Electronic Structure of $(EDO\text{-}TTF)_2PF_6$ in its High Temperature Metallic Phase: Indications of Inherent Strong Electron Correlation

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$(EDO\text{-}TTF)_2PF_6$ has been attracting particular interest since the discovery of its photoinduced insulator-metal phase transition [1]. So far, this material has been regarded as a strong electron-lattice interaction system, based on its distinguished structural features such as tetramerization, molecular deformation, and anion ordering in its low-temperature phase with (0110)-type charge ordering. In the previous work based on first-principles calculations [2], however, we have argued the mechanism of this charge ordering by the combination of long-range Coulombic interaction and the tetramerization.

In this work, we focus on the high-temperature phase. Performing a DFT cluster calculation augmented by environmental self-consistent charges [2], we find a dimer-Mott type of spin polarization in the ground state. We do not insist on the existence of a long-range magnetic order in the high temperature phase but recognize the importance of electron correlation in this material although this point has not been emphasized in the past studies.

We have also calculated optical absorption spectra expected for this ground state, obtaining a peak structure around 0.4 eV in the mid-IR region. Since a mean-field type calculation overestimates the peak energy, we also try a mapping to the one-dimensional Hubbard model with bond-alternation. Exact diagonalizations then gives an value around 0.07 eV. Since the experimental peak is located around 0.2 eV [3], we think that the actual situaiton lies between the situations described by the two opposite treatments.

When we compare this material with other quasi-one-dimensional materials such as $(TMTTF)_2PF_6$ and $(TMTSF)_2PF_6$, we find that the observed mid-IR peak energies are linearly scaled by the degrees of bond alternation. This scaling suggests that, in spite of the peculiar (0110)-type charge ordering, $(EDO\text{-}TTF)_2PF_6$ shares a nature of a strongly correlated electron system, and that its photoinduced phase transition must be discussed in the light of this recognition.

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[2] K. Iwano and Y. Shimoi, Phys. Rev. B 77 (2008) 075120.
[3] O. Drozdova *et al.*, Synthetic Metals 133-134 (2003) 277.