0 = |<S^z>|$ (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 ($\omega$) and wavevector ($Q = [q_a, q_b, q_c]$) dependence of the spin correlation functions are in good agreement with 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 $\sim 27$ meV were not noticeably changed. 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 continuum with a maximum at 23.5 meV and a full-width-half-maximum (FWHM) of 24.0 meV. We note that at this wavevector 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 inter-chain interactions as a staggered field and applying the random phase approximation (RPA) \cite{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, $\Delta_L$, 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 dispersions 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 \cite{6}. The scan coverage and resolution in previous measurements \cite{10} was insufficient to differentiate between the RPA and SWT so new experiments probing the low-energy sector were 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 mosaic 10', mass 6.86 g and volume 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 $\mathbf{a}^*$ and $\mathbf{c}^*$ 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 for freedom from phonon background, and maximization of longitudinal magnetic intensity. With the collimation 48'-40'-40'-240' the transverse modes have 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-wavevector scans were made over the energies
7 to 27 meV and the wavevectors \( l = -1.65 \) to -1.35 r.l.u. These scans were measured at a number of temperatures ranging from well below \( T_N \) at 2 K to well above it at 200 K. Figure 2(a) shows two constant-wavevector 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 K to 10 K. To distinguish magnetic from nonmagnetic features, further measurements were made at 200 K (open circles). At such high temperatures magnetic scattering is washed out by thermal broadening identifying the peaks at 19.5 meV and 25 meV as phonons.

To display the magnetic contributions at \( T \ll T_N \) more clearly, we show the \( T \leq 11 \) K data in Fig. 2(b) with the non-magnetic contributions subtracted off (the 200 K lineshape was subtracted but with the phonon amplitudes adjusted for the thermal population factor). Two features dominate the scattering: a large signal at low energies and a broad peak centered near 16 meV. The low-energy scattering comes from the capture of the transverse modes by the resolution function; while the broad peak lies at the anticipated longitudinal mode energy. The solid line is the fit of the instrumental resolution convolved with the longitudinal mode dispersion \( [6] \) assuming a Gaussian profile and a \( 1/\omega \) structure factor. The fitted parameters were \( \Delta_L \), the peak width and an overall intensity. The fitted \( \Delta_L \) was 14.9±0.1 meV which, due to resolution effects, is lower than the apparent mode position of \( \sim 16 \) meV. This gap energy 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 a FWHM of 4.95±0.07 meV suggesting 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 should not 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. Previous experiments [3] have shown that the scattering above $T_N$ is consistent with that expected for an ideal 1D chain. As the AF order is reduced one expects the free spinon continuum scattering to fill in at lower energies.

It is important to eliminate the possibility that the feature at 16 meV is signal from the transverse branches that has been distorted by the resolution function to give the appearance of a mode. Fig. 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-wavevector 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. Additional scans at other wavevectors where the resolution is different were also used to confirm that the 16 meV scattering was not an artifact.

Finally a series of constant-wavevector scans were performed at 10 K to map out the scattering as a function of energy and wavevector over the ranges 8 to 22.5 meV and -1.65 to -1.35 r.l.u. The phonon at 19.5 meV (Fig. 2(a)) was modelled and subtracted using additional scans at 200 K and the resulting data is 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 magnetic scattering over the same energy and wavevector region. The longitudinal mode was assumed to follow the theoretical dispersion [6], except that the zone-centre energy gap was fixed at 14.9 meV as obtained
from fitting Fig. 2(b). The lineshape of the mode was Gaussian with a FWHM of 4.95 meV (also extracted from the fit), while the transverse modes were given a resolution limited profile. Care was taken to ensure that the lineshapes of the modes were properly normalized so that the integrated intensity of the transverse modes was four times greater than that of the longitudinal mode as predicted theoretically. The calculation includes the thermal correction, the neutron polarization term and the Cu\textsuperscript{++} magnetic form factor. The resemblance between Fig. 4(a) and Fig. 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.

In summary our experiments have established the presence of a novel mode in KCuF\textsubscript{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 spin systems that show significant zero-point reduction in their ordered moment. Interestingly, a similar longitudinal mode is also predicted when the spectrum of the S=1/2 HAFC in a staggered field is calculated using pseudo-boson dimer operators. The dimer basis approach preserves most of the quantum fluctuations that are discarded by SWT and underscores that the physical origin of the longitudinal mode is the zero point fluctuations that suppress the ordered moment in the coupled chain system. Finally, we note that an isolated longitudinal mode is also present in the Haldane-gapped (S=1) chain in the presence of a staggered field, but the physics there is somewhat different since this corresponds to a splitting of the well defined gap mode.

We are grateful to G. Shirane for the loan of the sample, and to A.M. Tsvelik for helpful discussion. D.A.T. would like to thank Risø National Laboratory for financial support and hospitality as a visiting scientist. ORNL is managed for the U.S. D.O.E. by Lockheed Martin Energy Research under contract no. DE-AC05-96OR22464.
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FIG. 1. Three theories of the magnetic correlations in KCuF$_3$ plotted as functions of wavevector 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 [8].
FIG. 2. Constant-wavevector 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 signal is 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 fit as described in the text and the dashed line is a guide to the eye showing the tail of the transverse scattering. (c) The magnetic scattering measured in the temperature range 30-40 K, again the phonons have been subtracted. The line is a guide to the eye.

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) shows an energy-wavevector contour map of the magnetic signal collected at 10 K; the colors indicate the relative scattering intensities. (b) shows a simulation of the magnetic signal over the same reciprocal space region using the theoretical dispersions for the transverse and longitudinal modes $^3$ convolved with the resolution function.
Intensity per monitor unit

Energy Transfer (meV)

(a) $T < 11$ K

(b) $T < 11$ K

(c) $T \sim 35$ K

magnetic scattering

phonons

magnetic scattering
Intensity per monitor unit

(a) $E = 10$ meV  
(b) $E = 16$ meV
Energy (meV)

Wavevector [0,0,q_c]