Equilibrium magnetization of $Tl_2Ba_2CaCu_2O_{8+\delta}$ single crystals

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The temperature dependence of the reversible magnetization of $Tl_2Ba_2CaCu_2O_{8+\delta}$ single crystals with T_c $\sim$105 K was measured with fields up to 2 T. We analyzed the reversible magnetization in terms of the Hao-Clem model [Phys. Rev. Lett **67**, 2371 (1991)] and the Bulaevskii-Ledvij-Kogan (BLK) model [Phys. Rev. Lett **68**, 3773 (1992)]. At low temperatures, we found anomalous temperature dependences for κ and H_{c2}, similar to the case of $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystals and explained this with a nonlocal contribution to the magnetization. The reversible magnetization showed pronounced thermal fluctuation effects from T_c down to 75 K. Using the BLK model, we calculated the penetration depth and found that its temperature dependence deviated from that obtained using the London and the Hao-Clem models.

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I. INTRODUCTION

Various vortex phases in high-T_c cuprates have been the focus of intensive experimental and theoretical investigations. The short coherence length $\xi(0)$ and the large anisotropy factor γ in these materials result in very pronounced thermal fluctuations. Also, these materials have many pinning centers which result from point defects of intrinsic oxygen vacancies. Therefore, to elucidate the vortex phase diagram, the thermal and the pinning energies should be properly considered. From recent studies,[1,2] different phases are under a delicate competition of different energies, such as the elastic, the pinning, and the thermal energies and the boundaries between each phases were determined by a balance of these energies. The shape of the vortex melting, which is characterized by a first-order thermodynamical transition, was determined by the competition between the elastic and the thermal energies, whereas the entangled line, which was manifested itself as an origin of the second peak in the M-H hysteresis loops, was described due to the competition between the pinning and the elastic energies.

Recently, a large number of experimental findings for the vortex phase diagrams of both $Bi_2Sr_2CaCu_2O_{8+\delta}$(Bi2212) and $YBa_2Cu_3O_{7-\delta}$ (YBCO) have been accumulated by using various techniques.[3,4] However, compared with these compounds, $Tl_2Ba_2CaCu_2O_{8+\delta}$ (Tl2212) has not been well studied due to its toxicity during synthesis and even the superconducting parameters, such as the critical field H_c, the penetration depth λ, the coherence length ξ, and the anisotrpy factor γ have not been well studied for the polycrystalline samples. However, these parameters are very important for characterizing the different vortex phases and investigating the driving mechanism of high-T_c copper-oxide superconductors (HTSC). Also, the temperature dependence of the penetration depth at low temperatures can reveal the gap symmetry.[5,6]

Tl2212 has an anisotropy factor a little smaller than that of Bi2212, but much larger than that of YBCO. Furthermore, the interlayer distance between CuO_2 bilayers is smaller than that of Bi2212. Thus, the physical properties related to the vortex phase diagram are expected to be in the intermediate region between those of Bi2212 and YBCO. In this regard, a determination of the superconducting parameters is the first step toward further study of the physical properties related to various vortex phases. However, studies of the superconducting parameters through reversible magnetization are rare due to the difficulties in synthesizing high-quality samples. Though Wahl *et al.*[7] reported superconducting parameters, which they obtained by using two-dimensional scaling developed by Ullah-Dorsey, scaling analyses are prone to give an error due to the small signal from the fluctuations in the magnetization. More rigorously, these results should be confirmed through different analyses.

In Bi2212, analysis of the reversible magnetization in terms of the Hao-Clem[8] model revealed unusual temperature dependences for the parameters κ and H_c.[9] The most extraordinary feature is that H_{c2} is temperature independent until thermal fluctuations begin. This temperature independence of H_{c2} is a consequence of the scaling behavior $M(H,T)=m(H)A(T)$. Later, a possible explanation of the temperature independence of H_{c2} and the anomalous temperature dependence of κ and H_c was suggested by Kogan *et al.*,[10,11] who considered the nonlocal contribution to the magnetization. In nonlocal theory, the London equation is modified, and from among the HTSC, Bi2212, and Tl2212 were possible systems for which the magnetization could be explained by this theory, while YBCO, Bi2223, and Hg1223 followed the local theory better. However, until now, there has been no supporting evidence for a nonlocal contribution to the magnetization in either Bi2212 or Tl2212.

