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AC susceptibility of the $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{0.95}Ag_{0.5})_4O_{10+\delta}$ superconductor

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ABSTRACT

In this work, the temperature, magnetic field and frequency dependence of the ac susceptibility of $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{0.95}Ag_{0.5})_4O_{10+\delta}$ were studied. The superconductivity still survives even at this amount of Ag. The magnetic field dependence of the irreversibility line (IL) and the flux pinning of this compound are discussed and compared with those of low Ag content. The IL exhibits thermally activated behaviour. A collective creep of the vortex bundle also occurs for this level of doping. A crossover from a two- to a three-dimensional system is suggested at $T/T_c = 0.75$ and a magnetic field, $H_{dc} = 0.04$ T. Based on vortex glass phase transition theory, the effective pinning energy, u_{eff}, was calculated. The change in the characteristic temperature of the studied compound and that of low Ag content samples are summarised. Comparisons with similar materials are discussed.

1. Introduction

The study of the ac susceptibility of high-T_c superconductors (HTSCs) provides us information about their characteristic properties, such as the superconducting transition temperature, T_c, the inter-grain and intra-grain effects. By changing the applied dc magnetic field, H_{dc}, frequency, and temperature, one obtains useful information for understanding the mechanisms of HTSCs. In addition, the frequency dependence of the ac susceptibility is a suitable technique to throw light on vortex dynamics, and to give information about vortex mobility, flux diffusion, and relaxation mechanism using dynamic critical exponents [1–4].

Similar to our previous report on $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{1-x}Ag_x)_4O_{10+\delta}$, $0.10 \le x \le 0.30$ [5], a trail has been done to study the irreversibility line, IL when $x =$ 0.50 by the evaluation of the peak values of the imaginary parts of the ac susceptibility (χ''_{ac}). It was found that T_c of the compounds with x=0.1, 0.2 and 0.3 are 121.7 K, 126.7 K and 126.8 K, respectively. While T_c decreased with an increasing applied field, H_{dc}, two peaks were observed for field below 1.0 T. This behaviour was explained based on different pinning mechanisms. Moreover, the irreversibility temperature, T_{irr} and irreversible magnetic field (H_{irr}) which depend on x, show a crossover from two- to three-dimensional (2D–3D) as the temperature varies. Matsushita's formula, is well verified for the compounds of $0.10 \le x \le 0.30$, indicating thermally activated flux creep behaviour.

In this article, we attempted to extend our previous work for higher Ag content $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{1-x}Ag_x)_4O_{10+\delta}$, ($x$=0.50) to determine whether superconductivity still exists and provide important superconducting parameters. Motivated by the ac magnetic susceptibility, which was measured for different applied dc fields, we investigated how this high level of nonmagnetic Ag impurities influenced the irreversibility line and flux pinning of the bulk properties of the Hg-1234 system. The temperature dependent of χ''_{ac} at different frequencies are analysed by thermally activation mode.

2. Experimental methodology

A mercury-based compound, $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{0.5}Ag_{0.5})_4O_{10+\delta}$, was prepared using the conventional solid state reaction technique in a non-evacuated sealed quartz tube, as reported previously [6]. The ac susceptibility as a function of both temperature ($5\,K < T < 130\,K$) and magnetic field ($0\,Oe \le H_{dc} \le 3600\,Oe$) was carried out using a quantum design PPMS model 6000 magnetometer. At different values of the dc magnetic field, a set of measurements was taken of the sample when the temperature was cooled down to 5 K either in zero field cooling (ZFC) or field cooling (FC). The ac susceptibility was also measured with varying temperatures and frequencies.

