Optical Contrast Through the Field-driven Transition in a Quantum Heisenberg Spin Ladder

J.L. Musfeldt, J.M. White (Tennessee); S.A. Crooker, J. Singleton, R. McDonald (NHMFL); C. Landee, M. Turnbull (Clark), M.-H. Whangbo (NCSU)

Introduction

Physical manifestations of quantum spin ladders are relatively rare, especially in the theoretically interesting “strong rail” range and when the ratio of magnetic exchange constants $J_{\text{rung}}/J_{\text{rail}} \sim 1$. At the same time, many compounds that have been confirmed as spin ladders are complex oxides, which are difficult to study in conventional powered and pulsed magnets due to their relatively high energy scales. Copper halides, on the other hand, present a weaker ligand environment and overall lower exchange constants, allowing saturation of the magnetization in experimentally realizable magnetic fields. In this work, we take advantage of these considerations and employ the molecular spin ladder 2,3-dimethyl pyridinium tetrabromo cuprate ($\text{(2,3-dmpyH}_2\text{)}\text{CuBr}_4$, $J_{\text{rung}}/J_{\text{rail}} \sim 0.5$) to probe color property changes through the field-driven antiferromagnetic to ferromagnetic transition. The ability to saturate magnetization makes copper halides excellent model materials.

Results and Discussion

The optical properties of $(\text{2,3-dmpyH}_2\text{)}\text{CuBr}_4$ are shown in Fig. 1(b). Here, the $\text{CuBr}_4^{2-}$ anion acts as a chromophore, with a well-known series of on-site $d$ to $d$ excitations between crystal field split levels of the site $\text{Cu}^{2+}$ center. Strikingly, application of a magnetic field reduces the absorption coefficient between ~2 and 2.8 eV, with small changes in the triplet structure at ~2.15 eV and a larger shift of the absorption edge centered near 2.6 eV. We visualize these field-induced color property changes by plotting the absolute value of the absorption difference ($\alpha(H) - \alpha(H=0 \, \text{T})$), and we quantify these effects by integrating the absorption difference data (Fig. 1(c)). Strikingly, the field-induced optical contrast tracks the magnetization, albeit with a substantial “lag”. Based upon the magnitude of peak shifts in the field, the relatively small $g$-factor anisotropy and shift (which rule out mechanisms involving Zeeman splitting of $\text{Cu} d$ levels), and the observed color property modifications, we propose that the applied field modifies intermolecular exchange interactions and, as a consequence, the local structure of the chromophore. We also compare our values of $H_{c1}$ and $H_{c2}$ (2.9 and 30 T) obtained from magnetization data with various theoretical predictions and extract a value for the spin gap ($\Delta \sim 3.6$ K).

Figure 1: (a) Crystal structure of $(\text{2,3-dmpyH}_2\text{)}\text{CuBr}_4$ showing the bromine−bromine connections that form the rungs and rails of the ladder, (b) the optical absorption spectrum of $(\text{2,3-dmpyH}_2\text{)}\text{CuBr}_4$, and (c) the magneto-optical contrast which tracks the magnetization.

Acknowledgements

We thank the Division of Materials Research, National Science Foundation for financial support.