Superconducting fluctuations in Tl-based single crystals: Evidence of universal features and derivation of basic parameters

A. Wahl, A. Maignan, C. Martin, V. Hardy, J. Provost, and Ch. Simon
Laboratoire Crismat, CNRS URA 1318, Institut des Sciences de la Matière et du Rayonnement, 6 Boulevard du Maréchal Juin, 14050 Caen Cédex, France
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The critical fluctuations of the magnetization near the $H_{c2}(T)$ line have been studied in four different single crystals of Tl-based compounds ($Tl_{0.5}Pb_{0.5}Sr_2CaCu_2O_7$, $Tl_2Ba_2Ca_2Cu_3O_{10}$, $Tl_2Ba_2CaCu_2O_8$, and $TlBa_2Ca_2Cu_3O_9$) with the magnetic field applied along the c axis. It is shown that the magnetization can be scaled in reduced units according to the two-dimensional scaling developed by Ullah-Dorsey and Tesanovic *et al.*, exhibiting the universality of the fluctuations. From this analysis, we deduce the values of the Ginzburg parameter (G_i), the coherence length [$\xi_{ab}(0)$], and the penetration depth [$\lambda_{ab}(0)$] of each phase. The values of the Ginzburg fluctuation parameter in the different compounds are put in relation with the relative location of their irreversibility lines.

INTRODUCTION

A remarkable feature of high-critical-temperature superconducting oxides is the effect of thermodynamic fluctuations near the superconducting phase transition. These effects are generally small, but they can be measured especially in compounds of low dimensionality such as high-temperature superconductors (HTSC's) where this phenomenon has drawn a great interest and many theories have been developed.

In that way, Ullah and Dorsey[1] showed scaling behaviors of the temperature and field dependence of thermodynamical quantities in this critical fluctuation region; Bulaevskii, Ledvij, and Kogan[2] and Kogan *et al.*[3] have proposed a model predicting a spontaneous excitation of flux lines in weakly Josephson-coupled superconductors at a temperature $T < T_{c0}$ as well as a crossover of magnetization at the characteristic temperature T^*. This theory includes the contribution of thermal distortions to the free energy and more particularly to the temperature and field dependence of the magnetization below T_c. Besides, many attempts have been made to interpret the scaling behaviors of the transport and thermodynamic quantities by various approaches based on Ginzburg-Landau fluctuation theory.[4] A different approach to this problem has been developed by Tesanovic *et al.*;[5] these latter authors have obtained an explicit form of the scaling function for the fluctuations contribution to thermodynamic quantities in two-dimensional (2D) superconductors on the basis of the lowest-Landau-level approximation. Up to now, several experiments have pointed out the importance of these effects in HTSC's, particularly in Bi-based components.[6-11] Welp *et al.*[12] showed, in $YBa_2Cu_3O_{7-\delta}$, that the critical fluctuation contribution to magnetization displays a 3D scaling behavior of the form $M/(HT)^{2/3}$ versus $[T-T_c(H)]/(HT)^{2/3}$ near the $H_{c2}(T)$ line, whereas Li *et al.*,[13] in $Bi_2Sr_2Ca_2Cu_3O_{10}$, described the contribution to the magnetization with the 2D version of the scaling $M/(HT)^{1/2}$ versus $[T-T_c(H)]/(HT)^{1/2}$ where $T_c(H)$ is the mean-field transition temperature.

The present paper is a detailed study of magnetic measurements for a set of high-quality superconducting thallium single crystals of the series $Tl_nBa_2Ca_{m-1}Cu_mO_{2(m+1)+n}$ (where $n=1$, $m=2$, and $n=2$ with $m=2,3$) and for the lead-substituted thallium monolayer $Tl_{0.5}Pb_{0.5}Sr_2CaCu_2O_7$, in the critical region near the $H_{c2}(T)$ line. It will be shown that the critical fluctuations near the $H_{c2}(T)$ line display a 2D scaling behavior. Furthermore, through the 2D nonperturbative approach scaling theory developed by Tesanovic *et al.*, we have derived the values of basic superconducting parameters $H_{c2}(0)$, $\xi_{ab}(0)$, $\lambda_{ab}(0)$ and T_{c0} and the value of G_i, the Ginzburg fluctuation parameter.