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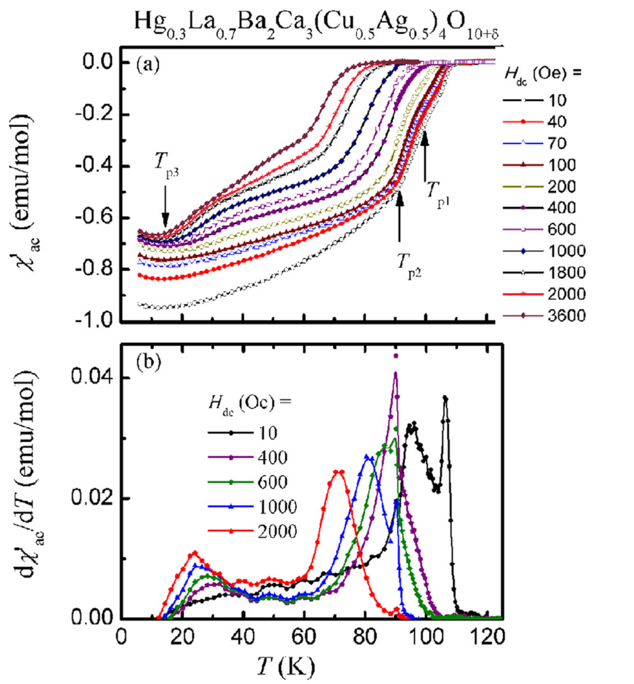

Fig. 1. (a) The real part of ac susceptibility, χ'_{ac}, in the temperature range 5-130 K at different dc fields and ac field amplitude of 1.0 Oe for $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{0.5}Ag_{0.5})_4O_{10+\delta}$. (b) The change of $d\chi'_{ac}/dT$ with temperature at some selected applied fields.

Table 1
The estimated transition peaks of the $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{0.5}Ag_{0.5})_4O_{10+\delta}$ compound at different applied fields based on the $d\chi'_{ac}/dT$ curves.

H_{dc} (Oe)	T_{p1} (K)	T_{p2} (K)	T_{p3} (K)
10	106	95	–
100	103	93	–
200	100	92	90
400	–	90	30
600	90	86	28
1000	90.5	81	25
2000	90	72	24

3. Results and discussion

Noting that the differences between the ZFC and FC measurements were very small, Fig. 1(a) depicts the field dependence of the real part of the FC susceptibility, χ'_{ac}, versus the temperature of $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{0.5}Ag_{0.5})_4O_{10+\delta}$, in the temperature range 5–125 K at different dc applied fields, H_{dc}. As shown in this figure, there is an abrupt change in the diamagnetic susceptibility with temperature and H_{dc}. Different characteristic temperatures are also characterised as T_{p1}, T_{p2}, and T_{p3}. Both T_{p1} and T_{p2} shift to lower temperatures with increasing H_{dc}. While T_{p2} could no longer be identified, T_{p1} spread out with increasing applied H_{dc}. Also, an increase in χ'_{ac} at the lower temperature (T_{p3}) was noticed at $H_{dc} > 100$ Oe. This behaviour can be seen from the variation of $d\chi'_{ac}/dT$ with temperature as shown in Fig. 1(b). The estimated values of these peaks are given in Table 1. In addition, the values of χ'_{ac} at $T \approx 5$ K suggest that $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{0.5}Ag_{0.5})_4O_{10+\delta}$ also exhibits Meissner state (MS), which decreases with the increase in H_{dc}. Even the applied field reaches 3600 Oe, χ'_{ac} remained negative. As seen in Fig. 1(b), T_c decreased with increasing of H_{dc}. The observed upturn in χ'_{ac} could be attributed to the relative increase in the magnetic contribution of the Cu planes.

