Exotic Properties of $Pd(dmit)_2$ Salts Based on Various Degrees of Freedom (dmit=1,3-dithiole-2-thione-4,5-dithiolate)

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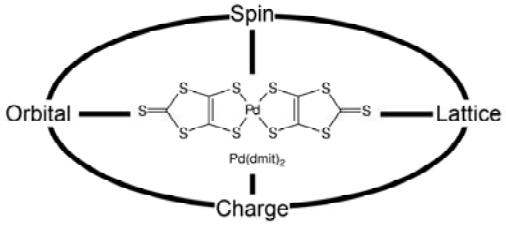
Metal-dmit complexes, $M(dmit)_2$ (dmit = 1,3-dithiole-2-thione-4,5-dithiolate; M=Ni, Pd), have provided a variety of molecular conductors. The electronic state depends on the central metal ion and the counter cation.



A series of anion radical salts of $Pd(dmit)_2$ with closed-shell cations, $(Cation)[Pd(dmit)_2]_2$, where cation is $Et_xMe_{4-x}Z^+$ (Z=N, P, As, Sb; x=0, 1, 2), have conducting anion layers. The $Pd(dmit)_2$ units are strongly dimerized. An important aspect of the Pd salts is a HOMO-LUMO interplay which originates from strong dimerization and a small HOMO-LUMO energy splitting. The conduction band is a narrow and half-filled HOMO band. Most of the Pd salts belong to a strongly correlated two-dimensional system with a quasi-triangular lattice formed by $[Pd(dmit)_2]_2^-$ dimers, and are Mott insulators at ambient pressure. The anisotropy of the inter-dimer interactions can be tuned by the choice of the cation. The localized electrons exhibit frustrated paramagnetism. At low temperatures, the spin frustration is removed by various transitions, including antiferromagnetic ordering, (intra- and inter-dimer) charge ordering, and valence bond ordering (spin gap formation). These phenomena are coupled with various degrees of freedom, charge, spin, lattice, and orbital. The Mott insulating state can be suppressed by the application of hydrostatic or uni-axial pressure, which leads to metallic behavior accompanied by superconductivity.