Muon Spin Relaxation on Layered Cobaltites $RBaCo_2O_{5.5}$

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The $RBaCo_2O_{5+x}$ system possesses an extremely rich structural, electronic and magnetic phase diagram. The properties of these layered perovskites are controlled by the oxygen concentration and the size of the rare earth ion R. Particularly interesting are the compounds with $x = 0.5$ where the Co ions nominally possess a valence of 3+. In these materials the Co^{3+} ions have two different environments, namely CoO_5 square pyramids and CoO_6 octahedra which alternate along one of the crystallographic axes. Since the crystal electric field (CEF) splitting of the Co $3d$-states is of the same order of magnitude as the Hund's exchange coupling, the Co ion may exist in three different spin states (low-spin state (LS), intermediate-spin state (IS) and high-spin state (HS)). The spin-state of the Co ion is determined by their coordination due to the different CEFs in the octahedral and pyramidal and by the temperature. The thermally driven spin-state transitions (SST) provide the background to a metal-insulator transition at higher temperatures and to three different magnetic phases below room temperature. The complexity of the magnetic system arises from the fact that the SST may be accompanied by both spin-structure and spin-state ordering (SSO) transitions.

Here, we report magnetic structures of $RBaCo_2O_{5+\delta}$ with $\delta \approx 0.5$ determined by means of muon spin relaxation (μSR). Our main result is that a homogenous FM phase with ferrimagnetic SSO of IS and HS states develops through two first order phase transitions into phase separated AFM1 and AFM2 phases with different types of antiferromagnetic SSO. We argue, that the SSO and the phase separation play a similar role as intrinsic inhomogeneities like doping do in cuprates and manganites. The specific SSO in this cobaltite is also responsible for its unusual transport properties.