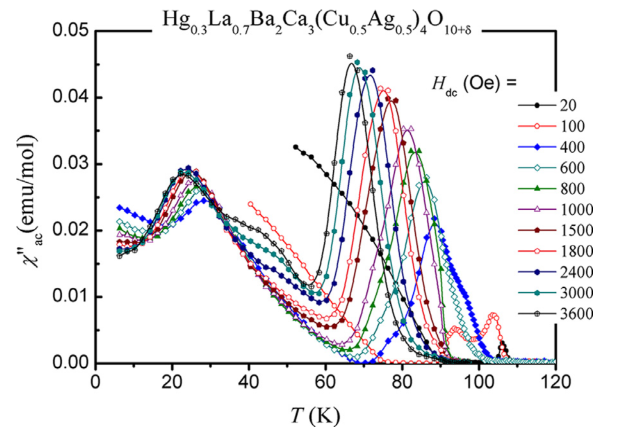

Fig. 2. The imaginary part of ac susceptibility, $\chi''_{ac}(T)$, in the temperature range 5-130 K at different dc fields and ac field amplitude of 1.0 Oe for $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{0.5}Ag_{0.5})_4O_{10+\delta}$, compound.

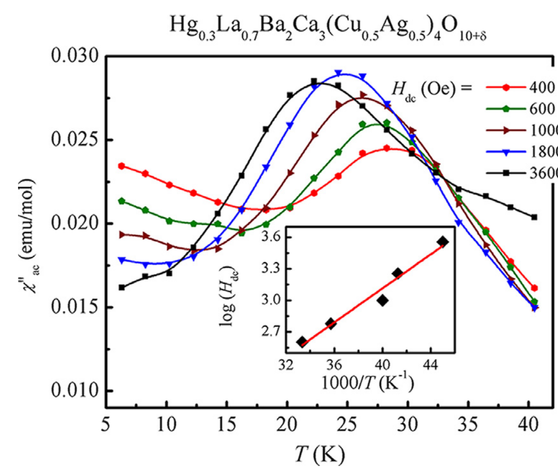

Fig. 3. The imaginary part of ac susceptibility, $\chi''_{ac}(T)$, in the temperature range 5-40 K at different dc fields and ac field amplitude of 1.0 Oe for $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{0.5}Ag_{0.5})_4O_{10+\delta}$, compound. The inset shows the variation of log (H_{dc}) against 1000/T, where the solid red line represents the fit according to Eq. 1.

Fig. 2 displays the temperature dependence of the imaginary part of the ac susceptibility, χ''_{ac}, measured at different H_{dc}, and ac amplitude of 1.0 Oe. Two peaks, T_{P1} (nearest T_c) and T_{P2}, were observed consistent with the inflection points in $\chi'_{ac,}$ (see Fig. 1(a)). These double peaks can be attributed to intergranular and intragranular effects [5] or different pinning mechanisms consistent with that observed for fluorine, F-doped Hg-1223 [7]. The increase in H_{dc} caused both peaks to shift to lower temperatures, with T_{P1} shifting faster than T_{P2}, until they completely merged into a single peak at $H_{dc} \geq 200$ Oe. The intensity of T_{P1} and T_{P2} initially increased up to $H_{dc} \sim 200$ Oe, and their relative intensities started to decrease and then increase in width until they completely merged at $H_{dc} \approx 400$ Oe. The resulting single peak is associated with T_{P1}, T_{P2} or both. A third broad peak T_{P3}, showing hysteresis, was found at $T \sim 30$ K. This peak was well developed at a field value $H_{dc} = 400$ Oe. Furthermore, it shifted slowly towards lower temperatures with increasing H_{dc} as seen in Fig. 3. The inset in Fig. 3 shows the thermally activated behaviour of the H_{dc} shift of T_{p3}:

$$H_{dc} \approx \exp\left(E/K_B T\right) \quad (1)$$

where K_B is Boltzmann's constant and E is the activation energy. Within the dc applied magnetic field range 400 Oe $\leq H_{dc} \leq$ 3600 Oe, the E value of the studied compound was 0.07 eV, which is higher than that of low Ag content sample [5].

