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Quantum spin liquid in the spin-1/2 triangular antiferromagnet $\text{EtMe}_3\text{Sb}[\text{Pd}(\text{dmit})_2]_2$

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$\text{EtMe}_3\text{Sb}[\text{Pd}(\text{dmit})_2]_2$ ($\text{Et}=\text{C}_2\text{H}_5^-$, $\text{Me}=\text{CH}_3^-$, $\text{dmit}=\text{C}_3\text{S}_5$) is one of molecular conductors derived from an anion radical of the $\text{Pd}(\text{dmit})_2$ molecule and closed-shell monocations $(\text{Et}_x\text{Me}_{4-x}\text{Z})^+$ ($\text{Z}=\text{N}, \text{P}, \text{As}, \text{Sb}$; $x=0, 1, 2$) [1]. A common feature of these $\text{Pd}(\text{dmit})_2$ salts is a conducting anion layer where the $\text{Pd}(\text{dmit})_2$ anions form a dimer unit $[\text{Pd}(\text{dmit})_2]_2^-$. Electronic structure around the conduction band can be described by a simple tight-binding calculation based on the dimer unit. The conduction band is half-filled and two-dimensional. At ambient pressure, all the $\text{Pd}(\text{dmit})_2$ salts behave as Mott insulators where one spin is localized on each dimer. Interdimer transfer integrals indicate that the dimers form a quasi (isosceles) triangular lattice. Interdimer transfer integrals can be tuned by the choice of the cation, which deeply affects the electronic state. The EtMe_3Sb salt has a nearly regular-triangular lattice. The EtMe_3Sb cations are located between conduction layers and exhibit orientational disorder. The temperature dependence of the magnetic susceptibility is described in terms of the Padé approximant expression based on the high temperature series expansion of χ of the antiferromagnetic spin-1/2 Heisenberg model on the triangular lattice with an exchange interaction $J=220\text{-}250$ K. The ^{13}C -NMR measurements show no indication of either spin ordering/freezing or an appreciable spin gap down to 1.37 K, which is lower than 1% of J [2]. The specific heat measurements indicate gapless spin excitation. These results strongly suggest that the ground state of the EtMe_3Sb salt is a gapless spin liquid state. On the other hand, the $\text{Et}_2\text{Me}_2\text{Sb}$ salt, which has also a nearly regular-triangular lattice, shows a first-order transition toward a charge separation state ($2\text{Dimer}^- \rightarrow \text{Dimer}^0 + \text{Dimer}^{2-}$) at 70 K [3].

References

- 1) R. Kato, *Chem. Rev.*, **104**, 5319 (2004).
- 2) T. Itou et al., *Phys. Rev. B*, **77**, 104413 (2008).
- 3) M. Tamura and R. Kato, *Chem. Phys. Lett.*, **387**, 448 (2004).