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Transport phenomena in polycrystalline bulk samples of $Ru_{1-x}Sb_xSr_2(Eu_{0.7}Ce_{0.3})_2Cu_2O_{10-\delta}$

G. ILONCA[*a], F. BEIUŞEANU[b], V. TOMA[a], T. JURCUT[b], E.-V. MACOCIAN[b]
[a]*Babeş – Bolyai University, Faculty of Physics, Cluj – Napoca, Romania*
[b]*Faculty of Sciences, University of Oradea, Oradea, Romania*

Samples in the system $Ru_{1-x}Sb_xSr_2(Eu_{0.7}Ce_{0.3})_2Cu_2O_{10-\delta}$, were prepared by conventional solid state reaction method. The structural analysis was investigated using X-ray diffraction. Electrical resistivity, Hall effect, and magnetic susceptibility measurements on Ru:1222 doped with Sb are presented, together with results in the temperature range 5 K-300 K. Transition temperature decreases from 43 K for x=0.00 to 20 K for x=0.06 Sb. This may be due to a distortion of RuO_6 octahedral, which is responsible of the increase in hole localization. The Hall effect anomalous decreases below $T_{magnetic}$ which may be explained within a simple two-band model by a transition from localized to more itinerant behavior in the RuO_2 layer at $T_{magnetic}$. The behavior of magnetic susceptibility is caused by an antisymetric exchange coupling of the Dzyaloshinsky – Morya type between neighboring Ru moments.

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1. Introduction

The coexistence of superconductivity (SC) and ferromagnetic (FM) in some compounds $RuSr_2LnCu_2O_{10}$ (Ru:1212) and $RuSr_2(Ln_{1-x}Ce_x)_2Cu_2O_{10-\delta}$ (Ru:1222) where Ln=Gd, Eu, Sm brings up the question how these two antagonistic states of matter can accommodate each other. The discovery of the coexistence of SC and FM in class of polycrystalline layered materials of Ru:1212 and Ru:1222 represents a remarkable new development in the study of competing magnetism and SC [1-11]. The physical nature of superconducting and ferromagnetism states is far from being understood.

In Ru:1212 low field antiferomagnetic order was observed and spin-flop transition is increased. Ru:1222 has displayed low and high field ferromagnetic order. These compounds have a layered crystalline lattice with copper-oxygen layers where some carriers are created by Ru^{5+}/Ru^{4+} charge transfer being responsible for the superconductivity around T_c=45 K, while the magnetic order is proposed to originate from the Ru atoms which have an octahedral coordination in $(Ru^{5+}O_6)^{7-}$ [12-13], and the magnetic ordering temperature T_M vary between 60 K and 180 K. Recently, it was observed the coexistence of superconductivity and magnetism in simple compounds like $EuNbO_{3-x}$ [14] and in complex borides which do not have layered structures [15].

Since the RuO_2 layer in Ru:1222 is at the origin of FM and also, at the creation mechanism, the cation substitution for Ru in Ru:1222 should provide some important insights into the understanding of the novel properties in Ru:1222. It is known that the Sb^{3+} substitution for Ru^{5+} should charge the hole concentration in CuO_2 planes and might change the superconducting and magnetic properties in Ru:1222 system, how was reported by L. Shi *et al.* [5].

In this paper we present the results for samples of composition $Ru_{1-x}Sb_xSr_2(Eu_{0.7}Ce_{0.3})_2Cu_2O_{10-\delta}$ (where $0 \le x \le 0.06$) which have been investigated by X-ray powder diffractions, electrical resistivity, Hall effect and magnetic measurements.

2. Experimental

Polycrystalline samples with a nominal composition $Ru_{1-x}Sb_xSr_2(Eu_{0.7}Ce_{0.3})_2Cu_2O_{10-\delta}$ (where $0 \le x \le 0.06$) were synthesized by quantitatively mixing high-purity powders of RuO_2, Sb_2O_3, $SrCO_3$, Eu_2O_3, CeO_2, CuO and fired at 950 ^oC in air for 25 hours. The mixture was grounded and pressed into pellets before preliminary reaction at 1020 ^oC and 1040 ^oC in air for 30 hours at each temperature with intermediate grindings.

The resulting samples were reground, repelleted and fired in a flow of oxygen at 1060 ^oC for 120 hours, and finally over a span of 24 hours cooled slowly down to room temperature. These samples were placed in sealed gold capsules and annealed at 1050 ^oC for 80 hours in high pressure oxygen of 60 atm and cooled slowly down to room temperature.

The X-ray powder diffraction pattern was taken of the examined samples using XD8 Advance – Bruker AXS diffractometer using CuK_α radiation. The resistance of samples and Hall effect were measured with a standard probe method DC – technique on bar shaped peaces of our samples using silver paint contacts, in the temperature range 4 – 300 K. Magnetic properties were measured with a SQUID magnetometer.

