Experimental Report

Proposal: **7-01-401** **Council:** 4/2014

Title: Study of multiple coupling betweenthe spin, charge, and lattice in amultiferroic organic charge-transfer salt k-(BEDT-TTF)2Cu[N(CN)2]Cl

This proposal is a new proposal

Researh Area: Physics

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Samples: k-(BEDT-TTF)2Cu[N(CN)2]Cl

Instrument	Req. Days	All. Days	From	To
IN8	5	8	10/12/2014	18/12/2014

Abstract:

Quasi-2D organic charge-transfer salts k-(BEDT-TTF)2Cu2(CN)3 has been in the focus of recent scientific attention as a candidate for a quantum spin liquid. After the discovery of a relaxor-like dielectric response in k-(BEDT-TTF)2Cu2(CN)3 and ferroelectricity in k-(BEDT-TTF)2Cu[N(CN)2]Cl, these compounds have gained further attentions as a new class of ferroelectrics of purely electronic origin. Since k-(BEDT-TTF)2Cu[N(CN)2]Cl shows long-ranged ferroelectric order and antiferromagnetic order simultaneously, it is an ideal system to study the coupling between the spin, charge, and lattice degrees of freedom. Here, we propose a neutron scattering experiment on deuterated single crystals of k-(BEDT-TTF)2Cu[N(CN)2]Cl in order to search for phonon anomalies coupled with electronic ferroelectricity.

The molecular dimer Mott insulator κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$ 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 $(BEDT\text{-}TTF)_2$ 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-$(BEDT\text{-}TTF)_2Cu[N(CN)_2]Cl$, which exhibits simultaneous occurrence of electronic ferroelectricity and antiferromagnetic spin order below T_{FE} ~T_N ~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-$(BEDT\text{-}TTF)_2Cu[N(CN)_2]Cl$ using the triple axis spectrometer IN8 at ILL. Two deuterated single crystals with size of 2 x 1.4 x 0.8 mm^3 were used for the experiments. The crystals were mounted so as to access (*h0l*) scattering plane. The final neutron energy was fixed to 14.7meV using. To obtain large neutron flux, doubly focused PG monochromator and Cu analyzer were used with fixed k_f at 2.66$Å^{-1}$.

Although our sample was relatively small (total mass of two single crystals is ~7mg) for an INS measurement, we succeeded to obtain clear phonon signals at T=100K with ~8min counting owing to the large neutron flux of IN8 and low background (Fig.1). Upon cooling, we observed softening and damping of the lowest optical mode in a wide temperature range 25<T<75K. Furthermore, we observed hardening of the soft mode at 25K (Fig.2), which indicates that these phonon modes are coupled to the multiferroic transition at

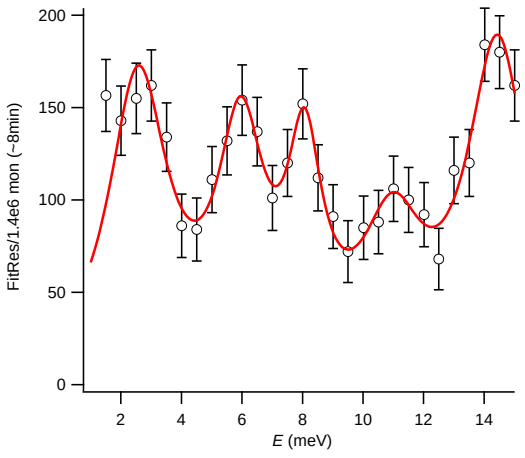

Fig.1 Constant-Q scans at (603) measured at T=100K.

25K. In the temperature region of the overdamped soft mode, the structural freezing of molecular rotational degrees of freedom occurs at T_g ~ 75K suggesting an intimate relation of both effects. Moreover, our experiments indicate also changes of higher energy phonon modes at the multi-ferroic transition at 25 K as e.g. seen around 6meV (Fig. 2 inset). Whereas this experiment focused on the phonon change at the multiferroic transition at 25K, further investigation at the glass-like structural transition at T_g ~75K and the charge localization at T_{ins} ~ 50 K would clarify the cross correlation between charge and lattice degrees of freedom.

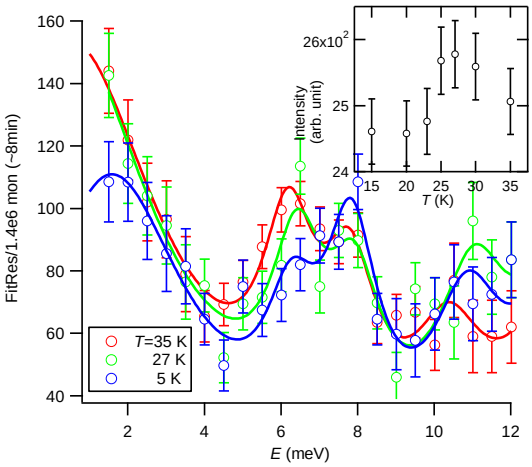

Fig.2 Constant-Q scans at (603) measured at T = 5, 27, and 35 K. Solid lines are fits assuming damped harmonic oscillator function. Inset shows thermal variation of intensity at E = 6.5 meV, which shows

References:

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