Multi-probe study on Charge Transport Transitions of PrBaCo$_2$O$_{5.5+x}$

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Charge Transport Transition (CTT) is a kind of phenomenon demonstrating huge resistivity changes, and has been widely observed in strongly correlated systems. The CTT in RBaCo$_2$O$_{5.5+x}$ can be induced by temperature, pressure and also hole doping. Most of the effort has been focused on the temperature induced MIT of stoichiometric RBaCo$_2$O$_{5.5}$, which is accompanied by an anomaly in inverse magnetic susceptibility. This anomaly is generally attributed to the spin state transition (SST) of Co$^{2+}$ ions. The competition between crystal field (CF), on-site Coulomb correlations, and the intra-atomic exchange energies leads to three possible spin states of Co$^{3+}$ ions: the low spin state (LS, t$_{2g}^6$e$_g^0$), the intermediate spin state (IS, t$_{2g}^5$e$_g^1$), and the high spin state (HS, t$_{2g}^4$e$_g^2$). However, the crystal structure phase transitions coupling with the CTTs in RBaCo$_2$O$_{5.5+x}$ are still in controversy and there is no consensus on the spin states below and above the transition temperature as well. Moreover, no satisfactory mechanism has been established for this system so far.

We conducted a multi-probe study PrBaCo$_2$O$_{5.5+x}$ polycrystalline samples with a combination of diffraction techniques (electron, X-ray and neutron) and X-ray spectroscopies (XPS and XAS). On discovering some weak super lattice peaks, the Pmma ($2a_p × 2a_p × 2a_p$) structure model of PrBaCo$_2$O$_{5.5}$ in insulating phase by single crystal X-ray diffraction study is justified, and a new Pmmm ($a_p × 2a_p × 2a_p$) structure model is constructed for hole doping sample, PrBaCo$_2$O$_{5.5+x}$ in the high-conductivity phase. Through magnetic structure analysis on neutron powder diffraction data, we directly observed the existence of low spin-state Co$^{3+}$ in PrBaCo$_2$O$_{5.5}$ in low-conductivity phase and for the first time found hole doping induced spin state transition in RBaCo$_2$O$_{5.5+x}$ system. It was finally recognized that the CTTs in PrBaCo$_2$O$_{5.5+x}$, induced by both temperature and hole doping, are coupling with the Pmma ($2a_p × 2a_p × 2a_p$) structure transition to Pmmm ($a_p × 2a_p × 2a_p$), which caused by spin-state transition of Co$^{3+}$ ions. Therefore, we propose a common scenario for the CTTs.