Excitations from a Bose-Einstein condensate of magnons in coupled spin ladders.

V. O. Garlea,1 A. Zhelev,1 T. Masuda,2 H. Manaka,3 L.-P. Regnault,4 E. Ressouche,4 B. Grenier,4 J.-H. Chung,5 Y. Qiu,5 K. Habicht,6 K. Kiefer,6 and M. Boehm7

1Neutron Scattering Sciences Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6393, USA.
2International Graduate School of Arts and Sciences, Yokohama City University, 22-2, Seto, Kanazawa-ku, Yokohama City, Kanagawa, 236-0027, Japan.
3Graduate School of Science and Engineering, Kagoshima University, Kormoto, Kagoshima 890-0065, Japan.
4CEA-Grenoble, DFRMC-SPSMS-MDN, 17 rue des Martyrs, 38054 Grenoble Cedex 9, France.
5NCNR, National Institute of Standards and Technology, Gaithersburg, Maryland 20899, USA.
6BENSC, Hahn-Meitner Institut, D-14109 Berlin, Germany.
7Institut Laue Langevin, 6 rue J. Horowitz, 38042 Grenoble Cedex 9, France.

(Dated: March 23, 2022)

The weakly coupled quasi-one-dimensional spin ladder compound (CH₃)$_2$CHNH$_2$CuCl₃ 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 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.

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 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 mainly studied in materials composed of coupled structural spin clusters [6, 7, 8, 9, 10, 11]. The condensing quasiparticles are then local triplet excitation that propagate due to inter-cluster interactions [14]. 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 [15, 16]. 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 (CH₃)$_2$CHNH$_2$CuCl₃ (IPA-CuCl₃). This compound almost exactly realizes the original theoretical model of Ref. [3]. Its spin ladders are built of magnetic $S = 1/2$ Cu²⁺ ions and run parallel to the a axis of the triclinic $\overline{P}$T crystal structure. Conveniently, each one can be viewed as a “composite” Haldane spin chain [17]: pairs of $S = 1/2$ spins on each rung are strongly ferromagnetically (FM) correlated and
act as effective $S = 1$ objects. Coupling along the legs of the ladders is antiferromagnetic (AF), and translates into AF interactions between effective spins in the “composite” $S = 1$ chains. Such chains are gapped and are spin liquids with only short-range correlation.

For IPA-CuCl$_3$, the energy gap is $\Delta = 1.2$ meV. Excitations from this quantum-disordered ground state are revealed in inelastic neutron scattering (INS) experiments that directly probe the pair spin correlation function $S(q, \omega)$. Fig. 1 shows a time-of-flight (TOF) spectrum collected on a 3 g deuterated IPA-CuCl$_3$ single crystal sample at $T = 100$ mK in zero magnetic field using the Disc Chopper Spectrometer (DCS) at 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. $\Delta$ is small compared to the chain-axis magnon bandwidth, but is considerably larger than transverse bandwidths along the $b$ (0.4 meV) and $c$ axes ($< 0.1$ meV). 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$_3$, a prototypical AF spin-dimer compound. There $\Delta$ is small compared to magnon dispersion bandwidths in all 3 directions. 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 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, focusing Pyrolitic graphite analyzer and a BeO filter after the sample. As the field is turned on, the single peak at $H = 0$ (Fig. 2a) becomes divided into three equidistant components (Fig. 2b). The peak widths are resolution-limited. The measured field dependencies of the gaps are plotted in Fig. 3a, which also includes points obtained using the cold neutron 3-axis spectrometer FLEX at HMI. 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.

As the gap softens, commensurate long-range AF order sets in and gives rise to new magnetic Bragg reflections of type $(h+1/2, k, l)$, $h$, $k$ and $l$-integer. The magnetic structure was determined at $H = 12$ T in a neutron diffraction experiment at the D23 lifting counter diffraction beamline of the ILL in Grenoble.
ometer at 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. 3b, lower curve. To estimate the order parameter critical exponent $\beta$ we performed power-law fits to the data in a progressively shrinking field window (Fig. 3b, insert). The extrapolated value is $\beta > 0.45$, in agreement with expectations. Indeed, at $T \to 0$, due to vanishing magnon density, one should recover the mean field (MF) result $\beta = 0.5$ [3, 23]. 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. 3b, upper curve). BEC critical indexes have also been observed under appropriate conditions in the dimer compound BaCuSi$_2$O$_6$ [11]. However, recent work showed that the BEC universality of the transition in TiCuCl$_3$ is compromised by deviations from the Heisenberg model. Anisotropy [12] and magnetoelastic coupling [13] 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 the high-field phase. Data collected above the critical field are shown in Fig. 2b (SPINS) and in Fig. 1b (DCS). Three distinct excitation branches, two gapped and one gapless, are shown in Fig. 2c (SPINS) and in Fig. 1b (DCS). Three high-field phase. Data collected above the critical field $H_c$.

