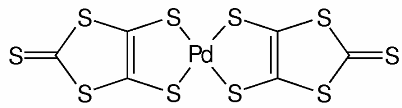
Molecular Mott insulators with various spin states and their release under pressure

R. Kato

RIKEN / JST-CREST, Japan



$Pd(dmit)_2$

A series of anion radical salts $Et_xMe_{4-x}Z$ $[Pd(dmit)_2]_2$ (dmit=1,3-dithiol-2-thione-4,5-dithiolate, Z = P, As, Sb and x = 0, 1, 2) are Mott insulators at ambient pressure.[1] Magnetic and conducting properties strongly depend on the counter cation. An important character of the $Pd(dmit)_2$ salts is a two-dimensional (2D) quasi triangular lattice formed by $[Pd(dmit)_2]_2^-$ dimers. In this Mott insulating state, one electron is located on each dimer. The localized electrons exhibit frustrated paramagnetism in the high temperature region.[2] At low temperatures, the spin frustration can be released by various transitions, including antiferromagnetic ordering,[1] charge ordering (2 $Dimer^- \rightarrow Dimer^0 + Dimer^{2-}$),[3] and valence-bond ordering (spin Peierls-like transition in the 2D system).[4]

The Mott insulating state of the $Pd(dmit)_2$ salts can be suppressed by the application of hydrostatic or uni-axial pressure.[1] In the case of the Me_4P and Me_4As salts with higher Néel temperatures (T_N > 35 K), the application of hydrostatic pressure cannot suppress the non-metallic behavior. For the Me_4As salt, however, the uni-axial pressure (strain) effectively induces the superconductivity. The Me_4Sb, Et_2Me_2Z (Z=P, As), and $EtMe_3As$ salts with T_N = 16~23 K at ambient pressure, turn metallic and show superconductivity under hydrostatic pressure. The $EtMe_3Sb$ salt, which retains the frustrated paramagnetism down to 1.37 K at ambient pressure, also shows a metallic behavior under hydrostatic pressure, but no superconductivity is observed up to 15 kbar. The charge ordering transition in the Et_2Me_2Sb salt is accompanied by a sharp increase of the resistivity at ambient pressure. The application of hydrostatic pressure turns this resistivity anomaly to a metal-insulator transition. Recently, we have found that the valence-bond ordering state of the $EtMe_3P$ salt (Fig. 1) is situated in the vicinity of the superconducting phase under pressure.[5]

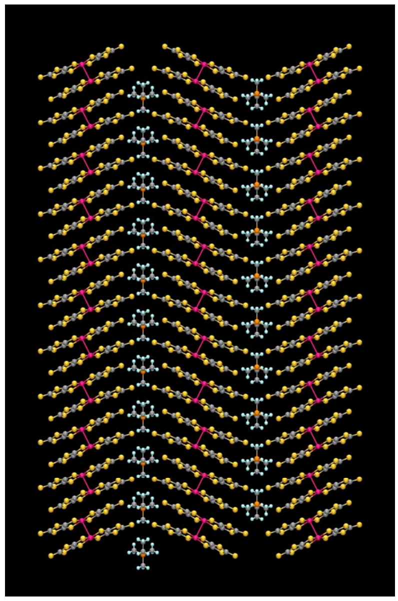


Figure 1: Crystal structure of $EtMe_3P[Pd(dmit)_2]_2$

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[2] M. Tamura, and R. Kato, J. Phys.: Condens. Matter, 14 (2002) L729
[3] M. Tamura, and R. Kato, Chem. Phys. Lett., 387 (2004) 448
[4] M. Tamura, A. Nakao, and R. Kato, J. Phys. Soc. Jpn. 75 (2006) 093701
[5] R. Kato, A. Tajima, A. Nakao, and M. Tamura, J. Am. Chem. Soc. 128 (2006) 10016

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Email: *reizo@riken.jp*