

Fluctuation diamagnetism around the superconducting transition in a cuprate crystal with a reduced Meissner fraction

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The magnetization around the superconducting transition was measured in a $\text{Tl}_{0.5}\text{Pb}_{0.5}\text{Sr}_2\text{CaCu}_2\text{O}_7$ crystal affected by a considerable reduction ($\sim 55\%$) of its effective superconducting volume fraction but still with a relatively sharp low-field Meissner transition, a behavior that may be attributed to the presence of structural inhomogeneities. By taking into account these inhomogeneities just through the Meissner fraction, the observed diamagnetism may still be explained, consistently above and below the superconducting transition, in terms of the conventional Ginzburg-Landau approach with fluctuations of Cooper pairs and vortices.

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The behavior of the magnetization around the Meissner transition provides an unavoidable constraint for any phenomenological description of a superconducting transition.¹ In the last few years, various groups have reported the observation of *strong anomalies* in the magnetization measured around the superconducting transition in high- T_C cuprate superconductors (HTSCs) with different doping levels. Among these anomalies are the observation under low fields of *giant* diamagnetism [with amplitudes of orders of magnitude larger than the one associated with superconducting fluctuations in the conventional Ginzburg-Landau (GL) scenario] and a seemingly nonlinear temperature behavior of the associated upper critical field $H_{C2}(T)$ near T_C .²⁻⁴ The origin of this unconventional (non-GL) behavior is at present a debated issue,²⁻¹⁴ the proposals including T_C inhomogeneities or vortex fluctuations even well above the measured T_C . The interest of this debate is enhanced by the fact that it also concerns other open aspects of the HTSCs, such as the pseudogap in the normal state or the possible existence of a vortex fluid over a wide temperature range above T_C .¹⁴⁻¹⁶

The magnetization measurements and analyses performed recently by our group in different HTSCs and *dirty* low T_C superconductors (without nonlocal electrodynamic effects) favor the presence of extrinsic T_C inhomogeneities, just associated with chemical inhomogeneities, as the origin of most of the observed magnetization anomalies.^{4,12,17,18} Nevertheless, there is another very common type of inhomogeneity whose influence on the magnetization also deserves a close inspection: the one associated with structural defects at different length scales, including those as smaller as a few times the superconducting coherence length amplitude $\xi(0)$. In extreme type II superconductors, even these short length inhomogeneities, difficult to be directly observed, may strongly decrease the effective superconducting volume fraction without enlarging the temperature width of the low-field Meissner transition. In this Brief Report, we will first present detailed magnetization measurements around the Meissner transition in a $\text{Tl}_{0.5}\text{Pb}_{0.5}\text{Sr}_2\text{CaCu}_2\text{O}_7$ (TlPb1212) crystal deeply affected by a reduction of the effective volume fraction, an effect which does not enlarge the temperature width of the low-field Meissner transition and that may be attributed to structural inhomogeneities. Then, it will be shown that if these anomalies are taken into account through the

Meissner fraction, the diamagnetism around T_C may still be explained in terms of the conventional Ginzburg-Landau approach with fluctuations of Cooper pairs and vortices. This agreement extends to all the different fluctuation regions in the H - T phase diagram, thus generalizing previous results for the so-called *crossing point* of the magnetization versus temperature curves.¹⁹

The TIPb1212 sample used in this work is a $1.10 \times 0.85 \times 0.192$ mm³ single crystal. Details of its growth procedure and subsequent structural characterization may be found in Ref. 20. Let us only mention that x-ray diffraction revealed that it was single phase, with a well defined c -crystallographic length of $c=12.1$ Å. The magnetization measurements were performed with a superconducting-quantum-interference-device magnetometer (Quantum Design). As a first magnetic characterization, we measured the temperature dependence of the field-cooled (FC) magnetic susceptibility with a 1 mT magnetic field applied perpendicularly to the ab crystallographic planes. The result is presented in the lower inset of Fig. 1, already corrected for demagnetizing effects. For that, we used the demagnetizing factor resulting from the sample dimensions by using the ellipsoidal approximation. For our sample, this leads to a factor of $D=0.75$. These last data show that this crystal has a narrow diamagnetic transition, the relative width being $\Delta T_C/T_C \sim 3 \times 10^{-2}$ with the midpoint at $T_C=77.1$ K. However, they also reveal a strong reduction (around 55%) of its effective superconducting volume fraction. These results provide a quite direct indication that this crystal is deeply affected by (temperature independent) structural inhomogeneities uniformly distributed in the sample volume, which, as noted before, are particularly effective in reducing the Meissner fraction of the extreme type II superconductor studied here.

