Specific heat provides new evidence for Bose-Einstein condensation of magnons in quantum magnets

Bose-Einstein condensation of magnons in Cs_2CuCl_4

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and

Bose-Einstein Condensation of $S = 1$ Ni spin degrees of freedom in $NiCl_2$-$4SC(NH_2)_2$

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The experimental realization of Bose-Einstein condensation (BEC) in ultracold dilute atomic gases has recently triggered experimental investigations on analogs of Bose condensation in quantum magnets. As early as 1956 it was shown by Matsubara and Matsuda that a quantum spin system can be mapped onto an interacting Bose gas and that the off-diagonal long-range order of the Bose-Einstein condensate corresponds to long-range magnetic order in the spin system. Recent theoretical work has specified the conditions for the analogy. Not all condensation of magnons (or any other Bosons) is a BEC. The critical exponents should be the same as for BEC. This in practise means that for magnetic transitions, it should be possible to map the model to an x-y aniferromagnet and if it is a quantum phase transition, the dynamical exponent z should be 2 chararcteristic of the Landau-Ginzburg-Pitaevskii dynamics.

Compounds currently under investigation in the context of search for analogs of Bose condensation contain either a lattice of dimerized $S = 1/2$ Cu^{2+} ions (e.g., $BaCuSiO_6$, $KCuCl_3$, $TlCuCl_3$), a two-dimensional anisotropic triangular lattice of $S = 1/2$ Cu^{2+} ions (Cs_2CuCl_4), or $S = 1$ Ni chains (e.g., $Ni(C_5H_{14}N_2)_2N_3ClO_4$, $NiCl_2$-$4SC(NH_2)_2$) and it is not always clear whether the requisite conditions of isotropy in the x-y plane and the correct dynamics are met.

Most thorougly investigated is $TlCuCl_3$ (Oosawa et al. 1999). The ground state of each Cu^{2+} dimer is a singlet ($S = 0$), the first excited state is a triplet ($S = 1$, $S_z = +1, 0, -1$). A magnetic field causes a Zeeman splitting of the triplet. At a critical field the lowest triplet state crosses the singlet state and the system undergoes a transition to a canted antiferromagnetically ordered state, the putative Bose-Einstein condensate. Inelastic neutron scattering experiments at fields above the critical field (Rüegg et al., 2003) detected a low-lying mode with linear dispersion, in agreement with one of the theoretical predictions for the Bose-Einstein condensation of spin-triplet states (Matsumoto et al., 2002). However, the second prediction, namely the universal power-law dependence between the critical density and temperature, $n_c \propto T_c^{\alpha}$ with $\alpha = 3/2$, has so far not been observed experimentally.

For Cs_2CuCl_4, neutron scattering experiments (Coldea et al., 2002) have revealed a fully spin aligned (effective ferromagnetic) state at fields above the critical field $B_c = 8.44$ T and an antiferromagnetically ordered state at fields below B_c. The excitations in the high-field phase are conventional gapped ferromagnetic magnons. The size of the magnon energy gap decreases with decreasing field and vanishes at B_c. The ordering of the transverse spin component below B_c is argued to correspond to the Bose-Einstein condensation of two contrarotating magnons.

The specific heat measurements down to 30 mK of Radu et al. provide further evidence for this scenario. Both the Néel temperature and the width of the gap in the magnon spectrum are extracted from temperature dependent specific heat data taken at various applied magnetic field values. The width of the gap decreases linearly with decreasing field and vanishes approximately at the same magnetic field where the magnetic order sets in. For the Néel temperature, the power law behaviour $H_c(T) = H_{c1} + aT^{\alpha}$ with α close to the predicted value 3/2 is observed. Thus, Cs_2CuCl_4 is the first compound for which both conditions necessary for a BEC, i.e., the gapless character of the spin excitations in the ordered state and the correct critical exponent are simultaneously fulfilled. However, it is stated that the precise value of α depends sensitively on the value chosen for H_{c1}. For a critical field of 8.51 T obtained from theoretical calculations $\alpha = 1.44$ results while for, e.g., $H_{c1} = 8.50$ T, $\alpha = 1.52$.

The work on $NiCl_2$-$4SC(NH_2)_2$ (DTN) by Zapf et al. addresses this latter point with great care. In DTN the Ni $S = 1$ spin state is split by the single ion anisotropy into an $S_z = 0$ ground state and two $S_z = \pm 1$ excited states.

Thus, here, a magnetic field induces a crossing between two triplet states. The temperature - magnetic field phase diagram has been determined by specific heat and magnetocaloric effect measurements at temperatures down to 100 mK. At a critical field H_{c1} of approximately 2 T applied parallel to the c-axis of the tetragonal compound, the onset of antiferromagnetic order is observed. At H_{c2} of about 12.7 T the antiferromagnetic order is again suppressed. The power law dependence $H_c(T) = H_{c1} + aT^\alpha$ is here determined by fitting the data up to different maximal temperatures and extrapolating to a vanishingly small temperature intervall. Also, a clever way of first determinimg H_{c1} and, in a second step, α was employed. In this way the power α is reliably determined to be 1.47 ± 0.06, in agreement with the predicted exponent 3/2 for a quantum phase transition to a Bose-Einstein condensate.

One point that might need further attention is the fact that the magnetocaloric effect data on DTN show heating on both entering and leaving the ordered phase. As pointed out by Zapf et al. this may indicate a coupling to the lattice leading to an apparent first-order phase transition. Similarly, in $TlCuCl_3$ ultrasound, NMR, thermal expansion, and magnetostriction experiments provide evidence for a strong magnetoelastic coupling (Sherman et al., 2003; Vyaselev et al., 2004; Johannsen et al., 2005). As for heavy fermion compounds the question of second vs. first order transition is crucial but can, on general grounds, not be definitely answered from *finite-temperature* experiments. A better theoretical understanding of first order quantum phase transitions is urgently needed.

Interestingly, in $TlCuCl_3$ antiferromagnetism cannot only be induced by a magnetic field but also by hydrostatic pressure (Rüegg et al., 2004; Johannsen et al., 2005). A detailed comparison of both types of experiments with theoretical predictions for the conventional pressure-induced and a magnetic field-induced quantum critical point (Fischer and Rosch, 2005) will be very interesting.