In this paper, we report the reversible magnetization for fields up to 2 T parallel to the c axis in order to obtain various superconducting parameters based on the London, the Hao-Clem, and the Bulaevskii-Ledvij-Kogan (BLK)[12] models. The measurements were carried out using high-quality Tl2212 crystals, which showed very sharp (00l) reflections characteristic of the 2212 structure with c $\sim$29.3 Å. We analyzed the experimental data based on the above-described models and observed the anomalous temperature dependences of κ, H_c, and H_{c2}, while comparing the data with the conventional prediction of the local theory. These anomalous dependences could be explained by a non-

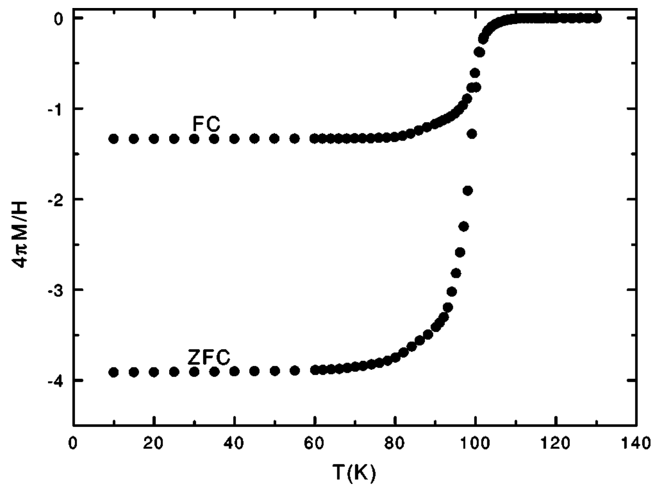

FIG. 1. Temperature dependence of the magnetization at 10 G. $T_c=105$ K and $\Delta T_c\sim 5$ K.

local contribution to the magnetization in Tl2212. Furthermore, positional fluctuations of the vortex, which are pronounced in HTSC, were observed in the magnetization and influenced the in-plane penetration depth (λ_{ab}) down to low temperatures.

II. EXPERIMENTS

Single crystals of Tl2212 were grown from a stoichiometric mixture of Tl_2O_3 and a precursor $Ba_2CaCu_2O_x$. The precursor was prepared by mixing the appropriate amounts of $BaCO_3$, $CaCO_3$, and CuO_2 (99.99% pure) to form the nominal stoichiometry of $Ba_2CaCu_2O_x$. The mixture was calcined in air at 840 °C for 24 h, ground and heated again for another 24 h at 910 °C. Then, the precursor and Tl_2O_3 were mixed and pressed into a pellet. A pellet of 5 gm was placed in an alumina crucible and sealed in a quartz ampoule. Sealing in the quartz ampoule prevented the evaporation of the thallium from the mixture and made the final products stoichiometric. The sealed tube was put into a preheated (950 °C) vertical furnace. The temperature was maintained for 15 min to melt the mixture, then it was cooled down to 900 °C at a rate of 3 °C/h, which was followed by fast cooled to room temperature.

Crystals, with typical dimensions of $1\times1\times0.2\times \text{mm}^3$, were extracted and annealed in oxygen for 48 h at a temperature of 400 °C. The phase was characterized using X-ray diffraction with 4 circle goniometers from 20° to 60° in steps of 0.05°. The X-ray diffraction pattern showed very sharp ($00l$) peaks characteristic of the 2212 structure with $c\sim 29.3$ Å.[13]

The shielding [zero-field-cooled (ZFC)] and Meissner [field-cooled (FC)] magnetizations measured at a field of 10 G applied parallel to the c-axis on a Tl2212 crystal are shown in Fig. 1. The value of $4\pi M/H$ for ZFC is around four. This large value is due to the demagnetization factor of the plate-shaped sample. The calculated demagnetization factor D is around 0.74, and this value is consistent with the above-mentioned dimensions of the sample. The onset of the transition is 105 K. The reversible magnetization was measured using a Quantum Designs superconducting quantum interference device magnetometer for fields applied parallel to the c axis.

