Self-organization and metallic conduction of Jahn-Teller polarons in perovskite Sr_{1-x}Ce_xMnO₃ single crystals

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Introduction

Double-exchange (DE) model has long been discussed as one of the most fundamental theories on the colossal magnetoresistance phenomena in hole-doped perovskite manganites. Due to the competition between ferromagnetic DE interaction induced by doped carriers and antiferromagnetic superexchange interaction among localized spins, there has still been theoretical controversy over the ground state (especially in the electron doping regime): a canted-antiferromagnetic metal, phase-separated state consisting of a ferromagnetic metal and an antiferromagnetic insulator, and anisotropic chainlike magnetic ordering. In this study, by utilizing a high-pressure (5-6 GPa) synthesis apparatus, we have successfully synthesized single crystals of cubic perovskite SrMnO₃ with controlled electron concentration, which can realize the most ideal (orbital-degenerate) DE system [1]. We here aim to experimentally elucidate its genuine electronic and lattice structures, by systematic transport, magnetic, and synchrotron x-ray diffraction measurements on these crystals.

Results and Discussion

Figure 1(a) displays the temperature profile of resistivity at 0 T for $Sr_{1-x}Ce_xMnO_3$ ($0\le x\le 0.1$) single crystals, respectively. The undoped SrMnO₃ is a G-type antiferromagnetic (AFM) Mott insulator. Ce substitution for Sr by only 0.5-1% (1-2% electron doping), however, makes the system metallic over the whole temperature range. For these compounds, the antiferromagnetic spins slightly cant at low temperatures. With further increasing *x* to 0.02, the metallic ground state is replaced by the insulating one. For *x*=0.03-0.1, a distinct metal-insulator transition shows up. The crystal structure simultaneously changes from cubic to tetragonal with the elongation of c axis [Fig. 1(b)], indicating the $3z^2-r^2$ -type orbital order (with C-type AFM).

The synchrotron powder x-ray diffraction was measured at the Beam Line 8A at the Photon Factory, KEK, Tsukuba. For $x \le 0.01$, the profiles are nicely indexed with the cubic structure (*Pm3m*), while those for $x \ge 0.02$ are indexed with tetragonal one. As shown in Fig. 1(b), the tetragonality (c_p/a_p) rapidly increases just below

 T_{OO} and then saturates toward the lowest temperature. Its value almost linearly increases with increasing x up to x=0.1.

Figure 1(c) summarizes an electronic phase diagram for Sr_{1-x}Ce_xMnO₃ ($0 \le x \le 0.1$), where a G-type AFM metal and a C-type AFM orbital-ordered insulator critically compete. Only ~4% electrons lead to the anisotropic orbital-ordered ground state in the originally isotropic (cubic) metal. This is probably due to the strong Jahn-Teller effect inherent in the degenerate e_g orbitals in the cubic system. In fact, detailed transport measurements have revealed significantly renormalized electron mass (~15 times heavier than band mass) for the adjacent metallic phase, indicating the substantial electron-phonon coupling. Thus, the dichotomous electronic/lattice phases in the lightly-doped DE system can be attributed to the keen competition between the self-organization and itinerancy of the Jahn-Teller polarons.



Fig. 1 Temperature profile of (a) resistivity and (b) tetragonality c_p/a_p for $Sr_{1-x}Ce_xMnO_3$ ($0 \le x \le 0.1$) single crystals. (c) Electronic phase diagram as a function of 2x (electron concentration). $T_N(G)$, T_{CA} and $T_N(C)$ indicate the G-type, canted G-type, and C-type AFM transition temperatures, respectively.

Reference

[1] H. Sakai et al., Phys. Rev. B 82, 180409(R) (2010).

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