

Symmetry-breaking phase transitions, dielectric and magnetic properties of pyrrolidinium-tetrahalidocobaltates

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We report the physicochemical characteristics of novel Co-based pyrrolidinium analogs: $(C_4H_{10}N)_2CoCl_4$ (**PCC**) and $(C_4H_{10}N)_2CoBr_4$ (**PCB**). Both compounds consist of a zero-dimensional (0D) anionic network and disordered pyrrolidinium cations. The structural origin of the anomalies observed in the electrical measurements has been confirmed by DSC thermograms and X-ray analysis. The crystals under investigations undergo a complex sequence of phase transitions: **PCC** at 347/353 K (phase I \leftrightarrow phase II) and 202/257 K (II \leftrightarrow III) and **PCB** at 380/381 K (I \leftrightarrow II) and 256/302 K (II \leftrightarrow III). The molecular motions of cations in both compounds were analysed based on proton magnetic resonance (1H NMR and M_2) measurements, while their magnetic properties were investigated in DC and AC modes. The DC data show magnetic anisotropy, which was confirmed by high-field HF EPR measurements. The AC susceptibility data of **PCC** reveal slow magnetic relaxation at the applied DC field with two relaxation channels. Given the above this organic–inorganic hybrid can be considered as a rare example of multifunctional materials which exhibit dielectric and magnetic activity.

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