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Superconducting and Magnetic Properties of Sn-Doped $EuBa_2Cu_3O_{7-\delta}$ Compound

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Influence of Sn-substitution on structural, superconducting and magnetic properties of EuBa₂Cu₃O_{7- δ} (Eu-123) compound was studied by X-ray diffraction and SQUID magnetic measurements. Transition process from normal to superconducting state and temperature dependences of zero field cooling and field cooling characteristics of magnetic moment and magnetization curves were measured and analysed. Samples of the nominal composition of EuBa₂Cu_{3-x}Sn_xO_{7- δ} with x ranging from 0.0 to 1.5 were prepared by the solid state reaction technique. The increasing Sn-content decreases the volume of the main superconducting phase (Eu-123) and for higher Sn-contents, semiconducting tetragonal Eu₃Ba₃Cu₆O₁₄(Eu-336) and Ba–Sn–O phases are the dominant ones.

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1. Introduction

In many studies of Sn doping of RE-123 family superconductors, most results were reported for the Y-123 superconductor [1, 2]. In this case, some inconsistent results were also reported, e.g. the question of Sn entering into the Y-123 phase or the question of an effect of increase of the Sn content on the critical transition temperature [3, 4]. Only several results were reported for the Eu-123 system, disregarding the Sn addition in the melt textured Eu-Ba-Cu-O compounds to increase the critical current density and to supply more oxygen in the crystal growth process using oxide precursors [5, 6].

In the paper, we studied structural, superconducting and magnetic properties of the $\text{EuBa}_2\text{Cu}_{3-x}\text{Sn}_x\text{O}_{7-\delta}$ samples.

2. Experimental

The polycrystalline samples of EuBa₂Cu_{3-x}Sn_xO_{7- δ}, with x = 0.00, 0.03, 0.1, 0.2, 0.7, and 1.5 were prepared by a standard solid-state reaction method from the commercial 99.99% purity oxide powders of Eu₂O₃, CuO and SnO₂ and BaCO₃. The powders were calcined at 930 °C for 40 h, sintered at about 1050 °C for 72 h in flowing oxygen, then oxygenated at 580 °C for 24 h. For more details, see [7]. The X-ray diffraction (XRD, Cu K_{α} radiation) measurements were performed on powdered samples and magnetic measurements on rectangular samples $\approx (2.2 \times 1.6 \times 8.4) \text{ mm}^3$ that were cut from pellet samples. The zero field cooling (ZFC) and field cooling (FC) temperature dependences of the magnetic moment M(T)and the mass magnetization M(H) were measured in the

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temperature range of 20 K–300 K and at the magnetic field of $0.8 \text{ kA m}^{-1} \div 5.8 \text{ MA m}^{-1}$ by the Quantum Design SQUID magnetometer MPMS XL-7.

3. Results

From the X-ray diffraction data, Fig. 1, it can be concluded that all peaks can be well ascribed to the Eu-123 superconducting phase for x = 0.00 and 0.03, however, for a higher content of Sn, x = 0.1 and 0.2, some new peaks could be identified that could be ascribed to the excess BaCuO₂ and CuO phases. For the maximum levels of Sn, the semiconducting tetragonal Eu₃Ba₃Cu₆O₁₄ and Ba–Sn–O phases are the significant ones. The cubic semiconductor BaSnO₃ phase is a dominant phase for x = 1.5 sample.

The temperature related to the onset of a diamagnetic decrease of the transition from the normal to the super-



Fig. 1. XRD patterns of $\text{EuBa}_2\text{Cu}_{3-x}\text{Sn}_x\text{O}_{7-\delta}$ samples with the nominal x values. Symbols designate the peaks that could be assigned to $\text{Ba}\text{Cu}\text{O}_2$ ($\mathbf{\vee}$), CuO ($\mathbf{\diamond}$), $\text{Eu}_3\text{Ba}_3\text{Cu}_3\text{O}_{14}$ ($\mathbf{\circ}$), Ba_2SnO_4 ($\mathbf{\Box}$) and BaSnO_3 (**a**).

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Fig. 2. Temperature dependences of the FC dc mass magnetic susceptibility χ of EuBa₂Cu_{3-x}Sn_xO_{7- δ} samples with x = 0 (•), 0.03 (•), 0.1 (•), 0.2 (•), 0.7 (•) and 1.5 (+). The insert shows dependences of the samples with the highest Sn contents.

conducting state of the FC dc mass magnetic susceptibility curves at magnetic field of 0.8 kA m⁻¹ is described as the critical transition temperature, T_c^{ON} , Fig. 2.

