Bose-glass phases at the boundary of Bose-Einstein Condensates in quantum magnets

The organic quantum magnet $NiCl_2$-*tetrakis* thiourea [$NiCl_2$-$4SC(NH_2)_2$] exhibits a magnetic field-induced phase transition that belongs to the universality class of Bose-Einstein condensation. The nickel magnetic spins forms an XY antiferromagnetic state in applied magnetic fields between 2 and 12.5 T that has been verified to correspond to a Bose-Einstein condensate (BEC) of spin degrees of freedom through extensive measurements of specific heat, the detailed phase boundary down to 1 mK, electron spin resonance, and elastic and inelastic neutron scattering [1-3].

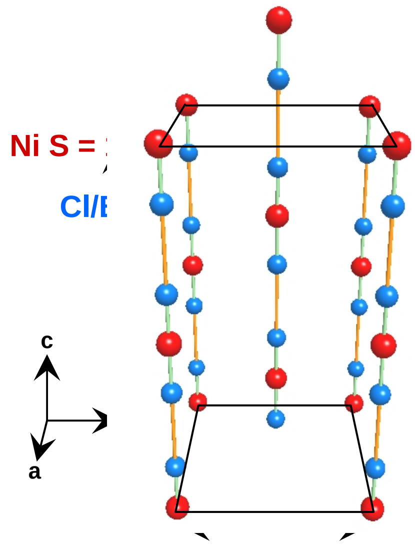

Fig. 1 Crystal structure of $NiCl_2$-$4SC(NH_2)_2$ showing the Ni and Cl atoms. Two unit cells are shown stacked along the c-axis, and the nominal Ni spin directions in the ordered state near H_{c1} are indicated by arrows.

The observation of Bose-Einstein Condensation (BEC) in the pure system $NiCl_2$-*tetrakis* thiourea gives us a unique opportunity to search for a boson-localization transition in the *doped versions of this compound*. Boson localization is analogous to the metal-to-insulator transitions in fermions, which has been the subject of extensive research in the past decades [4]. Boson localization is equally important and in fact easier to model theoretically [5] than the case for fermions. However, experimentally it has proven to be elusive. One route to boson localization is to induce disorder, creating a Bose glass (BG). A number of experiments have searched for the BEC-to-BG phase transition in systems including superfluid ^4He in porous glasses, and dilute gases of Alkali atoms in random potentials [6-8]. It has also been proposed to occur in high-T_c cuprates at the superconducting-to-normal phase transition at high fields [9, 10]. The possibility of observing this elusive state in a quantum magnet is now attracting increasing attention [11].

In the compound $NiCl_2$-*tetrakis* thiourea, we have preliminary evidence that inducing disorder by substituting Br for Cl creates a Bose glass at the boundaries of the BEC phase. A remarkable similarity between magnetization measurements and theoretical predictions suggest that this compound could be an experimental realization of the BEC-to-BG quantum phase transition (see Figs. 2 and 3). The width and the accessibility of the magnetic field range over which the BG phase occurs allows us to investigate it to an unprecedented level of detail. Thus, we propose to search for boson localization in $NiCl_{2-x}Br_x$-thiourea single crystals via magnetization, specific heat and magnetocaloric effect measurements at mK temperatures. The detailed phase boundary has been measured to 1 mK at the high B/T laboratory and the theoretical model has been developed and the phase boundary charted using Quantum Monte Carlo simulations. We now just need to complete the thermodynamic measurements, and this will be the *final data need to complete the P.I.'s user collaboration grant*.

The theoretical model that we have developed for this system [12-14] assumes bond disorder due to Br doping on the Cl site. In the pure compound, the dominant antiferromagnetic coupling occurs via super exchange mediated by the Ni-Cl-Cl-Ni bonds along the tetragonal c-axis. In the lightly doped system, the presence of two types of bonds, Ni-Cl-Cl-Ni and Ni-Cl-Br-Ni, creates a random distribution of antiferromagnetic coupling strengths.

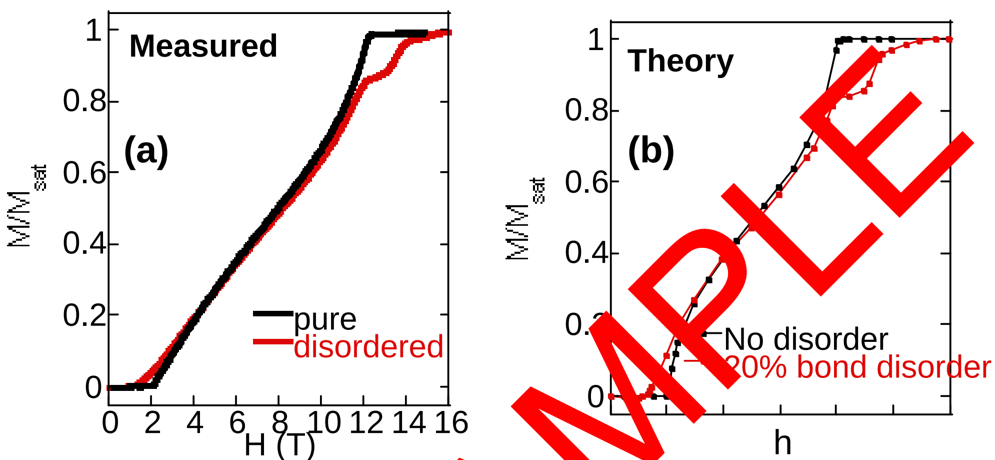

Figure 2 (a): Magnetization of pure $NiCl_2$-thiourea (black) and $Ni(Cl_{0.85}Br_{0.15})_2$-thiourea (red) (N. Oliveira, A. Paduan Filho, unpublished). **(b)** Theoretical prediction for the pure compound (black) and 20% bond disorder (T. Roscilde, unpublished).

