Spin Liquid, Spin Gap, and Superconductivity in the Triangular-Lattice Mott Insulators X[Pd(dmit)$_2$]$_2$

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The family of layered organic salts X[Pd(dmit)$_2$]$_2$ are Mott insulators and form scalene-triangular spin-1/2 systems. Most of them have significant difference between the magnitudes of exchange interactions on the three sides of the triangle and have antiferromagnetic ground states owing to less frustrated nature. However, the salts with X = EtMe$_3$Sb (space group $C2/c$) and EtMe$_3$P ($P2_1/m$) have nearly-regular triangular lattices and show quantum disordered spin states due to strong frustration effect.

The EtMe$_3$P salt has the parallel molecular stacking structure, where lattice is easy to distort [1]. Consequently, the EtMe$_3$P salt is reported to undergo a lattice dimerization at 25 K, below which the VBS state with a spin gap is realized [1]. Recent susceptibility [2] and resistivity [3] studies for this material report that superconductivity appears under pressure. We have performed ^{13}C-NMR measurements under pressure and obtained clear microscopic evidence that the spin-gapped VBS phase borders the superconducting phase without emerging a magnetically ordered phase [4]. This phase diagram contrasts strikingly with the case of the large majority of other correlated electron superconductors in which the superconducting phase borders a magnetically ordered phase. This implies the possibility that the present superconductivity in the EtMe$_3$P salt has an exotic origin and nature.

The EtMe$_3$Sb salt has the solid-crossing stacking structure, where lattice is hard to distort unlike the EtMe$_3$P salt. Actually, lattice distortion has not been observed so far. Therefore, this salt is a candidate material of the spin liquid. We performed ^{13}C-NMR measurements for this salt down to 19.4 mK using a dilution refrigerator and found that the ground state is certainly quantum disordered state [5]. The spin-lattice relaxation rate T_1^{-1} shows that spin excitation is gapless down to 1 K. Interestingly, however, T_1^{-1} shows a kink at 1 K and decreases rapidly below this temperature. This means that a second-order phase transition accompanied by a symmetry breaking occurs at this temperature. The instability of the spin liquid is a great issue and several exotic instabilities have been proposed theoretically. It is possible that the present transition in the EtMe$_3$Sb salt corresponds to one of these instabilities.

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