Neutral to Ionic Transition in Fluoranil based charge transfer crystals

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Mixed stack charge transfer (CT) crystals made up of alternating π-electron donor (D) and acceptor (A) molecules may undergo the neutral to ionic phase transition (NIT) at high pressure or low temperature. The NIT is both a valence and a structural instability, since the increase in the degree of charge transfer is associated with the dimerization of the stack due to Peierls instability. This breaks the inversion symmetry along the stack, possibly leading the system to a ferroelectric state, as in the prototypical Tetrathiafulvalene-Chloranil (TTF-CA).

The effects of the molecular properties of the components on NIT has been widely studied by pairing chemically substituted TTF or CA molecules. We recently expanded this TTF-quinone series with TTF-Fluoranil (TTF-FA), that undergoes a strong dirst-order temperature induced NIT[1]. Here we add a new system, DimethylTTF-FA, and compare the behavior of the two crystals. The infrared and Raman spectra recorded at high pressure and low temperature are presented, with focus in the low frequency spectral regions were lattice phonons and soft modes are expected.

[1] E. Ferrari, F. Mezzadri, and M. Masino, "Temperature-induced neutral-to-ionic phase transition of the charge-transfer crystal tetrathiafulvalene-fluoranil," *Phys. Rev. B*, vol. 105, no. 5, pp. 1–6, 2022, doi: 10.1103/physrevb.105.054106.