Study of Effect of Sn doping of $EuBa_2Cu_{3-x}O_{7-\delta}$ Compound on Superconducting Properties by Contactless Methods

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Abstract. *Effect of Sn addition on structural and superconducting properties in* $EuBa_2Cu_3O_{7-\delta}$ *(Eu-123) compound was studied using XRD and SQUID magnetometric measurements. Transition process from normal to superconducting state was analyzed by the measurement of temperature change of the magnetic moment of the combined ZFC and the remanence technique beside the usual ZFC and FC ones. Samples of the nominal composition* $EuBa_2Cu_{3-x}Sn_xO_{7-\delta}$ *with x ranging from 0.0 to 0.2 were prepared by the solid state reaction technique from* Eu_2O_3, $BaCO_3$, *CuO and* SnO_2 *precursors. The increasing Sn-content deteriorates the superconducting properties of the Sn doped samples.*

Keywords: High-Temperature Superconductors, Eu-123, Transition Characteristics, Sn Doping, Mass Magnetization, T_c^{on}

1. Introduction

In a new class of copper-based high-temperature superconductors (HTS), up to now, there is no generally accepted theory reasonably explaining all experimental results. All HTS are layered systems having the same basic crystallographic structure; they contain parallel two-dimensional layers of CuO_2 planes and a common characteristic dependence on doping. The chemical doping is a useful tool for revealing the HT superconductivity mechanism. With regard to the role of the $Cu\text{-}O_2$ planes, special attention is paid to chemical substitutions into copper positions.

At the study of Sn doping of RE-123 HT superconductors, where RE = Y, and rare earths or mixed rare earths elements, most results were reported for the Sn doping of Y-123 superconductor. In this case, some inconsistent results were also reported, e.g., such as the question of Sn entering the Y-123 phase and the formation of a solid solution or if the tin preferably enters the Cu(2) sites in $Cu\text{-}O_2$ layers or the Cu(1) sites in Cu-O chains [1-6] or the question of an effect of increasing of Sn content on the critical transition temperature. [4,6,7]. 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, in the crystal growth process, more oxygen using oxide precursors, [8-10]. In the paper, we studied sintered samples of the nominal composition of $EuBa_2Cu_{3-x}Sn_xO_{7-\delta}$.

2. Subject and Methods

The polycrystalline samples of $EuBa_2Cu_{3-x}Sn_xO_{7-\delta}$, where x = 0.00, 0.01, 0.03, 0.07, 0.10, and 0.20 were prepared by a standard solid-state reaction method from the commercial 99.99 % purity oxide powders of Eu_2O_3, CuO, SnO_2 and $BaCO_3$. The powders were carefully weighed in appropriate weight amounts and homogenized in air in an agate mortar for five minutes and calcined at 930 °C for 40 hours in air. The obtained precursors were again homogenized, pressed into the pellets and sintered in a horizontal tube furnace in flowing oxygen of 20 ml/min at about 1050 °C for 72 h, then cooled to 580 °C and held at this temperature for

24 h and thereafter cooled in the furnace to room temperature. The XRD measurements were performed on powdered samples and the magnetic measurements on cuboid samples ~ (2.2×1.6×8.4) mm cut from pellets. The four-point resistance measurement technique of the *R* vs. *T* dependence is the best-known standard method for determination of various characteristics of superconducting and normal states of superconductors. The other two contactless measuring methods - using a change of the self-inductance of a coil located in vicinity of the measured sample or using a change of the mutual inductance of two coils separated by a sample - were also used.

In the paper, we compared three characteristics of the transition from the normal to the superconducting state obtained by another contactless method. The method is based on the measurement of the temperature dependences of the magnetic moment (magnetization or magnetic susceptibility) of the studied samples. In our case, the temperature dependence of the DC magnetic moment of the superconducting compounds was measured by the Quantum Design SQUID magnetometer MPMS XL-7. The dependences were measured under three different conditions, at the zero field cooling (ZFC), field cooling (FC) and the technique combining the zero field cooling and the measurement of the remanent magnetic moment (ZFC-R). Before the measurement of each of the three characteristics, the shielding of the superconducting magnet has been demagnetized (using Degauss option of the MPMS), superconducting magnet was reset by driving into the normal state and ultra-low field procedure (using Ultra Low Field option) was used to set the zero field value and homogeneity. After stabilizing the temperature, the applied magnetic field of 795 Am^{-1} was set by using the No Overshoot mode.

