Excitations from a Bose-Einstein Condensate of Magnons in Coupled Spin Ladders

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The weakly coupled quasi-one-dimensional spin ladder compound $(CH_3)_2CHNH_3CuCl_3$ is studied by neutron scattering in magnetic fields exceeding the critical field of Bose-Einstein condensation of magnons. Commensurate long-range order and the associated Goldstone mode are detected and found to be similar to those in reference to spin-dimer materials. However, for the upper two massive magnon branches, the observed behavior is totally different, culminating in a drastic collapse of excitation bandwidth beyond the transition point.

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Bose-Einstein condensation (BEC), such as the superfluid transition in liquid ^{4}He [1], is the emergence of a collective quantum ground state in a system of interacting bosons. The condensate is characterized by a macroscopic order parameter that spontaneously breaks a continuous $U(1)$ symmetry. For BEC to occur at $T > 0$, the bosons should be able to freely propagate in 3 dimensions (3D) [2]. In one dimension, BEC is forbidden even at zero temperature. In a striking example of dimensional crossover, even weak 3D coupling can enable BEC in 1D systems, where the normal state itself results from the unique 1D topology. A realization of this peculiar quasi-1D case was proposed only recently [3] and involves the condensation of magnetic quasiparticles in weakly coupled antiferromagnetic (AF) spin ladders.

Magnetic BEC can occur in a variety of spin systems [4]. In gapped quantum magnets, for example, an external magnetic field drives the energy of low-lying magnons to zero by virtue of the Zeeman effect, prompting them to condense at some critical field H_c [5]. This transition is fully equivalent to conventional BEC. The rotational $O(2) \equiv U(1)$ symmetry is spontaneously broken by the emerging AF long-range order. At $T = 0$, the density of magnons is zero for $H < H_c$ and vanishingly small just above the transition. The phenomenon is therefore described in the limit of negligible quasiparticle interactions. To date, such transitions were studied mainly in materials composed of coupled structural spin clusters [6–11]. The condensing quasiparticles are then local triplet excitation that propagate due to intercluster interactions [12]. This is in contrast to the original model of Ref. [3] that deals with coupled extended, translationally invariant objects. Their disordered "spin liquid" normal state is a direct consequence of 1D topology [13,14]. Even for weak coupling, the magnons are fully mobile in 1D rather than localized. Is the physics of the field-induced BEC in quantum AF spin ladders any different from that in local-cluster spin systems?

In the present work, we address this issue experimentally, through a neutron scattering investigation of a prototypical spin ladder material $(CH_3)_2CHNH_3CuCl_3$ (IPA-$CuCl_3$, where IPA denotes isopropyl ammonium). This compound almost exactly realizes the original theoretical model of Ref. [3]. Its spin ladders are built of magnetic $S = 1/2$ Cu^{2+} ions and run parallel to the a axis of the triclinic $P\bar{1}$ crystal structure. Conveniently, each one can be viewed as a "composite" Haldane spin chain [15]: Pairs of $S = 1/2$ spins on each rung are strongly *ferromagnetically* correlated and act as effective $S = 1$ objects [15,16]. Coupling along the legs of the ladders is AF and translates into AF interactions between effective spins in the composite $S = 1$ chains. Such chains are gapped [14] and are spin liquids with only short-range correlation. For IPA-$CuCl_3$, the energy gap is $\Delta = 1.2$ meV [15,17].

Excitations from this quantum-disordered ground state are revealed in inelastic neutron scattering (INS) experiments that directly probe the pair spin correlation function $S(\mathbf{q}, \omega)$. Figure 1(a) shows a time-of-flight spectrum collected on a 3 g deuterated IPA-$CuCl_3$ single crystal sample at $T = 100$ mK in zero magnetic field using the disk chopper spectrometer (DCS) at National Institute of Standards and Technology Center for Neutron Research (NCNR) and 6.68 meV fixed-incident energy neutrons. The magnon, with a steep parabolic dispersion along the a axis and gap at the 1D AF zone center $h = 0.5$, is clearly visible. Δ is small compared to the chain-axis magnon bandwidth but is considerably larger than transverse band-

