Ab initio Derivation of Low-Energy Model for κ-(BEDT-TTF)$_2X$ [X= Cu(NCS)$_2$ and Cu$_2$(CN)$_3$]

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One of the most significant challenges in condensed matter physics is to clarify physics of electron correlations from first principles. Among a variety of correlated materials, organic conductors such as BEDT-TTF compounds are unique and important families which have served in revealing fundamental aspects of electron correlation in a number of novel phenomena. However, in contrast to another family of typical strongly correlated electron systems such as transition metal compounds, the organic compounds are so complex with hundreds of atoms in a unit cell that it has blocked theoretical studies on the realistic basis. While competitions of magnetic, charge, superconducting ordered phases or unprecedented phases such as quantum spin liquid phase or unconventional Mott transitions observed in organic compounds have been fascinating issues of great interest and hot debates, most of theoretical/experimental analyses so far have been based on empirical modeling. Thus, in order to step forward to realistic calculation, a reliable *ab initio* effective model on the firm grounds is imperative.

In the present study, we have for the first time succeeded in deriving *ab initio* effective models for organic conductors, κ-(BEDT-TTF)$_2X$, from first principles [1]. They contain band dispersions of the highest occupied Wannier-type molecular orbitals with the nearest neighbor transfer $t \sim 0.067$ eV for a metal X = Cu(NCS)$_2$ and 0.055 eV for a Mott insulator X = Cu$_2$(CN)$_3$, as well as screened Coulomb interactions with constrained random phase approximation. We will show that the resulting *ab initio* model is strikingly different from the past empirical Hückel model, especially much stronger onsite interaction $U \sim 0.8$ eV ($U/t \sim$ 12-15) than the Hückel estimates ($U/t \sim$ 7-8), together with an appreciable longer-ranged interaction, and also a smaller geometrical frustration (the ratio of the next-neighbor transfer to the nearest-neighbor one) than the Hückel estimates. The presented low-energy model firmly establishes a reliable starting point of analyses and offers a basis for further reexamination on physics of this family of materials.

[1] K. Nakamura, Y. Yoshimoto, T. Kosugi, R. Arita, and M. Imada, arXiv:0903.5409.