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Superconducting and magnetic properties of Sn-doped Ru-1222

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Abstract

Samples with nominal compositions $Ru_{1-x}Sn_xSr_2Gd_{1.4}Ce_{0.6}Cu_2O_y$ ($0 \le x \le 0.2$) were synthesized and their superconducting and magnetic properties were investigated. A non-monotonic behaviour of the lattice parameters and T_c with the increase of the dopant content was observed. It was established that small doping levels ($0 \le x \le 0.05$) significantly increase the T_c of the Ru-1222 samples, prepared at the same conditions – from 20 K for the undoped sample to 35 K for the x = 0.03 one. The $0.1 \le x \le 0.2$ samples are not superconducting, i.e. the Sn-doping more rapidly destroys the superconductivity in Ru-1222 than in the conventional superconductors. The initial increase of T_c was associated with an increase of the hole concentration. The decrease of T_c and suppression of SC at higher doping levels may be explained by an enhanced disorder in the system, due to a possible presence of Sn in both Ru and Cu sites. The latter fact could also explain that the onset of the magnetic transition T_{mag} weakly depends on the dopant content. (© 2007 Elsevier B.V. All rights reserved.

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1. Introduction

Several years ago a coexistence of superconductivity (SC) and weak ferromagnetism (FM) was discovered in $RuSr_2R_{2-x}Ce_xCu_2O_{10}$ (R = Eu, Gd, Ru-1222) [1] and subsequently in $RuSr_2GdCu_2O_8$ (Ru-1212) [2]. It was established that the magnetism originates from the RuO_2 layers and superconductivity arises from the CuO_2 planes. The two states are practically decoupled, so that there is no pair breaking. More information about the SC and magnetic state of Ru-1222 can be obtained if Ru is partially replaced by other ions. The foreign ions could modify the carrier concentration and oxygen content of Ru-1222 and influence its superconducting and magnetic properties.

* Corresponding author. E-mail address: nbalchev@yahoo.com (N. Balchev). Some reports exist on $(Ru_{1-x}M_x)$ -1222 systems where M = Fe, Co, Nb, Mo [3–8]. The Sn-substitution attracted the attention of the investigators since it was established that Sn weakly affects the SC properties of YBaCuO and YBaSrCuO [9]. It was also shown that the Sn-doping favours the melt-texture-growth (MTG) of YBaCuO [10]. The authors of [11,12] studied the effect of the Sn-substitution on the superconducting properties of the Hg-1223 and (Pb, Cu)-1212 systems. It was established that Sn stimulates the Hg-1212 phase formation and enhances the diamagnetic volume fraction but also the weak link behavior. In [13,14] the substitution of Sn in Ru-1212 was investigated. A superconductivity exists up to Sn-concentrations of $x \leq 0.5$ for Hg-1223 and $x \leq 0.3$ for (Pb, Cu)-1212 and Ru-1212. Concerning the phase 1222, Luo et al. [15] synthesized (Cd, Sn)-1222 samples thus showing that Sn can be involved in this phase. The Sn⁴⁺ ionic radius

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(0.69 Å) is slightly higher than that of Ru⁴⁺ (0.62 Å), Ru⁵⁺ (0.565 Å) and Cu²⁺ (0.65 Å) [16]. This could allow a partial substitution of Sn in the Ru and/or the Cu-sites. Felner et al. [17] studied the magnetization and Mössbauer spectra of Sn-doped Ru-1222. A gradual decrease of T_c and the magnetic transition temperature with the increase of the dopant content was observed. The present work is a more detailed investigation of the Sn-doping on the structure, superconducting and magnetic properties of Ru-1222. We established a non-monotonic behaviour of the lattice parameters and T_c with the increase of the dopant content, a rapid suppression of SC and a weak dependency of the onset of the magnetic transition T_{mag} on the Sn-content. The observed phenomena were discussed.

2. Experimental details

The investigated samples with nominal compositions $Ru_{1-x}Sn_xSr_2Gd_{1,4}Ce_{0,6}Cu_2O_v$ ($0 \le x \le 0.2$) were prepared by a solid state reaction from starting products RuO₂, Gd₂O₃, SnO₂, CeO₂, SrCO₃ and CuO with a purity above 99.9%. They were mixed, homogenized, pressed into pellets and preheated at 600 °C for 48 h in air and at 1000 °C for 24 h in flowing oxygen. Subsequently they were reground, repressed and sintered at 1060 °C for 96 h in flowing oxygen. Finally they were annealed at 350 °C for 48 h in flowing oxygen. X-ray diffraction (XRD) was used to examine the samples using a TUR-M62 diffractometer and CoK_{α} radiation. A scanning electron microscope (SEM) with energy dispersive system (EDS) analysis was performed on the pellets using a SEM-525 Philips microscope combined with an EDAX 9900 device. The susceptibility and magnetization of the samples were measured by a SQUID magnetometer (Quantum Design: PPMS-9T and MPMS-XL7T). The resistivity of the samples ρ vs T was measured by the standard four probe method.

3. Experimental results

Fig. 1 shows the XRD patterns of $Ru_{1-x}Sn_xSr_2Gd_{1.4}$ - $Ce_{0.6}Cu_2O_v$ for x = 0, 0.05 and 0.1. It may be seen that the samples are nearly single phased. It was established that the x = 0.2 sample contains SrSnO₃ as impurity. The calculated lattice parameters of the investigated samples are given in Table 1. It may be seen that the parameters a and c have a non-monotonic behaviour with the increase of x and this will be discussed in the next section. In Fig. 2 the SEM pictures of the x = 0, 0.05 and 0.1 samples are given. In the undoped sample several tetragonal crystals can be seen but in most of the microcrystals the crystal boundaries are not well expressed due to incomplete crystallization. In the x = 0.05 and 0.1 samples the relative number of the tetragonal microcrystals increases. From this we may conclude that the Sn-doping enhances the crystal growth in Ru-1222. The calculated densities of the samples are 5.4 for x = 0.03, 6.12 for x = 0.1 and 6.3 g/cm³ for x = 0.2. For comparison, the authors of [10] obtained



Fig. 1. XRD patterns of $Ru_{1-x}Sn_xSr_2Gd_{1.4}Ce_{0.6}Cu_2O_y$ for x = 0, 0.05 and 0.1.

Table 1

Lattice parameters, remanent moments μ_R , saturated moments per Ru atom μ_{sat} and T_{on} of the investigated Ru_{1-x}Sn_xSr₂Gd_{1.4}Ce_{0.6}Cu₂O_y samples

x	a (Å)	c (Å)	$\mu_{\rm R}~(\mu_{\rm B})$	$\mu_{\rm sat}$ ($\mu_{\rm B}$)	$T_{\rm on}$ (K)
0	3.827	28.37	0.48	0.87	20
0.03	3.827	28.53	0.39	0.62	35
0.05	3.846	28.69			24
0.1	3.808	28.22	0.24	0.39	not SC

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