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.

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 Fig. 3b and Fig. 4 at $H = 11.5$ T, only 20% above $H_c$, their bandwidths are suppressed by over a factor of two. 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 TiCuCl$_3$, where the bandwidth of the two upper excitation branches evolves continuously with field, and is decreased by only 20% at tiny anisotropy gap in the latter system [12], 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. 3b). A bond-operator theoretical treatment of the dimer model [23] attributes all 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.
$H = 12$ T, which is more than twice $H_c$.\[23\]

The observed phenomenon can hardly be explained by a simple Zeeman shift of quasiparticle energies. Indeed, the latter is negligible, as small as $\sim 0.2$ 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 inter-cluster interactions that define the ordering vector, neither does this necessarily happen in spin cluster materials. In particular, in TiCuCl$_3$ 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 inter-ladder 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\[24\], 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 coupled dimer systems, and unlike in conventional BEC, the transition breaks an additional discrete symmetry. The result is a radical modification of the excitation spectrum.

We thank A. Chernyshev (U. of California, Irvine) for his theoretical insight and to I. Zaliznyak (Brookhaven National Laboratory) for stressing the significance of the Brillouin zone folding. Research at ORNL was funded by the United States Department of Energy, Office of Basic Energy Sciences- Materials Science, under Contract No. DE-AC05-00OR22725 with UT-Battelle, LLC. T. M. was partially supported by the US - Japan Cooperative Research Program on Neutron Scattering between the US DOE and Japanese MEXT. The work at NIST is supported by the National Science Foundation under Agreement Nos. DMR-9986442, -0086210, and -0454672.

* Department of Materials Science and Engineering, University of Maryland, College Park, Maryland, 20742, USA

[1] F. London, Nature 141, 643 (1938).
[2] E. M. Lifshitz and L. P. Pitaevski, Statistical Physics Part 2. (Nauka, Moscow, USSR, 1978), chap. 26.
[3] T. Giamarchi and A. M. Tsvelik, Phys. Rev. B 59, 11398 (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, 895 (2002).
[8] C. Ruegg, N. Cavadini, A. Furrer, H.-U. Gödel, K. Kramer, H. Mutka, A. Willes, K. Habicht, and P. Vorderwisch, Nature 423, 62 (2003).
[9] M. Jaime, V. F. Correa, N. Harrison, C. D. Batista, N. Kawashima, Y. Kazuma, G. A. Jorge, R. Stern, I. Heinmaa, S. A. Zvyagin, 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. F. Correa, L. Balicas, N. Kawashima, C. D. Batista, and I. R. Fisher, Phys. Rev. B 72, 100404(R) (2005).
[12] J. Sirker, A. Weie, and O. P. Sushkov, J. Phys. Soc. Jpn. 74 Suppl., 129 (2005).
[13] N. Johannsen, A. Vasiliev, A. Oosawa, H. Tanaka, and T. Lorenz, Physical Review Letters 95, 017205 (2005).
[14] O. Nohadani, S. Wessel and S. Haas, cond-mat/0411599.
[15] T. Kennedy and H. Tasaki, Phys. Rev. B 45, 304 (1992).
[16] F. D. M. Haldane, Phys. Lett. 93A, 464 (1983); Phys. Rev. Lett. 50, 1153(1983).
[17] T. Masuda, A. Zhleudev, H. Manaka, L.-P. Regnault, J.-H. Chung, and Y. Qiu, Phys. Rev. Lett. 96, 047210 (2006).
[18] H. Manaka and I. Yamada, Phys. Rev. B 62, 14279 (2000).
[19] H. Manaka, I. Yamada, and K. Yamaguchi, J. Phys. Soc. Jpn. 66, 564 (1997).
[20] N. Cavadini, G. Heigold, W. Hengseder, A. Furrer, H.-U. Gdel, K. Krmer, and H. Mutka, Phys. Rev. B 63, 172414 (2001).
[21] A. Oosawa, T. Kato, H. Tanaka, K. Kakurai, M. Muller, and H.-J. Mikeska, Phys. Rev. B 65, 094426 (2002).
[22] H. Manaka, I. Yamada, Z. Honda, H. A. Katori, and K. Katsumata, J. Phys. Soc. Jpn. 67, 3913 (1998).
[23] M. Matsumoto, B. Normand, T. M. Rice, and M. Sigrist, Phys. Rev. Lett. 89, 077203 (2002).
[24] J. M. Ziman, Principles of the Theory of Solids (Cambridge, 1972), chap. 2.2.
[25] For an excitation with gap $\Delta$ we define the velocity as $v = d(\sqrt{\hbar \omega})^2 - \Delta^2 / (2\pi \hbar)$, to be measured in energy units.