Frustrated spin system in molecular crystals has received attention because their triangular lattices hinder any spin ordering even below liquid helium temperature. Whereas magnetic and thermal properties have been discussed, no data has been presented concerning frustrations originating from charge, lattice and molecular orbital. Here, we present mechanism of spin frustration in EtMe₃Sb[Pd(dmit)₂]₂ revealed by vibrational spectroscopy¹. Fig. 1 shows schematic view of our mechanism. Because HOMOs and LUMOs at monomers constitute LUMO and HOMO in a tight dimer, respectively, electron-electron repulsion cooperates with inter-molecular bonding. Not only magnetic order as a Mott insulator but also non-magnetic orders due to formations of tetramer and octamer become candidates for a ground state. Result of vibrational spectroscopy shows that three kinds of ordered state are degenerate in EtMe₃Sb[Pd(dmit)₂]₂, which leads to frustration due to interplay between spin, charge, lattice and orbitals¹.


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**Figure 1** (a) Monomer of [Pd(dmit)₂] and Tight dimer (b) Energy diagram near Fermi energy (E_F) (c) Dimer, tetramer and octamer in triangular lattice