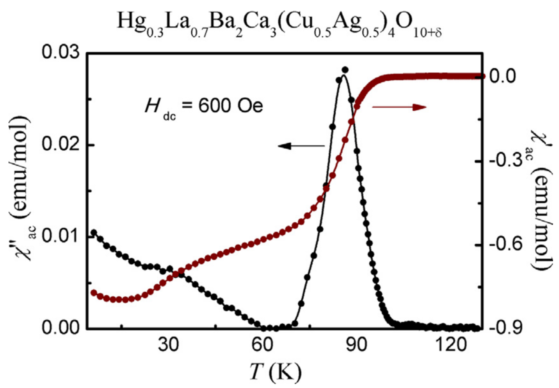

Fig. 4. The temperature dependence of $\chi'_{ac}(T)$ (right side) and $\chi''_{ac}(T)$ (left side) at $H_{dc} = 600$ Oe for $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{0.5}Ag_{0.5})_4O_{10+\delta}$, compound.

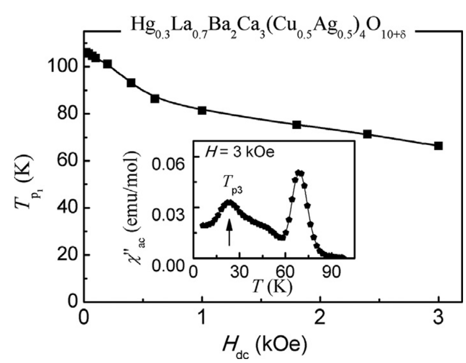

Fig. 5. The temperature dependence of the loss peak, T_{p1}, with applied H_{dc} for $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{0.5}Ag_{0.5})_4O_{10+\delta}$ compound. The inset displays the loss peaks at $H_{dc} = 3000$ Oe applied field.

The behaviour of $\chi'_{ac}(T)$ (right side), and $\chi''_{ac}(T)$ (left side), at 600 Oe is presented in Fig. 4. It was noted that $\chi''_{ac}(T)$ had peaks that coincided with the inflection points in the $\chi'_{ac}(T)$ curves (see Fig. 1(a)) similar to the results of the compounds with $0.10 \le x \le 0.30$ [5]. It is noticed that the two peaks were completely merged and T_c decreased with increasing H_{dc}. Peak losses occasionally cannot be easily distinguished in polycrystalline HTSCs. Similar to other HTSCs, all impurities located in the Cu planes increased electron scattering and then T_c decreased. It is also known that in conventional superconductors, magnetic impurities act as pair breakers and destroy the superconductivity, while nonmagnetic impurities affect T_c to a much lesser extent [8]. In addition, the scattering of impurities was complicated and depended on different parameters in the case of HTSCs. The outcomes of different studies concerning the effect of various impurities at the Cu site in many superconducting systems [9–11] emphasised that either the suppression or the decrease in superconductivity depended on the type of substituted cation. For instance, the superconductivity of the Ru-1212 HTSC was suppressed particularly quickly when low levels of nonmagnetic impurities were added at the copper site [12].

Fig. 5 displays the temperature dependence of T_{p1} with increasing magnetic field for $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{0.5}Ag_{0.5})_4O_{10+\delta}$ compound. As seen, T_{p1} decreased by about 40 K when H_{dc} changed from 0.01 to 3 kOe. Moreover, the inset of this figure depicts the decrease of T_c with increasing H_{dc} and shows the existence of T_{p3} even at $H_{dc} = 3$ kOe. This peak, T_{P3} can be attributed to intra-granular losses [5]. This intra-granular peak shift reflects the field-dependent behaviour of the thermal activation energy [13].

The irreversibility line (IL) $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{0.5}Ag_{0.5})_4O_{10+\delta}$ is determined by evaluating the peak values of χ''_{ac}. As mentioned

Table 2
Fitting parameters of $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{1-x}Ag_x)_4O_{10+\delta}$, $0.10 \le x \le 0.50$, according to Eq. 2.

x	H_{dc}(Oe)	T_c (K)	n_I ($H_{dc} < 400$ Oe)	n_{II} ($H_{dc} > 400$ Oe)
0.10	400	121.7	1.15	2.49
0.20	500	126.7	1.16	2.57
0.30	600	126.8	1.10	2.59
0.50	600	106.0	0.75	2.74

