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

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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 μ-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].

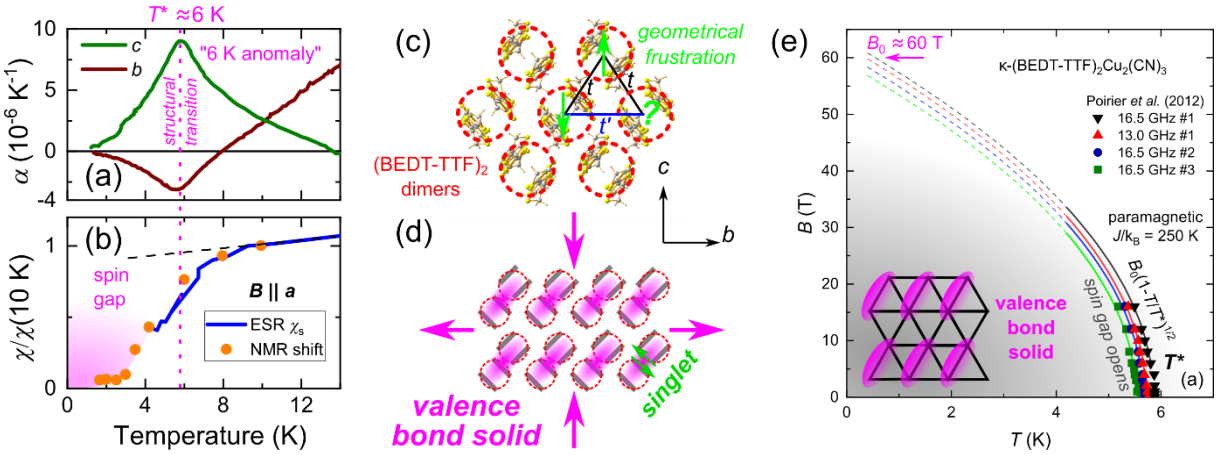

Fig. 1. (a) Anisotropic structural changes below T^* = 6 K were revealed by thermal expansion experiments [7]. (b) Drop of spin susceptibility determined from ESR and NMR data upon spin gap formation [4,6]. (c,d) The frustrated triangular lattice is anisotropically deformed at T^*, which is typical for a valence-bond-solid transition [5]. (e) The magnetic field dependence extrapolates to $B^* \approx 60$ T. Panels (a,b,c,e) taken from Ref. [5].

References

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