ESR Studies of the Molecular Spin Liquid System EtMe$_3$Sb[Pd(dmit)$_2$]$_2$

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The magnetic properties of the molecular spin liquid system EtMe$_3$Sb[Pd(dmit)$_2$]$_2$, where dmit denotes for 1,3-dithiol-2-thione-4,5-dithiolate, is studied by ESR measurements. The metal dithiolene complexes X[Pd(dmit)$_2$]$_2$, where X is a monovalent cation, show a variety of ground states at low temperature depending on the cation X. [1] These salts are basically a Mott insulator that forms a two-dimensional triangular lattice, and most of them show an antiferromagnetic order at low temperature. Meanwhile, other interesting ground state such as a non-magnetic charge ordered state is observed for the X=Et$_2$Me$_2$Sb salts, and a valence bond solid state is also observed for X=EtMe$_3$P salts. On the other hand, the title compound: EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ does not show any long range order down to 30 mK, and it is considered as a candidate material for the quantum spin liquid state. [2] Although the specific heat, thermal conductivity and magnetic torque measurements show a gapless excitation, NMR indicates a gapped-like behavior. [2-4] Moreover, the triangular lattice of this salt is strongly distorted at low temperature, and the nature of the ground state in this compound is still unclear.

Therefore, to have more microscopic information about the ground state of this salt, we have performed X-band and High-field ESR measurements. As shown in the figure below, a relatively large g-anisotropy was observed due to the d-orbit of Pd, and we have also observed the splitting of two EPR spectra coming from two different layers in which the stacking direction of the dimer of Pd(dmit)$_2$ is different. The splitting of the EPR signals suggests that the interlayer exchange interaction is infinitesimal. Moreover, a g-shift is observed for B//c-axis in the temperature region where a crossover behavior is observed in the specific heat measurement. [3] Detailed ‘in-plane’ angular dependence and the temperature dependence will be presented, and its characteristic ESR behavior will be discussed in contrast with other [Pd(dmit)$_2$]$_2$ salts that have different ground state.

References