The excess diamagnetism for $H \perp ab$ was obtained by subtracting to the raw data the normal-state contribution, determined by fitting a Curie-like function to the $M(T, H)$ curves well above T_C (between ~ 100 and ~ 200 K). An overview of the resulting $\Delta M(T)$ in all the reversible regions is presented in Fig. 1 for magnetic fields between 0.5 and 5 T. As may be seen in the upper inset, the curves for $\mu_0 H \geq 2$ T cross at a temperature $T_1^* \approx 75$ K, which is a signature of the thermal fluctuations in highly anisotropic supercon-

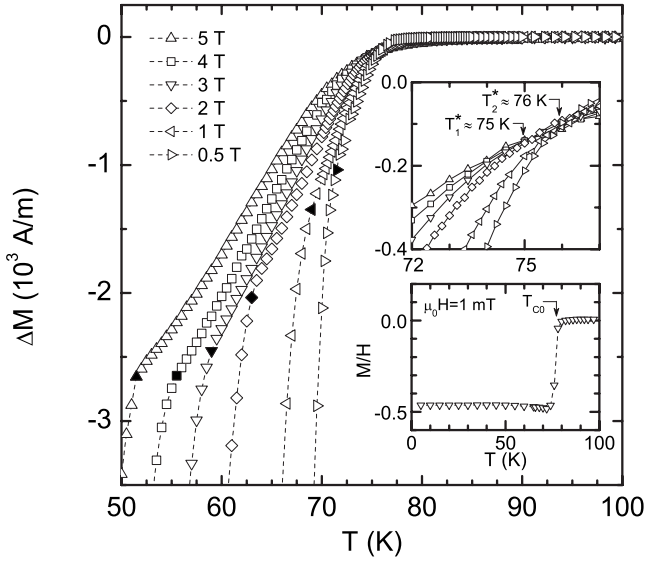


FIG. 1. Overview of the T dependence of the fluctuation magnetization for $H \perp ab$ in the reversible region around T_C . The solid symbols indicate the transition to the irreversible mixed state. Upper inset: detail of the crossing point which reveals its splitting, the $\Delta M(T)$ curves for $\mu_0 H \geq 2$ T cross at T_1^* , while for lower fields cross at T_2^* . Lower inset: T dependence of the low-field FC magnetic susceptibility, already corrected for demagnetizing effects.

ductors. For lower-field amplitudes, the crossing point shifts to a higher temperature ($T_2^* \approx 76$ K). This behavior, which may be seen more clearly in the $\Delta M(H)_T$ representation (see below), was already observed in other highly anisotropic superconductors^{17,21} and may be attributed to a change of the fluctuation regime induced by the magnetic field.

For temperatures and magnetic fields well above $T_C(H)$, the fluctuation magnetization of highly anisotropic superconductors when $H \perp ab$ predicted by the GL theory in the Gaussian approximation (GGL approach) and taking into account the total energy cutoff reads^{17,22–24}

$$\Delta M = -f \frac{k_B T N}{\phi_0 s} \left[-\frac{\varepsilon^c}{2h} \psi \left(\frac{h + \varepsilon^c}{2h} \right) - \ln \Gamma \left(\frac{h + \varepsilon}{2h} \right) + \ln \Gamma \left(\frac{h + \varepsilon^c}{2h} \right) + \frac{\varepsilon}{2h} \psi \left(\frac{h + \varepsilon}{2h} \right) + \frac{\varepsilon^c - \varepsilon}{2h} \right], \quad (1)$$

where the notation is the same as for Eq. (1) in Ref. 17. Some examples of the measured ΔM above T_{C0} are presented in Fig. 2. The lines correspond to Eq. (1) with $N=2$, $s=c=12.1$ Å, $\varepsilon^c=0.55$, f approximated by the Meissner fraction $|\chi^{FC}(0)| \approx 0.45$, and $H_{C2}(0)$ as the only free parameter. As may be clearly seen, the agreement with the experimental data is excellent down to a few degrees above T_{C0} , where the Gaussian approximation is no longer valid, and it leads to $\mu_0 H_{C2}(0) \approx 170$ T, a value that is going to be used in the remaining analyses. Note that Eq. (1) is also in excellent agreement with the H dependence of ΔM (inset of Fig. 2). This behavior is expected to hold up to $h \sim 0.1$,²⁵ where fluctuation effects begin to decrease due to quantum effects associated with the shrinkage of the superconducting wave