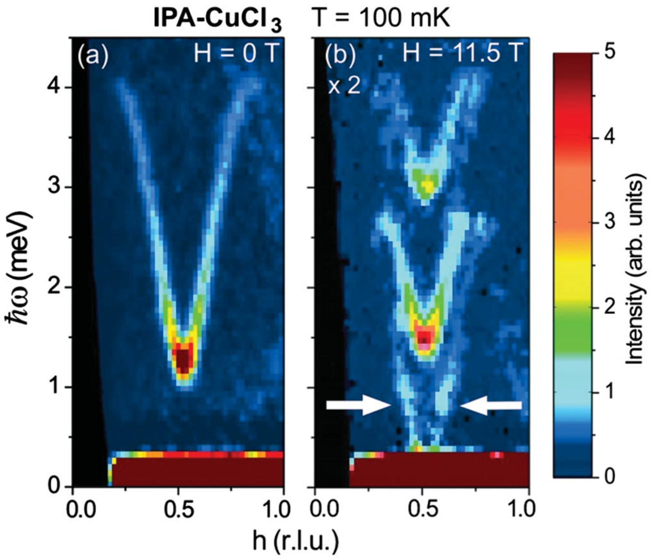

FIG. 1 (color). Time-of-flight neutron spectra measured in IPA-$CuCl_3$ in (a) the 1D Haldane-gap spin liquid phase and (b) the 3D ordered BEC phase, at $H = 0$ and $H = 11.5$ T, respectively. Solid white arrows indicate the linearly dispersive gapless Goldstone mode. r.l.u. denotes reciprocal-lattice units.

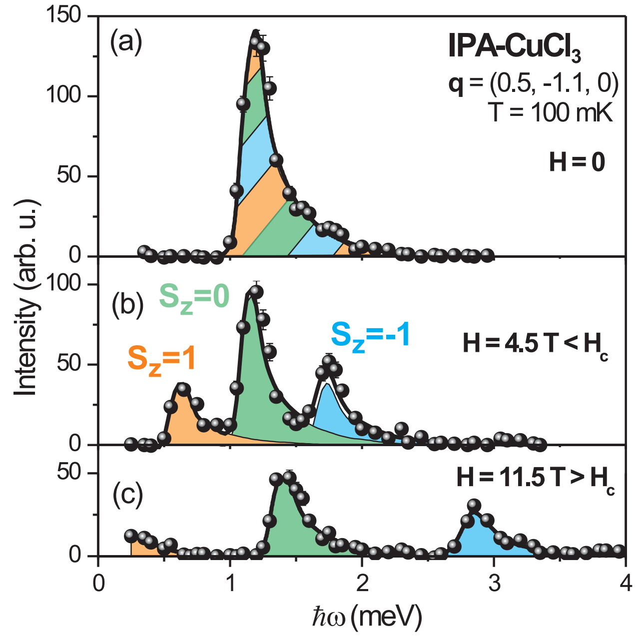

FIG. 2 (color online). Background-subtracted neutron spectra measured in IPA-$CuCl_3$ at the 1D AF zone center $h = 0.5$ in various applied magnetic fields (symbols). The field values correspond to (a),(b) the 1D Haldane-gap spin liquid phase and (c) the 3D ordered magnon BEC phase. Line shapes are entirely due to experimental resolution (solid lines).

widths along the c (0.4 meV) and b axes (<0.1 meV) [15]. Since our main purpose will be to understand the special role that the AF spin ladder structure plays in the BEC phase of IPA-$CuCl_3$, we shall be comparing our results to those found in literature for $TlCuCl_3$ [8], a prototypical AF spin-dimer compound. There, Δ is small compared to magnon dispersion bandwidths in all 3 directions [18,19]. To date, $TlCuCl_3$ is the only material for which the spectrum of spin excitations in the BEC phase has been measured experimentally.

Figure 2 illustrates the effect of the magnetic field on IPA-$CuCl_3$. It shows spectra collected at the 1D AF zone center $h = 0.5$ in several fields using the SPINS 3-axis spectrometer at NCNR, with 3.7 meV fixed-incident energy neutrons, a focusing pyrolitic graphite analyzer, and a BeO filter after the sample. As the field is turned on, the single peak at $H = 0$ [Fig. 2(a)] becomes divided into three equidistant components [Fig. 2(b)]. The peak widths are resolution-limited. The measured field dependencies of the gaps are plotted in Fig. 3(a), which also includes points obtained using the cold neutron 3-axis spectrometer FLEX at the Hahn-Meitner Institute. The gap in the lower mode extrapolates to zero at $H_c = 9.6$ T, where a BEC of magnons was previously detected in bulk measurements [20].

As the gap softens, commensurate long-range AF order sets in and gives rise to new magnetic Bragg reflections of the type $(h + 1/2, k, l)$, with h, k, and l integers. The magnetic structure was determined at $H = 12$ T in a neutron diffraction experiment at the D23 lifting counter diffractometer at Institut Laue Langevin (ILL), on a $3 \times 2 \times 9$ mm^3 single crystal sample, using $\lambda = 1.276$ Å neutrons. A good fit to 48 independent magnetic reflections measured at $T = 50$ mK was obtained using a collinear model with spins perpendicular to the field, aligned parallel to each other on the rungs and antiparallel along the legs of the ladders. The refined value of the ordered moment is $0.49(1)\mu_B$.

