

Study of multiple coupling between the spin, charge, and lattice in a multiferroic organic charge-transfer salt κ -(BEDT-TTF)₂Cu[N(CN)₂]Cl

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The molecular dimer Mott insulator κ -(BEDT-TTF)₂Cu₂(CN)₃ has been in the focus of scientific research efforts as a candidate for a quantum spin liquid due to strong geometrical spin frustration on a triangular lattice based on (BEDT-TTF)₂ dimers[1]. Recently, these materials have gained additional attention as a new class of ferroelectrics of purely electronic origin [2-4]. In the dimer-Mott insulator picture, one hole carrier with $S = 1/2$ is localized uniformly on the dimers. The ferroelectricity indicates broken inversion symmetry, which can be induced by a charge disproportionation within a dimer, resulting in an electric dipole. Therefore, a close coupling between the charge, spin, and lattice degrees of freedom is expected.

κ -(BEDT-TTF)₂Cu[N(CN)₂]Cl, which exhibits simultaneous occurrence of electronic ferroelectricity and antiferromagnetic spin order below $T_{FE} \sim T_N \sim 25$ K [3] is an ideal system to study such cross coupling between charge, spin and lattice. We performed inelastic neutron scattering (INS) on κ -(BEDT-TTF)₂Cu[N(CN)₂]Cl using the triple axis spectrometer IN8 at ILL. Two deuterated single crystals with size of $2 \times 1.4 \times 0.8$ mm³ were used for the experiments. To obtain large neutron flux, doubly focused PG monochromator and Cu analyzer were used with fixed k_f at 2.66 \AA^{-1} .

Although our sample was relatively small (total mass of two single crystals is ~ 7 mg) for an INS measurement, we succeeded to obtain clear phonon signals at $T = 100$ K with ~ 8 min counting owing to the large neutron flux of IN8 and low background. Upon cooling, we observed softening and damping of the lowest optical mode in a wide temperature range, which

indicates anharmonic lattice coupled to the other degree of freedom. Now, the detailed mechanism of overdamped phonon modes are analyzed in the view point of cross coupling.

References:

- [1] Y. Shimizu et al., Phys. Rev. Lett. 91 (2003) 107001.
- [2] Majed Abdel-Jawad et al, Phys. Rev. B 82 (2010) 125119.
- [3] P. Lunkenheimer et al., Nature Materials 11, (2012) 755.
- [4] S. Iguchi et al. Phys. Rev. B 87 (2013) 075107.