Low temperature ESR in spin ladder

\((C_7H_{10}N)_2Cu_{(1-x)}Zn_xBr_4\)

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Abstract. Spin-gap magnet \((C_7H_{10}N)_2Cu_{(1-x)}Zn_xBr_4\) (DIMPY) is an example of a strong-leg spin ladder. We report here the results of the ESR study of pure and diamagnetically diluted DIMPY. ESR study of the pure system \((x=0)\) revealed that the spin dynamics of DIMPY is affected by uniform Dzyaloshinskii-Moriya (DM) interaction. We observe narrowing of the ESR absorption line in diamagnetically diluted DIMPY pointing to suppression of DM channel of spin relaxation by doping.

1. Introduction

Spin ladder is a model low-dimensional spin system. It consists of two spin chains (called ‘legs’) coupled by interchain exchange bonds (which are the ‘rungs’ of spin ladder). The ground state of such system is a singlet and exited states are triplets. Energy spectrum of spin chain has a gap for arbitrary ratio between in-chain and inter-chain antiferromagnetic exchange constants \([1]\). Recently discovered \([2, 3]\) metallo-organic compound \((C_7H_{10}N)_2CuBr_4\) (called DIMPY for short) is a rare example of the strong leg spin ladder: in-chain exchange coupling \(J = 1.42\) meV exceeds inter-chain coupling \(J = 0.82\) meV \([4]\). Energy gap \(\Delta =0.33\) meV was directly observed with inelastic neutron scattering \([4]\) and in the specific heat and magnetization measurements \([2, 3, 5]\). Magnetic phase diagram of DIMPY features low temperature field-induced ordering below 300 mK in the fields above approx. 20 kOe \([5]\).

The ESR method has high energy resolution (about 0.005 meV) and it could be used to detect weak anisotropic spin-spin interactions. Such anisotropic spin-spin interactions exist in our system: DIMPY crystal symmetry allows Dzyaloshinskii-Moriya (DM) interaction. Because of translation and inversion symmetry present, DM interaction is forbidden on the rungs of the ladder, it is uniform along the leg of the ladder and has a different sign on different legs (figure 1). This allows to build a compact microscopic model which includes two exchange coupling constants and three components of DM vector only:

\[
\mathcal{H} = J \sum (\hat{S}_{i,a}\hat{S}_{i+1,a} + \hat{S}_{i,b}\hat{S}_{i+1,b}) + j \sum \hat{S}_{i,a}\hat{S}_{i,b} + D \sum (\hat{S}_{i,a} \times \hat{S}_{i+1,a} - \hat{S}_{i,b} \times \hat{S}_{i+1,b})
\] (1)
Figure 1. Model spin ladder with uniform Dzyaloshinckii-Moriya interaction along the legs of the ladder (Eqn.(1)). \(J, j\) — in-chain and inter-chain exchange couplings, grey arrows schematically show directions of DM vectors \(\vec{D}\); indices \(a\) and \(b\) enumerate legs of the ladder, \(i, (i+1)\) etc. enumerate rungs of the ladder. Crosses on the rungs mark positions of inversion centers \(I\).

Here \(i\) counts spins along the leg of the ladder and \(a, b\) are legs’ indices. Solving such a model remains a challenge, but supposedly it should yield both the fine structure of the excitations spectrum and relaxation of the spin excitations due to anisotropic spin-spin interactions.

Here we report on observations proving presence of DM interaction in pure DIMPY and on the effect of the diamagnetic dilution. We observe that diamagnetic dilution results in the narrowing of ESR absorption line, i. e. that introduction of structural disorder actually increases quasiparticles lifetime.

2. ESR in pure DIMPY

Figure 2. ESR absorption spectra of pure DIMPY at low temperatures. Magnetic field is applied at 45° from \(a\) to \(b\) axis, which corresponds to the maximum difference between absorption components. Vertical dashed lines show position of resonance absorption with certain \(g\) - factors, narrow line at \(g = 2.00\) is a DPPH marker.

ESR in pure DIMPY was described in details in [6]. We have measured ESR spectra for DIMPY at different temperatures (figure 2). The two main signals are because of two types of ladders in DIMPY, which are inequivalent for the chosen field direction. On cooling ESR absorption looses intensity as expected for the system with an energy gap. At the lowest temperature of 450 mK ESR signal vanishes. We have found additional splitting of ESR components at \(T \approx 1\) K (figure 2), which can be ascribed to the effect of zero field splitting of triplet sublevels. The estimate of zero field splitting is about 0.03 meV, as evaluated through maximum subcomponent splitting \(\Delta H = 150\) Oe.

DM interaction could lead to such a zero field splitting. Additional proof of the presence of DM interaction can be obtained through the analysis of the high temperature ESR linewidth
which is determined by anisotropic spin-spin interactions [7]. Analysis of linewidth angular dependences proved valuable contribution of this interaction, $D \approx 0.03$ meV [6]. Independently, high-frequency high-field ESR study [8] proved existence of DM interaction in DIMPY through observation of additional ESR mode which became allowed due to DM interaction.

Temperature dependence of the ESR linewidth was measured from 400 mK to 300 K [6]. Below 100 K ESR linewidth increases on cooling reaching its maximum value around 10 K. Below 10 K ESR line narrows with cooling as the gapped system enters to the regime of diluted quasiparticles gas (figure 3). Complete description of this temperature dependence still awaits theoretical effort, but we speculate that it can be explained in the compact model of Eqn.(1).

3. ESR in Zn-doped DIMPY

![Figure 3](image3.png)

**Figure 3.** Temperature dependences of the ESR linewidth for the 4% Zn-doped sample (triangles) and for the pure DIMPY (squares). Upper panel shows typical ESR absorption spectrum with resolved absorption signals from the inequivalent ladders. Left and right panel show data for left and right absorption components, respectively.

![Figure 4](image4.png)

**Figure 4.** Temperature dependences of the ESR intensity for the 4% Zn-doped sample (closed squares) and for the pure DIMPY (open squares). Intensities are scaled to the known static susceptibility. Upper panel shows typical ESR absorption spectrum with resolved absorption signals from the inequivalent ladders. Left and right panel show data for left and right absorption components, respectively.

Recently, samples of DIMPY with part of the copper ions substituted by non-magnetic zinc were grown [9]. This diamagnetic dilution resulted in the formation of multispin defects around the impurity ion. These multispin defects determine the low temperature magnetic properties of the system. To check effect of this doping on spin dynamics of DIMPY we have measured ESR absorption for the sample containing 4% Zn in wide temperature range (0.5 K - 25 K).

ESR absorption spectra for the doped DIMPY have the same two components corresponding to different types of spin ladders. Intensity of the ESR absorption can be scaled to static magnetization thus allowing to compare absorption intensities for different samples (figure 4). We have found that above 4 K scaled intensities for pure and 4% Zn-diluted sample practically coincides. This proves that at these temperatures triplet excitations dominate. Below 4 K the intensity of the ESR absorption for 4% Zn-diluted sample increases on cooling as paramagnetic multispin defects became dominant. ESR $g$-factors are the same in both regimes indicating that triplet excitations and multispin defects are formed from the same spins of Cu$^{2+}$ ions.
Surprisingly, ESR linewidth near maximum at $T \approx 10$K decreases with doping (figure 3). It means that excitations lifetime increases with doping at this temperature, where the magnetic response arises from collective delocalized triplet excitations. Recalling that the ESR linewidth for pure DIMPY is, supposedly, due to DM interaction, we conjecture that disorder created by diamagnetic dilution of the spin ladder suppresses relaxation channel due to DM interaction.

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