Effective model and spin/charge ordering in Pd(dmit)$_2$ salts

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Among the family of $\beta'$-type $X$[Pd(dmit)$_2$]$_2$ ($X$: monovalent cation) salts [1], many become a dimer-type Mott insulator and then at low temperatures they show magnetic order or spin-liquid behavior. On the other hand, pressure or chemical modification brings about competition with metallic/superconducting states, and also with a peculiar charge ordering involving multi-orbitals. In this study, we construct an effective low-energy model which take into account the multi-orbital degree of freedom, and investigate their electronic states systematically.

We take fragments of molecular orbital (fragment MO; fMO) as a basis set of the model, of which HOMO and LUMO of the Pd(dmit)$_2$ molecule are composed by linear combinations; each fMO becomes nearly localized on one of the dmit ligands (see Fig. 1). The inter-fMO transfer integrals are obtained for a series of salts $X = EtxMe_{4-x}Z$ ($x = 0-2$, $Z = P, As, Sb$) by fitting to the first-principles band calculations [2]. We find that all the intra-dimer transfer integrals illustrated in Fig. 1, including the diagonal ones, are appreciably large (~ 0.2-0.3eV); this results in a modification of the orbital scheme in strongly dimerized [Pd(dmit)$_2$]$_2$ discussed in the literatures, then to the effective dimer model. We calculate possible spin and charge ordering based on mean-field approximation to the extended Hubbard model incorporating the fitted parameters.

Fig. 1: [Pd(dmit)$_2$]$_2$ dimer. Green and purple ellipses schematically represent two kinds of fragments $a$ & $b$, and red arrows indicate inter-fragment transfer integrals.

References