Low Temperature Heat Capacity of Quantum Spin-Liquid State in Molecular Crystals
\(\text{EtMe}_2\text{Sb}[\text{Pd(dmit)}_2]_2\) and \(\kappa-(\text{BEDT-TTF})_2\text{Cu}_2(\text{CN})_3\)

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In the case of a planar antiferromagnetic triangular lattice consisting of \(S = 1/2\) spins, the geometric frustration prohibits the formation of ordering and therefore a liquid-like ground state is expected. Since it is considered as a macroscopic appearance of a quantum mechanical character, the physics of the spin-liquid has been discussed as one of the central topics of condensed matter science in the last thirty years \[1\]. By our single crystal calorimetry measurements for dimer-based organic charge transfer salt of \(\kappa-(\text{BEDT-TTF})_2\text{Cu}_2(\text{CN})_3\) down to the dilution temperatures, a \(T\)-linear heat capacity was clearly observed of which electronic heat capacity coefficient (\(\gamma\)) was about 15 m\(\text{JK}^{-2}\text{mol}^{-1}\). One of the characteristic feature suggested by the RVB (Resonating Valence Bond) model explaining the formation of spin-liquid state is the existence of such linearly temperature dependent behavior in heat capacity. The degeneration of energy state should give a gap-less excitations from the ground state like a Fermi liquid state in usual metals \[2\]. The present thermodynamic result seems to be consistent with the Fermionic excitations as are suggested by the RVB like scenario. However, we have observed another peculiar thermodynamic character which predicts a kind of crossover behavior into the quantum spin-liquid state. This is an unexpected result which has been unveiled by the thermodynamic measurement. The discussion is also expanded to another organic triangular system of \(\text{EtMe}_2\text{Sb}[\text{Pd(dmit)}_2]_2\) \[3\], which also has a \(T\)-linear term expressed by \(\gamma = 20\text{ mJK}^{-2}\text{mol}^{-1}\) in its low temperature heat capacity. The characteristic features in the spin-liquid state observed at extremely low temperatures and an importance of thermodynamic measurements to understand the ground state of quantum system are discussed.