The field dependence of the $(0.5, -1, 0)$ peak intensity measured at $T = 50$ mK is plotted in Fig. 3(b), lower curve. To estimate the order parameter critical exponent β, we performed power-law fits to the data in a progressively shrinking field window [Fig. 3(b), inset]. The extrapolated value is $\beta > 0.45$, in agreement with expectations. Indeed, at $T \rightarrow 0$, due to vanishing magnon density, one should recover the mean field (MF) result $\beta = 0.5$ [3,21]. Any discrepancies between the observed and MF behavior become more pronounced at elevated T, when magnon density increases, and their interactions become relevant. At $T = 500$ mK, for example, in IPA-$CuCl_3$ we get $\beta = 0.25(3)$ [Fig. 3(b), upper curve]. BEC critical indexes have also been observed under appropriate conditions in the dimer compound $BaCuSi_2O_6$ [11]. However, recent work showed that the BEC universality of the transition in $TlCuCl_3$ is compromised by deviations from the Heisenberg model. Anisotropy [22] and magnetoelastic coupling [23] modify the critical indexes and account for a small gap in the ordered phase. To date, in IPA-$CuCl_3$ we found no evidence of lattice distortions at H_c or deviations from BEC behavior.

A key result of this work is a direct measurement of excitations of the magnetic Bose-Einstein condensate in

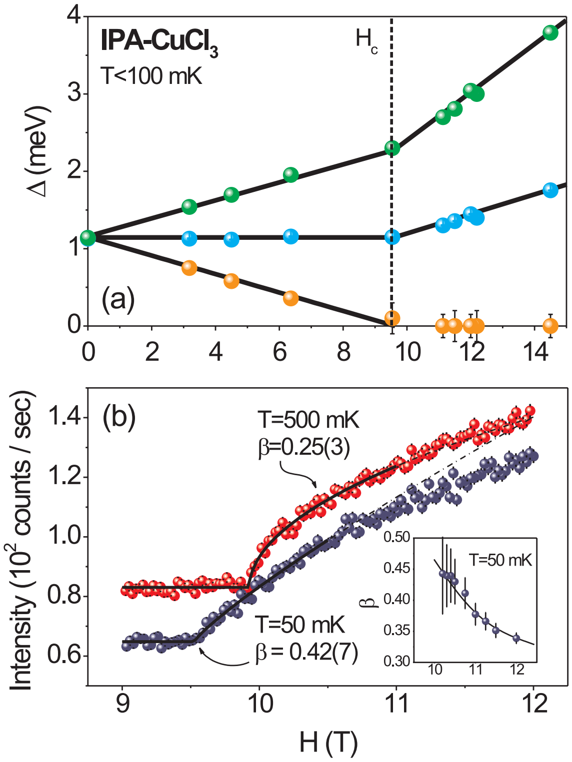

FIG. 3 (color online). (a) Measured gap energies in IPA-$CuCl_3$ as a function of applied magnetic field. (b) Measured field dependencies of the $(0.5, 0, 0)$ magnetic Bragg reflection at two different temperatures (symbols). The y offset corresponds to the actual background for $T = 50$ mK and is arbitrary for the $T = 500$ mK data. Lines are power-law fits over a range of 1 T. Inset: Critical exponent β as a function of the field window used in the least squares fit at $T = 50$ mK.

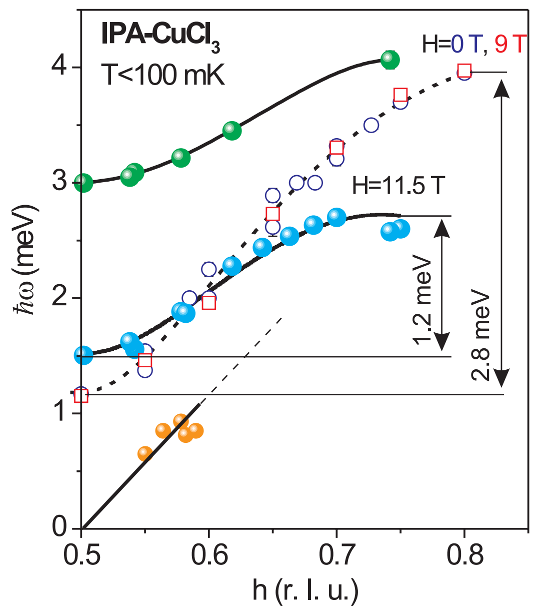

FIG. 4 (color online). Dispersion relation of the three excitation branches measured in IPA-$CuCl_3$ at $H = 11.5$ T $> H_c = 9.7$ T (solid symbols). Open circles: Magnon dispersion at $H = 0$ [15]. Open squares: Dispersion of the middle branch at $H = 9$ T. Lines are guides for the eye. r.l.u. denotes reciprocal-lattice units.

