Glass-like transitions in the frustrated charge systems θ-(BEDT-TTF)$_2$X revealed by thermal expansion measurements

Y. Saito,[1] T. Thomas, [1] Y. Agarmani, [1] T. Thyzel, [1] M. Lonsky, [1] K. Hashimoto, [2, 3] T. Sasaki, [3] J. Müller[1], and M. Lang[1]

[1]Institute of Physics, Goethe University Frankfurt, 60438 Frankfurt (M), Germany, E-mail: saito@physik.uni-frankfurt.de
[2]Department of Advanced Materials Science, University of Tokyo, 277-8561 Chiba, Japan
[3]Institute for Materials Research, Tohoku University, 980-8577 Sendai, Japan

Geometrical frustration causes degenerate states, giving rise to intriguing quantum phenomena such as a quantum spin liquid. In addition to a frustrated spin system, a frustrated charge system is proposed in θ-type (BEDT-TTF)$_2$X salts where the ratio of inter-site Coulomb interactions within their triangular lattice is close to unity [1, 2]. It is expected that charge ordering is suppressed and may be possibly replaced by a charge-glass state. θ-(BEDT-TTF)$_2$*MM'*(SCN) $_4$ (*MM'* = CsZn, CsCo, and RbZn) are good candidates to investigate the frustrated charge system: whereas the CsZn and CsCo salts exhibit no charge-ordered transition, slowly-cooled RbZn salt shows the transition at around 200 K. On the other hand, rapidly-cooled RbZn crystals do not exhibit this transition. As in these salts, the charge-order transition accompanies a structural transition, investigations of their elastic properties in both the charge-ordered and non-charge-ordered states are of fundamental interest. For that purpose, we performed thermal expansion measurements that are sensitive to lattice changes. For the CsZn and CsCo salts the thermal expansion coefficient $\alpha = L^{-1}dL/dT$ reveals evidence for a glassy transition with a pronounced hysteresis at 90-100 K [4] (see Fig. 1 for the CsZn salt). This behavior is reminiscent of the freezing of the ethylene end-groups of the BEDT-TTF molecules with activation energies similar to those of κ-type salts [3, 4]. For the slowly-cooled RbZn salt we observe a metal-insulator transition at 210 K and also an ethylene-group related glass-like transition at 80-100 K. Therefore, the glass-like anomaly around 90 K appears as a common feature in these θ-type salts regardless of whether long-range charge ordering exists or not. Moreover, we find yet another glassy transition at 120-130 K for the CsZn and CsCo salts where the development of a superlattice structure was reported [1]. These results point to the importance of the lattice degrees of freedom in the frustrated charge system.

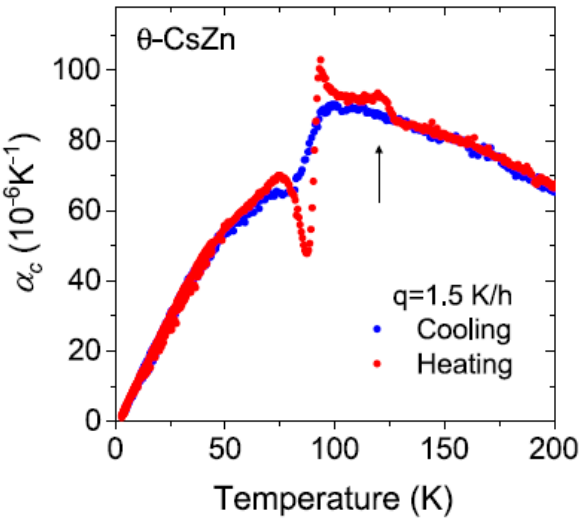

Fig. 1. Thermal expansion coefficient $\alpha(T)$ of θ-CsZn along the *c* axis upon heating and cooling at a rate ±1.5 K/h. The arrow indicates 120 K anomaly. [4]

References

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