

## Infrared Investigations of Low-Dimensional Molecular Solids

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**Introduction:** Use of the infrared beamline facilities at Brookhaven National Lab enabled us to complete two projects. The first involved polarized far infrared measurements to support our more extended middle infrared studies of an inorganic molecular magnet material at SUNY-Binghamton. The second also exploited the high brightness of the synchrotron source to investigate the far infrared response of a prospective spin Peierls material. These works are summarized below.

**Results and Discussion:** We have investigated the temperature dependent infrared spectra of pure, deuterated, Zn-doped, and dimethyl-substituted samples of the  $S=1$ , one-dimensional quantum Heisenberg antiferromagnet copper pyrazine dinitrate ( $\text{Cu}(\text{C}_4\text{H}_4\text{N}_2)(\text{NO}_3)_2$ ). Of the more than 100 vibrational modes observed in the spectra, nearly one third of them unexpectedly soften throughout the temperature range of investigation (300 – 5 K). We discuss the temperature dependence of the vibrational spectra in terms of several different modes for mode softening. Based on detailed structural information and a comparison of the infrared spectra between pure and chemically modified copper pyrazine dinitrate, we conclude that the unusual softening observed in this low-dimensional molecular magnet is due to enhanced interchain hydrogen bonding with decreasing temperature.

We also measured the polarized infrared reflectance of  $\beta\text{-(ET)}_2\text{SF}_5\text{CF}_2\text{SO}_3$  as a function of temperature and analysed the spin-exchange interactions of this salt by calculating the spin-orbital interaction energies between adjacent spin sites. We examined the vibrational features below the 45 K transition that support a lattice distortion in combination with a spin gap in this material. The spin-exchange interaction is predicted to occur primarily along the (a+b) diagonal direction, in good agreement with the spectral data. In addition, notable frequency shifts of vibrational modes point to charge ordering between 100 and 200 K. The low-lying electronic excitation in the stack direction (assigned as a charge-transfer feature based upon our electronic band structure calculations) shows distinct changes with temperature that are consistent with the trends in the vibrational spectrum. The pattern of a high temperature charge ordering transition preceding the spin-Peierls transition in  $\beta\text{-(ET)}_2\text{SF}_5\text{CF}_2\text{SO}_3$  is discussed in relation to other low-dimensional organic and inorganic materials.

**Conclusions:** Two different infrared beamline setups at Brookhaven were used to obtain detailed far infrared spectra on very small single crystal samples of low-dimensional inorganic and organic molecular materials, allowing us to explore the magnetoelastic correlations in two prototypical magnetic solids.

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