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Magnetization, Susceptibility and Critical Currents of $(Tl_{2-x}Re_x)Ba_2CaCu_2O_y$ Thin Films

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The magnetization and the AC susceptibility vs. the temperature as well as the applied magnetic field of the thin film $(Tl_{2-x}Re_x)Ba_2CaCu_2O_y$ with $x = 0$ and 0.15 on R-plane sapphire substrate with CeO_2 buffer layer were measured and analyzed. XRD measurements show c-axis as well as a–b plane oriented Tl-1212 and superconducting pure phase. The zero critical temperature of the Tl–Re sample is 99.9 K and is practically the same as the critical temperature of the rhenium free sample: 99.5 K. The Tl–Re superconductor exhibits two peaks of the absorption part of AC susceptibility in the vicinity of the critical temperature in contrary to the rhenium free sample. The first peak placed in higher temperature is related to intragranular properties while the second peak is connected with the intergranular one. The critical current densities versus temperature were calculated from AC susceptibility as well as from the magnetization loops measurements using the Bean's critical state model. The Tl–Re film exhibits the higher critical current in comparison to the rhenium free thallium based film.

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1. Introduction

The thin films of the thallium based high temperature superconductors (HTS) are promising candidates for practical applications as the coated conductors and the digital electronics [1]. From the point of view of these applications the important matter is to improve the weak links between the adjacent superconducting grains. It causes increasing the basic superconducting parameters like critical currents and irreversibility fields [2–4].

It is well known that Re-doped precursors and pellets enhance the formation and stability of the Hg-based superconductors [5–7]. The first experiments with Tl-based superconductors showed that the Re doping also improves the chemical stability of the thallium pellet and the target used for the sputtering and did not have significant impact on the phase composition and on the values of the critical temperatures [8]. However, in the next investigation we have registered a noticeable difference in the grain size of the prepared films. Using the rhenium the prepared films consist of smaller grains. The results of the microwave measurements indicate that the films without the rhenium are more convenient for the application in cryoelectronics [9]. On the other hand, the rhenium stabilizes the sputtering target thus facilitates the process of the superconducting film fabrication [10]. On the base of these results the further detailed study of the samples with and without Re is necessary.

In this paper the dispersion and absorption parts of AC susceptibility as a function of temperature for several amplitudes of the AC applied magnetic field amplitude and the magnetization as a function of applied magnetic field for several temperatures of the Tl, Re-2212 film prepared on the R-sapphire substrate with CeO_2 buffer layer were measured and analyzed.

From the absorption parts of AC susceptibility and the width of the hysteresis loops the critical current densities as a function of temperature were calculated using the Bean critical state model [11] and these results were compared with the rhenium free thallium based sample.

2. Experiment

2.1 Sample preparation

Thallium based thin films on the CeO_2 buffered R-plane sapphire substrates were prepared by a two step process. First, a Tl-free precursor was deposited on the top of the substrate using an RF magnetron sputtering. In the second step, the precursor films were annealed in the presence of the source of the thallium.

Before the deposition of the precursor films, the 60 nm thick CeO_2 buffer layer was deposited on the top of

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R-plane sapphire substrates using RF magnetron sputtering [12, 13]. The 300 nm thick precursor films of a nominal composition of $Re_{0.15}Ba_2Ca_2Cu_3O_x$ and $Ba_2Ca_2Cu_3O_x$ were deposited by RF magnetron sputtering from a single target of the same stoichiometry. The films were sputtered at a total pressure of 10 Pa in Ar atmosphere at room temperature with a deposition rate of 0.3 nm/s [14, 15].

The precursor $Re_{0.15}Ba_2Ca_2Cu_3O_x$, $BaCuO_2$ and Ca_2CuO_3 powders for the preparation of the sputtering targets and thallium sources were prepared by a sol–gel method [16]. The prepared powders were fine (diameter of the grains — 0.1–0.3 μm) and did not contain carbonates.

