Interplay and coupling of charge and spin degrees of freedom in k-(BEDT-TTF)$_2X$ triangular Mott insulators

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A family of organic quasi-two-dimensional Mott insulators with a general formula k-(BEDT-TTF)$_2X$, where BEDT-TTF is bis(ethylenedithio)tetrathiafuvalene and *X* denotes a suitable anion was suggested to host a quantum spin liquid with S=1/2 on a triangular lattice. Breaking of an inversion symmetry of a molecular orbital of (BEDT-TTF)$_2$ dimer lattice sites can result in a ferroelectric state in these Mott insulators, with a quantum paraelectric state found in proximity. While the charge degree of freedom of members of this family κ-ET$_2$Hg(SCN)$_2$Cl (T_{CO}=30 K) and k-ET$_2$Hg(SCN)$_2$Br is relatively easy to probe by vibrational spectroscopy [1-2], understanding of the magnetic state of these materials still presents a challenge. Understanding the magnetic state will allow to elucidate how the charge degree of freedom determines magnetism in these materials.

Here we summarize the results of different probes such as on magnetic Raman scattering, magnetic torque and ESR applied to study magnetism in of κ-ET$_2$Hg(SCN)$_2$Cl and k-ET$_2$Hg(SCN)$_2$Br. We demonstrate how the symmetry of charge on a (BEDT-TTF)$_2$ dimer controls magnetic state. Magnetic Raman scattering clearly shows two-magnon excitations only in compounds where charge asymmetry is low or absent. Two-magnon excitations of S=1/2 on triangular lattice are absent in the charge ordered κ-ET$_2$Hg(SCN)$_2$Cl. Magnetic susceptibility and torque measurements allow us to show that the system develops spin-singlet correlations in the charge ordered state at T= 24 K, which are destroyed on further cooling by melting of charge order below T=15 K, suggesting a spin liquid candidate. Fluctuating dipoles in k-ET$_2$Hg(SCN)$_2$Br also prevent a development of two-magnon. Our ESR data confirm the presence of inhomogeneous fluctuating behavior in this system suggested in Ref. [3] as a source of exotic ferromagnetic correlations.

References

[1] N.M. Hassan *et al*., *Science* **360**, 1101-1104 (2018).
[2] N.M. Hassan *et al*., *npj Quantum Mater.* **5**, 15 (2020).
[3] M Yamashita *et al*., *npj Quantum Mater.* **6**, 1 (2021).