The measurement of the magnetic moment in each measured point followed after the temperature was stabilized by the system and after the expiration of the waiting time of 90 s. The measured data were obtained using the RSO option. (The amplitude of 4 cm, frequency of 1.5 Hz, 5 cycles of the sample oscillation and two sets of such measurement were averaged to represent one point of the presented data (curve)). The ZFC procedure consists of cooling the sample from the room temperature to ~2 K in the zero applied magnetic field, $H_a = 0$. Thereafter, H_a was set to 795 Am^{-1} and the magnetic moment of the sample was measured at an increasing temperature to 300 K. The FC curve was obtained by the measurement of the magnetic moment at $H_a = 795$ Am^{-1} at a decreasing temperature. In the ZFC-R procedure, the sample was zero-field cooled to 2 K, subsequently, after the application of $H_a = 795$ Am^{-1} for 180 s, the remanent magnetic moment ($H_a = 0$) was measured at an increasing temperature. The temperature resolution was 0.001 K. The DC mass magnetization loops were measured at 77 K and 20 K by the Quantum Design SQUID magnetometer MPMS XL-7 whose differential sensitivity is 10^{-11} Am^2 from 0T to 1T. The phase composition was studied by X-ray diffraction measurements (CuKα radiation).

3. Results and Discussion

From X-ray diffraction data, Fig. 1, it can be concluded that all peaks can be well ascribed to the Eu123 superconducting phase up to $x \leq 0.03$, however, for higher contents of Sn, some new peaks could be identified that could be ascribed to the excess $BaCuO_2$ and CuO and $Eu_2Sn_2O_7Cu$ phase at an increasing of Sn content. The ZFC, FC, and ZFC-R mass magnetization transition curves are in Fig.2. In the case of ZFC-R conditions, the positive values of magnetization in the range of the superconducting state result from the opposite direction of the circulation of the super conducting shielding current as a

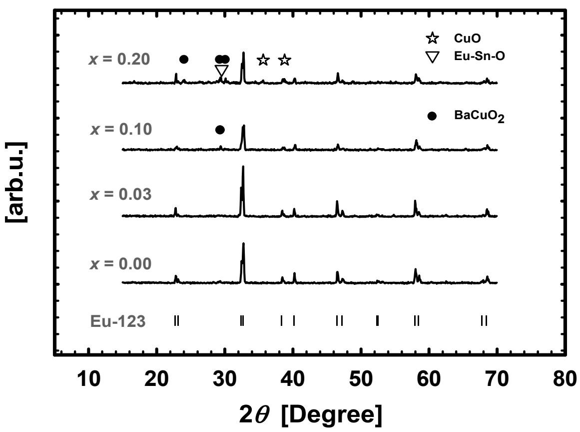


Fig.1. XRD patterns of $EuBa_2Cu_{3-x}Sn_xO_{7-\delta}$ samples showing the nominal x - values.

reaction on the set of $H_a = 0$.

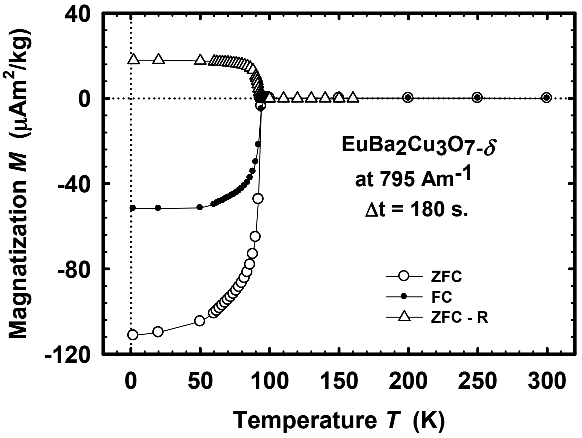


Fig. 2. ZFC, FC, and ZFC-R mass magnetization vs. temperature of the undoped $EuBa_2Cu_3O_{7-\delta}$ sample measured at H_a = 795 Am^{-1}. The H_a was applied for 180 s and then set to zero before the measurement of data in the ZFC-R -procedure.