EXPERIMENTS

Single crystals have been synthesized from nominal composition $Tl_{0.5}Pb_{0.5}Sr_2CaCu_2O_x$, $Tl_2Ba_2Ca_2Cu_3O_x$, $Tl_2Ba_2CaCu_2O_x$, and $TlBa_2Ca_2Cu_3O_x$. These stoichiometric mixtures were prepared from the oxides Tl_2O_3, $BaCuO_2$, CaO, PbO, $SrCuO_2$, and CuO. The powders were ground in an agate mortar, placed in an alumina crucible, and sealed in silica ampoules. The sealed tubes were heated in a vertical furnace whose temperature gradient was controlled according to thermal cycles described elsewhere.[14-16] Large crystals were extracted from each preparation with the typical dimensions presented in Table I. The crystals were first characterized by ac susceptibility measurements using a Lake Shore susceptometer in the temperature range 5–200 K with $H_{ac}=0.5$ Oe. This preliminary test has allowed several crystals to be selected in each preparation for their high onset critical temperature and their sharp transition (10%–90%) (see Table I for their transition widths). The selected crystals were investigated by x-ray diffraction by means of Weissenberg photographs, which revealed, for each chosen crystal, only one kind of reflection.

TABLE I. Sizes, transition width, and T_c of the studied crystals.

	$T_{c\ \mathrm{onset}}$ (K)	Transition width (K)	Dimension (μm^3)
Tl,Pb-1212	80	3	900×770×360
Tl-1223	114	3	800×480×130
Tl-2212	101	3	800×695×155
Tl-2223	124	2	650×540×77

The magnetization measurements were performed by means of a Quantum Design superconducting quantum interference device (SQUID) magnetometer with fields applied parallel to the c axis. The data $\mu_0 M(T,H)$ were obtained from measurements registered for temperature up to 250 K and applied fields 5.5, 4.5, 2.5, 1.5, and 0.2 T. A 2-min delay was introduced after each temperature installation in order to stabilize the system. The reliability of the experimental results concerning the contribution of the magnetization due to superconductivity in these types of materials depends on the magnitude of the normal-state background. For the four samples we found experimentally that the normal-state magnetization for each field value may be well fitted by

$$\chi(T)=\chi_0+\frac{C}{(T+\theta)}\ ,$$

where χ_0, C, and θ are the fitting parameters.

The normal-state background at each applied field was taken into account by fitting the experimental data in the high-temperature region. Typically, the extrapolated fitting of the temperature dependence of the normal-state magnetization is shown for the Tl-1223 (Fig. 1).

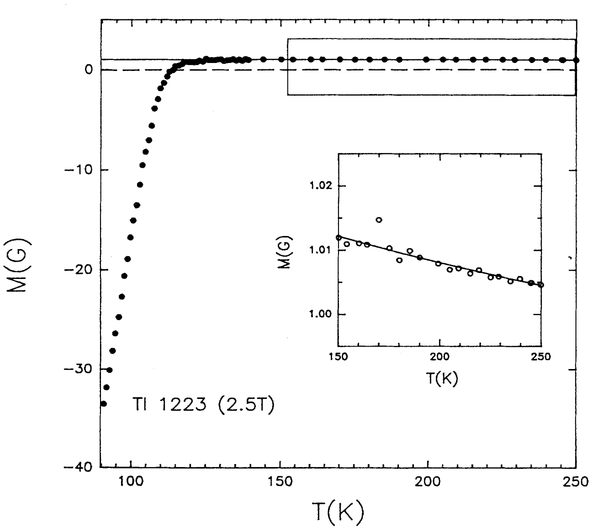

FIG. 1. Example of temperature dependence of the normal-size magnetization for Tl-1223 at 2.5 T. The solid line is the extrapolated fitting of the normal-state magnetization. The inset shows the temperature dependence of the magnetization between 150 and 250 K.