3. Results and discussion

The samples microstructure was investigated by X-powder diffraction. The XRD patterns recorded at room temperature for samples $Ru_{1-x}Sb_xSr_2(Eu_{0.7}Ce_{0.3})_2Cu_2O_{10-\delta}$, (where x=0, 0.02, 0.04, 0.06) reveal the presence of the main of Ru:1222 materials, as shown in Fig. 1.

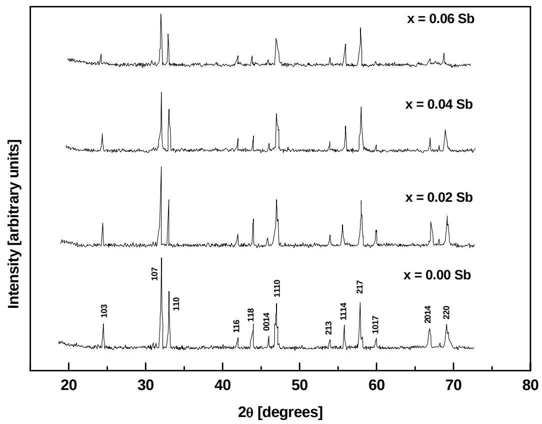

Fig. 1. XRD pattern of $Ru_{1-x}Sb_xSr_2(Eu_{0.7}Ce_{0.3})_2Cu_2O_{10-\delta}$ samples with $0 \le x \le 0.06$.

The lattice parameters were calculated by a least-squares method using external standard (α-Al_2O_3 with a=4.7588(1) Å, c=12.993(2) Å). The results are presented in Table 1.

The Sb^{3+} ion substitutes for Ru in solid solution has a dramatically effect on the transport and magnetic properties what can be see in previously work [11,16]. The temperature dependence of the resistivity ρ(T) for samples $Ru_{1-x}Sb_xSr_2(Eu_{0.7}Ce_{0.3})_2Cu_2O_{10-\delta}$ (where x=0, 0.02, 0.04, 0.06) are shown in Fig. 2.

Table 1. Structural parameters derived from X-ray powder diffraction data for $Ru_{1-x}Sb_xSr_2(Eu_{0.7}Ce_{0.3})_2Cu_2O_{10-\delta}$.

x	a (Å)	c (Å)	Volume (Å^3)
0	3.8362(1)	28.5235(1)	419.7641(0)
0.02	3.8340(3)	28.5387(0)	419.6156(3)
0.04	3.8341(5)	28.5340(2)	419.4590
0.06	3.8425(0)	28.5285(4)	421.1217(7)

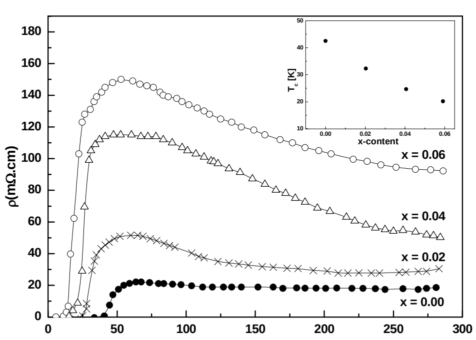

Fig. 2. Temperature dependence of the resistivity for $Ru_{1-x}Sb_xSr_2(Eu_{0.7}Ce_{0.3})_2Cu_2O_{10-\delta}$ samples with $0 \le x \le 0.06$. Insert T_c vs. content of Sb.

The superconducting transition is observed in all the samples. The superconducting transition temperature T_c (onset) decreases with increasing Sb content from T_c=43 K for x=0.00 to 20 K for x=0.06 and all samples are semiconductor like above T_c (onset).

The hole concentration in the CuO_2 plane decreases with Sb doping, in agree with Ref. [5]. The possible origin of this results is due to the distortion of RuO_6 octahedron in Ru:1212 was proved through neutron powder diffraction experiments and synchrotron X-ray diffraction experiment by Refs. [4] and [17]. The distortion of RuO_6 octahedron might lead to strong narrowing of the bands to result in the localization for Ru:1212 and Ru:1222. The Sb substitution for Ru increases the distortion of RuO_6 octahedron and leads too more localization of carriers, so the transport behavior show typical semiconductor-like and the superconducting transition temperature decreases with increasing Sb substitution for Ru.

This effect is confirmed by Hall effect measurements what can see in Fig. 3.

The magnetic susceptibility χ obeys Curie-Wais law in the temperature range 150-300 K.

The curves of the temperature dependence of magnetic susceptibility show the cusps at around ~ 142 K what can see in Fig. 4. These are the Néel temperatures corresponding to the canted antiferomagnetic ordering of the Ru magnetic moments along the c-axis in $Ru_{1-x}Sb_xSr_2(Eu_{0.7}Ce_{0.3})_2Cu_2O_{10-\delta}$ samples, as in the case of the isostructural gadolinium-rutheno-cuprate reported by Lynn et al. [17].

