Magnetic Superconductor Based on BETS Radical Cation Salt with Polymeric Dicyanamidomanganate (II) anion: κ-$(BETS)_2Mn[N(CN)_2]_3$

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The anion dicyanamide (dca) complexes of divalent first-row transition metals are specified by a wide variety of polymeric structures and are of interest as magnetic counter ions for synthesis of bifunctional materials based on radical cation salts combining conducting and magnetic properties. However, so far the magnetic dca complexes were little used to attain these purposes. In this talk, we present the synthesis, structure and properties of the radical cation salt based on π-donor BETS with paramagnetic tri(dicyanamido)-manganate (II) anion: $(BETS)_2Mn[N(CN)_2]_3$ which exhibits a metal-insulator transition at $T_{MI} \approx 21$ K and becomes superconducting near 5 K under a moderate pressure of ~ 0.3 kbar. The crystal structure of the salt is characterized by the alternation of κ-type organic radical cation layers with layers composed of polymeric $Mn[N(CN)_2]_3$ anions along the *a* axis. In an anion layer, each Mn^{2+} ion with S = 5/2 has an octahedral environment and is linked with six neighboring metal ions via dca bridges. X-ray structure analysis showed that the BETS ethylene groups are disordered at room temperature and continuously order on cooling, without any indication on characteristic temperature of ordering; at 90 K a completely ordered staggered conformation is detected. Around 102 K there is a phase transition associated with an incommensurate superstructure formation: below 102 K X-ray diffraction patterns show weak superstructure reflections which can be described by an incommensurate wave vector q = 0.42b*. The superstructure is retained down to 15 K. There is a relationship between the occurrence of the superstructure near 100 K and a behavior of the interlayer resistance, which exhibits a distinct anomaly in the derivative and a hysteresis in the range of 80-100 K.

Magnetic susceptibilty violates the Curie-Weiss law at $T < 10$ K due to strong antiferromagnetic interactions in the Mn^{2+} lattice. Although ^1H NMR data does not have clear indications of a long range magnetic order, the temperature dependences of the linewidth and spin-lattice relaxation rate suggest a formation of spin glass in the frustrated Mn^{2+} lattice with an onset temperature close to $T_{MI.}$ This fact implies a significant role of π-d interaction in the salt.