Experimental report

09/09/2015

Proposal: 4-01-1393 **Council:** 4/2014

Title: Spin dynamics of a bond disorderedspin liquid at z=1 quantum phase transition

Research area: Physics

This proposal is a new proposal

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Samples: Ni(Cl1-xBrx)2-4SC(NH2)2

Instrument	Requested days	Allocated days	From	To
IN5	5	5	01/10/2014	06/10/2014

Abstract:

Disorder can have a profound effect on quantum spin liquids. New phases such as the Bose glass or new modes inside the excitation gap can appear. While most work was done on field driven quantum phase transitions with the dynamic critical exponent z=2 we propose an experiment on a z=1 quantum critical point. The aim of the proposed experiment is to search for in gap modes predicted by a recently published numerical study.

Spin dynamics of a bond-disordered spin liquid at $z = 1$ quantum phase transition

There is a fundamental interest in studying the properties of bosons in a disordered potential. As the Heisenberg Hamiltonian can be exactly mapped onto an ensemble of bosons, chemically disordered quantum magnets offer a convenient possibility of experimental research of "dirty bosons" physics [1]. The present experiment probes the spin dynamics in the bond-disordered modification of a $S = 1$ quantum paramagnet $NiCl_2{\cdot}4SC(NH_2)_2$ [2]. The structure of this material, schematically shown in Fig. 1, consists of two interpenetrating tetragonal lattices of $S = 1$ Ni^{2+} ions. The strong easy-plane anisotropy $D \simeq 0.78$ meV results in a quantum-disordered ground state with a gap $\Delta \simeq 0.3$ meV in the magnetic excitation spectrum. The peculiar finding of previous studies [3, 4] was the decrease of this gap induced by chemical substitution on halogen site. This implies that introduction of disorder drives the system towards the quantum critical point — an intriguing case, numerically investigated by Vojta [5], but not yet encountered in a real magnetic material. In such a case the disorder-induced changes in the spectrum including the in-gap states are expected to be especially pronounced. This is why studying the zero-field dynamics of bond-disordered $NiCl_2{\cdot}4SC(NH_2)_2$ is of interest. However, due to 3D character of magnetic interactions and excitation dispersion in $NiCl_2{\cdot}4SC(NH_2)_2$, the possibility of collecting the data with high energy resolution over a wide momentum range is crucial for this study. This makes the time-of-flight neutron spectroscopy the tool of choice.

Our sample consisted of two huge (~ 1 g) co-aligned fully deuterated single crystals of $NiCl_{2-2x}Br_{2x}{\cdot}4SC(NH_2)_2$ with $x = 0.06$ disorder concentration. All data were collected at $T = 60$ mK, which is much lower than all the relevant energy scales of the material under investigation. The principal experimental scattering plane was $(1, -1, 0)$, providing access to scattering vectors of type (h, h, l) as shown in Fig. 1. Two data sets were collected, using incident neutron energies $E_i = 2.26$ and 1.17 meV, respectively. The

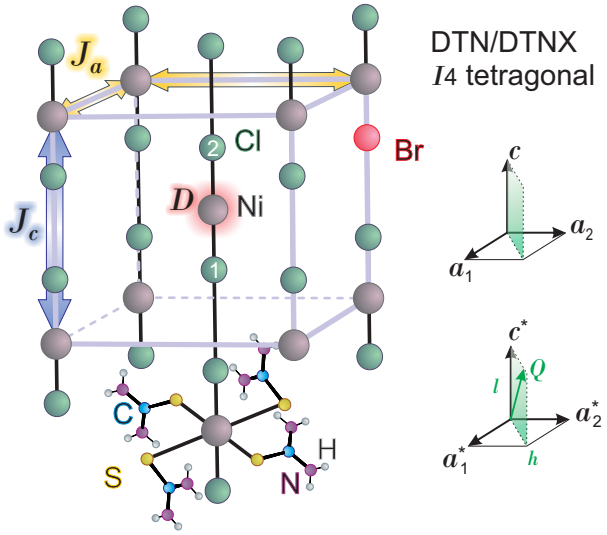

Figure 1: A sketch of the $NiCl_{2-2x}Br_{2x}{\cdot}4SC(NH_2)_2$ structure with the main magnetic interactions for $S = 1$ nickel ions labeled. The scattering plane is shown on the right, in both real (top) and reciprocal (bottom) space representations.

