The Role of Charge Degrees of Freedom in Mott Insulators: Coupling of Dielectric and Magnetic Properties in Organic Cr-trimer Complexes.

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Materials that are insulating owing to strong electron correlations are pervasive in condensed matter physics – from the organics to the parent phase of high-T_C cuprates, colossal magnetoresistive manganites and quantum magnets. All are characterized by a large onsite coulomb repulsion relative to the dominant electron hopping ($U/t >> 1$). As such, at half-band filling the charge is localized (one electron per site). The physical properties of these materials are therefore commonly described solely in terms of their remaining spin degrees of freedom (magnetic exchange interactions) with little attention given to any further role of the charge degrees of freedom. Certain classes of Mott insulator are predicted to break this paradigm. For finite U/t, the kinetic energy gain due to the overlap with neighboring sites of opposite spin orientation allows for the possibility of real currents resulting from hopping around a closed loop. This provides a direct correlation between the magnetic spin texture and the dielectric properties of a material. The simplest loop is an isolated triangle such as the Cr-trimer systems investigated, where the size of the organic packing molecule can be used to minimize the intertrimer interactions.

We observe a correlation between the magnetic and dielectric responses of Cr-trimer systems, which combined with recent theoretical developments, indicates a purely electronic mechanism for multiferroic behavior. Magnetic field strengths of the order of the exchange interaction strongly perturb the spin texture, which is evident as steps and plateau in the magnetization. The corresponding shifts in dielectric properties reveal the role of the charge degrees of freedom. Electron Spin Resonance (ESR) results and the prospect of novel dipole-active ESR giving rise to the possibility of negative refractive indices will also be discussed.