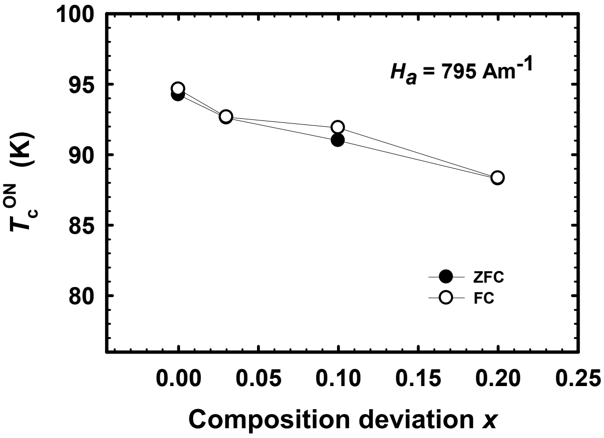
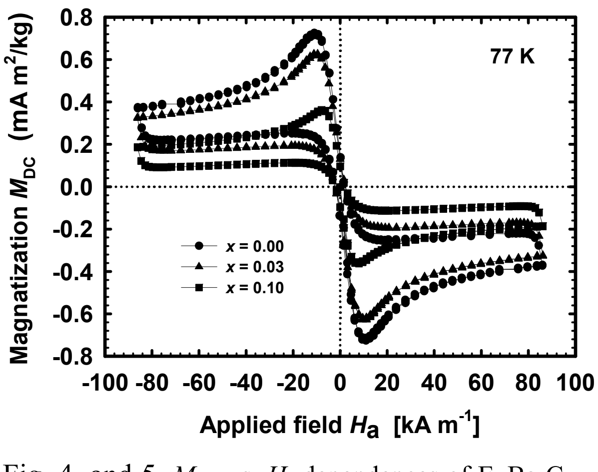


Fig. 3. ZFC and FC T_c^{on} - values of the $EuBa_2Cu_{3-x}Sn_xO_{7-\delta}$ samples vs. the nominal composition deviation of x.

The temperature related to the onset of a diamagnetic change of the transition from the normal to the superconducting state of the ZFC and FC magnetization curves is described as the critical transition temperature, T_c^{on}. The ZFC and FC T_c^{on} - values of some samples are shown in Fig. 3. It can be seen that the T_c^{on} values are still higher than 88 K up to x = 0.20; whereas the ZFC and ZFC-R values are practically the same with respect to the temperature resolution of 0.001K, the FC T_c^{on} - values slightly differentiate; see Fig. 3. In the FC case, the applied magnetic field in the normal state penetrates into the sample volume before the superconducting state (the superconducting shielding current) is present, while in the former cases, the H_a is applied to the sample at a low temperature (in the superconducting state), so the field cannot penetrate into the sample volume, if its value is lower than the mean value of the first penetration magnetic field of the intergrain junctions of the H_{p1}^{IJ} sample. From the magnetization data at 77 K, the estimated value of H_{p1}^{IJ} for the undoped sample is more than one order higher than H_a = 795 Am^{-1}. In the case of the ZFC-R magnetization curve, the T_c^{on} - value was determined as an onset of a paramagnetic trend. In general, the ZFC-R technique can be more useful for verification of the superconducting state and its differentiation from other diamagnetic effects by the contactless methods.



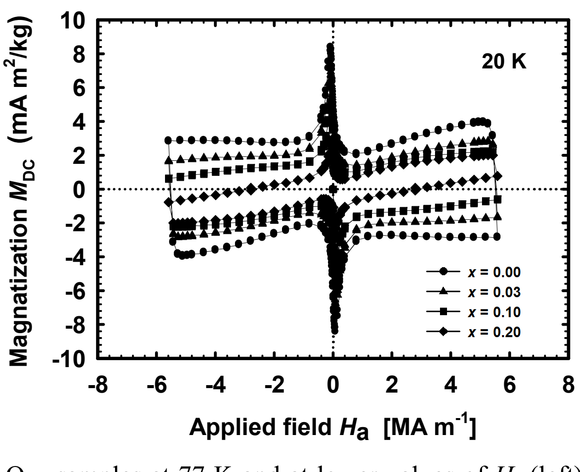


Fig. 4. and 5. M_{DC} vs. H_a dependences of $EuBa_2Cu_{3-x}Sn_xO_{7-\delta}$ samples at 77 K and at lower values of H_a (left) and at 20 K and at higher values of the applied magnetic field H_a (right), respectively.

