ESR studies of the valence bond solid state

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The valence bond solid (VBS) state of the anion radical salt EtMe₃P[Pd(dmit)₂]₂ is studied by electron spin resonance (ESR) measurements. The anion layer of EtMe₃P[Pd(dmit)₂]₂ is composed of Pd(dmit)₂ dimers which is S=1/2, and the dimers form an antiferromagnetic triangular lattice. Although the anion layer is expected to be frustrated, this sample shows a VBS state below 25 K [1]. Hence, we have performed the ESR measurements of EtMe₃P[Pd(dmit)₂]₂ to study its spin correlations and spin dynamics.

Firstly, the ESR linewidth shows an unconventional angular dependence, at 4 K which can only be explained by the lack of interlayer interaction between the Pd(dmit)₂ dimers (Fig. 1). However, this lack of interlayer interaction contradicts with recent ab-initio calculations. Meanwhile, the integrated intensity of ESR signals, which is proportional to the spin susceptibility, decreases gradually by lowering the temperature and becomes constant at 25 K (Fig. 2). This behavior suggests that spin singlet is gradually created by lowering the temperature, but some spins remain below 25 K. We suppose the lack of the interlayer interaction is due to the long distance between the residual spins. Detailed analysis of the ESR measurements will be presented and discussed.


Fig. 1. Angular dependence of linewidth at 4 K for B//ab-plane.

Fig. 2. Temperature dependence of the ESR integrated intensity for B//b*-axis.