Observation of “Spin Blockade” in a Bulk Single Crystal

– Electron-Spin-Controlled Charge Conduction in a Layered Cobalt Oxide –

Background

Recently, layered cobalt perovskite oxide $R\mathrm{BaCo_2O_{5+x}}$ (R is a rare-earth ion) has attracted a lot of attention owing to such fascinating features as the spin-state and metal-insulator transitions, charge and orbital ordering phenomena, and giant magnetoresistance. This material belongs to the family of strongly-correlated electron materials where the Coulomb interactions play important roles, and a central problem of the current condensed matter physics is to understand the various novel physical properties (such as high-temperature superconductivity and giant magnetoresistance) that are found in those strongly-correlated electron materials. As a result of our research efforts on this front, we have previously succeeded in growing high-quality single crystals of $\mathrm{GdBaCo_2O_{5+x}}$ (GBCO, Fig.1) and established a complicated electronic phase diagram in the whole available doping range, providing a basis for general understanding of charge carrier transport in this interesting compound.

Objectives

Based on the advanced understanding of GBCO, we address the problem of coupling of spin and charge degrees of freedom in strongly-correlated electron materials, by examining the details of the charge transport phenomena in GBCO.

Principal Results

By using high-quality GBCO single crystals in which the oxygen contents are precisely controlled, we have studied detailed doping dependences of resistivity, Hall effect, and thermoelectric power, and obtained the following results:

(1) We have found a remarkable asymmetry between the electron-doped ($x < 0.5$) and the hole-doped ($x > 0.5$) sides manifested in all the transport properties studied (Figs. 2 and 3). In particular, whereas an eventual development of a metallic state is observed for hole doping, the system remains an insulator upon electron doping.

(2) This finding provides strong evidence for the curious quantum-mechanical phenomenon of “spin blockade[*1]” occurring in this material. Due to this effect, the electrons doped to the parent-insulating GBCO ($x = 0.5$) become immobile, which gives rise to a behavior contrasting to that observed for doped holes (Fig.4). The spin-blockade phenomenon has already been observed in nano-scale systems such as quantum dots, but this is the first time that this quantum-mechanical effect is observed in a bulk system where the strong electron correlations are obviously responsible for its occurrence. This result presents a new and interesting example of how the quantum-mechanical phenomena govern macroscopic properties of materials, and hence is of fundamental importance in condensed matter physics.

Future Developments

This study has significantly advanced our understanding of the doping behavior of charge carriers in the $\mathrm{RBaCo_2O_{5+x}}$ system. We plan to further address the origin of large thermoelectric power observed in this class of materials, which may give us a clue to where to search for new promising materials for thermoelectric applications.

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Reference

A. A. Taskin & Yoichi Ando, “Electron-Hole Asymmetry in $\mathrm{GdBaCo_2O_{5+x}}$: Evidence for Spin Blockade of Electron Transport in a Correlated Electron System”, Physical Review Letters **95** (2005) 176603.

[*1] ：Spin blockade is a phenomenon where spatial arrangements of spin states prohibit hopping of electrons to neighbouring sites.

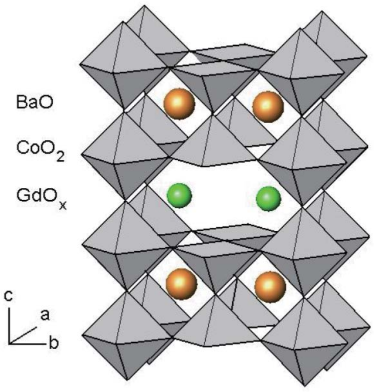

Fig.1 A schematic picture of the crystal structure of GBCO at x = 0.5. At this parent composition, GBCO is an insulator with a well-defined gap. Decreasing x from 0.5 causes electron doping, while increasing x from 0.5 causes hole doping

