Magnetic relaxation and the flux diffusion barrier for $TlSr_2Ca_2Cu_3O_y$ doped with Pb and Ba determined by complex ac susceptibility measurements

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Complex ac susceptibility has been measured on the $TlSr_2Ca_2Cu_3O_y$ superconductor doped with Pb and Ba, which was prepared by the partial melt processing. The flux creep barriers $U(J,B,T)\propto J^{-\mu}B^{-0.75}(1-T^2/T_c^2)^{1.5}$ with $\mu=0.64$ and current decay $S=-d\ln J/d\ln[t/t_0]=1/(\mu\ln[t/t_0])$ with $t_0=2\times10^{-6}$ s have been determined at various temperatures, ac frequencies f, dc fields B, and current densities J in a time window 10^{-5} s-10^{-3} s, which extends considerably the very limited one (typically 1 s-10^4 s) used by the conventional dc magnetometer. The result that $U(J)\propto J^{-\mu}$ is an evidence of the vortex-glass phase or collective-creep mechanism predicted by theories from the ac susceptibility measurements.

It is well known that the Tl-based superconductors have very high T_c ($=125$ K), which is attractive for both technical application and an academic study standpoint. Unfortunately, the Tl-2223 ($Tl_2Ba_2Ca_2Cu_3O_x$) material of 125 K T_c is a layered compound and has a critical current density J_c that decreases too rapidly with increasing magnetic field, e.g., J_c is drastically reduced in a field of, say, 1 T at liquid nitrogen. However, the Tl-1223 ($TlSr_2Ca_2Cu_3O_y$) phase which has only one insulating Tl-O plane per unit cell, has an important advantage of stronger flux pinning, resulting probably from a shorter distance between the neighbor conducting Cu-O planes and less anisotropy.[1] Consequently, the Tl-1223 material has a higher J_c than Tl-2223 in applied fields of about 1 T. Furthermore, it has been demonstrated that flux pinning centers can be introduced into the Tl-1223 phase using its derivative (Tl/Pb)-(Sr/Ba)-Ca-Cu-O with the nominal composition of (0.5/0.5)-(1/6/0.4)-2-3.[2] However, the J_c of the Tl-1223 is still not high enough in fields and the underlying pinning mechanism is not understood so

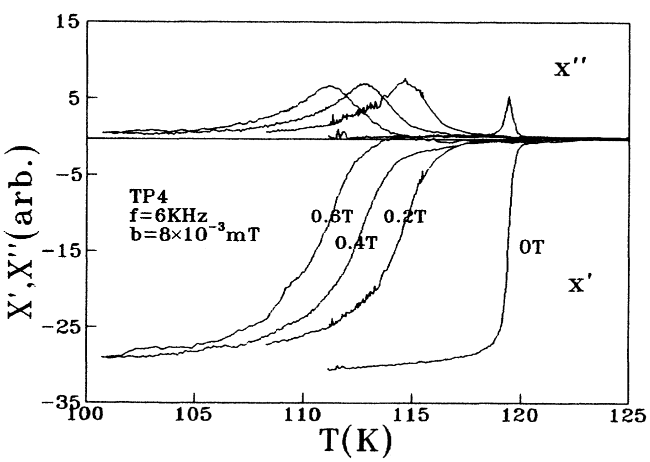

FIG. 1. The real (X') and imaginary (X'') parts of acs in various applied dc fields B and at frequency f given in the figure for the specimen.

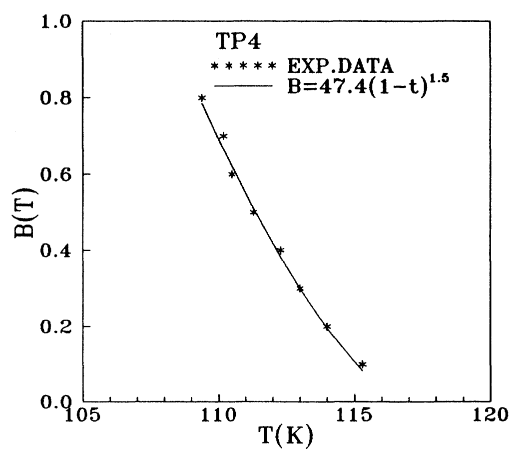

FIG. 2. The irreversibility line of the specimen, where the solid line fits the experimental data (stars) well with reduced temperature $t=T/T_c$, and $T_c=117$ K is a fitting parameter. $f=6$ KHz.