THEORETICAL BACKGROUND

Critical fluctuations in the magnetization of type-II superconductors exhibit a scaling behavior near $Hc_2(T)$ which was proposed by Ullah and Dorsey[1] and developed by Tesanovic *et al.*[5] This scaling behavior has two limit forms for 3D and 2D systems:

$$\frac{M(T,H)}{(TH)^{2/3}}=F_{3D}\left[A\frac{T-T_c(H)}{(TH)^{2/3}}\right]\ , \qquad (1)$$

$$\frac{M(T,H)}{(TH)^{1/2}}=F_{2D}\left[A\frac{T-T_c(H)}{(TH)^{1/2}}\right]\ . \qquad (2)$$

In this model, A is a field- and temperature-independent coefficient and F the scaling function, with F_{2D} for the 2D case and F_{3D} for the 3D one. The contribution of Tesanovic *et al.* to this model has been used to evaluate the scaling function of magnetization in a closed form in the critical region near the $Hc_2(T)$ line for the 2D systems:

$$\frac{M}{M^*}=\frac{1}{2}\left[1-\tau-h+\sqrt{(1-\tau-h)^2+4h}\right]\ , \qquad (3)$$

where

$$\tau=\frac{T-T^*}{Tc_0-T^*}\ ,$$

$$h=\frac{H}{H'_{c2}(Tc_0-T^*)}\ ,$$

with

$$H'_{c2}=-\left.\frac{dH_{c_2}}{dT}\right|_{T_{c_0}}$$

and T_{c_0} is the mean-field transition temperature. This expression is valid at $h\geq|1-\tau|/3$. For each phase, the temperature T^* is the temperature where all the $\mu_0 M(T)$ curves registered for different magnetic fields cross at a single point. For instance, the existence of such a crossing point has been pointed out for $Bi_2Sr_2CaCu_2O_8$,[7] $Bi_2Sr_2Ca_2Cu_3O_{10}$,[13] and $Tl_2Ba_2CuO_6$.[8] Although the existence of such a crossing point, where magnetization is independent of the field (M^*) is considered as a manifestation of the critical superconducting fluctuations in Josephson-coupled layered superconductors, it could also be observed in Y-123,[12] which is believed to behave as an anisotropic 3D superconductor.

If we define $T_c(H)=T_{c0}-H/H'_{c2}$ as the critical temperature, one can rewrite Eq. (3) with the reduced parameters x and y in order to outline a universal scaling behavior of the critical fluctuations in a 2D system. The relation leading to such a feature is given by

$$y=\tfrac{1}{2}[-x+\sqrt{x^2+4}]\ , \qquad (4)$$

where

$$x=\left[\frac{T-T_c(H)}{T_{c_0}}\right]\left[\frac{T_{c_0}}{T_{c_0}-T^*}\right]\left[\frac{H}{H'_{c_2}(T_{c_0}-T^*)}\right]^{-1/2}$$

and

$$y=\frac{M}{M^*}\left[\frac{H}{H'_{c_2}(T_{c_0}-T^*)}\right]^{-1/2} .$$

There are only two reduced parameters in this new expression leading to the same formula for each phase.

According to Bulaevskii, Ledvij, and Kogan,[2] we have the relation

$$T_{c0}-T^*=\frac{4\pi\mu_0\lambda_{ab}^2(0)k_BT_{c0}^2}{\phi_0^2 s} , \tag{5}$$

where s is the interlayer spacing of the structure given in the framework of this 2D model by

$$s=\alpha\frac{k_BT^*}{M^*\phi_0} . \tag{6}$$

α is an adjustable parameter of this model and M^* the magnetization at the field-independent crossing point. The crossing point T^* lies in a region of strong (critical) fluctuations which could be characterized by G_i, a field-independent Ginzburg fluctuation parameter for quasi-2D superconductors defined in Refs. 5 and 17 as $G_i=(T_{c0}-T^*)/T^*$.