The thallination of the precursor films were performed in a one zone configuration where the sources of the thallium and the precursor films were kept at the same temperature. The thallination temperature and time were 860°C and 30 min, respectively. The samples were put in the furnace at the annealing temperature. We used oxygen atmosphere for the annealing. The precursor film was wrapped in a silver foil face to face with the source of the thallium and placed into an alumina crucible. The thallium sources — the unreacted pellet with $Tl_{1.85}Re_{0.15}Ba_2Ca_2Cu_3O_{10.3}$ composition — were prepared by mixing Tl_2O_3 + $Re_{0.15}Ba_2Ca_2Cu_3O_x$ in stoichiometric ratio. The precursor films of the composition $Re_{0.15}Ba_2Ca_2Cu_3O_x$ and $Ba_2Ca_2Cu_3O_x$ were sputtered on the top of the CeO_2 buffered R-plane sapphire and thallinated with the sources of thallium having $Tl_{1.85}Re_{0.15}Ba_2Ca_2Cu_3O_{10.3}$ composition. In all cases the prepared films were black, homogenous, without visible defects and their thickness is about 300 nm.

Using XRD analyses c-axis as well as a–b plane oriented Tl-1212 superconducting pure phase were identified. The morphology of the films was studied using scanning electron microscopy. The prepared Tl–Re film consists of the smaller grains with an average grain size about 0.5 μm while the Re-free film contains large epitaxial blocks (3–5 μm) [9].

2.2. Measuring techniques

The dispersion χ' and absorption χ'' parts of the AC susceptibility as a function of temperature for several AC magnetic field amplitudes were measured by a standard mutual inductance bridge operating at the frequency of 189 Hz. A Stanford SR 830 lock-in nanovoltmeter served both as a source for the AC current for the coil which produced the AC magnetic field and as a voltmeter of the bridge. The AC magnetic field was applied parallel to the a–b plane of the films. The temperature from 77 K to 110 K was monitored by a Lake Shore temperature controller employing a chromel–gold — 0.07% Fe thermocouple with an accuracy of $\pm$0.05 K. The superconducting histeresis loop of magnetization for several temperatures were done using the Quantum Design PPMS apparatus with superconducting magnet.

3. Results and discussion

The selected curves of the dispersion (χ') and absorption (χ'') parts of the $Tl_2Ba_2CaCu_2O_y$ film as a function of temperature for several AC magnetic fields are shown in Figs. 1a and b, respectively. The selected curves of

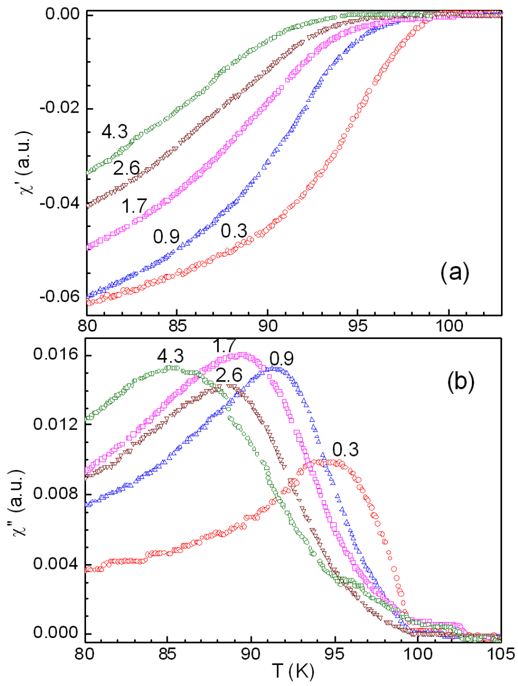

Fig. 1. The selected curves of the dispersion (a) and the absorption (b) parts of AC susceptibility as a function of temperature for different AC magnetic fields for $Tl_2Ba_2CaCu_2O_y$ thin film on R-plane sapphire substrate with CeO_2 buffer layer. The AC applied magnetic field is in Oe.

the χ' and χ'' parts of the $Tl_{1.85}Re_{0.15}Ba_2CaCu_2O_y$ film as a function of temperature for several AC magnetic fields are shown in Figs. 2a and b, respectively. The intra-granular critical temperatures were defined as the onset of transition to the superconductor state of the dispersion part of AC susceptibility. These temperatures are close to the 50% critical temperature that can be obtained from resistive measurements without the applied magnetic field e.g. the widths of the transition are sufficiently small. The critical temperature of the intergranular links was found out as the beginning of the second peak on the absorption part of AC susceptibility. These critical temperatures are collected in Table I.