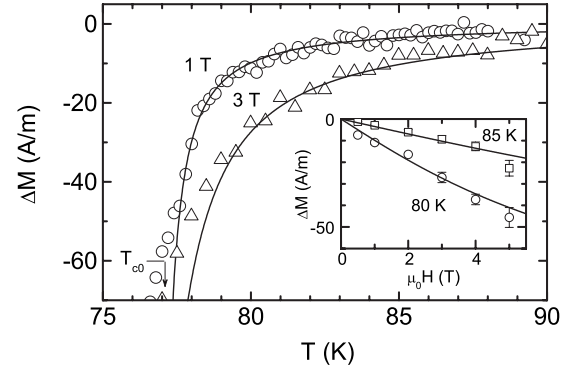


FIG. 2. Fluctuation magnetization vs T and vs H (inset) in the Gaussian region above $T_C(H)$. The lines are the GGL result for finite H [Eq. (1)].

function.^{23,26} If an ideal superconducting volume fraction is assumed ($f=1$), the agreement is also good, but leads to a different $\mu_0 H_{C2}(0)$ value (~ 370 T), which has consequences in the subsequent analysis.

For temperatures closer to $T_C(H)$, in the so-called critical region [see Fig. 3(a)], the Gaussian approximation is no longer valid. In a sufficiently strong magnetic field, in which the Cooper pairs are limited to the lowest Landau level, this critical region is bounded by the so-called field-dependent Ginzburg criterion,²⁷ which for two-dimensional systems is given by²⁸

$$|T - T_C(H)|/T_{C0} \lesssim \sqrt{4\pi k_B \mu_0 H / \phi_0 s \Delta c}, \quad (2)$$

where Δc is the specific heat jump at T_{C0} . In this regime, the GL theory predicts that the fluctuation induced magnetization follows a scaling behavior in the variables²⁹

$$m \equiv \Delta M / \sqrt{HT}, \quad t \equiv [T - T_C(H)] / \sqrt{HT}. \quad (3)$$

By using a nonperturbative approach to the GL free energy in the lowest Landau-level approximation [Ginzburg-Landau lowest Landau-level (GL-LLL) approach], Tesanović *et al.* obtained an explicit equation for the scaling function, which may be written as³⁰

$$m = f \frac{A}{H'_{C2}} \frac{k_B}{\phi_0 s} (At - \sqrt{A^2 t^2 + 2}), \quad (4)$$

where $A \equiv [H'_{C2} T_1^* / 2(T_{C0} - T_1^*)]^{1/2}$ and $H'_{C2} \equiv H_{C2}(0) / T_{C0}$, and T_1^* corresponds to the limit of the critical region below T_{C0} when $H=0$. This expression predicts the crossing of the $M(T)_H$ curves at T_1^* and gives for the crossing point magnetization,

$$\Delta M_1^* = -f \frac{k_B T_1^*}{\phi_0 s}. \quad (5)$$

This equation allows a direct comparison with the experiments. As may be easily checked, the high-field crossing point observed at T_1^* falls into the critical region bounded by Eq. (2) and should be described by Eq. (5). By using $f = |\chi^{FC}(0)|$, it leads to $\Delta M_1^* \approx -190$ A/m, in relatively good agreement with the experimental value (-150 A/m) taking