Using Eqs. (5) and (6), the London penetration depth is written as

$$\lambda_{ab}^2(0)=\alpha\frac{G_i\phi_0T^*}{4\pi\mu_0M^*T_{c0}^2} , \tag{7}$$

where the following temperature dependence[18] is assumed:

$$\lambda_{ab}(T)=\frac{\lambda_{ab}(0)}{\sqrt{2(1-T/T_{c0})}} .$$

Moreover, in order to link the $\lambda_{ab}(0)$ values obtained through the fluctuational analysis with the ones measured from the classical analysis of the reversible magnetization (London model) performed far enough from T_c to neglect the fluctuation effects, the adjustable parameter of the model (α) has been set equal to 4 [see the $\lambda_{ab}(0)$ values of Table II]. Using the Werthamer-Helfand-Hohenberg[19] and Ginzburg-Landau theories, we have

$$H_{c_2}(0)=-0.69T_c\frac{dH_{c_2}}{dT}\bigg|_{T_{c_0}}=\frac{\phi_0}{2\pi\xi_{ab}^2(0)} . \tag{8}$$

From the scaling theory developed in Eq. (3), we obtain T_{c_0} and H'_{c_2}. It is then possible to obtain a precise determination of $H_{c_2}(T)$ and hence $\xi_{ab}(0)$ from Eq. (8) and $\lambda_{ab}(0)$ from Eq. (7).

RESULTS

In the following are presented our high-field magnetization data for the single crystals $Tl_{0.5}Pb_{0.5}Sr_2CaCu_2O_7$ (Tl,Pb-1212), $Tl_2Ba_2Ca_2Cu_3O_{10}$ (Tl-2223), $Tl_2Ba_2CaCu_2O_8$ (Tl-2212), and $TlBa_2Ca_2Cu_3O_9$ (Tl-1223), which correspond to Figs. 2(a)–2(d), respectively. For each sample, the data have been plotted in reduced scales according to the 3D and 2D scaling forms expressed by Eqs. (1) and (2). For both cases, the only free parameter for each applied field is $T_c(H)$. The data for Tl-2212 and Tl-2223 cannot be scaled with the form of the 3D case [Eq. (1)] but concerning the phases Tl,Pb-1212 and Tl-1223, this 3D scaling works quite well; nevertheless, the $H_{c2}(t=T/T_c)$ obtained in these cases leads to incoherent results as seen in Fig. (3) for Tl,Pb-1212. The procedure has been completed with the 2D scaling form for all the phases. All the data at various magnetic fields converge to a unique curve [see the insets of Figs. 2(a)–2(d)]. The $H_{c2}(t=T/T_c)$ obtained through this 2D scaling procedure are presented in Fig. 4 and have been fitted by the two parameters H'_{c2} and T_{c0}, which are presented in Table II. This procedure does not lead to incoherent results for Tl,Pb-1212 and Tl-1223.

We now compare the temperature dependence of the magnetization in the fluctuation region with the explicit scaling function given by Eq. (3). The lines shown in Figs. 2(a), 2b–2c, 2(d) are the theoretical fitting curves based on this model. The critical region near the $H_{c2}(T)$ line where the scaling function can be applied corresponds approximately to $T\geq T_{c0}-3H/H'_{c2}$ at a fixed applied field H. Once again, the least-squares minimization based on this model involves the two free parameters T_{c0} and H'_{c2}. So, for each phase, in order to perform the initial fitting procedure, the experimental data for tempera-

TABLE II. Basic superconducting parameters.

	H'_{c2} (T/K)	T_{c_0} (K)	$\xi_{ab}(0)$ (nm)	$H_{c_2}(0)$ (T)	T^* (K)	M^* (G)	s (nm)	c (nm)	G_i	$\lambda_{ab}(0)$ fluctuation (nm)	$\lambda_{ab}(0)$ London (nm)
Tl,Pb-1212	1.7	77.7	1.93	90	77.0	1.35	19	1.21	10^{-2}	209	214
	±0.1	±0.1	±0.05	±5	±0.1	±0.1	±0.5			±5	±5
Tl-1223	2.0	111.7	1.45	160	109.6	3.2	11.5	1.59	1.9×10^{-2}	188	186
	±0.1	±0.1	±0.05	±5	±0.1	±0.1	±0.5			±5	±5
Tl-2223	3.0	123.6	1.13	260	120.8	3.5	12	3.56	2.3×10^{-2}	202	207
	±0.1	±0.1	±0.05	±5	±0.5	±0.1	±0.5			±5	±5
Tl-2212	2.6	103.1	1.34	180	99.3	2.75	12	2.94	3.8×10^{-2}	266	287
	±0.1	±0.1	±0.05	±5	±0.1	±0.1	±0.5			±5	±5