One can notice that the intragranular critical temperatures of the films with and without rhenium are almost the same. The intergranular critical temperature of the Tl–Re film is almost 7 K lower than for intragranular ones. In comparison to the critical temperatures of the Hg–Re sample is several degrees higher than in rhenium free Hg samples [17].

At the low magnetic fields one can observe the two peaks of χ'' for $Tl_{1.85}Re_{0.15}Ba_2CaCu_2O_y$ (see Fig. 2b)

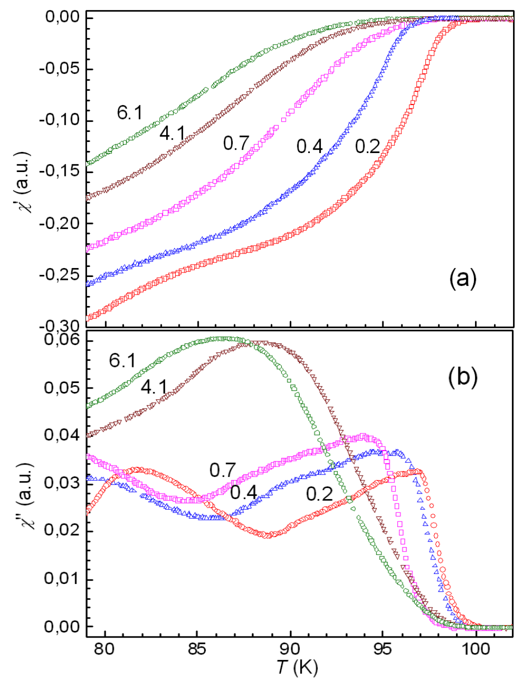

Fig. 2. The selected curves of the dispersion (a) and the absorption (b) parts of AC susceptibility as a function of temperature for several AC magnetic fields of $Tl_{1.85}Re_{0.15}Ba_2CaCu_2O_y$ thin film on *R*-plane sapphire substrate with CeO_2 buffer layer. The AC applied magnetic field is in Oe.

TABLE I

The critical temperatures of the superconducting thin films.

Sample	T_c [K]
$Tl_2Ba_2CaCu_2O_y$ on the *R*-plane sapphire with CeO_2 buffered layer	99.5 ± 0.3
$Tl_{1.85}Re_{0.15}Ba_2CaCu_2O_y$ on the *R*-plane sapphire with CeO_2 buffered layer	intra: 99.9 ± 0.3 inter: 93 ± 1.5

The first one, at the higher temperatures corresponds to the absorption of grains and the second one, at the lower temperatures corresponds to the absorption of the inter-grain links. The inter-grain peaks move to the lower temperatures when the applied magnetic field increases.

>From the absorption part of AC susceptibility the so-called contact-less critical current densities were calculated. The critical currents can be derived from the position of the absorption peaks employing Bean's critical state model and its extensions [11]. At the peak temperature the AC field amplitude H_{ac} penetrates into the sample and the current induced by the magnetic field flows uniformly throughout the entire sample and is equal to the critical current density. The Bean's model yields the following equation for the critical current density [11]:

$$J_c = \frac{2H_{ac}}{d}, \tag{1}$$

where H_{ac} is the value of the applied AC magnetic field and d is the sample dimension perpendicular to the AC magnetic field. The thikness $d = 300$ nm was taken to calculate the critical current densities for both the grains as well as the inter-granular links.

The critical currents as a function of temperature for intra-granular regions of $Tl_2Ba_2CaCu_2O_y$ and $Tl_{1.85}Re_{0.15}Ba_2CaCu_2O_y$ films were calculated using Eq. (1) and are shown in Fig. 3 (open triangles and open circles respectively). For the $Tl_{1.85}Re_{0.15}Ba_2CaCu_2O_y$ film the critical current density as a function of temperature for inter-grain regions was also calculated (see inset in Fig. 3, open diamonds). The critical temperature as well as the critical current at 77 K of $Tl_2Ba_2CaCu_2O_y$ on the sapphire with CeO_2 buffered layer are the same order as presented in the papers [18, 19]. For the sample without Re the second maximum of χ'', responsible for the absorption within the intergrain weak links, are not seen even in the very small magnetic field. That is why only the intragrain critical currents were determined for the specimen (cf. Fig. 3).

