Thirty-Year Anniversary of κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$: Reconciling the Spin Gap in a Spin-Liquid Candidate

Andrej Pustogow

Institute of Solid State Physics, TU Wien, 1040 Vienna, Austria, e-mail: pustogow@ifp.tuwien.ac.at

In 1991, the Argonne group led by Jack Williams [1] reported the first synthesis of κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$. Although, originally, the focus was on the superconducting properties under pressure, this frustrated Mott insulator with a triangular lattice (Fig. 1c) has been the most promising quantum-spin-liquid candidate for almost two decades [2], widely believed to host gapless spin excitations down to $T = 0$ [3]. The recent observation of a spin gap by the Stuttgart group [4] rules out a gapless spin liquid with itinerant spinons and puts severe constraints on the magnetic ground state. Here I evaluate magnetic, thermal transport, and structural anomalies around $T^* = 6$ K [5]. The opening of a spin gap yields a rapid drop of spin susceptibility [4], NMR Knight shift [6] (Fig. 1b), spin-lattice relaxation rate, and $\mu$-SR spin fluctuation rate, but is often concealed by impurity spins [5]. The concomitant structural transition at $T^*$ manifests in thermal expansion (Fig. 1a,d) [7], THz phonons and $^{63}$Cu NQR relaxation. Based on the field dependence of $T^*$, a critical field of order 60 T (Fig. 1e) is estimated for the underlying spin-singlet state [5]. Overall, the physical properties are remarkably similar to those of spin-Peierls and valence-bond-solid phases. Thus, a strong case is made that the ‘6K anomaly’ in κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ is the transition to a valence-bond-solid state and it is suggested that such a scenario is rather the rule than the exception in materials with strong magnetic frustration [5].

References
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