tures and fields were chosen according to an estimated validity range of Eq. (3) to get an approximate value of T_{c0} and of H'_{c2}. Obviously, the measurements performed at low fields (1.5 and 0.2 T) are far outside the critical region allowed by the theory. Indeed, experimentally, we do not find excellent agreement between our low-field data and the theory. Then, in the next step, we performed the same fitting procedure to refine the parameters, restricting our magnetization data to field 5.5 $\mathrm{T} \geq H \geq 2.5$ T. Finally, the fitting has been achieved using only data in the validity range of the model. It should be pointed out, as mentioned in Ref. 10, that the relation given by Eq. (3) works quite well at temperatures far outside the range of validity. The H'_{c2} and T_{c0} obtained with this procedure based on the 2D scaling function of Tesanovic *et al.* are identical with those obtained from a previous magnetization analysis based on the Ullah-Dorsey scaling procedure. This is a positive test for the reliability of the 2D scaling function of Tesanovic *et al.*

Another important point is to report superconducting basic parameters of four Tl-based single crystals. Considering the presented data (Table II), one can remark that all the values of $\xi_{ab}(0)$ and $\lambda_{ab}(0)$ are in reasonable agreement with published results.[11,14,16,20,21,22] The values of the Ginzburg fluctuation parameter found for