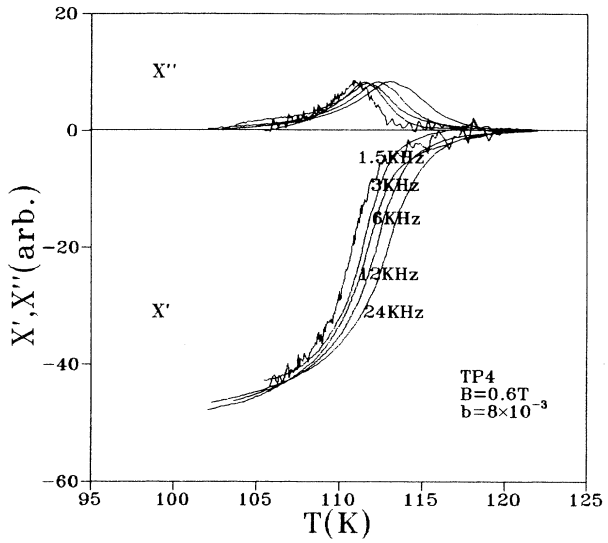

FIG. 3. The typical frequency effect of the acs $X'(f,T)$, $X''(f,T)$ at dc field B, ac field amplitude b and frequencies f given in the figure, showing effect of frequency on X'' peak temperature T.

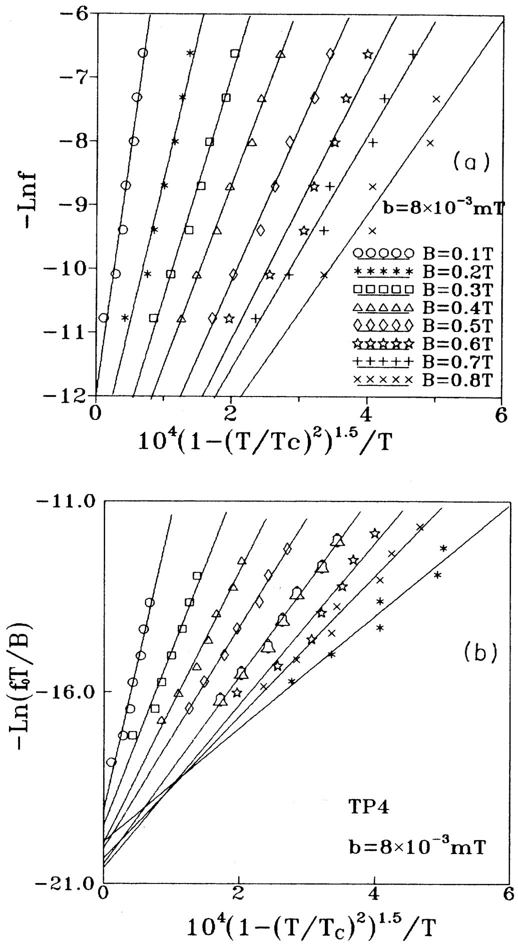

FIG. 4. (a) The experimental data $f(B,T)$ (the symbols) and their fitting straight lines (the solid lines) from the measurements as shown in Figs. 1 and 3, a summary on the effect of dc field B and ac field frequency f on the X'' peak temperature T at an ac field amplitude b given in the figure. (b) The data shown in (a) plotted in another way.

far. The dc magnetization experiments are widely used to study flux pinning and the thermally activated flux motion to estimate the barrier height U impeding flux motion. However, the usual relaxation experiments using conventional magnetometers are restricted to times larger than tens of seconds because of instrumental integral times. This precludes getting information occurring at earlier times. A lot of effort has been made, e.g., Gao *et al.* and Brawner *et al.* were able to measure relaxations at times larger than about 1 s.[3,4] The ac susceptibility (acs) technique is a powerful method not only in characterizing high-T_c superconductivity, but in studying flux dynamics because of its ability to get information in a time window of 10^{-2}–10^{-4} s.[5,6] In this paper we report recent experimental results on the preparation of partial melt processing and on flux dynamics for Tl-1223 doped with Pb and Ba superconductor with the ac technique.

