Valence Bond Physics in the Frustrated Quantum Antiferromagnets, [Pd(dmit)$_2$] Salts

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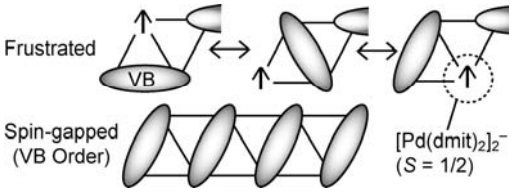
A series of the [Pd(dmit)$_2$] salts provide triangular antiferromagnets, in which the S =1/2 Heisenberg spins localized on the dimeric radical anion units $[Pd(dmit)_2]_2^-$ are frustrated [1]. In the $EtMe_3P$ salt (the $P2_1/m$ phase), spin-gapped phase appears below 25 K accompanied by spontaneous breaking of lattice translational symmetry, indicating that the spin gap is formed by the spatially ordered spin-singlet pairs (valence bonds, VB's) [2]. Suppression of the VB ordering by pressure affords superconductivity [3,4]. Some other salts with larger spatial anisotropy undergo antiferromagnetic long-range ordering. Crossover from a frustrated paramagnetic state to an antiferromagnetically correlated state is observed slightly above the transition temperature, indicating that low-lying short-range spin excitations are crucial role for the release of frustration leading to magnetic long-range order at low temperatures [5]. All these features can be systematically explained in terms of the ordering, resonance and flipping of the VB's over the 2D triangular network formed by the spin-1/2 units $[Pd(dmit)_2]_2^-$.



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