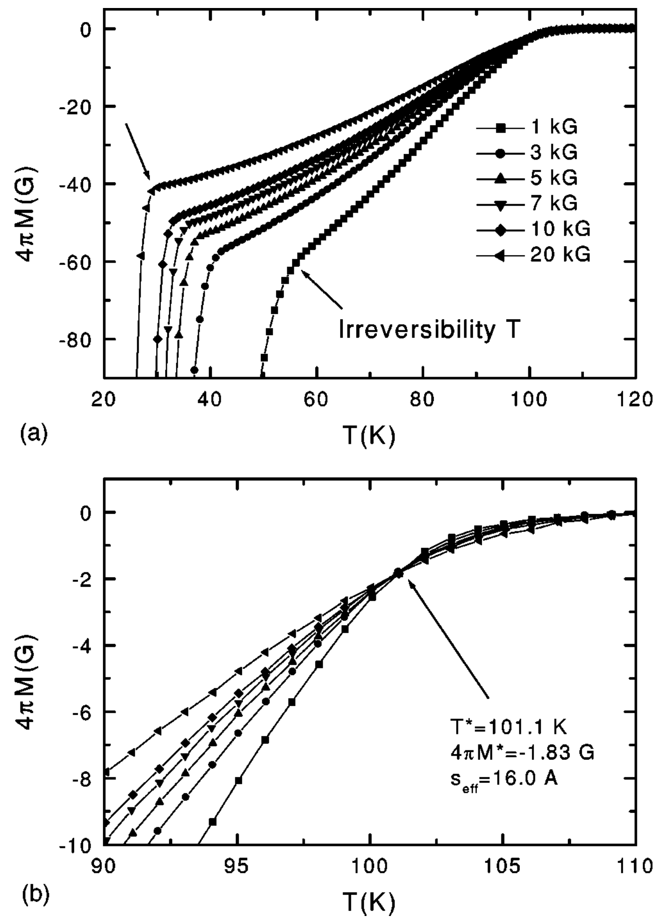

FIG. 2. (a) Temperature dependence of the magnetization at high fields. (b) Figure (a) for temperatures between 90 and 110 K. Temperature independence of the magnetization, $4\pi M^*=-1.84$ G at $T^*=101.1$ K can be clearly seen.

III. RESULTS AND DISCUSSION

Figure 2(a) shows the temperature dependence of the magnetization $M(T)$ for different fields applied parallel to the c axis. The irreversible temperature, where the ZFC and the FC magnetizations begin to split, is indicated by the arrowhead lines in this figure. The shape of the irreversibility curve obtained here exactly overlaps the data measured from the M(H) hysteresis loops.[13]

Figure 2(b) is the $M(T)$ in the temperature range from 90 to 110 K. As shown in this figure, there is a crossing point at which all the data for the different fields overlap. The values of the magnetization and the temperature at which this crossing point occurs are $4\pi M^*=-1.84$ G and $T^*=101.1$ K. This crossing point for HTSC was explained earlier by using the BLK approach.[12] BLK calculated the free energy in the mixed state within the frame of the Lawrence-Doniach model by using a harmonic approximation. The calculated free energy consisted of both the London contribution and a contribution from the positional fluctuations of pancake vortices. For $H_{cr} \approx \phi_0/s^2\gamma^2 \ll H \ll H_{c2}$, the magnetization was given by the relation

$$M(T,H) = -\frac{\phi_0}{32\pi^2\lambda_{ab}^2}\ln\frac{\eta H_{c2}}{eH} + \frac{k_B T}{\phi_0 s}\ln\frac{16\pi k_B T\kappa^2}{\alpha\phi_0 s H\sqrt{e}}, \quad (1)$$