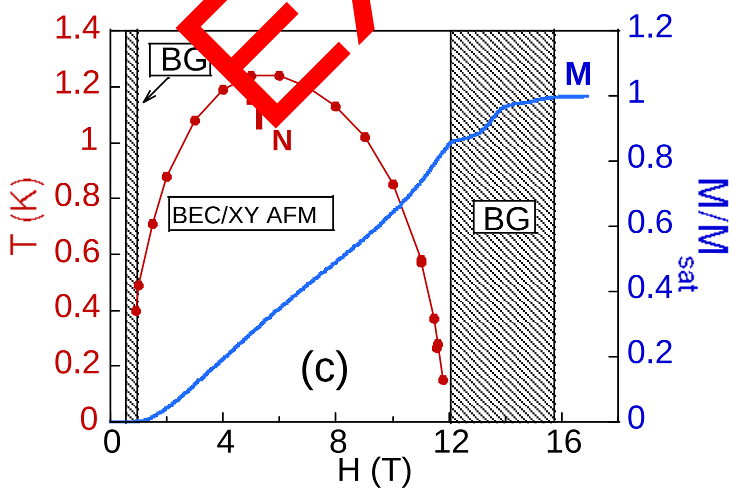

Fig. 3. Phase diagram showing the region of AFM/BEC in the dome-shaped region determined from specific heat (red). Magnetization is shown in blue, and proposed region of BG behavior is indicated in the hashed regions.

Thus the critical fields H_{c1} and H_{c2} vary across the crystal. Starting in the ordered phase, if the magnetic field is increased until the critical field H_{c2} of the Ni-Cl-Cl-Ni bond is reached, these weaker bonds decouple while the Ni-Cl-Br-Ni remain intact, leading to a disordered Bose Glass. The region of proposed Bose-Glass behavior is shown in Figs 3 and 4 between 12 and 16 T. When the field is increased further, the larger critical field for the Ni-Cl-Br-Ni bonds is reached and the short-range disorder vanishes and the magnetic spins saturate at ~ 16 T. The phase diagram showing predicted regions of Bose-Glass behavior is depicted in Fig. 3.

In order to test this model experimentally, we plan to measure the specific heat, magnetocaloric effect and magnetization, and map the detailed phase diagram of the doped DTN system as a function of magnetic field and temperature for varying concentrations of Br. These will all be compared with the theoretical calculations. Ac susceptibility at varying frequency has already been completed to investigate the dynamics of the bose-glass phase and will confirm and extend the phase boundary to low temperatures. Specific heat and ac susceptibility are sensitive to different phase boundaries and thus both are needed for a complete picture.

Broader Impacts

This work will involve, depending on when it is scheduled, either a graduate student Farzana Nasreen, who is a female New Mexico State University student and near the end of her Ph.D., or if it is scheduled later, a post-doctoral associate. Development of a new and improved specific heat capacity system will be continued through this work, and a new Faraday magnetometer that is part of the newly 20 T magnet system will also be developed to function down to 20 mK in conjunction with this work.

This work will enhancing our understanding of the fundamental quantum-mechanical states of matter including Bose-Einstein condensates and insulating to conducting transitions at the border of a Bose Glass.

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Dr. Zapf completed her Ph.D. at UCSD with Brian Maple in 2003 on crystal growth of heavy-fermion and high-T_c materials, in particular substitution studies leading to frustration and spin glass phases, and measurements of thermodynamic properties down to dilution refrigerator temperatures including magnetization, specific heat, and transport. She was then awarded a Milliken Post-doctoral fellowship at Caltech to study infinite-layer superconducting cuprates, and finally joined the National High Magnetic Field Lab (MPA-NHMFL) first as a director-funded post-doc and then as a staff member in 2006. There she worked on quantum magnets, establishing the existence of Bose-Einstein Condensation in the (frustrated) quantum magnet $NiCl_2$-$4SC(NH_2)_2$ (DTN), discovering organic-transition metal hybrid magnetoelectric materials, and developing a calorimeter for specific heat and magnetocaloric effect measurements for the dilution refrigerator in the 20 T magnet at NHMFL-LANL. She gained experience measuring Hall effect, dielectric constant, dilatometry, magnetic torque, and magnetoelectric current as well as measurements in the 33 T resistive and 45 T hybrid magnets at NHMFL-Tallahassee and in pulsed fields up to 65 T at the NHMFL in Los Alamos. She has published more than 50 papers on topics including heavy-fermion, high-Tc and quantum magnets and given 18 invited talks.

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