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Non-resonant microwave absorption in $Pr_{1-x}Ba_xCoO_{3-\delta}$

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Abstract

In this work we report magnetically modulated microwave absorption experiments performed at 9.4 GHz from 110 up to 300 K on samples of $Pr_{1-x}Ba_xCoO_{3-\delta}$ ($0.10 < x \leqslant 0.50$) which were prepared by the "Pechini" method.

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1. Introduction

The rare-earth cobaltates are always of interest due to their peculiar magnetic and transport properties. These properties change with temperature, upon doping and oxygen stoichiometry [1]. These compounds are also excellent candidates to study thermally induced spin transitions (low, high and intermediate spin) [2]. The complex magnetic properties of these compounds have been mainly studied by means of DC and AC magnetometries. Nevertheless, electron spin resonance—which is a powerful technique to investigate the nature of magnetic phases in magnetic materials at different temperatures [3], and that could be very helpful—has not been used in these studies. In this work we report the basic electrical and magnetic properties of $Pr_{1-x}Ba_xCoO_{3-\delta}$ ($0.10 < x \leqslant 0.50$) cobaltates. A comparison between DC magnetic susceptibility and microwave absorption experiments reveal the complementarity of these two techniques to study complex magnetic materials and in the case of the $x = 0.4$ sample the coexistence of two closely related magnetic phases with different Curie temperature in the $Pr_{0.6}Ba_{0.4}CoO_{3-\delta}$ compound.

2. Experimental

Powder samples were prepared by the "Pechini" method as described elsewhere [4]. The final heating temperature was 1000/54 h or 1100°C/72 h in air, depending on the samples. The samples were characterized by powder X-ray diffraction using a Siemens D-5000 diffractometer and Cu (K_α) radiation. Electron spin resonance (ESR) spectra were measured at 9.4 GHz (X-Band) between 110 and 300 K with an EMX Bruker spectrometer. DC magnetic properties were studied in a SQUID Quantum Design magnetometer. Zero-field-cooling (ZFC) and field-cooling (FC) magnetic susceptibility data were obtained from 5 to 300 K at 1000 Oe. The DC four-probe electrical resistivity, ρ, was measured as a function of temperature in the range $78 \leqslant T(\mathrm{K}) \leqslant 300$ using a homemade device.

3. Results and discussion

According to the X-ray diffraction results, we have obtained single-phase materials for $0.10 < x \leqslant 0.50$. The average particle size of these polycrystalline materials, d is 1–2 μm, as shown by SEM micrographs. The X-ray

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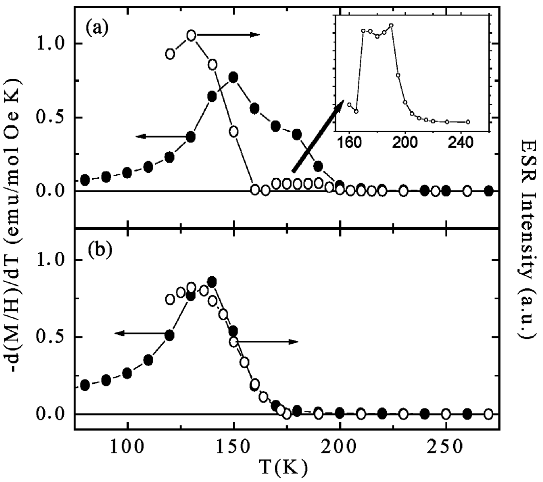

Fig. 1. First derivative of χ_m with respect to temperature (left axis) and microwave absorption at 50 G (right axis) as a function of temperature for $Pr_{0.6}Ba_{0.4}CoO_{3-\delta}$ (a) and $Pr_{0.5}Ba_{0.5}CoO_{3-\delta}$ (b).

