Bond randomness induced magnon decoherence in a spin-1/2 ladder compound

B. Náfrádi[1], T. Keller[1], H. Manaka[2], U. Stuhr[3], A. Zheludev[4], B. Keimer[1]

[1]Max-Planck-Institut für Festkörperforschung, Stuttgart, Germany
[2]Kagoshima University, Graduate School of Science and Engineering, Kagoshima, Japan
[3]Paul Scherrer Institute, Laboratory for Neutron Scattering, Villigen, Switzerland
[4]ETH Zürich, Laboratorium für Festkörperphysik, Neutron Scattering and Magnetism Group, Zürich, Switzerland

Quantum systems with long-living coherent states are proposed for use in quantum computing. However, finite coherence lengths and thus decoherence limit the time frame for information storage and manipulation. We have used a combination of neutron resonant spin-echo (NRSE-TAS) and triple-axis spectroscopies (TAS) to determine the magnon decoherence in IPA-$Cu(Cl_{0.95}Br_{0.05})_3$, a model spin-1/2 ladder antiferromagnet where Br substitution induces bond randomness. We find that the bond defects induce a blueshift, $\delta\Delta$, and broadening, $\delta\Gamma$, of the magnon gap excitation compared to the pure compound. At temperatures exceeding the energy scale of the inter-ladder exchange interactions, $\delta\Delta$ and $\delta\Gamma$ are temperature independent. Upon cooling, $\delta\Delta$ and $\delta\Gamma$ become temperature dependent and saturate at values lower than those observed at higher temperature, consistent with the crossover from one-dimensional to two-dimensional spin correlations with decreasing temperature previously observed in pure IPA-$CuCl_3$.

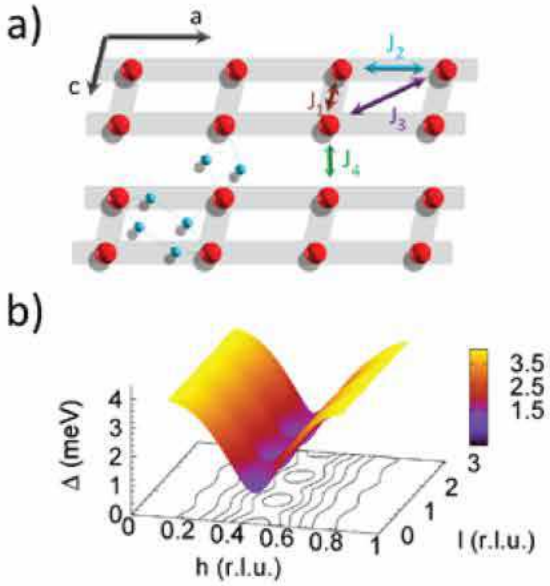

Figure 1: (a) Schematic view of the a-c plane of IPA-$Cu(Cl{:}Br)_3$. The ladder structure is highlighted in gray. Cu atoms are represented by red. Cl and Br positions represented by blue. The arrows show the exchange interactions: J1 = −2.3 meV, J2 = 1.2 meV, J3 = 2.9 meV and J4 = −0.3 meV. (b) Color plot of the magnon dispersion.

Magnon in a potential box

One-dimensional (1D) spin liquids, such as $S = 1$ chains or $S = 1/2$ ladders with antiferromagnetic interactions, have emerged as important model systems for the study of long-range quantum coherence. These systems have collective spin-singlet ground states. Their magnetic excitations are mobile "magnon" quasiparticles, with a gap Δ_0 even for temperature $T \rightarrow 0$. In an ideally clean system, the magnon mean free path is infinitely long at $T = 0$. At $T > 0$, theoretical work based on the non-linear sigma model (NLσM) predicts that the T-dependent coherence length, and thus observables such as the energies, Δ, and linewidths, Γ, of the magnons depend solely on T/Δ_0 [1,2,3].

A simple yet effective picture is that magnons are confined into finite 1D potential "boxes", defined by both static and dynamic defects. The goal of our experiments is to test the applicability of this simple rule for the energy, Δ, and width, Γ, of the magnon resonance in a disordered $S = 1/2$ ladder system.