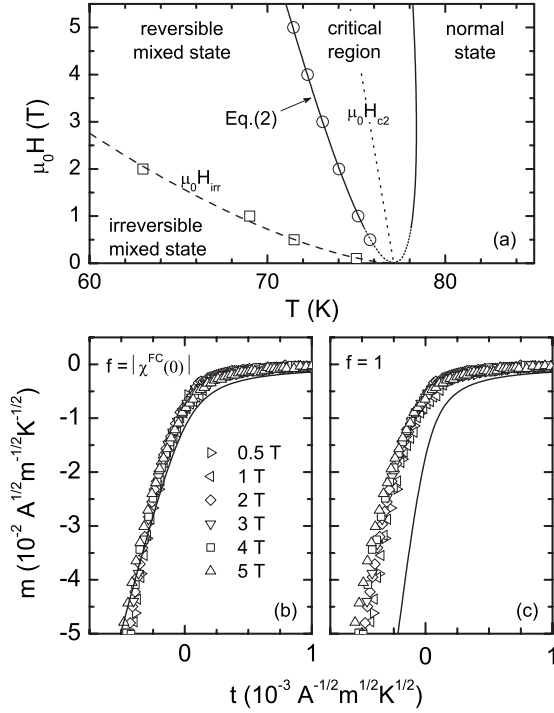


FIG. 3. (a) H - T superconducting phase diagram indicating the different fluctuation regions around $H_{C2}(T)$. Circles are the low-temperature limit of the region where the scaling approach holds, and the solid line is the best fit of the H -dependent Ginzburg criterion. Squares are the irreversibility line as deduced from Fig. 1 and the dashed line a fit to a $(T_C - T)^{3/2}$ power law. (b) Scaling of the m vs t curves in the critical region around $H_{C2}(T)$. The line is the GL-LLL scaling function [Eq. (4)]. In obtaining the scaling variables and function, we used the $H_{C2}(0)$ value resulting from the ΔM analysis in the Gaussian region well above T_C (Fig. 2). (c) The same analysis assuming an ideal effective superconducting volume fraction ($f=1$).

into account the experimental uncertainties in $\chi^{FC}(0)$ and in the normal-state $M_B(T)$ contribution. A similar agreement was also found in a variety of highly anisotropic HTSCs with different $\chi^{FC}(0)$ values.¹⁹ However, by imposing $f=1$ as would correspond to an ideal sample, the disagreement is well beyond these uncertainties.

In Fig. 3(b), we present the scaling of the $\Delta M(T)_H$ data in the critical region according to Eq. (3). The scaling variable t is obtained by assuming a linear H dependence of the critical temperature, $T_C(H) = T_{C0}[1 - H/H_{C2}(0)]$, and by using $\mu_0 H_{C2}(0) = 170$ T, as results from previous analysis in the Gaussian region above T_C . The line in this figure is the scaling function [Eq. (4)] calculated with the same $H_{C2}(0)$ value. As may be clearly seen, the scaling of the $m(t)$ curves is excellent and the scaling function is also in good agreement with the data. The low-temperature limit of the region where the scaling holds is represented as circles in the H - T phase diagram of Fig. 3(a). The fit of Eq. (2) to these data (solid line) is excellent and leads to $\Delta c \approx 1.7 \times 10^5 \text{ J/K m}^3$, which is close to the value found in other highly anisotropic HTSCs.³¹ For completeness, in Fig. 3(c), we present the $m(t)$ data evaluated by using $\mu_0 H = 370$ T (the value resulting