the high-field phase. Data collected above the critical field are shown in Fig. 2(c) (SPINS) and in Fig. 1(b) (DCS). Three distinct excitation branches, two gapped and one gapless, are clearly visible, though the overall inelastic intensity is reduced compared to lower fields. For each mode, the dispersion relations at $H = 11.5$ T were obtained by fitting Gaussian profiles to constant-h cuts through the data in Fig. 1(b). The results are plotted in Fig. 4 in solid spheres. For comparison, we also plot the dispersion relation of the triplet at $H = 0$ (open circles) [15]. Open squares show the dispersion of the middle magnon branch measured just below the transition at $H = 9$ T, on the IN14 3-axis spectrometer at ILL under similar conditions.

The gapless mode observed at $H > H_c$ at low energies [arrows in Fig. 1(b)] is fully analogous to the phonon in conventional BEC, being the Goldstone mode associated with the spontaneous breaking of $O(2)$ symmetry. It is a *collective* excitation of the magnon condensate and has a linear dispersion relation [21]. It takes the place of the massive quadratically dispersive *single-magnon* excitation below H_c [Fig. 1(a)]. At $H = 11.5$ T, the fitted velocity of the Goldstone mode $v_G = 1.74(3)$ meV is reduced compared to the spin wave velocity [24] in zero field $v = 2.9(2)$ meV [15]. This behavior is qualitatively similar to that in $TlCuCl_3$ [8,21], if one neglects the tiny anisotropy gap in the latter system [22], which is too small to be detected with INS anyway. The similarity also extends to the field dependence of the gap energies in those magnon branches that do not soften at the transition point. In both compounds, the corresponding slope increases abruptly at H_c [Fig. 3(a)]. A bond-operator theoretical treatment of the dimer model [21] attributes all of these effects to an admixture of the higher-energy triplet modes to the condensate. This interpretation can be qualitatively extended to our case of coupled spin ladders.

While the long-wavelength spectral features in IPA-$CuCl_3$ are very similar to those in simple spin-dimer systems, the *short-wavelength* spin dynamics at the zone boundary is strikingly different. We find that the two massive excitations in IPA-$CuCl_3$ undergo a qualitative change upon the BEC transition. As seen in Figs. 1(b) and 4, at $H = 11.5$ T, only 20% above H_c, their bandwidths are suppressed by over a factor of 2. The collapse occurs abruptly at the critical point: For the middle mode, there is virtually no change of dispersion between $H = 0$ and $H = 9$ T (Fig. 4). Nothing of the sort happens in the spin-dimer compound $TlCuCl_3$, where the bandwidth of the two upper excitation branches evolves continuously with field and is decreased by only 20% at $H = 12$ T, which is more than twice H_c [21].

The observed phenomenon can hardly be explained by a simple Zeeman shift of quasiparticle energies. Indeed, the latter is negligible, as small as ~0.2 meV between H_c and 11.5 T. Instead, we suggest that the abrupt spectrum restructuring is related to the translational invariance of the ground state wave function for an AF spin ladder or chain below H_c. At $H > H_c$, the emergence of long-range AF order breaks an *additional* discrete symmetry operation, namely, a translation by the structural period of the ladder. There is no analogue of this in conventional BEC. Depending on intercluster interactions that define the ordering vector, neither does this necessarily happen in spin cluster materials. In particular, in $TlCuCl_3$, the induced magnetic structure retains the periodicity of the underlying crystal lattice. However, in a uniform AF spin ladder, the extra symmetry breaking is unavoidable, regardless of interladder coupling.

For IPA-$CuCl_3$, the spontaneous doubling of the period implies that at $H > H_c$ the wave vectors $h = 0$, $h = 0.5$, and $h = 1$ all become equivalent magnetic zone centers. At the same time, $h = 0.25$ and $h = 0.75$ emerge as the new boundaries of the Brillouin zone. The result is a formation of anticrossing gaps for all magnons at these wave vectors [25], where each branch interacts with its own replica from an adjacent zone. This translates into a reduction of the zone-boundary energy for the visible (lower) segments of the two gapped magnons in IPA-$CuCl_3$. The additional violated symmetry operation is a microscopic one and, therefore, plays no role in the long-wavelength physics probed at $h = 0.5$.