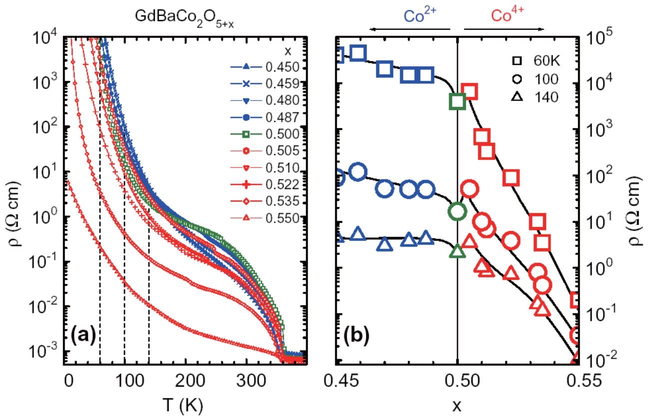

Fig.2 Temperature dependences of the in-plane resistivity ρ(T) of GBCO crystals with oxygen contents close to x = 0.5. (b) Doping dependences of the in-plane resistivity ρ (x) of GBCO crystals at several temperatures (shown by dashed lines in the left panel) ; ρ does not change much for x < 0.5 (electron doping), while it decreases exponentially for x > 0.5 (hole doping)

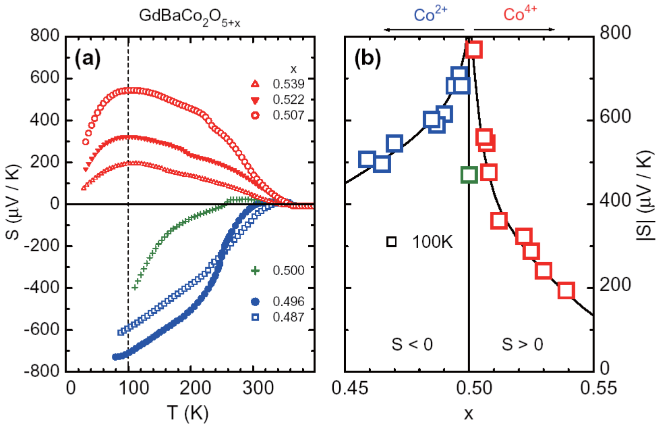

Fig.3 (a) Temperature dependences of the Seebeck coefficient S(T) of GBCO crystals with oxygen contents close to x = 0.5. (b) The doping dependence of the absolute value of the Seebeck coefficient |S(x)| in GBCO at T = 100 K. These data confirm that electrons and holes are indeed doped to GBCO in the regimes of x < 0.5 and x > 0.5, respectively

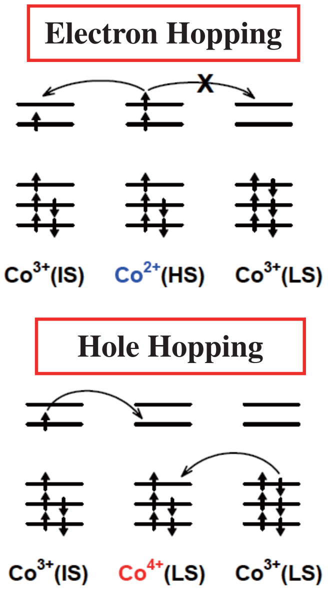

Fig.4 The scheme of electron (Co^{2+}) and hole (Co^{4+}) hopping through the intermediate-spin state (IS) and low-spin state (LS) of Co^{3+} ions, illustrating the phenomenon of the spin blockade for electron hopping. At the parent composition x = 0.5, due to the peculiar crystal structure shown in Fig. 1, IS-Co^{3+} ions and LS-Co^{3+} ions form alternating rows in the CoO_2 planes. When electrons are doped, a part of the Co^{3+} ions turn into Co^{2+} in the high-spin state (HS), but the process of exchanging an electron between HS-Co^{2+} and LS-Co^{3+} is hard to occur due to the mismatch in the spin arrangements (spin blockade). On the other hand, the LS-Co^{4+} ions created upon hole doping can easily exchange an electron with either IS-Co^{3+} or LS-Co^{3+} ions, making it easy for holes to hop around