The increasing Sn-content deteriorates the superconducting properties of the Sn doped samples. T_c^{ON} decreases from ≈ 95 K to ≈ 88 K from x = 0 to x = 0.7, likewise the magnization hysteresis at 20 K, Fig. 3, see inset in Fig. 4.



Fig. 3. The dc mass magnetization M_{DC} vs. the applied magnetic field H_a of EuBa₂Cu_{3-x}Sn_xO_{7- δ} samples with x = 0 (•), 0.03 (\blacktriangle), 0.7 (\bigstar) and 1.5 (+) at 20 K.

At 20 K, the magnetization loops with $x \leq 0.03$ indicate the so-called second peak effect [8], while the ones with the highest Sn contents show an evident magnetic "tail", paramagnetic contribution.

All the samples show superconducting ordering at the field of 0.8 kA m⁻¹, except for the one with x = 1.5. However, the sample with x = 1.5 also shows a weak hump at 90 K that could be ascribed to the presence of superconducting phase(s), see inset of Fig. 2 and Fig. 4.



Fig. 4. Temperature dependences of the FC dc mass magnetic susceptibility χ of the sample with x = 0.0 (•), x = 0.2 (•), x = 0.7 (★) and 1.5 (+) at the field of 0.8 kA m⁻¹. Inset shows values of the T_c^{ON} vs. the nominal composition deviation of x.

The temperature dependence of the FC mass magnetic susceptibility of all samples moves up to positive values by the appplication of magnetic field of 4 MA m⁻¹, see Fig. 5. In this case, the presence of the superconducting phase is signalized only by a diamagnetic decrease at 90 K e.g. the stoichiometric sample with x = 0. The inset shows that the ZFC characteristic is more sensitive for identification of superconductivity. In this case, there is a diamagnetic decrease at about 8 K of the sample with x = 0.7.



Fig. 5. Temperature dependences of the FC dc mass magnetic susceptibility χ of the sample with x = 0.0 (•), x = 0.7 (★) and 1.5 (+). The inset shows the corresponding the ZFC dependences of the samples with x = 0.7 and 1.5 at low temperatures.

The results given above indicate the presence of superconducting and (para)magnetic components. For better understanding, it is necessary to focus on the temperature range over the critical temperature T_c^{on} , where an effect of the superconducting phase is absent. Dependences of the dc mass magnetization M_{DC} vs. the applied magnetic field H_a for the samples with x = 0.0, 0.7 and 1.5 are shown in Fig. 6. At 300 K, all the samples show non-hysteresis linear dependences that are confirmed also at low field values. The insets prove the presence of a superconducting component in the sample with x = 0.7 at 77 K and 20 K, while the x = 1.5 sample shows paramagnetic dependence.

More detailed information can be obtained from the temperature dependences of the FC mass magnetic susceptibility that are shown in Fig. 4. In the temperature range above T_c^{ON} , Fig. 4, a good correlation can be observed between the level of the susceptibility curves and the reported magnetic properties of the emerging phases identified by XRD. The lift of the curve with x = 0.2 can be ascribed to the presence of the Eu-336 phase (probably also the paramagnetic one) based on results of Agilandeswari et al. [9]. The increase of tin content to x =0.7 can be ascribed to the decrease of the content of the superconducting phase and to the presence of the paramagnetic Ba_2SnO_4 phase [10]. The significant decrease of the x = 1.5 curve can result from the presence of the diamagnetic $BaSnO_3$ phase [11]. We remark that the magnetic properties of all of the excess phases are sensitive to oxygen nonstoichiometry.



Fig. 6. M_{DC} vs. H_a dependences of EuBa₂Cu_{3-x}Sn_xO_{7- δ} samples with x = 0 (•), 0.7 (★) and 1.5 (+) at 300 K. Insets show the M_{DC} vs. H_a dependences at 77 K and 20 K at small field for samples with x = 0.7 (★) and 1.5 (+).

4. Conclusions

Influence of Sn-substitution on structural, superconducting and magnetic properties in $EuBa_2Cu_3O_{7-\delta}$ (Eu-123) compound was studied by XRD, The received results confirm the usefulness of ZFC and FC magnetic characteristics at the study of the mutual presence of superconducting and magnetic components and at the separation of their effects.

Acknowledgments

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