The DC magnetization hysteresis curves of M_{DC} vs. H_a for $EuBa_2Cu_{3-x}Sn_xO_{7-\delta}$ samples at 77 K and 20 K are shown in Fig. 4 and Fig. 5, respectively.

All the samples show the Z-shape of magnetization curves at the low H_a typical for the superconducting polycrystalline samples. The increasing content of Sn decreases the volume superconducting properties, as the values of M_{DC} and the magnetization hysteresis decrease. At 20 K, the magnetization loops with the Sn content $x \leq 0.03$ indicate the so-called second peak effect, while the M_{DC} vs. H_a curves with a higher Sn content of $x \geq 0.10$ show an evident (para) magnetic "tail" - magnetic contribution indicated by the slope of the curves pointing to the first and third quadrants. The magnetic component can be associated with the origination of excess phases, namely with the $BaCuO_2$ phase [11,12].

4. Conclusions

Effect of the Sn- addition on the structural and superconducting properties in $EuBa_2Cu_{3-x}Sn_xO_{7-\delta}$ compounds with the nominal value of x from zero to 0.20 was studied using the XRD and SQUID magnetometric measurements. The polycrystalline samples were sintered in a horizontal tube furnace in flowing oxygen of 20 ml/min at about 1050°C for 72 h. In the paper, the combined ZFC-R technique was used to study the transition process from the normal to the superconducting state together with the usual ZFC and the FC contactless techniques. The ZFC-R procedure can be more useful for verification of the superconducting state and its differentiation from other diamagnetic effects by the contactless methods. It can be concluded, based on the results and the XRD data, that the superconducting Eu-123 phase is dominant in all our samples and that the solution limit of Sn in Eu-123 is limited to $x = 0.03$, if at all.

Acknowledgements

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References

[1] J. Feng, K.K. Yeung, K.W. Wong, Eric C.L. Fu, C. C. Lam, *Supercond. Sci. Technol.,* 13: 215-224, 2000.

[2] H. Zhang, G. M. Wang, Q. R. Zhang, *Physics Letters A*, 138: 517-522, 1989.

[3] Y. Matsumoto, T. Nishida, M. Katada, *Physica B*, 194-196: 2127-2128, 1994.

[4] F. Licci, L. Raffo, T. Besagni, C. Frigeri, C. Paris, *Physica C*, 196: 307-314, 1992.

[5] T.Saito, K. Mizumo,H. Watanabe, K. Koto, *Physica C*, 171: 167-173, 1990.

[6] Y. Maeno, M. Kato, Y. Aoki, T. Nojima, T. Fujita, *Physica B*, 148: 357-359, 1987.

[7] H. Zhang, X. Y. Zhou, Y. Zhao, S. H. Liu, Q. R. Zhang, *Sol. Stat. Comm.*, 72: 75-79, 1989.

[8] P. Boolchand, R. n. Enzweiler, I. Zitkovsky, J. Wells, W. Bresser, Darl McDaniel, R. L. Meng, P. H. Hor, C. W. Chu, C. Y. Huang, *Phys. Rev. B,* 37; 3766-3769, 1988.

[9] E. Kuzmann, Z. Homonnay, A.Vertes, *Phys. Rev. B*, 39: 328-333, 1989.

[10] C. Jiang, Y. Mei, S. M. Green, H. L. Luo, C. Polits, *Z. Phys. B -Cond. Matt.*, 68: 15-18, 1987.

[11] N. Guskos, V. Likodimos, C. A. Londos, V. Psycharis, C. Mitros, A. Koufoudakis, H. Gamari-Seale, W. Windsch, H. Metz, *J. Sol. State. Chem.*, 119: 50-61, 1995.

[12] Yamamoto, T. Mito, S. Wada, R. A. Fisher, N. E. Phillips, Z. P. Wang, D. C. Johnston, *J. Magnetism and Magnetic Materials,* 272-276: 88-89, 2004.