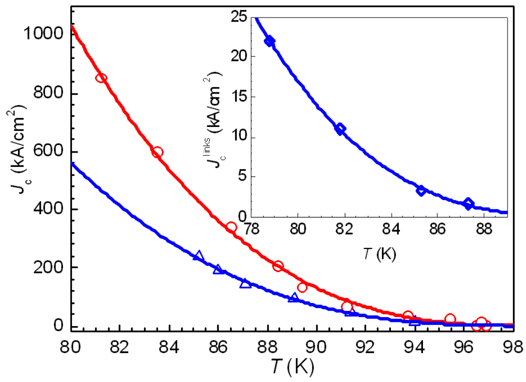

Fig. 3. The temperature dependence of the critical currents of intra-grain regions of $Tl_2Ba_2CaCu_2O_y$ (open triangles) and $Tl_{1.85}Re_{0.15}Ba_2CaCu_2O_y$ (open circles) on CeO_2 buffered *R*-plane sapphire. Inset: the inter-grain critical current of $Tl_{1.85}Re_{0.15}Ba_2CaCu_2O_y$ (open diamonds). Solid lines are the fit to Eq. (2).

The temperature dependencies of the critical current data were successfully fitted using the Ginzburg–Landau strong coupling limit approach expressed by the following equation [20]:

$$J_c = J_{c0}\left(1 - \frac{T}{T_{c0}}\right)^n, \tag{2}$$

where T_{c0} is the temperature in which the whole sample stayed superconducting. T_{c0} depends on the applied magnetic field. J_{c0} is the critical current extrapolated to 0 K. Originally, the exponent n was determined to be 1.5 but it may vary within a wide range [21, 22]. If the exponent n is greater than unity then the strong pinning and a vortex glass structure exist. In this case the function expressed by Eq. (2) has an upward curvature which is typical for HTS. If the exponent n is less than unity the pinning force is weak and limits the critical current. Then the function expressed by Eq. (2) has a downward curvature typical for low temperature superconductors. The fit of the temperature dependencies of the critical current densities using the Eq. (2) are shown in the Fig. 3 (solid lines). The fit parameters: J_{c0} and n as well as the critical currents J_c (77 K) that were calculated from Eq. (2) are collected in Table II. The temperatures T_{c0} were taken from experiment as shown in Table I.

TABLE II

Fit parameters J_{c0} and n for the temperature dependence of the critical current densities according to Eq. (2). The parameters J_c at 77 K were calculated from Eq. (2) using J_{c0} and n.

Sample		J_{c0} [MA/cm^2]	J_c(77K) [kA/cm^2]	n
$Tl_2Ba_2CaCu_2O_y$ on the R-plane sapphire with CeO_2 buffered layer		52 ± 12	845 ± 40	2.71 ± 0.18
$Tl_{1.85}Re_{0.15}Ba_2CaCu_2O_y$ on the R-plane sapphire with CeO_2 buffered layer	intra	95 ± 13	1550 ± 78	2.72 ± 0.08
	inter	5.7 ± 0.9	31.4 ± 1.6	2.95 ± 0.08

One can see that the critical currents at 77 K of $Tl_{1.85}Re_{0.15}Ba_2CaCu_2O_y$ is about two times higher than for the thallium free film. The substitution of the rhenium enhances the number of pinning centers and as a consequence it can improve the critical currents in the grains of this film. The same effects were observed in Re substitution of Hg-based superconductors [7, 17]. The critical currents of the inter-grain links is about 30 kA/cm^2 at 77 K. The intragranular critical current of Tl–Re film is almost the same as in (Hg,Re) – 1223 films reported in the paper [23]. The exponents n in the Eq. (2) are very large in comparison to other thallium-based superconductors which usually are not larger than two [3, 4, 24]. This means that the vortices are being arranged in the glassy-like structure with the strong pinning force [22]. As a consequence the large values of the critical currents were found out for the investigated thallium-rhenium thin films (see Table II).

