

Role of the antisymmetric exchange in quantum spin liquids

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The quantum critical state of organic quantum spin liquids (QSL) exhibits large sensitivity even to weak perturbations. For example, the antisymmetric exchange, the Dzyaloshinskii-Moriya (DM) interaction, which is present in all spin systems without inversion symmetry, could result in a phase transition from the quantum critical phase to an antiferromagnetic phase already at moderate magnetic fields. Using the combination of multi-frequency Electron Spin Resonance spectroscopy (ESR) in the 1-500 GHz frequency range and muon spin rotation (mSR), we studied the influence of the DM interaction in two-dimensional and quasi-one-dimensional organic QSL candidates.

In the triangular lattice QSL, $k\text{-(ET)}_2\text{Ag}_2\text{(CN)}_3$ ($J'/J=0.94$, $J=175$ K), our ESR measurements found a static staggered moment of 6×10^{-3} mB at $T=1.5$ K and at $B=15$ T [1]. The magnetic field dependence of the ESR linewidth, which measures the spectral density of the antiferromagnetic fluctuations, proves that this staggered moment stems from the DM interaction ($DM_0=4$ K) in a perfectly crystalline two-dimensional structure. In a new quasi-one-dimensional QSL candidate, $(\text{EDT-TTF-CONH}_2)_2+\text{BABCO-}$, which is a weak Mott insulator with a distorted triangular lattice ($J'/J=3$, $J=360$ K), our combined ESR and mSR study confirmed the absence of magnetic ordering down to 20 mK [2]. This remarkable observation is partially attributed to a unique structural motif of the $(\text{EDT-TTF-CONH}_2)_2+\text{BABCO-}$ salt. Here, the $(\text{EDT-TTF-CONH}_2)_2+$ conducting layers are separated by the highly disordered BABCO- molecular rotors. Importantly, despite the presence of a sizable DM interaction ($DM_0=0.6$ K), the staggered moment is smaller than 4×10^{-4} mB at $T=1.5$ K and $B=15$ T. The magnetic field dependence of the ESR linewidth does not show the effect of the DM interaction. Instead, the linear dependence is indicative of the presence of fast spin fluctuations, which is supported by longitudinal-field mSR measurements that reveal the spin excitations to possess one-dimensional diffusive character. The quenching of the effect of the DM interaction is explained by the strong disorder introduced by the anion layer.

Despite the fact that the magnitude of the DM interaction is 2 to 3 orders of magnitude weaker than the symmetric exchange, it can substantially alter the phase diagram of QSLs. Our work gives a novel explanation to the field-induced phase transitions, and it demonstrates that high-frequency ESR is a powerful technique to study the spin dynamics of QSLs.

Résumé en anglais

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