Heat Capacity in the 20 T Dilution Refrigerator

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The recent addition of a Heat Capacity capability to the 20 T superconducting magnet at the NHMFL/LANL has spawned a remarkable discovery of Bose-Einstein Condensation (BEC) in an organic material. The collaboration between in-house scientists and international users from Switzerland and Brazil is a shining example of how scientists from around the world can come together at the NHMFL and produce exemplary science. The 20 T dilution refrigerator Heat Capacity and Magnetocaloric effect system, developed by Vivien Zapf, is currently on-line and available to users.

Bose-Einstein Condensation Induced by Magnetic Fields in the Quantum Magnet $NiCl_2$-$4SC(NH_2)_2$

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In traditional Bose-Einstein Condensates, such as dilute gases of cold ^{40}K atoms, the bosons form a coherent ground state as the temperature is lowered below the critical temperature T_{BEC}. The Bose-Einstein Condensate (BEC) is formed when the lowest energy state of the system has a finite occupation of bosonic particles. A new set of materials is receiving an increasing amount of attention in which Bose-Einstein condensation can be induced by magnetic fields. Here the magnetic field, not the temperature, tunes the number of bosons from zero to nonzero across a critical field. The quantum magnet $NiCl_2$-$4SC(NH_2)_2$ (see Fig. 1) is one such material, in which the BEC can be induced by magnetic fields, and corresponds to an XY antiferromagnetic state. The $S = 1$ spin triplet of the Ni^{2+} ion is split at zero field by spin-orbit coupling into an $S_z = 0$ ground state and an $S_z = \pm 1$ excited doublet. In applied magnet fields, the $S_z = +1$ level is lowered linearly with field until it crosses the ground state. In the region where the $S_z = 0$ and $S_z = +1$ overlap, between $H_{c1} = 2.1$ T and $H_{c2} = 12.6$ T,

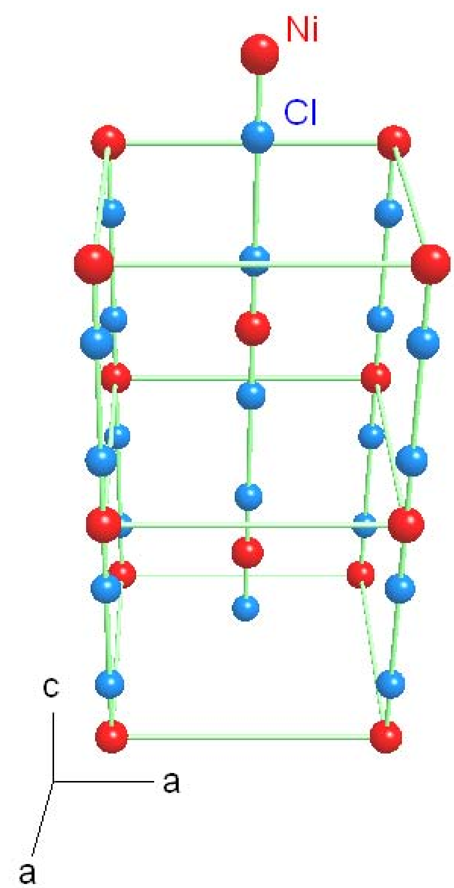

Figure 1. Two unit cells of the tetragonal crystal structure showing Ni (red) and Cl (blue).