NRSE spectroscopy

IPA-$Cu(Cl_{0.95}Br_{0.05})_3$ is an excellent model system to experimentally study the effect of bond disorder on the Δ and Γ of the magnon resonance in gapped quantum spin liquids. The structure is

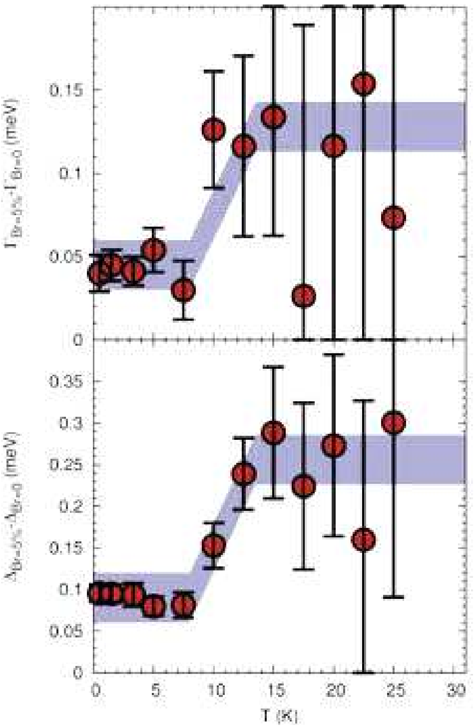

Figure 2: (a) Temperature dependence of the bond randomness induced magnon blueshift of IPA-Cu($Cl_{0.95}Br_{0.05}$)$_3$. (b) Temperature dependence of the magnon decoherence induced by bond randomness of IPA-Cu($Cl_{0.95}Br_{0.05}$)$_3$. The broad shaded lines are a guide for the eyes. Line used in panel-b is the same as in panel-a but divided by a factor 2.

depicted in figure 1a [4]. The Cu^{2+} ions carry a spin S = 1/2 and form ladders along the a axis. Exchange interactions within these Cu^{2+} ions are mediated by Cl di-bridges. Substituting Cl with Br results in a substantial change in the interaction strength [4], but IPA-Cu($Cl_{0.95}Br_{0.05}$)$_3$ remains in a gapped quantum spin liquid state (fig. 1b).

NRSE-TAS experiments at TRISP

The NRSE-TAS experiments were performed on the TRISP spectrometer at MLZ in Garching, Germany. The energy resolution of these measurements was about 1 μeV [3]. The TAS measurements were carried out at the TASP spectrometer at PSI in Villigen, Switzerland. The energy resolution of this setup was about 0.16 meV.

Magnon decoherence and dimensional crossover

In IPA-Cu($Cl_{0.95}Br_{0.05}$)$_3$, Δ and Γ is always larger, but its temperature evolution is similar to that of the pure compound (fig. 2a and fig. 2b respectively). Furthermore, $\delta\Delta$ and $\delta\Gamma$ show very similar behavior in the entire T- range. For $T > 13$ K, $\delta\Delta$ and $\delta\Gamma$ is T-independent within our experimental precision. Upon lowering T, both $\delta\Delta$ and $\delta\Gamma$ drop in a narrow T-range of $8 < T < 13$ K. For $T < 8$ K, $\delta\Delta$ and also $\delta\Gamma$ saturate at ~0.1 meV and ~0.05 meV respectively.

The observation of approximately T-independent values of $\delta\Delta$ and $\delta\Gamma$ in IPA-Cu($Cl_{0.95}Br_{0.05}$)$_3$ at high-T is qualitatively consistent with the NLσM, and with Matthiessen's rule[5]. Written in terms of the magnon mean free path, L, this rule stipulates that $L^{-1} = L^{-1}_d + L^{-1}_s$, where L_d and L_s arise from magnon-magnon and magnon-defect collisions, respectively. At high temperatures, the T-dependence of L arises solely from L_d, which is well described by the NLσM. We can thus compare our experimental data to the results of Monte Carlo calculations for finite S = 1 chains, which yield a chain length of $L_s \sim 10a$. A similar estimate can be made on the basis of the broadening, which translates into $L_s \sim 22a$ [5].

In the low-T limit, L is expected to be dominated by L_s. The observation of T-independent values of $\delta\Delta$ and $\delta\Gamma$ for $T \leq 8$ K is consistent with this expectation. The reduced values of $\delta\Delta$ and $\delta\Gamma$ at low, compared to those observed in the high-T regime, are in qualitative agreement with the dimensional crossover picture developed for IPA-$CuCl_3$[3].

At temperatures $8 \text{ K} < T < 13$ K, $\delta\Delta$ and $\delta\Gamma$ are strongly T-dependent, which indicates that the simple Matthiessen's rule no longer applies in this regime, where dimensional crossover phenomena occur. We hope that our data will stimulate further experimental and theoretical work on dimensional crossover phenomena and their influence on the magnon spectrum and magnon-mediated thermal transport in IPA-$CuCl_3$ and other quasi-1D quantum magnets.

[1] D. Senechal, Phys. Rev. B 48, 15880 (1993).
[2] G. Y. Xu et al., Science, 317, 1049 (2007).
[3] B. Náfrádi et al., Phys. Rev. Lett., 106, 177202 (2011).
[4] H. Manaka et al., Phys. Rev. B, 73, 104407 (2006).
[5] B. Náfrádi et al., Phys. Rev. B, 87, 020408(R) (2013).