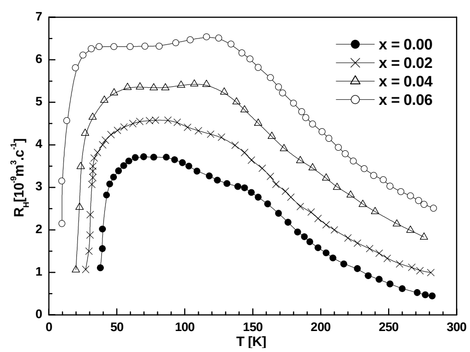

Fig. 3. Temperature dependence of Hall coefficient for $Ru_{1-x}Sb_xSr_2(Eu_{0.7}Ce_{0.3})_2Cu_2O_{10-\delta}$ samples with $0 \le x \le 0.06$.

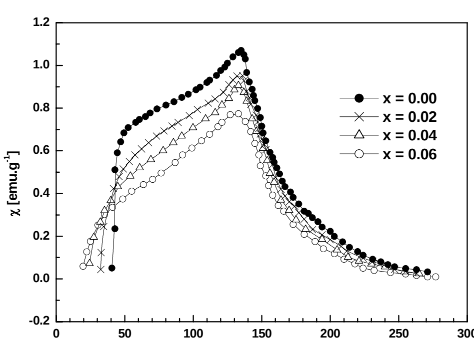

Fig. 4. Temperature dependence of magnetic susceptibility of $Ru_{1-x}Sb_xSr_2(Eu_{0.7}Ce_{0.3})_2Cu_2O_{10-\delta}$ samples with $0 \le x \le 0.06$.

A cusp observed below 142 K for the magnetic susceptibility levels off due to the nonmagnetic Eu^7F_0 ground term, so that the non-diagonal interaction of the Van Vleck equation are solely responsible of the well known temperature independent paramagnetic susceptibility in agreement with data reported for Ru:1212 by R. Ruiz-Bustos *et al.* [18].

The magnetization curves, corresponding to the $Ru_{1.92}Sb_{0.02}(Eu_{0.7}Ce_{0.3})_2Cu_2O_{10-\delta}$ sample at 30 K and 120 K are plotted in Fig. 5.

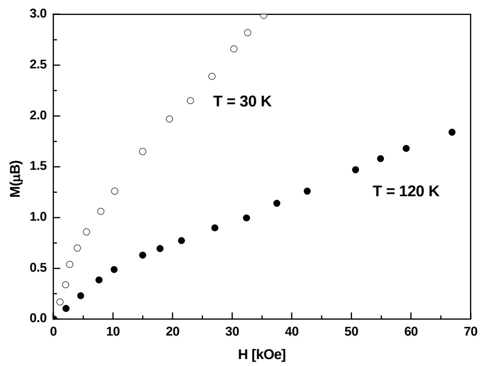

Fig. 5. Field dependence of magnetization for $RuSr_2(Eu_{0.7}Ce_{0.3})_2Cu_2O_{10-\delta}$ sample.

The values of the ferromagnetic components extrapolated from magnetization versus magnetic field at 120K is 0.10 μ_B/Ru in agreement with the values Ref. 17 obtained for the isostructural Gd compounds (Ru:1212) by neutron diffraction. The ferromagnetic order is caused by an antisymmetric exchange coupling of the Dzyaloshinsky-Moriya (DM) type between neighboring Ru moments, induced by a local distortion that breaks the tetragonal symmetry of the RuO_6 octahedra. Due to this DM interaction the applied magnetic field causes the adjacent spins to cant slightly out of their original direction and to aligned a component of the moments with the direction of applied magnetic field, as can see in χ versus T in our systems. At low temperature the Ru-Ru interaction begins to dominate and leads to reorientation of the Ru moments, what can be see around 100 K in plotted χ versus T.

The coexistence of superconductivity and magnetism survives in these systems based on Ru because the Ru moment is probably align in the basal planes and exchange coupling between the superconducting carriers and the Ru moments is weak, so that there is no pair breaking in the CuO_2 planes.

4. Conclusions

The bulk single phase samples of copper ruthenium oxides with composition close to $Ru_{1-x}Sb_xSr_2(Eu_{0.7}Ce_{0.3})_2Cu_2O_{10-\delta}$ (Ru:1222) with $0 \le x \le 0.06$ were synthesized by a solid-state reaction method. The X-ray diffraction analysis has shown only small differences in the lattice parameters when Sb content varies.

Magnetic susceptibility measurements reveal the existence of the cusps below 150 K, in χ versus T, which are difficult to explain due to the presence of magnetic impurities. The Sb - doping can increase the distance between paramagnetic Eu^{3+} ions, which is evidenced by the increase of lattice parameters with Sb doping.

The Sb substitution for Ru in the ferromagnetic superconductor $Ru_{1-x}Sb_xSr_2(Eu_{0.7}Ce_{0.3})_2Cu_2O_{10-\delta}$ systems reduces the onset of the superconducting transition from 43 K for x=0 to 20 K for x=0.06. This demonstrates the increase in hole localization due a distortion of RuO_6 octahedra.

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*Corresponding author: gilonca@phys.ubbcluj.ro