where $e = 2.718, \ldots$, $\eta = 1.4$,[8,14,15] s is the distance between the superconducting layers, and α is a constant of the order of unity. The first term results from the unperturbed line vortex and the second one from the positional fluctuations of the pancake vortices. The crossing point in the BLK approach was determined by the condition $\partial M/\partial(\ln H) = 0$ at $T = T^*$, and at this temperature T^* the magnetization becomes

$$M^*(T^*) = -\frac{k_B T}{\phi_0 s}\ln\frac{\eta\alpha}{\sqrt{e}}. \quad (2)$$

There have been several reports where $\ln\eta\alpha/\sqrt{e} = 1$ was used to determine the interlayer distance s by using the value of magnetization at $T = T^*$, but the calculated value of s was found to be larger than the actual value obtained from the structural characterization. To remove this discrepancy, Koshelev[16] calculated the fluctuation contributions to the magnetization by considering higher Landau levels. From this consideration, the calculated magnetization at $T = T^*$ was given as $M^* = -m_\infty k_B T^*/\phi_0 s$, where $m_\infty \approx 0.346$. This result is the same as in Eq. (2) except the coefficient of $k_B T^*/\phi_0 s$. Using Koshelev's result, we obtained $s = 1.60$ nm, which is very close to the value ~ 1.47 nm, obtained from x-ray analysis.

To investigate the superconducting parameters of the Tl2212 crystal, we applied the Hao-Clem variational method to the $M(T)$ data. The principle of the Hao-Clem theory is to minimize the Ginzburg-Landau free energy of the mixed state for a high-κ type-II superconductor by using a trial wave function. The $|\psi|^4$ term and the vortex core contribution are included in the free energy. However, this model does not consider the effects of thermodynamic fluctuations in the presence of magnetic fields, which causes an anomalously large increase in $\kappa(T)$, while calculated by using this model, near T_c in many HTSCs.[17–21]. The temperature dependences of κ and the critical field H_c can be evaluated using this model. A detailed description of this method is given in Refs. 8 and 14. By fitting the experimental data to Eqs. (20) and (21) in Ref.14 at each temperature, one can obtain $\kappa(T)$ and $H_c(T)$.

Figure 3 shows $\kappa(T)$, $H_c(T)$, and $H_{c2}(T)$ obtained from the Hao-Clem analysis. $H_{c2}(T)$ was calculated using the relation of $H_{c2}(T) = \sqrt{2}H_c(T)\kappa(\mathrm{T})$. In Bi2212 crystals,[9] the temperature dependence of H_c was observed to be quite different from the usual parabolic temperature dependence predicted by the two-fluid model $H_c(T) = H_c(0)[1-(T/T_c)^2]$. An empirical relation $H_c(0)\sqrt{1-(T/T_c^*)^2}$ was observed to fit the data much better. Similarly, $H_c(T)$, as shown in the inset of Fig. 3(a), also deviates strongly from the usual parabolic temperature dependence, giving an anomalously large T_c of ~ 125 K. The solid line in the inset of Fig. 3(a) is a

