Magnetic, Transport and Spectroscopic Properties of $PrBaCo_2O_{5.35}$

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Abstract. Oxygen deficient double perovskites $PrBaCo_2O_{5.35}$ has been studied by XRD, magnetization, resistivity, infra-red (IR) and Raman spectroscopy. Spectroscopic properties reveal that the local structure around Co ions plays an important role in deciding magnetic and transport properties of such perovskites.

Keywords: Layered Perovskites, Magnetic oxides , Infra-red and Raman spectroscopy
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INTRODUCTION

"Non-stoichiometry" and "doping" are important concepts in layered perovskites like $RBaCo_2O_{5+\delta}$ (R=rare earth). "Oxygen engineering" is therefore one of the most important tool for on-demand tailoring functional oxides to get desire properties. Due to wide accessible of oxygen non-stoichiometry and a strong tendency of oxygen ions to order in 112-type cobaltites, one can expect a dependence of structural, transport and magnetic properties not only upon oxygen content itself, but also upon the synthesis history [1]. The spin-state degrees of freedom of Co ions introduce new effects in these narrow band oxides [2]. The existence of several possible spin states in given oxidation state makes cobaltites a rich but challenging system to study. In this paper we report detailed investigations of magnetic, transport and spectroscopic of $PrBaCo_2O_{5.35}$.

EXPERIMENTAL

Polycrystalline sample of $PrBaCo_2O_{5.35}$ was prepared by sol-gel method. The oxygen content of as synthesized sample was reduced by annealing in argon atmosphere followed by ice quenching. X-ray diffraction (XRD) pattern was recorded in the range of $20° \leq \theta \leq 80°$ using Cu Kα radiation. Low temperature (10K – 320K) resistivity measurements were carried out using four-probe method. The magnetization measurements were carried out as a function of temperature M(T) and magnetic field M(H) using a Quantum Design SQUID magnetometer in the temperature range of 10 K to 300 K. IR spectra were recorded on FTIR-8900 Shimadzu spectrophotometer in the temperature range 80K – 350K. HORIBA JOBIN YVON HR 800 with 488 nm laser was employed for Raman spectrometry in the temperature range 80 K – 350 K.

RESULTS AND DISCUSSION

Rietveld refinement of XRD pattern confirmed the formation of single phase sample with 122 type orthorhombic unit cell belonging to *Pmmm* space group. Resistivity exhibits a semiconducting behaviour as shown in Figure 1(a). Change in slop of resistivity curve has been observed around 70K and 300K. M(T) at 100 Oe is depicted in the Figure 1(b**)** , shows large difference between zero field cooled (ZFC) and field cooled (FC) curves indicating complex magnetic ground state. Several magnetic transitions are observed with decrease in temperature in the ZFC curve. Around 300K (T_{R1}) there appears to be weak magnetic transition whose origin is not yet clearly known. This transition is also reflected in the resistivity curve as indicated by an arrow. At T_C = 240K, the sample undergoes paramagnetic (PM) to ferromagnetic (FM) transition followed by a FM to antiferromagntic (AFM) transition at T_N = 180K. Further, at lower temperature two new transitions are observed at 60K (T_{R2}) and 20K (T_{R3}) respectively. These transitions have been associated with structural phase transitions [3]. M(H) curve recorded in the range ±7T just above T_C at 250K (top left inset of Figure 1(b)) shows a narrow hysteresis loop indicating the transition at

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300K to be FM in nature. FM can be attributed to Co^{2+} - O – Co^{3+} super-exchange interaction.

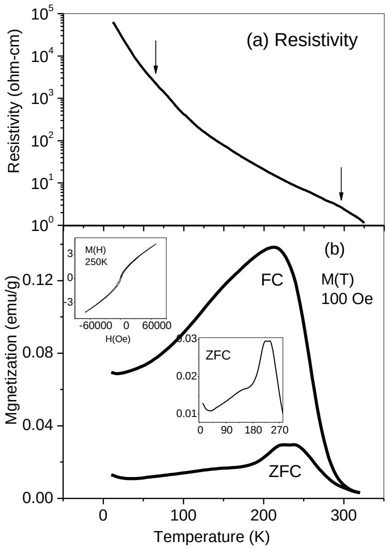

Figure 1. (a) Resistivity (b) Magnetization behavior of $PrBaCo_2O_{5.35}$. Insets show M(H) at 250K and ZFC curve.

Figure 2(a) depicts IR modes due to stretching and bending of Co-O bonds are observed in the range of 400-700 cm^{-1} at 80K.

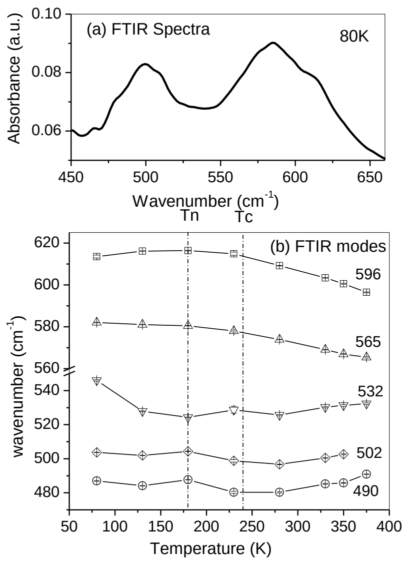

Figure 2 (a) IR spectra of $PrBaCo_2O_{5.35}$ at 80K, (b) temperature evolution of IR modes.

The spectra can be fitted with five Gaussian peaks corresponding to two bending and three stretching modes. The temperature evolution of various mode frequencies is shown in Figure 2(b). Generally IR modes harden with decrease in temperature, but a marked softening of some of the modes is observed near the magnetic transition temperatures. Furthermore, the mode at 502 cm^{-1} disappears at 350K, which can be related to the insulator to metal transition. A typical Raman scattering spectra recorded at 80K is presented in Figure 3(a). The temperature evolution of Raman modes due to Co-O stretching (400-650 cm^{-1}) is shown in Figure 3(b).

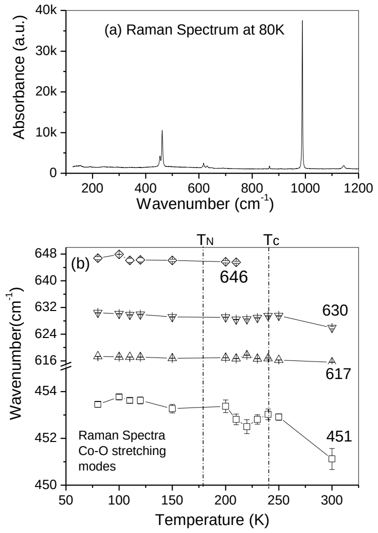

Figure 3 (a) Raman spectra of $PrBaCo_2O_{5.35}$ at 80K, (b) temperature evolution of IR modes.

It can be clearly seen that near T_C there is shift in Raman modes towards lower wavenumbers, while a new mode (646 cm^{-1}) has been observed below T_C.

Therefore from the spectroscopic studies it can be concluded that local structure around Co ions plays an important role in magnetic and transport properties of oxygen deficient $PrBaCo_2O_{5+d}$.

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