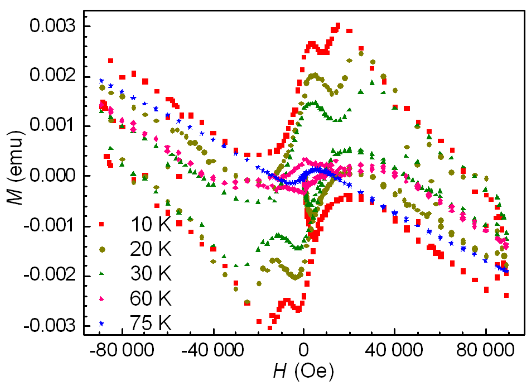

Fig. 4. The hysteresis loops of the magnetization for selected temperatures of $Tl_{1.85}Re_{0.15}Ba_2CaCu_2O_y$ film.

The hysteresis loops of the magnetization of $Tl_{1.85}Re_{0.15}Ba_2CaCu_2O_y$ film were measured and the results obtained at various temperatures are shown in Fig. 4. Take advantage of the Bean's model the critical currents were calculated using the loops width at

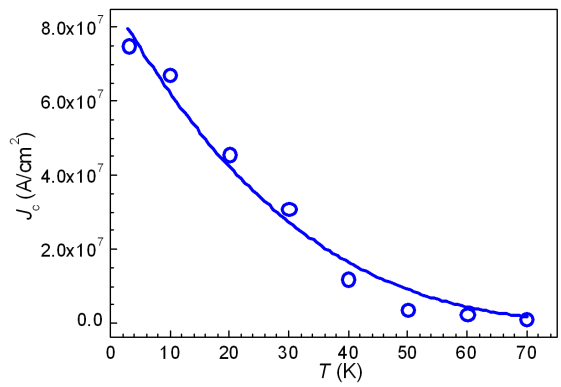

Fig. 5. The temperature dependence of the critical current density of $Tl_{1.85}Re_{0.15}Ba_2CaCu_2O_y$ film determined from the magnetization loops presented in Fig. 4. Solid line is the fit to Eq. (2).

the applied magnetic field $H = 5$ T because above this value the width of the loops does not seriously depend on the applied magnetic field. The obtained critical current density as a function of temperature are shown in Fig. 5. This dependence was well fitted using Eq. (2) and the fit parameters are as follows: $J_{c0} = 8.8 \times 10^7$ A/cm^2 and $n = 3.3$. This large exponent is consistent with the exponents that have got from fitting of the critical currents obtained from susceptibility measurements. The parameters T_{c0} were taken from experiment as shown in Table I. Using the fit parameters the critical current densities were calculated at 77 K and 4.2 K and they are $J_c = 500$ kA/cm^2 and $J_c = 75$ MA/cm^2 at 5 T applied magnetic field respectively.

4. Conclusions

The results of the paper can be concluded as follows:

1. The critical temperatures of the films with and without rhenium are almost the same.

2. The inter-grain peaks on χ'' were observed for $Tl_{1.85}Re_{0.15}Ba_2CaCu_2O_y$ film and these experimental data were used to calculate the inter-grain densities of critical currents.

3. The intra-granular critical current density obtained from susceptibility measurements of Tl–Re film is 1.6 MA/cm^2 at 77 K and it is two times higher than for the thallium free film.

4. The intra-granular critical current density obtained from magnetization loops for Tl–Re film is about $J_c = 500$ kA/cm^2 at 77 K and at 5 T of the applied magnetic field. The critical current densities obtained from both the susceptibility and magnetization loops measurements are comparable.

5. The rhenium substitution improves the critical currents of the grains due to introduction of some effective pining centers.

6. The critical current densities as a function of temperature were well described by the Ginzburg–Landau theory expressed by Eq. (2). The large exponents obtained from the fit of the Eq. (2) to experimental data suggest that the glassy-like vortex structures and strong pinning exist in the films.

Acknowledgments

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