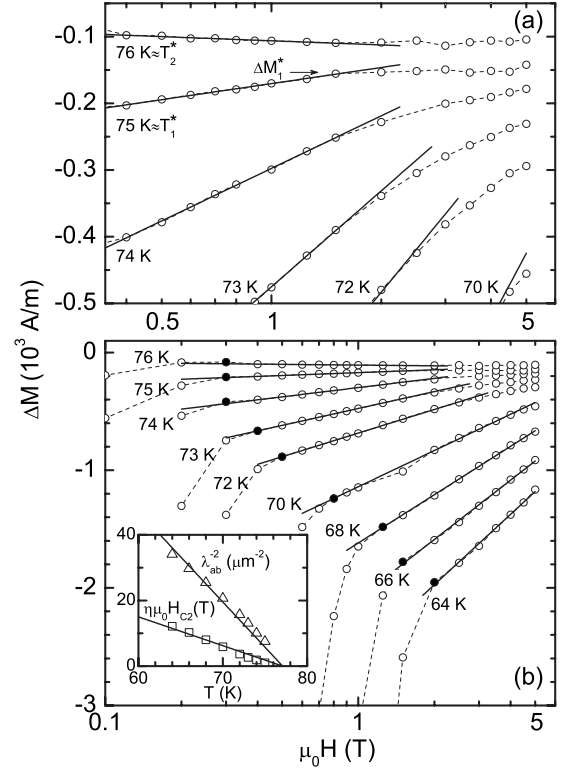


FIG. 4. ΔM vs H in the Gaussian region below $H_{C2}(T)$. (a) Detail around the crossing point temperatures. (b) Overview of the reversible mixed state (solid symbols indicate the transition to the irreversible region). The lines are fits of the Bulaevskii-Ledvig-Kogan theory [Eq. (6)], with λ_{ab} and ηH_{C2} as the free parameters. The resulting $\eta \mu_0 H_{C2}$ and λ_{ab}^{-2} (inset) follow the GL prediction (solid lines) (see main text for details).

from previous analysis if $f=1$ is imposed). As may be clearly seen, the scaling is considerably worsened, and also the scaling function is far from the data points.

For temperatures well below $T_C(H)$, outside the critical region delimited by Eq. (2), the fluctuations of the order parameter amplitude are negligible. However, the highly anisotropic nature of this compound leads to a contribution to the magnetization associated with thermal fluctuations of the two-dimensional vortex positions. This contribution has been calculated by Bulaevskii *et al.* in the framework of the GL theory.³² For $H \perp ab$, it may be expressed as

$$\Delta M(T, H) = -f \frac{\phi_0}{8\pi\mu_0\lambda_{ab}^2(T)} \ln\left(\frac{\eta H_{C2}(T)}{H}\right) + f \frac{k_B T}{\phi_0 s} \ln\left(\frac{8\pi\mu_0 k_B T \lambda_{ab}^2(T) H_{C2}(T)}{\alpha s \phi_0^2 \sqrt{e} H}\right). \quad (6)$$

The first term on the right is the conventional London magnetization, while the second one is associated with vortex fluctuations. In this equation, λ_{ab} is the magnetic penetration length in the ab planes, and η and α are constants around the unity. This equation also predicts the crossing of the $\Delta M(T)_H$ curves at a temperature $T_2^* < T_{C0}$, the magnetization at the crossing point being

$$\Delta M_2^* = -f \frac{k_B T_2^*}{\phi_0 s} \ln(\eta \alpha \sqrt{e}). \quad (7)$$

This expression is analogous to Eq. (5) for the crossing point in the critical region, except for a constant around the unity. By combining both expressions, we obtained $\ln(\eta \alpha \sqrt{e}) = \Delta M_2^* T_1^* / \Delta M_1^* T_2^* \approx 0.73$, as expected. The comparison of Eq. (6) with the experimental data is presented in Fig. 4 where, for convenience, ΔM is represented against the magnetic field for several constant temperatures. For each isotherm, the only free parameters are ηH_{C2} and λ_{ab} . The fit quality is excellent for isotherms up to $\sim T_2^*$, except for data under high fields [$H \sim H_{C2}(T)$] which are already inside the critical region. The resulting $\eta \mu_0 H_{C2}(T)$ and $\lambda_{ab}^{-2}(T)$ are presented in the inset. They follow the GL linear temperature dependence, tending to zero at $T_{C0} \approx 77$ K in agreement with the precedent analysis. It is worth noting that the London theory alone fits the data of Fig. 4 as well (it follows the same H dependence). However, it would lead to an anomalous temperature dependence of H_{C2} and λ_{ab} , mainly close to T_2^* .³³ The same happens if an ideal effective volume fraction ($f=1$) is assumed.

Summarizing, the diamagnetism anomalies observed around the Meissner transition in a cuprate superconductor deeply affected by a low effective superconducting volume fraction may be easily overcome by just normalizing the magnetization through the low-field Meissner fraction: The resulting diamagnetism on both sides of T_C may be explained in terms of the conventional Ginzburg-Landau approach with fluctuations of Cooper pairs and vortices. A remarkable result of our present work when compared with previous magnetization measurements in other cuprate single crystals is the unambiguous demonstration of the need of a normalization through the Meissner fraction to eliminate the temperature independent anomalies in the diamagnetism amplitude.³⁴ Our results provide then a further confirmation that the Meissner transition in cuprate superconductors is a conventional GL transition, although in some cases entangled with chemical, structural, or electronic inhomogeneities and disorder.

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