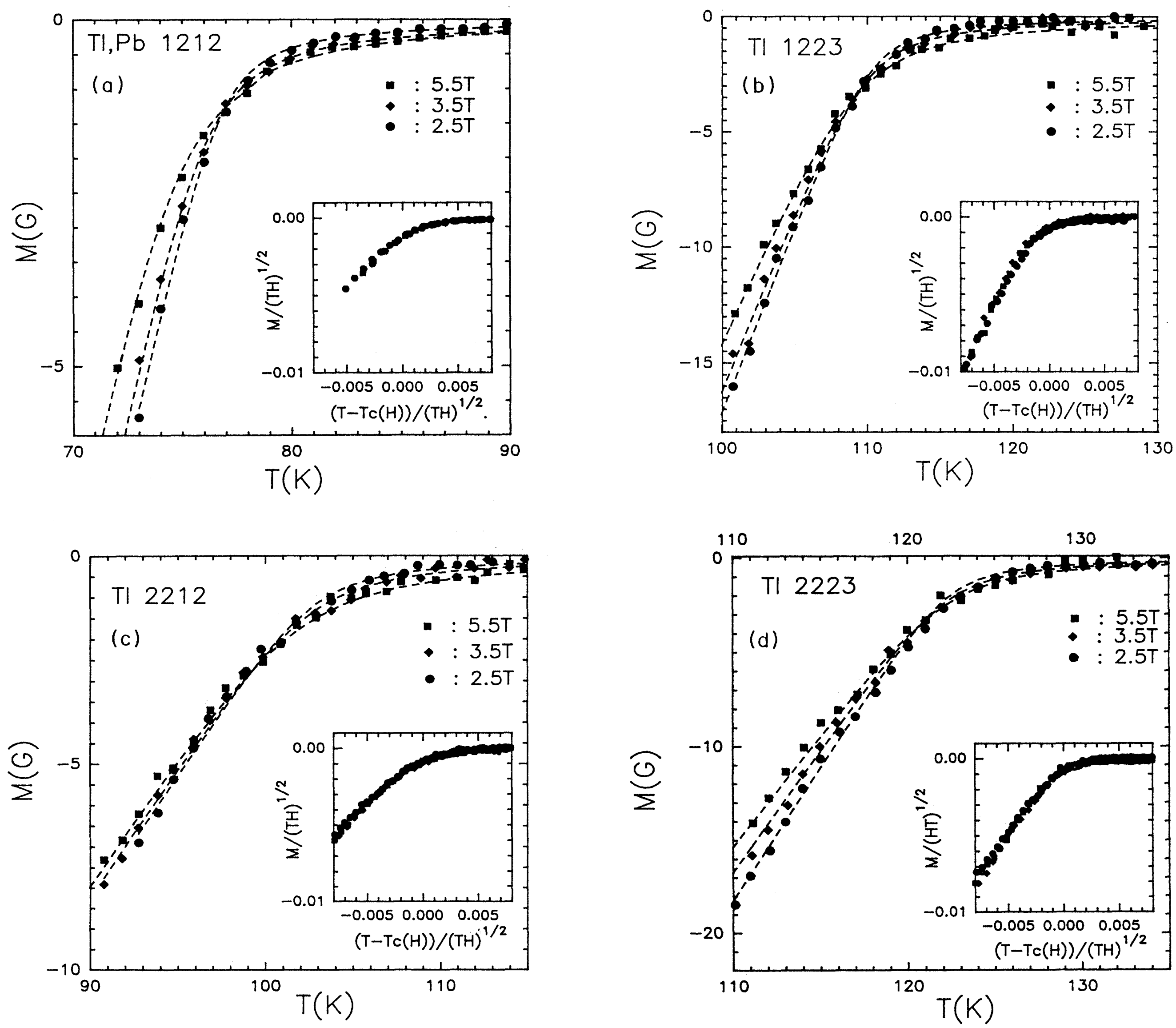

FIG. 2. Temperature dependence of the magnetization at various magnetic fields (5.5, 3.5, and 2.5 T for single crystals of (a) Tl, Pb-1212, (b) Tl-1223, (c) Tl-2212, and (d) Tl-2223 [the lines correspond to the theoretical fitting procedure based on Eq. (3)]. The insets correspond to the 2D scalings of the magnetization data based on Eq. (2). The units of the variables $M/(HT)^{1/2}$ and $[T-T_c(H)]/(HT)^{1/2}$ are $\mathrm{G}^{1/2}\,\mathrm{K}^{-1/2}$ and $\mathrm{G}^{-1/2}\,\mathrm{K}^{1/2}$, respectively.

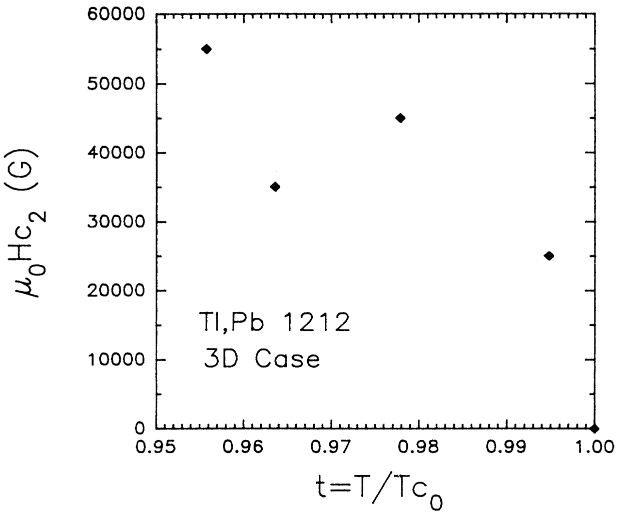

FIG. 3. Temperature dependence of the mean-field upper critical field H_{c2} obtained from the 3D scaling plot of magnetization for Tl,Pb-1212.

each phase are of the order of 10^{-2}, as reported in specific-heat measurements,[23] normal-state susceptibility,[24] or resistivity[25] on high-T_c materials. All the parameters collected in Table II allow us to perform a general scaling by Eq. (4) of the magnetization in the critical region where fluctuations occur: Figure 5 presents the final results of this scaling function for the four samples together, showing the universality of the phenomenon of the fluctuation in these quasi-2D superconductors belonging to the Tl system.