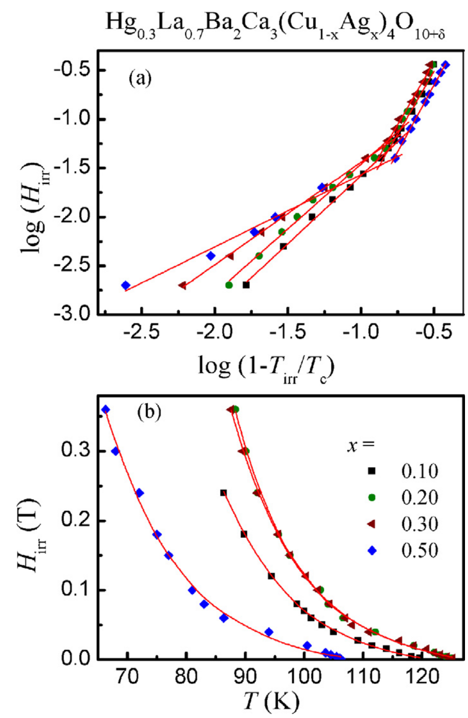

Fig. 6. (a) Analysis of the irreversibility line for $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{1-x}Ag_x)_4O_{10+\delta}$, $0.10 \le x \le 0.50$, based on creep of pancake vortices Eq. (2) and (b) Analysis of the irreversibility line based on Matsushita's formula. The solid red lines represent the best fit according Eq. 3.

previously, double loss peaks near T_c were observed in $\chi''_{ac}(T)$ and at $H_{dc} \le 200$ Oe. Above this value of H_{dc}, these double loss peaks merged into one as shown in Fig. 2. A similar behaviour has been observed for the compounds of lower content of Ag. It was found that the IL follows a power law:

$$H_{irr} = \left[\frac{1 - T_{irr}}{T_c(0)}\right]^n \tag{2}$$

where n is related to the degree of anisotropy in superconducting materials [14–16]. Also, the exponent n varies from one HTSCS to another. Blatter and Ivlev reported that the value of n depends on the reduced temperature as well as on the value of the quantum fluctuation [17]. According to Eq. 2, the fitting parameters for all compounds are given in Table 2. While T_c and n_I ($H_{dc} < 400$ Oe) decreased with increasing of Ag content, n_{II} ($H_{dc} > 400$ Oe) increased. This means the degree of anisotropy of Hg-1234 system increased when the content of doping at cupper planes increased. The manner of changing either n_I or n_{II} with doing needs theoretical interpretations.

A logarithmic plot of H_{irr} versus $\log(1-T_{irr}/T_c)$ for $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{1-x}Ag_x)_4O_{10+\delta}$, $0.10 \le x \le 0.50$, is shown in Fig. 6(a). It is noticed that the difference between the samples is small in the high temperature region. For x = 0.10 and 0.20, a crossover at $T_{irr}/T_c \approx 0.86$ from a 2D to a 3D system was observed, where a distinct break was noted. On the other hand, such crossover shifts at ≈0.75 for the compounds of $x = 0.30$

Table 3
Free fitting parameters of $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{1-x}Ag_x)_4O_{10+\delta}$, $0.10 \le x \le 0.50$, samples according to Eq. 3

x	k	m	γ
0.10	76.3 ± 0.4	0.37 ± 0.06	1.13 ± 0.02
0.20	80.9 ± 0.5	0.25 ± 0.04	1.18 ± 0.01
0.30	81.6 ± 0.8	0.20 ± 0.06	1.21 ± 0.02
0.50	276 ± 1.2	0.69 ± 0.08	1.69 ± 0.05