To summarize, any *long-wavelength* characteristics of the field-induced magnetic BEC transition and the magnon condensate, such as critical indexes, emergence of the Goldstone mode, and behavior of gap energies, appear to be universal. They are not affected by the 1D topological nature of the normal state in spin chains and ladders and are very similar to those in local-cluster spin systems. In contrast, the *short-wavelength* properties can be significantly different in these two classes of materials. In coupled AF spin chains or ladders, unlike in many couple dimer systems and unlike in conventional BEC, the transition breaks an additional discrete symmetry. The result is a radical modification of the excitation spectrum.

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[1] F. London, Nature (London) **141**, 643 (1938).
[2] E. M. Lifshitz and L. P. Pitaevski, *Statistical Physics Part 2* (Nauka, Moscow, 1978), Chap. 26.
[3] T. Giamarchi and A. M. Tsvelik, Phys. Rev. B **59**, 11 398 (1999).
[4] E. G. Batyev and L. S. Braginski, Sov. Phys. JETP **60**, 781 (1984).
[5] I. Affleck, Phys. Rev. B **43**, 3215 (1991).
[6] W. Shiramura, K. Takatsu, H. Tanaka, K. Kamishima, M. Takahashi, H. Mitamura, and T. Goto, J. Phys. Soc. Jpn. **66**, 1900 (1997).
[7] K. Kodama, M. Takigawa, M. Horvatic, C. Berthier, H. Kageyama, Y. Ueda, S. Miyahara, F. Becca, and F. Mila, Science **298**, 395 (2002).
[8] C. Ruegg, N. Cavadini, A. Fürrer, H.-U. Güdel, K. Kramer, H. Mutka, A. Wildes, K. Habicht, and P. Vorderwisch, Nature (London) **423**, 62 (2003).
[9] M. Jaime *et al.*, Phys. Rev. Lett. **93**, 087203 (2004).
[10] V. S. Zapf, D. Zocco, B. R. Hansen, M. Jaime, C. D. Batista, M. Kenzelmann, C. Niedermayer, A. Lacerda, and A. Paduan-Filho, Phys. Rev. Lett. **96**, 077204 (2006).
[11] S. E. Sebastian, P. A. Sharma, M. Jaime, N. Harrison, V. Correa, L. Balicas, N. Kawashima, C. D. Batista, and I. R. Fisher, Phys. Rev. B **72**, 100404(R) (2005).
[12] O. Nohadani, S. Wessel, and S. Haas, Phys. Rev. B **72**, 024440 (2005).
[13] T. Kennedy and H. Tasaki, Phys. Rev. B **45**, 304 (1992).
[14] F. D. M. Haldane, Phys. Lett. **93A**, 464 (1983); Phys. Rev. Lett. **50**, 1153 (1983).
[15] T. Masuda, A. Zheludev, H. Manaka, L.-P. Regnault, J.-H. Chung, and Y. Qiu, Phys. Rev. Lett. **96**, 047210 (2006).
[16] H. Manaka and I. Yamada, Phys. Rev. B **62**, 14 279 (2000).
[17] H. Manaka, I. Yamada, and K. Yamaguchi, J. Phys. Soc. Jpn. **66**, 564 (1997).
[18] N. Cavadini, G. Heigold, W. Henggeler, A. Furrer, H.-U. Güdel, K. Krämer, and H. Mutka, Phys. Rev. B **63**, 172414 (2001).
[19] A. Oosawa, T. Kato, H. Tanaka, K. Kakurai, M. Muller, and H.-J. Mikeska, Phys. Rev. B **65**, 094426 (2002).
[20] H. Manaka, I. Yamada, Z. Honda, H. A. Katori, and K. Katsumata, J. Phys. Soc. Jpn. **67**, 3913 (1998).
[21] M. Matsumoto, B. Normand, T. M. Rice, and M. Sigrist, Phys. Rev. Lett. **89**, 077203 (2002).
[22] J. Sirker, A. Weie, and O. P. Sushkov, J. Phys. Soc. Jpn. **74**, Suppl., 129 (2005).
[23] N. Johannsen, A. Vasiliev, A. Oosawa, H. Tanaka, and T. Lorenz, Phys. Rev. Lett. **95**, 017205 (2005).
[24] For an excitation with gap Δ, we define the velocity as $v = d\sqrt{(\hbar\omega)^2 - \Delta^2}/d(2\pi h)$, to be measured in energy units.
[25] J. M. Ziman, *Principles of the Theory of Solids* (Cambridge University Press, Cambridge, England, 1972), Chap. 2.2.