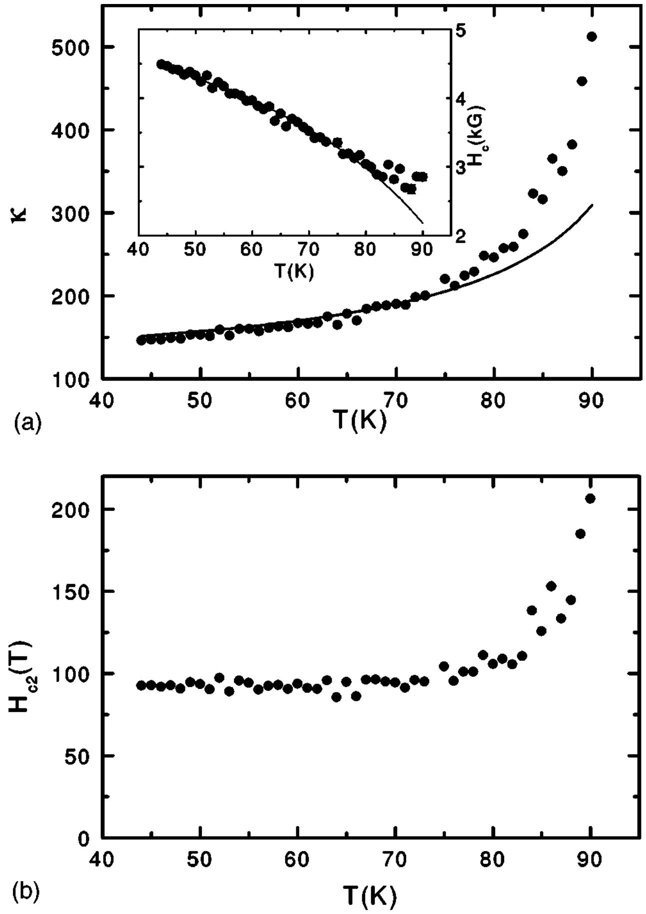

FIG. 3. (a) Temperature dependence of κ. The solid line is the fitting curve $\sim 1/\sqrt{1-(T/T_c^*)^2}$. The inset shows $H_c(T)$. $H_c(T)$ is proportional to $\sqrt{1-(T/T_c^*)^2}$. (b) Temperature dependence of H_{c2}. H_{c2} is constant where $H_c \propto \kappa^{-1}$.

fitting of the empirical formula to our data for temperatures below 75 K. Values of $T_c^* = 100.2 \pm 0.7$ K and $H_c(0) = 4950 \pm 24$ G were obtained. The behavior of $\kappa(T)$ is also remarkable. Instead of being constant in magnitude, as observed in many HTSCs,[20,21] κ increased gradually from $\kappa(0) = 136$ as the temperature increased up to 75 K and then diverged near T_c as shown in Fig. 3(a). In Fig. 3(b), the temperature dependence of H_{c2} is shown. An interesting, and unexpected behavior is its temperature independence below around 75 K, which implies that κ is inversely proportional to H_c. The solid line of Fig. 3 gives a temperature dependence of $\kappa \propto H_c^{-1}$.

Kogan *et al.*[10] suggested the modified London equation which explained this anomalous temperature dependences of κ, H_c, and H_{c2} and which resulted in a scaling behavior of the magnetization: $M(H,T) = m(H)A(T)$. In their theory, they included the nonlocality of the microscopic current-field relation by using a parameter, known as the nonlocality range ρ in their equation. The modified London equation is given by

$$-\frac{M}{M_0} = \ln\left(\frac{H_0}{B}+1\right) - \frac{H_0}{H_0+B} + \zeta(T), \quad (3)$$

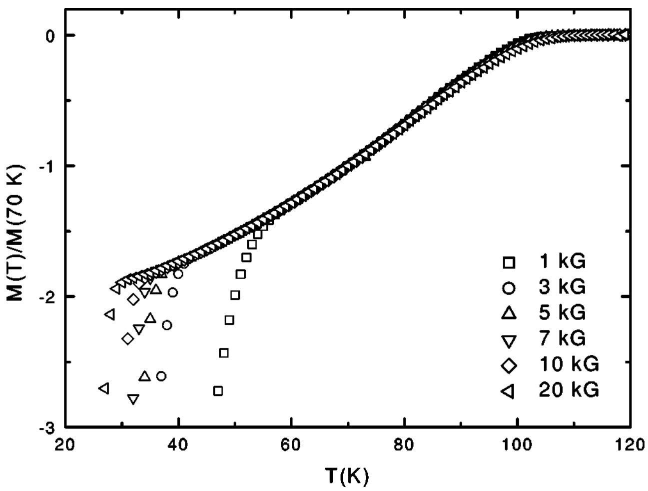

FIG. 4. Normalized magnetization, $M/M(70\text{ K})$, vs temperature.

where $M_0=\phi_0/32\pi^2\lambda^2$, $H_0=\phi_0/2\pi^2\sqrt{3}\rho^2$, and $\zeta(T)$ is a quantity that slowly decreases with increasing temperature. For a clean system far away from T_c, the right-hand side of Eq. (3) is temperature independent, resulting in the previously mentioned scaling behavior $M(H,T)\approx m(H)A(T)$.