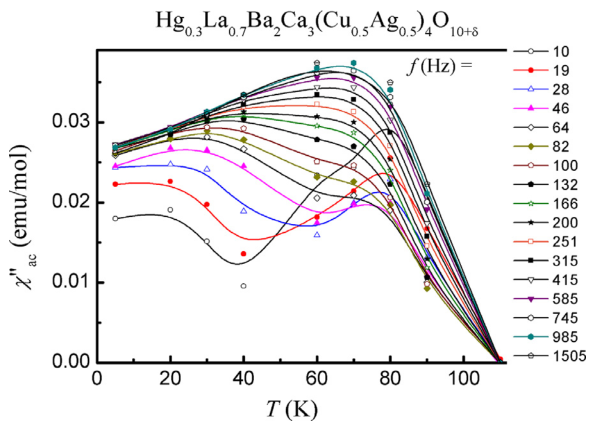

Fig. 7. Temperature dependence of $\chi''_{ac}(T)$, at different frequencies for $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{0.5}Ag_{0.5})_4O_{10+\delta}$ compound.

and 0.50. These signify the possibility of another mechanism. In other words, two linear parts with different slopes n_I and n_{II} were identified. The values of n_I and n_{II} of this investigated sample are given in Table 2. For the sample of $x = 0.50$, the slope of region I gives $n_I = 0.75$ for $H_{irr} < 0.04$ T while the slope of region II gives $n_{II} = 2.74$ for $H_{irr} > 0.04$ T. We compared the values of n_I and n_{II} for $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{0.50}Ag_{0.50})_4O_{10+\delta}$, with those of $0.10 \le x \le 0.30$ compounds [5]. When n_I decreased with increasing Ag content, n_{II} increased.

Matsushita model is also applied to fit the data of $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{0.5}Ag_{0.5})_4O_{10+\delta}$ as [18,19]:

$$H_{irr} \approx \left(\frac{k}{T}\right)^{4/(3-2\gamma)} [1 - \left(\frac{T}{T_c}\right)^2]^{2m/(3-2\gamma)} \tag{3}$$

where k, m and γ are numerical parameters that depend on the creep free pinning force and current density [7]. The resulting parameters of the fit for the compound of $x = 0.50$ along with that of other x are listed in Table 3, and the fitted data are shown in Fig. 6(b). This graph clearly indicates the low value of the irreversible field. It was noted that the values of k, m and γ depend on the Ag content that was substituted for Cu in Hg-1234 [5].

Fig. 7 depicts the temperature dependence of χ''_{ac} on selected frequencies. Below T_c, more than one peak representing energy losses was observed. At a frequency of $f < 64$ Hz, two peaks can be distinguished: one at the low temperature, T_{p3}, and the second at T_{p2} (see Fig. 1(b) and Fig. 2). For $f > 64$ Hz, these two peaks merged and the intensity of the merged peak increased with increasing f. It was proposed [20] that the resistance arising from thermally activated flux motion was the same as the case of the thermally activated phase motion in a heavily damped Josephson junction [21]. The behaviour of the resulting loss peak was dependent on temperature, magnetic field history and AC frequency.

The effective pinning energy, u_{eff}, can be calculated as [22]:

$$u_{eff} = TK_B\frac{T_c - T}{T_c - T_g}, \tag{4}$$

where T_g is the glass transition temperature. Fig. 8(a) shows the variation in u_{eff}/K_B (left side) with T_{p2} and K_BT (right side). It was