The specimen in the present study was synthesized by means of partial melt processing. A precursor powder with a nominal composition of $(Sr_{0.2}Ba_{0.2})_2Ca_2Cu_3O_x$ was prepared by mixing $BaCo_3$, $SrCO_3$, and CuO, and sintering at 900 °C for 30 h. Then the powder Sr-Ba-Ca-Cu-O was pulverized, mixed with Tl_2O_3 and PbO according to the nominal composition of $(Tl_{0.5}Pb_{0.5})(Ba_{0.2}Sr_{0.8})_2Ca_2Cu_3O_y$ and pressed into long blocks. This was sintered at 940 °C for 10 h, followed by furnace cooling to room temperature. Slabs cut from the blocks were partially melted at 1010 °C for about 15 min, then kept at 940 °C for 10 h, followed by furnace cooling to room temperature. A specimen named TP4 hereafter was ready. Magnetic measurements were carried out in an ac susceptometer with sensitivity of 10^{-7} emu at different ac frequencies f. An applied magnetic field $B(t)=B+b\cos[2\pi ft]$ was used, where the dc field $B \gg b$.

Shown in Fig. 1 are typical $X'(B,T)$ and $X''(B,T)$

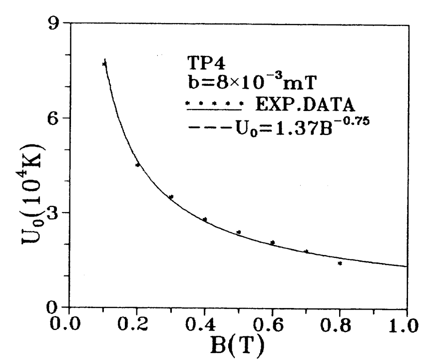

FIG. 5. The dependence of U_0 on dc fields B at $T=0$ K and ac field amplitude b given in the figure. The stars (data) are the slopes of the straight lines in Fig. 4(a), the solid curve is a fitting one.

curves where X' and X'' are the real and imaginary parts of the acs, respectively, and where the superconducting onset temperature $T_{c0}=120$ K and the temperature T_p at which the X'' peak occurs is about 119 K ($B=0$). From Fig. 1 and those measurements not shown here for clarity, the irreversibility line has been reconstructed and shown in Fig. 2, where T_p has been denoted by T for simplicity, and where the best fitting curve of the data is illustrated by a solid line $B=47.4\times(1-t)^{1.5}(T)$ with $t=T/T_c$ and $T_c=117$ K (a fitting parameter). Shown in Fig. 3 are typical $X'(f,T)$ and $X''(f,T)$ curves which display the effect of frequency f on the X'' peak temperature T. Similar measurements have been conducted at various dc fields B. In Fig. 4(a) we summarize the measured data from such measurements with different symbols to show the effects of frequency f and dc field B on the X'' peak temperature T in terms of $f(b,B,T)$ at $b=8$ μT, where the plot $-\ln f$ versus $[1-(T/T_c)^2]^{1.5}/T$ with the common fitting parameter $T_c=117$ K has been made. In Fig. 4 it is clear that those solid straight lines are fitting the data very well and can be described in terms of $f=(1/t_0)e^{-U_0(b,B)U(T)/T}$, or the following linear equation

$$-\ln[ft_0]=U_0(b,B)[U(T)/T]\ , \quad (1)$$

where $f_0t_0=1$ and $-\ln[ft_0]$ are these line's intercepts on the $Y(=-\ln f)$ axis and hence determined by the variable $X\{=[U(T)/T]\}=0$ (i.e., $T=T_c$) and $U(T)=(1-T^2/Tc^2)^{1.5}$, U_0 are the slopes of these straight lines. From the intercepts in Fig. 4(a) we determined 10^{-6} s $<t_0<5\times10^{-6}$ s with a mean value $t_0=2\times10^{-6}$ s. Figure 4(b) shows $T/(Bt_0)$ is nearly a constant, which means $t_0\propto T/B$. Figure 5 illustrates the slopes of the straight lines in Fig. 4(a) by stars and the best fitting function $U_0(B)$ by the solid line. The above results show the barrier is $U(b,B,T)=U_0'(b)U_0(B)U(T)$. The $U_0'(b)$ can be obtained by the measurements with data as shown in Fig. 4 but at different ac amplitude b (i.e., amplitude effect). Shown in Fig. 6 is an example of such amplitude effect data at $f=6$ KHz. We show in Fig. 7 the experi-

