Unusual electronic and magnetic properties in cobaltites with low dimensionality

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The cobaltites properties depend on the cobalt oxidation- and spin-states. Combined to the low dimensionality and geometrical frustration, this gives rise to complex physical properties. In the case of the 1D Ca₃Co₂O₆ compound, the "Co-O" chains are made of alternating face-sharing octahedra (*oct.*) and trigonal biprisms (*bip.*). The ferromagnetic (F) chains separated by the calcium are set on a triangular lattice and, due to the interchain antiferromagnetic (AF) coupling, Ca₃Co₂O₆ can be viewed as a triangular Ising AF system. In the chains, low-spin (*LS*) and high-spin (*HS*) Co³⁺ cations alternate in the *oct.* and *bip.*, respectively. This 1D system exhibits remarkable magnetization (M) plateaus, at low T, with one plateau corresponding to the ferrimagnetic (FI) configuration of the chains on the triangle. Interestingly, the *oct.* coordinated *LS* cations can be totally or partially substituted by either magnetic (Ir⁴⁺, Rh⁴⁺, S = 1/2) or diamagnetic (Sc³⁺: 3d⁰) cations. For both Ca₃Co₂O₆ and Ca₃CoRhO₆, a 'partially disordered AF' state is realized, but, for Ca₃CoIrO₆ the M plateau at M_{saturation}/3 (FI) is suppressed. The properties of these materials (M, ρ , Seebeck) will be compared to those of Sr₃NiIrO₆. In the latter, the intrachain coupling between Ni²⁺_{bip.} (S=1) and Ir⁴⁺_{oct.} (S = 1/₂) is AF contrasting with the F ones in Ca₃CoMO₆ (M = Co, Ir, Rh). The low-T magnetic properties of the Sr₃NiIrO₆ compound show a spin disordered state similar to that of the Ca₃CoMO₆ compounds (left fig.).

The 'misfit' cobaltites possess 2D structures with two aperiodic substructures built of rocksalttype separating layers sandwiching CdI₂.type CoO₂ layers. The latter is made of edge-shared CoO₆ octahedra leading to an unusual coexistence of LS $\text{Co}^{3+}/\text{Co}^{4+}$ cations. In this hexagonal layer, the cobalt form a triangular plane and due to the rhombohedral crystalline-field, the t_{2g} orbitals are believed to be splitted in two subbands with heavy and light holes responsible for metallicity and large thermoelectric power (TEP), respectively. The structural features of new misfit cobaltites will be shown and discussed in connection to their magnetoresistance and magneto-TEP properties (right fig., from *A.Maignan,S. Hébert, M.Hervieu, C. Michel, D. Pelloquin and D. Khomskii, J. Phys.: Condens. Matter, 15 (2003) 2711*).



