Coupling of charge and spin order in organic charge transfer salts

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There is a growing number of organic charge-transfer salts, such as TTF-CA, TMTTF, and BEDT-TTF salts, where electronic degrees of freedom and electronic interactions are directly responsible for electric polarization and ferroelectric transition. Recently, it was discovered that charge order in TMTTF salts not only produces electronic ferroelectricity but also breaks the symmetry of the magnetic degree of freedom in these organic quantum spin chains [1]. The interaction of charge order and spin order naturally involves also the underlying lattice. In two-dimensional BEDT-TTF salts, charge order is well known in $\alpha$- and $\theta$-phase systems, with a strong dielectric response due to domain-wall motion [2]. Fluctuating charge order was shown to drive superconductivity in $\alpha$- and $\beta'$-salts [3]. Currently an intense debate takes place for the $\kappa$-phase compounds, where dimers form a triangular lattice. For $\kappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl and $\kappa$-(BEDT-TTF)$_2$Cu$_2$CN$_3$ vibrational spectroscopy rules out any charge disproportionation required for electric dipoles on the dimers [4]; alternative explanations for the dielectric response have to be discussed. For the highly frustrated system $\kappa$-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl the on-site Coulomb repulsion is reduced making inter-site interaction more important and paving the way to quantum electric dipoles: in fact, the splitting of the vibrational modes proves that charge order drives the metal-insulator transition at 30 K [5]. We discuss the possible existence of fluctuating spin singlets.