

## Spin-State Transition in $R\text{CoO}_3$ ( $R = \text{La}, \text{Pr}, \text{ and Nd}$ ): Single-Crystal $^{59}\text{Co}$ NMR Measurements

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A thermally-induced transition from a nonmagnetic to a paramagnetic state, so-called spin-state transition, in  $R\text{CoO}_3$  ( $R$ : rare earth) is a long-standing issue of strongly correlated electron system. It has attracted much interests whether the transition from a low-spin ( $S = 0$ ) to a high-spin ( $S = 2$ ) state takes place through an intermediate spin ( $S = 1$ ) state. We address a microscopic study of the orbital state in  $R\text{CoO}_3$  ( $R = \text{La}, \text{Pr}, \text{ and Nd}$ ) by single-crystal  $^{59}\text{Co}$  NMR measurements. The  $^{59}\text{Co}$  nuclear spin-lattice relaxation rate and Knight shift measurements revealed the spin-state transition with the critical slowing down of fluctuations at 50 K in  $\text{LaCoO}_3$ . On the other hand,  $\text{PdCoO}_3$  and  $\text{NdCoO}_3$  exhibit no clear transition but a small continuous change in the local spin susceptibility at high temperatures above 300 K. Subtracting the contribution from the magnetic rare-earth spins to the  $^{59}\text{Co}$  Knight shift, we obtained the magnetic hyperfine coupling constant between a  $^{59}\text{Co}$  nuclear spin and  $3d$  spins in the paramagnetic state with the finite  $3d$  spin susceptibility in  $\text{PrCoO}_3$  and  $\text{NdCoO}_3$ . The hyperfine coupling tensors of  $\text{PrCoO}_3$  and  $\text{LaCoO}_3$  are axially symmetric and dominated by a sum of dipole hyperfine fields from the  $3d$  orbitals theoretically expected in the high-spin state.