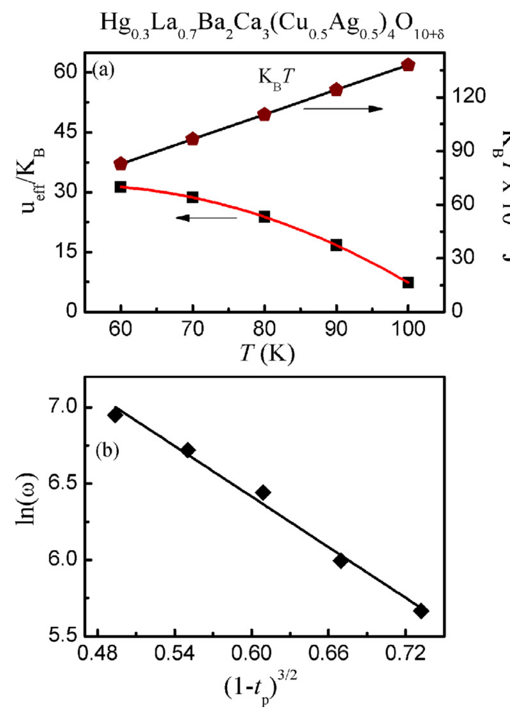

Fig. 8. (a) The temperature dependence of effective pinning energy, u_{eff}, for $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{0.5}Ag_{0.5})_4O_{10+\delta}$ sample. The red solid line is a fit with Eq. (4) while the black solid line is the thermal activation energy in unit of K_B. (b) Plot of $\ln(\omega)$ versus $(1-t_p)^{3/2}$ for the studied sample with ac amplitude 1 Oe.

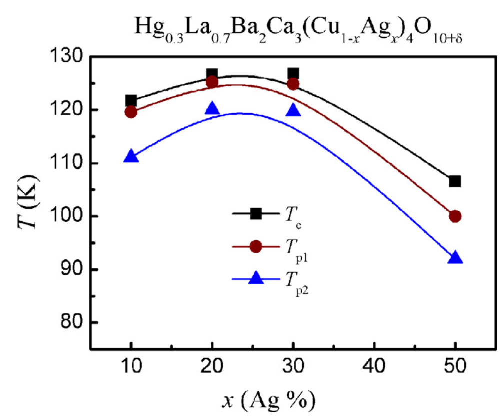

Fig. 9. The variation of the characteristic peaks; T_c, T_{p1} and T_{p2} against x (Ag%) of $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{1-x}Ag_x)_4O_{10+\delta}$ compounds.

noted that u_{eff} decreased with increasing temperature. The fitting, according to Eq. 5, yielded the value of $T_g = 17.42$ K. This means that the frequency-dependent vortex dynamics follow a model using the critical slowing down of the spin glass and the extended thermal activation model. At constant AC amplitude ($H_{ac} = 1$ Oe) and taking T_{p2} into account, the angular frequency ($\omega = 2\pi f$) is related to $(1 - t_p)^q$, where the value of q is chosen to be 3/2 similar to other superconducting materials [23,24]. The peak temperature of χ''_{ac} can be written as:

$$(1 - t_p)^{3/2} \approx \frac{H_{ac}^{\mu}H^n}{A}\ln\left(\frac{\omega_p}{\omega_o}\right) \tag{5}$$

where $H = H_{dc} + H_{ac}$, t_p is T_{p2}/T_c, and ω_o is the characteristic angular frequency. The results of the plot of $\ln(\omega)$ against $(1 - t_p)^{3/2}$ for the $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{1-x}Ag_x)_4O_{10+\delta}$ compound are shown in Fig. 8(b).

It is useful to summarise the results of this work along with our previously published data in Fig. 9. We note that superconductivity still survives even until x = 0.50 Ag. These results are in contrast to other HTSCs such as Ru-1212 [12], because the low level of nonmagnetic impurities is enough to suppress superconductivity. This difference can be attributed to the large distance between the copper planes in the Hg-1234 system. The other peaks, T_{p1} and T_{p2}, decreased for the studied compound compared with those of lower x (Ag) samples.

4. Conclusions

The Ag-doped Hg-1234 of $x = 0.5$, $Hg_{0.3}La_{0.7}Ba_2Ca_3(Cu_{0.5}Ag_{0.5})_4O_{10+\delta}$, is still a superconductor with $T_c = 106.57$ K. It was observed that T_c decreased with an increasing applied dc field. Study of the irreversibility line of this level of Ag-doping in the Hg-1234 superconductor showed different pinning mechanisms. Thermally activated pinning mechanism was verified according to Matsushita's formula. A crossover from a 2D to a 3D system is possible around a 0.04 T applied dc field and $T/T_c \approx 0.75$. The effective pinning energy, u_{eff}, decreased with increasing temperature because the thermal energy increased.

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