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Thermodynamic Properties of Non-ordered Spin States in Molecular Compounds with Geometric Frustration

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We present unusual low-temperature thermodynamic properties of two-dimensional organic triangle lattice compounds by single crystal calorimetry technique. The target compounds of this study are κ -(BEDT-TTF)₂Cu₂(CN)₃, EtMe₃Sb[Pd(dmit)₂]₂ and κ -H₃(Cat-EDT-TTF)₂, which are known as typical dimer Mott insulators with strong electron correlations. The absence of any kind of long-range orders down to dilution temperature range was confirmed by heat capacity measurements. Instead, the low temperature heat capacity shows a typical feature explained by gapless excitations due to the formation of a liquid-like ground state. The scaling between the electronic heat capacity coefficients and magnetic susceptibilities with Wilson ratio's close to ~ 1 was observed, which means that the spin degrees of freedom of localized pair-electrons have strong quantum fluctuations and carries entropy at nearly zero energy region like a Fermi liquid state. These features are common in three compounds, though the structures and the magnitude of transfer energies between dimers are different with each other.

We also discuss chemical pressure and magnetic fields effects in the low energy excitations. Throughout the detail studies of X[Pd(dmit)₂]₂ of which counter-cation sites expressed as X are chemically controlled by making mixed crystals of different cations, we succeeded to derive information on the systematic variation of electronic ground states. We observed that the spin-liquid state exists as a distinct phase and a kind of quantum phase transition to AF and CO phases occurs with some criticality peculiar for spin liquids. The relation with spin properties coupled with other degrees of freedoms is discussed.

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