Anion radical salts of Pd(dmit)$_2$, (Et$_x$Me$_{4-x}$Z)[Pd(dmit)$_2$]$_2$ (Et=C$_2$H$_5^-$, Me=CH$_3^-$, dmit=C$_3$S$_5^{2-}$, Z=N, P, As, Sb; $x=0, 1, 2$) belong to a dimer Mott system [1]. All the Pd(dmit)$_2$ salts are characterized by a dimer unit [Pd(dmit)$_2$]$_2$ and a conducting anion layer where the dimer units form a quasi-triangular lattice. The anion layers are separated from each other by an insulating and nonmagnetic cation layer. The electronic structure near the Fermi level can be described by the dimer-based tight-binding picture, because the dimerization is very strong. Interdimer transfer integrals can be tuned by the choice of the cation. The conduction band, which originates from HOMO (Highest Occupied Molecular Orbital) of the Pd(dmit)$_2$ molecule, is two-dimensional, half-filled, and narrow.

At ambient pressure, all the Pd(dmit)$_2$ salts are in the Mott insulating state and are the spin-1/2 Heisenberg antiferromagnets where the spin frustration operates [2]. When the deviation from the regular-triangular lattice is large (for example, Me$_4$P and Me$_4$As salts), the antiferromagnetic long-range order is dominant and the frustration is removed at low temperature. On the other hand, the EtMe$_3$Sb salt with a nearly regular-triangular lattice does not show any spin ordering/freezing down to 19.4 mK [3, 4]. Since this temperature is smaller than 0.01% of $J$, the absence of spin ordering/freezing is attributed to quantum fluctuations.

In the system with a nearly regular-triangular lattice, the above-mentioned spin liquid state appears to compete with the valence bond formations. The Et$_2$Me$_2$Sb salt undergoes a first-order transition toward a charge separation state (2Dimer$^-$ $\rightarrow$ Dimer$^0$ + Dimer$^{2-}$) at 70 K [5]. In the EtMe$_3$P salt, a second-order transition from the frustrated paramagnetic state to the valence bond solid (VBS) state with a spin gap occurs at 25 K [6]. The former is an “intradimer” valence bond formation, and the latter an “interdimer” one. In these processes, charge, lattice, and orbital degrees of freedom play important roles.

The Mott insulating state in the Pd(dmit)$_2$ salts can be removed by application of pressure. The antiferromagnetic long-range order turns to a superconducting state under hydrostatic or uni-axial pressure. The EtMe$_3$P salt with the VBS ground state undergoes a first-order insulator-metal (I-M) Mott transition under hydrostatic pressure. With increasing pressure, the system shows a reentrant (I-M-I) transition and eventually a metal-superconductor transition [7, 8]. The VBS state survives over the whole insulating region, not replaced by a magnetically ordered state [9, 10]. That is, the spin-gapped VBS phase borders the superconducting phase.

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