Tuning Interactions in κ-BEDT-TTF salts via Strain

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In triangular lattice systems with strong electron interactions a ground state of a system is defined by lattice frustration and strength of electronic interactions. If a system is close to a phase transition, then small perturbations of these parameters can result in a large transformation of the macroscopic ground state. The BEDT-TTF[1] based materials κ-$ET_2Hg(SCN)_2X$ (X=Br, Cl) constitute an anisotropic triangular lattice of dimers of ET molecules with +0.5e per molecule on average. In the low temperature Mott insulating state of these materials the competition between Coulomb interactions and intermolecular hopping within and between ET^{1e+} dimers lead to a complex phase diagram, where ferroelectric, dipole-liquid, and spin-liquid behavior are all possible [1,2,3,4,5]. Previously we have shown how an anion substitution changes a ground state of these materials from dipole liquid in $ET_2Hg(SCN)_2Br$)[4], to a charge order ferroelectric state in (κ-$ET_2Hg(SCN)_2Cl$)[1,3] .

This work is the first step to demonstrate that uniaxial strain is a tuning parameter for a ground state of these triangular correlated systems through a phase border of a charge ordered state. Using Raman scattering spectroscopy, we have access to local properties such as charge disproportionation within dimers through vibrational spectroscopy, as well as collective phenomena through the observation of collective charge excitations. Through uniaxial strain of 0.5% along the c-axis we are able to tune κ-$ET_2Hg(SCN)_2Br$ into a charge ordered state with a transition temperature of 40K, above the charge-ordering temperature of κ-$ET_2Hg(SCN)_2Cl$ at 30K[1,3]. Moreover, we suppress the ferroelectric transition in κ-$ET_2Hg(SCN)_2Cl$ down to 10K applying uniaxial strain of 1.7% along the b-axis. We are also able to follow collective charge excitations of these charge states at frequencies of about 50 cm^{-1} as a function of strain. A comparison with theoretical predictions of Ref. [2,5,6] suggests a critical value of effective interdimer Coulombic interactions can be estimated, V_C, at which point the Dimer-Mott Insulator to Charge-Ordered Insulator transition occurs.

[1] (bis(ethylenedithio)tetrathiafuvalene, referred to as ET)

References

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