Paramagnetic nature of the layered cobalt dioxide with a double rocksalt-type block

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Layered cobalt dioxides with a triple or a quadruple rocksalt-type block, such as ($[A_2BO_3]_z^{RS}[CoO_2]$ and $[A_2B_2O_4]_z^{RS}[CoO_2]$, where A stands for Ca, Sr, and/or Ba, B for Tl, Bi, Pb, Co, and/or Cu, and RS for a rocksalt-type block, exhibit magnetic transitions due to the intra- and/or inter-plane interaction of the CoO_2 plane. Thanks to the power of μ^+SR, we found that $[Ca_2CoO_3]_{0.62}^{RS}[CoO_2]$ undergoes a transition from a high-T paramagnetic state to an incommensurate spin density wave (IC-SDW) state with short-range order at ~ 100 K [1]. As T decreases from 100 K, short-range order develops, long-range IC-SDW order completes at 30 K, and finally a ferrimagnetic state appears below ~ 19 K. Similar magnetic transitions were also found for $[Ca_2Co_{4/3}Cu_{2/3}O_4]_{0.62}^{RS}[CoO_2]$ [2].

Although the layered cobalt dioxides with a double RS were not prepared so far, very recently $[Ca_{0.85}OH]_{1.16}^{RS}[CoO_2]$ has been synthesized using a high-pressure technique at 6 GPa [3]. According to susceptibility (χ), specific heat (C_p), resistivity, and thermopower measurements, $[Ca_{0.85}OH]_{1.16}^{RS}[CoO_2]$ is assigned as a Curie-Wess insulator, in spite of the effort for hole-doping. Furthermore, the detailed analysis of the $\chi(T)$ and $C_p(T)$ curve suggests the existence of a small transition at 13 K ($=T_m$). In order to clarify the origin of T_m, we have measured μ^+SR spectra for $[Ca_{0.85}OH]_{1.16}^{RS}[CoO_2]$ using a pressed powder sample.

The ZF-μ^+SR spectrum at 1.8 K is well fitted by a static Kubo-Toyabe function due to a nuclear magnetic field of ^{59}Co and ^{1}H, together with a slow cosine oscillation caused by a formation of μ^+-OH bond. The volume fraction of the former signal is about 80%, whereas the latter approximately 20%. Furthermore, all the μ^+SR parameters, such as the field distribution width (Δ), the two asymmetries (A_{KT} and $A_{\mu-OH}$), and the oscillation frequency due to the μ^+-OH bond, are independent of T both above and below T_m. This is a clear indication that T_m is not magnetic but structural and/or electronic in origin.

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