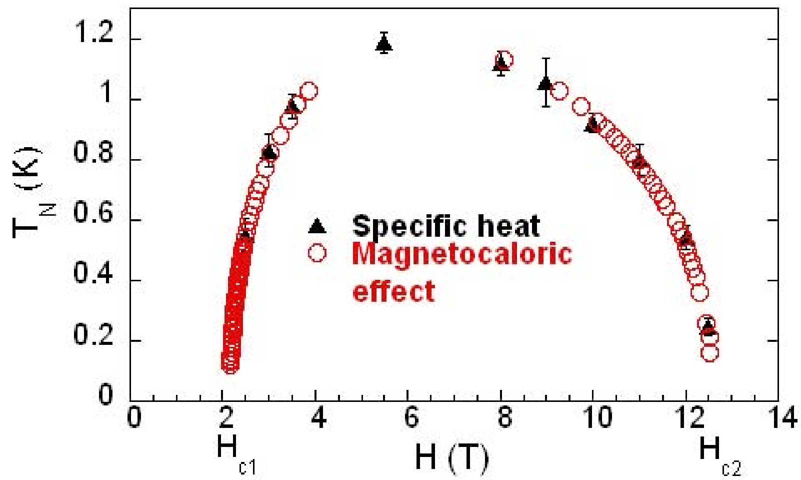

Figure 2. Phase diagram of $NiCl_2$-4SC$(NH_2)_2$ showing the region of antiferromagnetism/BEC determined from specific heat (black triangles) and magnetocaloric effect (red circles).

antiferromagnetism occurs. The region of overlap is more than 10 T wide due to dispersion of the energy levels by antiferromagnetic interactions among neighboring Ni ions. The antiferromagnetic state can be mapped onto a Bose-Einstein condensate, e.g, by treating the $S_z = 0$ state as an empty state, and the $S_z = 1$ state as an occupied boson. A rigorous theoretical description can be found in Ref. [1]. The relative occupancy of the spin levels depends on the applied magnetic field, which acts as the chemical potential of the system. Due to the tetragonal symmetry of the crystal, the Ni magnetic spins do not have any preferred orientation in the plane perpendicular to the magnetic field and the antiferromagnetic order is XY-like. It is this same symmetry that creates a number conservation among the bosons. Number conservation forces the bosons to remain in the system and macroscopically occupy the ground state at low temperatures, rather than vanishing as the temperature is lowered.

We have investigated single crystals of the organic magnet $NiCl_2$-4SC$(NH_2)_2$ for the phenomena of BEC using specific heat and magnetocaloric effect measurements taken in a dilution refrigerator and a 20 T magnet at the NHMFL in LANL.[2] The single crystals were grown by A. Paduan-Filho at the Universidade de São Paulo in Brazil. The phase boundary determined by these methods is shown in Fig. 2, where the antiferromagnetism/BEC occurs in the dome-shaped region under the symbols. The key prediction of Bose-Einstein condensation that we have been able to verify is the power-law dependence of the critical field line $H_c - H_{c1} \sim T^{\alpha}$. The power-law exponent α is predicted to be $\alpha = 3/2$ for a 3-D BEC, $\alpha = 1$ for a 2-D BEC and $\alpha = 2$ for an ordinary Ising antiferromagnet. The difficulty in measuring this exponent, which has created problems in many previous investigations of BEC in quantum magnets, is the fact that it only occurs as T approaches absolute zero. We have used dilution refrigerator temperatures and an extrapolation technique to determine α as T approaches 0, and our results from two separate measurements are both consistent with $\alpha = 3/2$, and inconsistent with $\alpha = 1$ or $\alpha = 2$. Thus we have direct experimental verification of Bose-Einstein condensation in this compound. This is the second-ever compound for which this exponent has been conclusively determined. The other compound, $BaCuSi_2O_6$,[3] found $\alpha = 1$, consistent with a 2-D BEC.

Our results have also benefited greatly from a collaboration with Michel Kenzelmann and colleagues, who have conducted inelastic neutron diffraction measurements on large deuterated single crystals of $NiCl_2$-4SC$(NH_2)_2$ at the Paul-Scherrer Institute in Switzerland. The neutron scattering measurements probed the antiferromagnetic dispersion relation, and showed that the antiferromagnetic interactions in the plane J_a are ten times weaker than along the tetragonal c-axis J_c. Thus $NiCl_2$-4SC$(NH_2)_2$ can be thought of as a weakly coupled 1-D chain system at high temperatures, crossing over to 3-D antiferromagnetic order below ~1 K.

Figure 3. The 20 T superconducting magnet system at the NHMFL-Los Alamos.

1 H.-T. Wang and Y. Wang, *Phys. Rev. B*, **71**, 104429 (2005); and K.-K. Ng and T.-K. Lee, cond-mat/0507663.
2 V.S. Zapf, *et al*., *Phys. Rev. Lett*., **96**, 077204 (2006).
3 S. Sebastian, *et al*., *Nature* (2006), in press.