DISCUSSION

For superconducting materials, the importance of the fluctuations can be directly related to the value of the

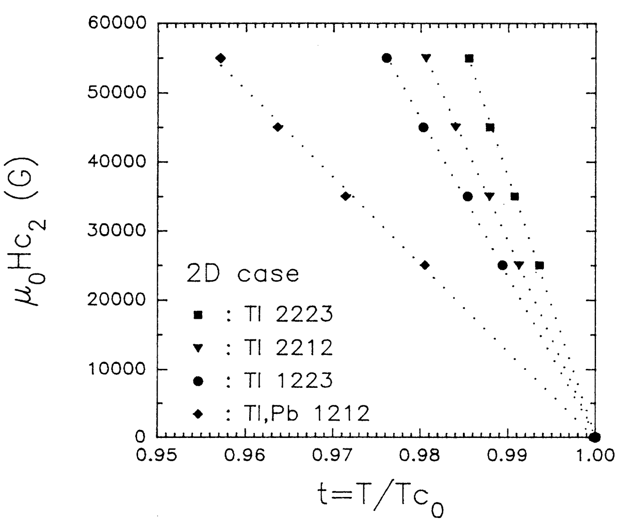

FIG. 4. Temperature dependence of the mean-field upper critical field H_{c2} obtained from the 2D scaling plots for Tl,Pb-1212, Tl-1223, Tl-2212, and Tl-2223.

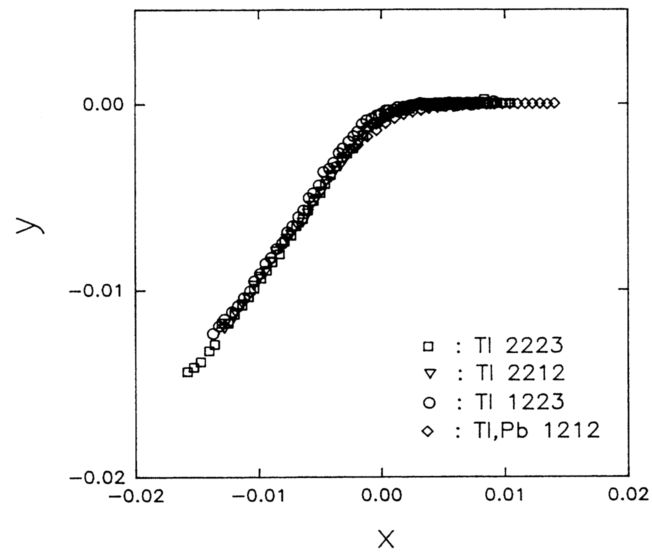

FIG. 5. Universal scaling of the fluctuations for Tl,Pb-1212, Tl-1223, Tl-2212, and Tl-2223 (see text for the definition of x and y).

Ginzburg fluctuation parameter G_i. In high-T_c materials, the high critical temperatures and the high λ values as well as the small s enhance the effects of the critical fluctuations as is shown in Table II. Indeed, we have compared the values of G_i obtained for the different compounds. In the case of Tl,Pb-1212, the smallest value of G_i (10^{-2}) is related to the largest value of s in these compounds (19 nm). However, the largest value of G_i is obtained for Tl-2212 (3.8×10^{-2}) and is due to a large $\lambda_{ab}(0)$ rather than to a small value of s (12 nm) if one compares to the other compounds. The influence of T_c could be evidenced considering the high critical temperatures of Tl-2212 ($T_{c_0}=103.1$ k), Tl-2223 ($T_{c_0}=123.6$ K), and Tl-1223 ($T_{c_0}=111.7$ K) compared to Tl,Pb-1212 ($T_{c_0}=77.7$ K), which exhibits the smallest value of G_i ($G_i=10^{-2}$).

