Study of spin-state polarons in LaCoO₃ with diluted impurity by magnetization and heat capacity measurements

Y. Kubota, K. Tomiyasu*, and K. Iwasa Department of Physics, Tohoku University, Sendai 980-8578, Japan * tomiyasu@m.tohoku.ac.jp

The perovskite-type LaCoO₃ is a nonmagnetic insulator with low spin state (Co³⁺, d^6 , S = 0) below about 100 K. By lightly doping Sr²⁺ (hole) at La³⁺ sites into this system, colossal magnetic moment over 10µ_B/hole appears [1]. A ferromagnetic spin cluster, in which six nearest-neighbor Co³⁺ ions change to intermediate spin state (S = 0) around Co⁴⁺ with a hole, probably generates the moment (spin-state polaron model) [2]. This model is based on ferromagnetic double exchange interaction, caused by e_g orbital electrons of intermediate spin state in the polaron.

In this study, to examine the double exchange mechanism, we comparatively studied other analogue systems La(Co_{0.99} $M_{0.01}$)O₃ (M = Cr, Mn, Fe, Ni). In particular, the double exchange should be forbidden for Fe³⁺ (d^5 , S = 5/2) doping because its e_g orbital of majority spin is fully occupied.

Magnetization and heat capacity measurements were performed by MPMS (Tanigaki group in Tohoku Univ.) and PPMS (muon group in KEK), respectively. Polycrystalline samples of LaCoO₃, $(La_{0.99}Sr_{0.01})CoO_3$, and $La(Co_{0.99}M_{0.01})O_3$ (M = Cr, Mn, Fe, Ni) were synthesized in the standard solid-state reaction method.

Figure 1 shows the magnetic field dependence of magnetic moment per a doped ion (0.01 formula unit) at 1.8 K. All the doped systems exhibit the values over $5\mu_B$ at 7 T, the maximum value of *d* electron systems. Surprisingly, in the Fe doped system, the value is as large as $18 \mu_B$ /hole. Thus, the double exchange is not the only mechanism to form the polaron.

Figure 2 shows the temperature dependence of heat capacity of Cr and Fe doped systems. A Shottky anomaly appears from the low temperature side with increasing the magnetic field, suggesting that the spin polaron states are degenerated as ground states in zero field and are split by external field. On the other hand, the excited state with 0.8 meV is directly observed in Sr doped system by inelastic neutron scattering [2]. The interpretation is in progress.

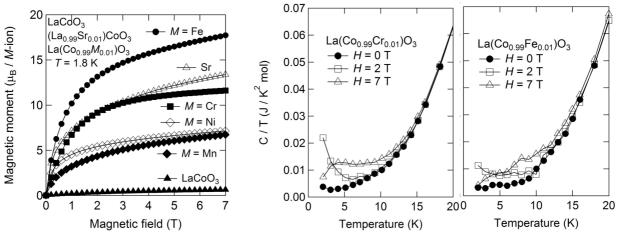


Fig. 1: Magnetization process at 1.8 K. Fig. 2: Heat capacity in zero and non-zero magnetic field.
[1] S. Yamaguchi *et al.*, PRB **53**, 6 (1996). [2] A. Podlesnyak *et al.*, PRL **101**, 247603 (2008).