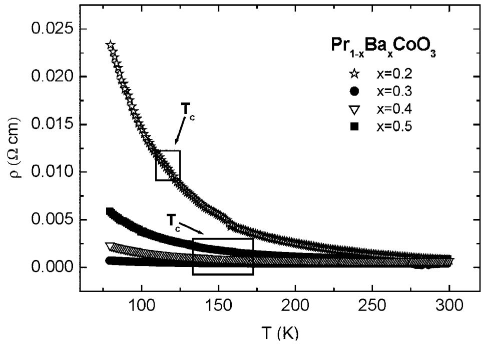

Fig. 2. Resistivity versus temperature for the compounds $Pr_{1-x}Ba_xCoO_{3-\delta}$ ($0.10 < x \leq 0.50$). The rectangles show the relative positions of the Curie temperatures.

diffraction patterns with $x \leq 0.30$ can be indexed on the basis of an O-type ($a \leq b/\sqrt{2} \leq c$) orthorhombic $GdFeO_3$-like perovskite structure with cell parameters $a \approx \sqrt{2}a_c$, $b \approx 2a_c$, and $c \approx \sqrt{2}a_c$, with $a_c = 3.8$ A (S.G. Pnma [5]). The $Pr_{0.5}Ba_{0.5}CoO_{3-\delta}$ sample presents a tetragonal symmetry with cell parameters $a = b \cong a_c$ and $c \cong 2a_c$ (S.G. P4/mmm [6]) and the structure of the $Pr_{0.6}Ba_{0.4}CoO_{3-\delta}$ consists of a mixture of an orthorhombic and a tetragonal phase.

From the magnetic point of view, DC magnetometry reveals that while $PrCoO_3$ is paramagnetic, upon Ba-doping the materials evolve towards a ferromagnetic behavior. It is worth noting that the Curie temperature—estimated from the first derivative of FC susceptibility with respect to temperature (see Fig. 1)—increase slowly with x achieving a maximum value for $x = 0.30$ and decreases thereafter [4]. Very remarkably, in the case of the $x = 0.40$ sample a first magnetic onset is seen at ~ 180 K followed by a second inflection at ~ 160 K (see Fig. 1a).

From ESR experiments, above the magnetic ordering temperature, T_c, no resonant absorption is observed, indicating a fast spin–lattice relaxation rate. However, below T_c there is a microwave absorption signal at low fields. When we plot this microwave absorption signal at 50 G against temperature, we notice that it follows a similar behavior to the magnetic susceptibility temperature derivative (Fig. 1). In the case of the $x = 0.4$ sample two maxima are observed in the ESR measurements, that correlate well with those observed in the corresponding magnetic susceptibility results.

Previously microwave absorption has been used to study the onset of superconductivity (SC) [7]. In the SC case the observed ESR absorption is related to the resistivity anomaly. In order to investigate the origin of this phenomenon, we measured the resistivity of these compounds as a function of temperature (Fig. 2). We did not observe any anomaly in the behavior of the resistivity so we can conclude that the maximum in the ESR spectra can be completely attributed to magnetic processes.

These results illustrate the potentiality and complementarity of ESR and DC magnetic measurements to study complex magnetic systems. On the other hand, ESR is a good technique to determinate the onset of the ferromagnetic behavior and it allow us to distinguish the two closely related magnetic phases with different Curie temperature present in the $Pr_{0.6}Ba_{0.4}CoO_{3-\delta}$ sample.

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References

[1] P.M. Raccah, et al., Phys. Rev. 155 (1967) 932.
[2] M.A. Señarís-Rodríguez, et al., J. Solid State Chem. 118 (1995) 323.
[3] G.E. Pake, Paramagnetic Resonance, W.A. Benjamin, INC, New York, 1962.
[4] B. Rivas-Murias, et al., NATO Sci. Ser. II: Math., Phys. Chem. 61 (2002) 577.
[5] P. Radaelli, et al., Phys. Rev. B 56 (1997) 8265.
[6] A. Maignan, et al., J. Solid State Chem. 142 (1999) 247.
[7] F. Prado, et al., Solid State Commun. 94 (1995) 75.