Raman scattering signatures of quantum dipole liquid in an organic Mott insulator.

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Mott insulators are commonly pictured with electrons localized on lattice sites. Their low-energy physics involves spins only. Recent theoretical work suggests that in molecular systems a new on-site charge degree of freedom can emerge. On a frustrated lattice with charge-spin coupling it would result in a new quantum spin liquid state [1-3].

We experimentally demonstrate a presence of this fluctuating charge degree of freedom in a molecule-based Mott insulator k-(BEDT-TTF)$_2$Hg(SCN)$_2$Br. When electrons localize on a triangular lattice of molecular dimers of this compound at temperatures below 100 K, they form electric dipoles which do not order at low temperatures and fluctuate, resulting in a so-called quantum dipole liquid state. A frequency of dipole fluctuations of 40 cm^{-1} is detected experimentally in our Raman spectroscopy experiments through an observation of a related collective mode. We show that this spectroscopic response of a quantum dipole liquid is qualitatively different from a response of molecular Mott insulators with no on-site charge degree of freedom. The Raman spectra of the latter show two-magnon excitations at frequencies below 500 cm^{-1} expected for a S=1/2 antiferromagnet on a triangular lattice with J≈250 K. Our results can be a key to understanding of organic triangular lattice spin liquid candidates.

References

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