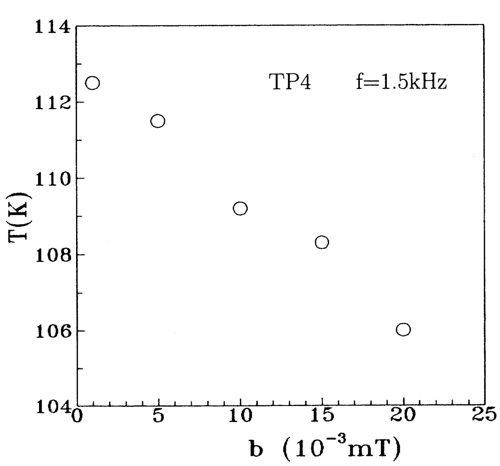

FIG. 6. The amplitude effect of X'' peak temperature T, showing the effect of ac field b on the X'' peak temperature T at frequency given in the figure.

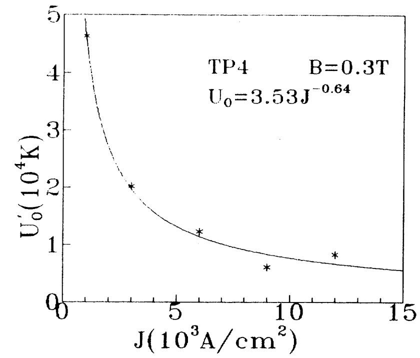

FIG. 7. The flux diffusion barrier heights $U(J)$ at $T=0$ K and dc field given in the figure. The stars are experimental data, see text; the solid curve depicted by function U_0 given in the figure is a fitting curve.

mental $U_0'(J)=U(J,0.3\text{ T},0\text{ K})$, i.e.,

$$U_0'(J)=A_jJ^{-\mu}\ , \quad (2a)$$

where $\mu=0.64$, $A_j=3.53\times10^4$ K $[10^3\text{ A/cm}^2]^{\mu}$, and current density

$$J=Cb/d \quad (2b)$$

has been used with $C=1$ and d being a half grain dimension. In general, in a sample with thickness $2d$ in an ac applied field with amplitude b, a screen current will be induced with penetration depth D. At X'' peak $D=d$, which means the screen current fronts from the opposite sides meet at the sample center, and hence the mean current density $J=Cb/d$. For a thin slab in an ac field parallel to its wider faces, $C=1$, otherwise C is 1 in order of magnitude. We here take the thin slab approximation as most cases in references, where $J=J_c$ for the static Bean critical state case and J is nearly a constant as Eq. (2b) for the self-organized criticality case as shown by Vi-

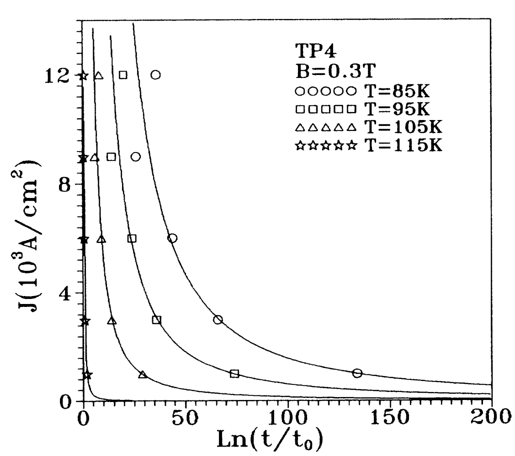

FIG. 8. The plot J versus $\ln[t/t_0]$ according to data (symbols) and Eq. (3) (the solid lines) in the text at various temperatures T and dc field B given in the figure, showing current relaxations in the time window 10^{-3} s-10^{-5} s determined by the ac technique.

