Spin-State Polarons in Lightly Hole-Doped $LaCoO_3$

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Physical properties of nanostructured magnetic materials are extensively studied because of their fundamental interest and potential applications. A naturally occurring analog to the artificially fabricated heterostructures are hole-doped cobaltites $La_{1-x}Sr_xCoO_3$ with intrinsic inhomogeneities, i.e. with a spatial coexistence of magnetic clusters in a nonmagnetic matrix.

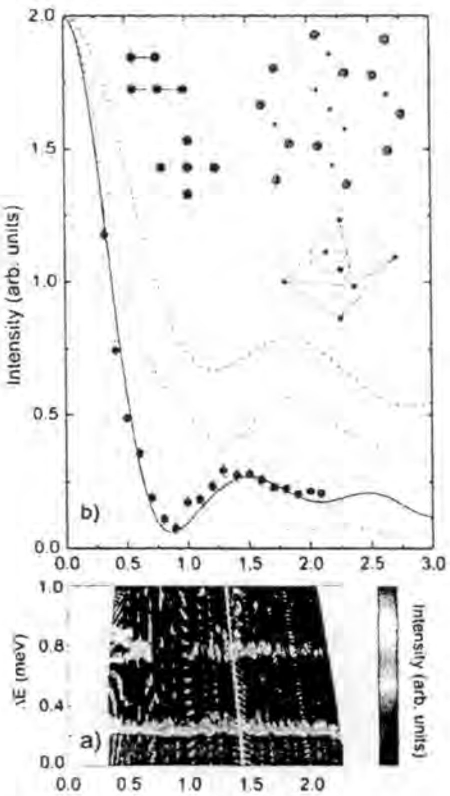

Fig. 1. a) Excitation INS spectrum from $La_{0.998}Sr_{0.002}CoO_3$ at $T = 1.5$ K. b) Circles: experimental Q dependence of the intensity of the peak observed at 0.75 meV. Lines: Calculated Q dependence of the neutron cross section for different Co multimers (visualized in the Figure) in the cubic perovskite lattice of $LaCoO_3$ and for |Si> -> |Si> transitions

In this work, we elucidate the mechanism of how already the light hole doping $x \sim 0.002$ dramatically affects magnetic properties of $LaCoO_3$. Using inelastic neutron scattering (INS) data, obtained with and without external magnetic field, we find that the charges introduced by substitution of Sr^{2+} for La^{3+} do not remain localized at the Co^{4+} sites. Instead, each hole is extended over the neighboring Co^{3+} ions, transforming them to higher spin state and thereby forming a magnetic seven-site (heptamer) polaron. Spin-state polarons behave like magnetic nanoparticles embedded in an insulating nonmagnetic matrix. The present data give evidence for two regimes in the lightly hole-doped samples: i) $T < 35$ K dominated by spin polarons; ii) $T > 35$ K dominated by thermally activated magnetic Co^{3+} ions. Additional charge carriers increase the number of such spin-state polarons, which form a percolative network resulting in a metallic state with long-range ferromagnetic order at the critical concentration $x_c = 0.18$.