Complex magnetic order in quasi-one-dimensional compound \( \text{Ca}_3\text{CoIrO}_6 \)

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In the compound with the general formula \( \text{Ca}_3\text{B}_{\text{Co}}\text{O}_{6} \) (\( \text{B} = \text{Co}, \text{Rh}, \) or \( \text{Ir} \)), the crystal structure is made up of \( \text{CoBO}_{6} \) linear chains parallel to the \( c \)-axis, spaced apart by \( \text{Ca}^{2+} \) ions which fill the space between them. The chains themselves are made up of alternating face-sharing pairs of blocks: \( \text{CoO}_{6} \) trigonal prisms and \( \text{BO}_{6} \) octahedra. The chains form a hexagonal array in the \( ab \) plane so each chain is surrounded by six other structurally identical chains. For \( \text{Ca}_3\text{Co}_{2}\text{O}_{6} \), the spins of \( \text{Co}^{3+} \) ions in the prism tend to order ferromagnetically (FM) along the chain, while the interchain interaction is weaker and favors antiferromagnetic (AF) order. Note that the \( \text{Co}^{3+} \) ions in the octahedron are non-magnetic, because they are in a low-spin state. The balance of these competing interactions gives rise to interesting magnetic properties, such as the partially disordered AF (PDA) state below 28 K, while short-range AF order persists up to \( \sim 100 \) K \([1,2]\).

According to a very recent \( \mu^{+}\)SR experiment on \( \text{Ca}_3\text{CoRhO}_{6} \) \([3]\), although the charge distribution in the chain changes to \( \text{Co}^{2+} \) and \( \text{Rh}^{4+} \), the compound exhibits essentially the same magnetic transitions; that is, short-range AF order appears below \( \sim 180 \) K, the PDA state completes below \( \sim 90 \) K, and finally the ferrimagnetic state emerges below \( \sim 20 \) K. The increase in the transition temperatures compared with those of \( \text{Ca}_3\text{Co}_{2}\text{O}_{6} \) suggests the effect of the wide spread \( 4d \)-orbitals of \( \text{Rh}^{4+} \) ions.

In order to determine whether the magnetic behavior is similar for \( \text{Ca}_3\text{CoIrO}_{6} \), we have measured a \( \mu^{+}\)SR spectrum using a polycrystalline sample. We found that short-range AF order appears below \( \sim 300 \) K, while long-range order sets in below \( \sim 110 \) K. Moreover, the ZF spectrum exhibits a very clear oscillation (at \( \sim 50 \) MHz) below 100 K, but its volume fraction is only \( \sim 20\% \) even at 1.8 K. Since the oscillation for \( \text{Ca}_3\text{Co}_{2}\text{O}_{6} \) and \( \text{Ca}_3\text{CoRhO}_{6} \) is strongly damped, and eventually only the first minimum is seen in the ZF-spectrum, the magnetic nature of \( \text{Ca}_3\text{CoIrO}_{6} \) is likely to be rather different from that of the others.