nokur, Feigel'man, and Geshkenbein.[7] Therefore, the sample's grains are slabs with mean thickness of $2d$(= the mean grain size), which is about 2 μm according to our scanning electron microscopy (SEM) observations and $C=1$ in Eq. (2b). It should be emphasized that Eq. (2a) implies the flux motion barrier height $U(\propto J^{-\mu})$ will increase infinitely as J decreases. This is evidence from the present acs experiment, showing the collective creep or vortex-glass phase predicted by theories[8-10] operating in the Tl-1223 superconductor. Since $ft=1$ at the X'' peak, it is easy to obtain the current decay function $J(t)$ by substituting Eq. (2a) into (1):

$$J(t)=A(\ln[t/t_0])^{-1/\mu}=A(-\ln[ft_0])^{-1/\mu}\ , \qquad (3)$$

where $A=[A_j U(T)/T]^{1/\mu}(B=0.3\ \text{T})$, and μ is called the vortex-glass exponent. Figure 8 displays the plot J versus $\ln[t/t_0]$ according to the data and Eq. (3) at several temperatures. The $ft=1$ means that we have carried out a relaxation experiment in a time window 1 ms (1 KHz)−20 μs(50 KHz). From Eq. (3) the normalized relaxation rate $S=-d\ln J/d\ln t=1/(\mu\ln[t/t_0])$, which confirms that S is universal according to the collective creep or vortex-glass theories and the dc relaxation experiments.[7-11] What is more important is the flux relaxation experiment has been extended to a much earlier time window. However, with $t/t_0=10^3$ and $\mu=0.64$, the $S=0.16$, which is larger than for $YBa_2Cu_3O_x$ (Ref. 11) obtained from dc relaxation measurements.

In summary, we have carried out a relaxation experiment using the acs method and overcame the usual difficulty in conventional relaxation measurements of having only a very limited time window, 1 s-10^4 s, by extending the latter to the earlier one, 2×10^{-5} s-10^{-3} s. This increases the dynamic range of $\ln[t/t_0]$ considerably to 10^{-4} s-10^4 s. For Tl-1223 we show the important parameter $t_0(d=1\ \mu\text{m},\ T=117\ \text{K},\ B=1\ \text{T})$ is about 2×10^{-6} s. We have also determined the important flux pinning barriers $U(J,B,T)\propto J^{-0.64}B^{-0.75}(1-T^2/T_c^2)^{1.5}$, and the flux relaxation $J\propto(\ln t/t_0)^{-1/0.64}$. Therefore, we have presented independent evidence showing the vortex-glass phase or the collective creep mechanism in the high-T_c superconductors.

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[1] R. S. Liu, D. N. Zheng, J. W. Loram, K. A. Mirza, and A. M. Campbell, Appl. Phys. Lett. **60**, 1019 (1992).
[2] Tomoichi Kamo, Toshiya Doi, Atsukoo Soeta, Toyotaka Yuasa, Naomi Inoae, Katsuzo Aihara, and Shi-Pri Matsuda, Appl. Phys. Lett. **59**, 3186 (1991).
[3] L. Gao, Y. Y. Xue, P. H. Hor, and C. W. Chu, Physica C **177**, 438 (1991); Y. Y. Xue, L. Gao, Y. T. Ren, W. C. Chen, P. H. Hor, and C. W. Chu, Phys. Rev. B **44**, 12 029 (1991).
[4] D. A. Brawner, N. P. Ong, and Z. Z. Wang, Phys. Rev. B **47**, 1156 (1993).
[5] C. J. van der Beek, V. B. Geshkenbein, and V. M. Vinokur, Phys. Rev. B **48**, 3393 (1993).
[6] G. Blatter, M. N. Feigel'men, V. B. Geshkenbein, A. I. Larkin, and V. M. Vinokur (unpublished).
[7] V. M. Vinokur, M. N. Feigel'man, and V. B. Geshkenbein, Phys. Rev. Lett. **67**, 915 (1991).
[8] M. N. Feigel'man, V. B. Geshkenbein, A. L. Larkin, and V. M. Vinokur, Phys. Rev. Lett. **63**, 2303 (1989).
[9] T. Natterman, Phys. Rev. Lett. **64**, 2454 (1990).
[10] M. P. A. Fisher, Phys. Rev. Lett. **62**, 1415 (1989).
[11] A. P. Malozemoff and M. P. A. Fisher, Phys. Rev. B **42**, 6784 (1990).