Motivated by this theory, we attempted a scaling of $M(T)$. As shown in Fig. 4, the curves measured at different fields merge into a single curve, as expected from Eq. 3, while the magnetization at 70 K is fixed. Therefore, the abnormal temperature dependences of κ, H_c, and H_{c2} and the scaling property of $M(T)$ for the Tl2212 crystal, which are unexpected in the local theory, can be explained when a nonlocal contribution to the magnetization is considered. A scaling behavior of magnetization was also observed in Tl2212 polycrystal[22] and later, that scaling behavior was understood in the frame of the nonlocal theory,[10] giving further support of our results.

Now, let us concentrate on the behavior above ~75 K. Above ~75 K, the temperature dependences of H_c and κ deviate from solid lines as shown in Fig. 3, and $H_{c2}(T)$ increases. As mentioned above, since in the analysis of the reversible magnetization based on the Hao-Clem model, the contribution from thermal fluctuations is ignored, an anomalously large increase in κ occurs near T_c, while calculated by using this model. In this context, the deviation at ~75 K in Fig. 3(a) is thought to come from thermal fluctuations.

To investigate the effect of thermal fluctuation, we compared the results for $\lambda_{ab}(T)$ calculated from both the BLK and the London models. Figure 5(a) shows the temperature dependence of the penetration depth, $\lambda_{ab}(T)$. Near T_c, a pronounced deviation is observed in $\lambda_{ab}(T)$ obtained from the BLK and the London models and this deviation continues down to low temperatures. The unphysical divergence of $\lambda_{ab}(T)$ near T_c in the London model may be due to a lack of positional fluctuation of vortices in the mean-field picture. For example, the contribution from these fluctuations was essential to calculate the $\lambda_{ab}(T)$, as in the Bi2212 case.[15] For comparison, the values of $\lambda_{ab}(T)$ from the Hao-Clem model and Kogan's nonlocal model are presented in the same figure. $\lambda_{ab}(T)$ calculated from the Hao-Clem model is almost the same as that calculated from the London model, and this implies that for the field ranges we investigated here, the vortex-core contribution to the free energy, which is ignored in the London model, is negligibly small. $\lambda_{ab}(T)$ in Kogan's nonlocal model is not much different from that obtained by using the London model.[23]