It is tempting to relate the irreversible behavior to this large fluctuation effect. However, it must not be forgotten that these two phenomena occur in totally different parts of the H-T pane, the fluctuations lying in the reversible part of the magnetization near H_{c2}. The determination of the irreversibility lines, the boundary separating the reversible and irreversible magnetic behavior, has been performed using the closing point of the hysteresis loops. The hierarchy shown in Fig. 6 can be related to the values of G_i for the different compounds. The combination of s, T_c, and λ affects in the same way the reversible and irreversible properties of the compounds.

An interesting prediction of the model developed by Bulaevskii, Ledvij, and Kogan and Tesanovic *et al.* is that the value of the parameter s [Eq. (6)] is supposed to coincide with the crystal structure parameter (c or $c/2$). The large values of s that we have extracted from Eq. (6) (from 11.5 to 19 nm) are very far from c or $c/2$ (Table II). Many authors found smaller values ($s=1.53$ nm in Bi-2212 in Ref. 7 and 1.8 nm in Bi-2223 in Ref. 13) and at-

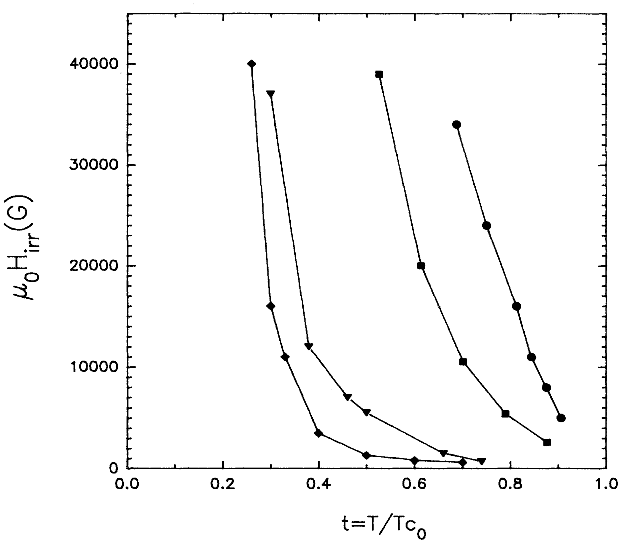

FIG. 6. Irreversibility lines of Tl,Pb-1212 (circle), Tl-1223 (square), Tl-2223 (triangle), and Tl-2212 (diamond).

tempted to correlate them with the periodicity of the superconducting sandwich as predicted by the models,[2,3,5] but the discrepancy that we have observed in these values has also been found by other groups (Ref. 10 for Bi,Pb-2223 and Ref. 17 for La-Sr-Cu-O). They have tried to explain this difference by an ambiguity in the estimation of the absolute value of the magnetization or a contribution from a higher Landau level. This raises the question of the meaning of s in this model. As it was already pointed out, s does not correspond to c (or $c/2$) and is much larger. If the superconductor is not in the 2D limit, s should be replaced by the c-axis coherence length (ξ_c). But the expected values of ξ_c are much smaller than the observed s. It should be interesting to verify if s is indeed proportional to ξ_c or to the electronic anisotropy γ ($\gamma = \xi_{ab}/\xi_c$); however, no reliable values of γ are available. Even for the most studied single crystal (Tl-2223), there are controversial values ranging from $\gamma = 4.6$,[14] derived from the longitudinal component of the reversible magnetization, to $\gamma = 900$,[26] obtained by torque measurements. Though this disagreement with the expected values of s raises the question of its significance, this model has provided an excellent framework for the analysis of the fluctuation data.

CONCLUSION

The magnetization of four high-quality single crystals (Tl,Pb-1212, Tl-1223, Tl-2212, and Tl-2223) was measured at various fields up to 5.5 T applied along the c axis in order to study the fluctuation effects. The reduced magnetization $M/(TH)^{+1/2}$ clearly exhibits a 2D scaling behavior in the variable $[T-T_c(H)]/(TH)^{+1/2}$, and the universality of the fluctuational phenomenon has been shown using the parameters deduced from the analysis. The simple 2D scaling function defined by Tesanovic *et al.* seems to be accurate for describing the critical fluctuations. Finally, the values of the Ginzburg parameters of each compound have been correlated with their pinning properties.

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