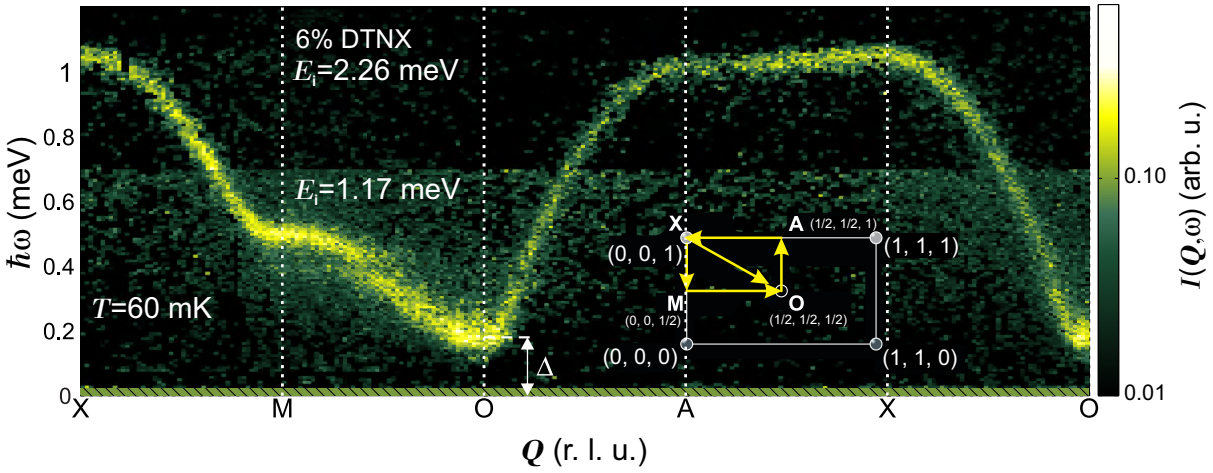

Figure 2: Complex cut of combined datasets of both E_i = 2.27 and 1.17 meV. Cut trajectory through the first Brillouin zone is shown on the figure.

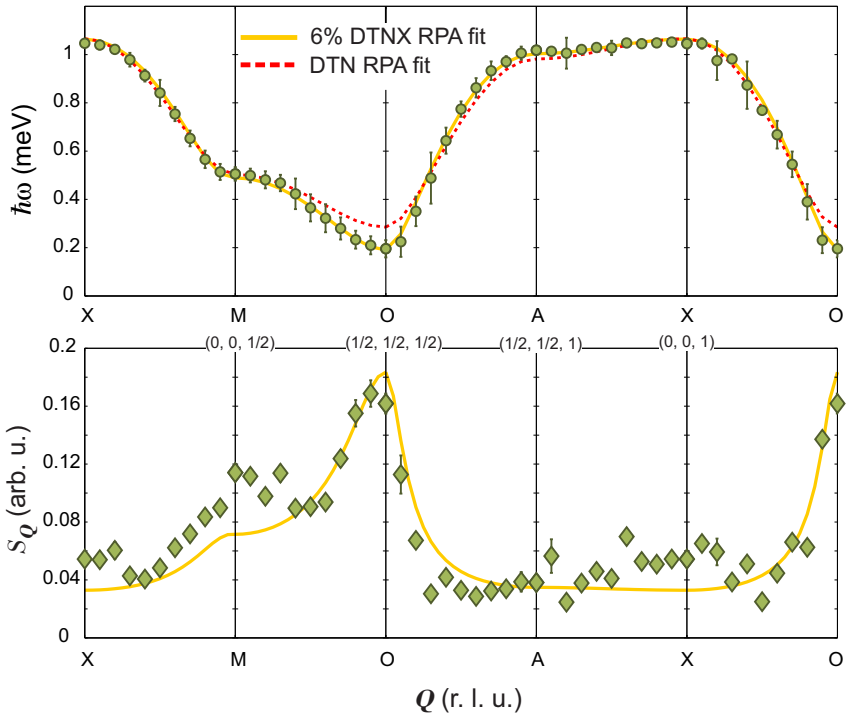

Figure 3: Top panel: Magnon dispersion relation measured in 6% $NiCl_{2-2x}Br_{2x}{\cdot}4SC(NH_2)_2$ with E_i = 2.26 meV. The solid line is a fit to the RPA prediction. The dashed line is the reference magnon dispersion in disorder-free DTN. Bottom panel: Integrated intensity corrected for the magnetic form factor and polarization factor, $\mathcal{S}_{\mathbf{Q}}$, as determined in fits to the same individual cuts. The solid line is the RPA result, which is simply the inverse of $\hbar\omega(\mathbf{Q})$.

angular coverage, step spacing of 1° and counting time of approximately 15 min/angle were identical in both datasets.

Figure 2 shows a quick overview of the data obtained. It represents a complex cut in the first Brillouin zones, composed of both datasets with different incident energies. It is

clear that no additional features emerge in the sub-gap range at least down to 25 μeV, somewhat in contrast with our initial expectations. At the same time, the gap itself is noticeably lowered by 100 μeV.

For a quantitative analysis we employed the following empirical approach. The collected data, spanning over a few Brillouin zones, was split into a series of $\mathcal{S}(\omega)$ cuts. Every cut was fitted to a resolution-convoluted damped oscillator response, assuming the single-mode approximation. The resulting intensities and excitation spectrum $\hbar\omega(\mathbf{Q})$ are shown in Fig. 3. The dispersion of $NiCl_{2-2x}Br_{2x}{\cdot}4SC(NH_2)_2$ is clearly different from the one in the parent material and 6% of disorder already induces the 30% change of the gap. Fitting the experimental data yields the effective Hamiltonian parameters different from the ones found in disorder-free case. We find that the key parameter for the red shift of magnetic excitations in $NiCl_{2-2x}Br_{2x}{\cdot}4SC(NH_2)_2$ is the diminishing ratio D/J_c, while both D and J_c actually increase. Here J_c is the antiferromagnetic exchange along the c direction, mediated by the pair of halogen ions. Hence, the halogen substitution modifies the relevant superexchange path and also the local surrounding of nickel, which in turn affects the single-ion anisotropy. Crude analogy with the model studied by Vojta [5] also explains why in this case no in-gap states should appear.

The results of the experiment are already published [6]. In this publication the above analysis is discussed in more details as well as some more subtle spectral features, being available for observation due to the exceptional quality of the data coming from IN5.

References

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