Phase transition and lattice distortion in the proposed spin-liquid system κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

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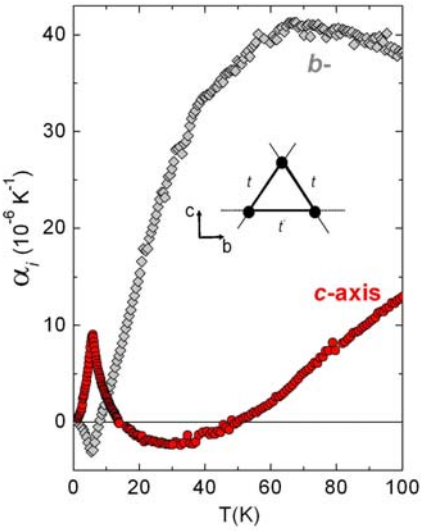
The charge-transfer salt κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ is a half-filled spin S = ½ Mott insulator on a two-dimensional triangular lattice. The material has been considered as a promising candidate for a quantum spin-liquid state owing to its high degree of magnetic frustration ($t'/t \sim 1$) and the absence of long-range magnetic order down to very low temperatures [1].

Issues of high current interest include the character of the low-lying elementary excitations, particularly whether or not there is a spin gap [2, 3], as well as the nature of the mysterious anomaly around 6 K [2, 3]. Here we report high-resolution thermal expansion and specific heat measurements focusing on the 6 K anomaly and the state above. The main observations are sharp and strongly anisotropic peaks in the uniaxial expansivities at 6 K, which lack any hysteresis upon cooling and warming, indicative of a second-order phase transition with a strong coupling to the lattice degrees of freedom. The phase transition is preceded by a wide temperature range characterized by a pronounced temperature-dependent in-plane lattice distortion, see the figure, i.e., an anomalously small *c*-axis expansivity which even turns negative below 50 K, and a large positive *b*-axis expansion coefficient. Our data demonstrate that cooling towards the 6 K transition is accompanied by a significant increase in the t'/t ratio.



Uniaxial expansion coefficient along the in-plane *b*- and *c*-axis of the title compound.

[1] Y. Shimizu et al., Phys. Rev. Lett. **91**, 107001 (2003)
[2] S. Yamashita et al., Nature Phys. **4**, 459 (2008)
[3] M. Yamashita et al., Nature Phys. **5**, 44 (2009)