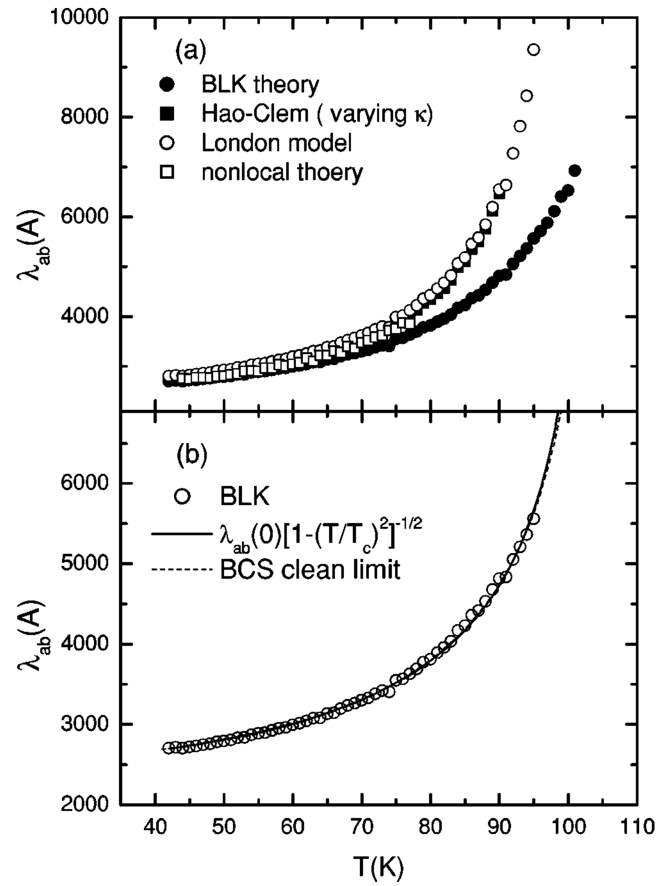

FIG. 5. (a) $\lambda_{ab}(T)$ calculated from the London model, the Hao-Clem model, the Kogan's nonlocal model, and the BLK approach. (b) $\lambda_{ab}(T)$ with the fitted curves. The formulas for the fitting curves, see the text.

To calculate the thermodynamic parameters at $T=0$, we used the results of the BLK model. We obtained $\lambda_{ab}=2614 \pm 5$ Å with $T_c=106.7$ and 2479 ± 6 Å with $T_c=105.6$ by using the BCS clean limit formula and an empirical relation, defined by $\lambda_{ab}(T)=\lambda_{ab}(0)(1-(T/T_c)^2)^{-1/2}$, respectively. Figure 5(b) shows the experimental data with the fitted curves, which were obtained by using both the BLK model and the above empirical relation. $H_{c2}(T)$ was calculated using Eq. (1) with $\eta=1.4$, $\alpha=1$, and $\kappa=150$. From these data, a value of $(dH_{c2}/dT)_{T_c}=-2.11$ T/K is obtained. In the Werthamer, Helfand, and Hohenberg theory, $H_{c2}(0) =0.5758(\kappa_1/\kappa)T_c|dH_{c2}/dT|_{T_c}$, and the ratio of κ_1/κ is given by 1.20 and 1.26 in the dirty and the clean limit, respectively.[24] The value of $H_{c2}(0)$ was estimated to be 161 ± 3 T in the clean limit; which in turn, the value of the coherence length $\xi_{ab}(0)$ becomes 1.43 nm, as deduced using the expression $H_{c2}(0)=\phi_0/2\pi\xi_{ab}^2$. This set of values is reasonable when we compare them with the previous results of thermal fluctuation theory.[7]

Thermal fluctuations influence the determination of $\lambda_{ab}(T)$ down to low temperatures in both Bi2212[15,25] and Tl2212 and the corrections due to such fluctuations are common among HTSC materials having $\gamma \gg 1$. The fundamental parameter, which determines the strength of the thermal fluctuations, is the Ginzburg number, Gi $=[T_c/H_c^2(0)\gamma\xi^3(0)]^2/2$. Among all HTSCs, though these three physical parameters T_c, $H_c(0)$, and $\xi(0)$ used in the Ginzburg number are of the same order of magnitude, a dissimilarity is observed due to the anisotropy factor γ. An estimate of the anisotropy factor from the transport measurement[26] showed that the γ of Tl2212 was slightly smaller than that of Bi2212. In this respect, we could expect a similarity of the thermal fluctuation effects in Bi2212 and Tl2212, which we did observe in our analysis of $\lambda(T)$.

IV. CONCLUSIONS

The reversible magnetization of Tl2212 single crystals in high fields parallel to the c axis was investigated in this paper. This paper showed that at low temperatures, the anomalous behavior of $H_c(T)$, $\kappa(T)$, and $H_{c2}(T)$ were consistent with anomalous scaling behavior in the magnetization and could be explained by a theory that considered nonlocal contributions to the magnetization. This study also revealed, through the BLK model, that thermal fluctuations influenced the determination of $\lambda_{ab}(T)$ significantly and that this effect continued down to at least 75 K.

ACKNOWLEDGMENTS

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