First Observation for a Cuprate Superconductor of Fluctuation-Induced Diamagnetism Well Inside the Finite-Magnetic-Field Regime

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(Received 30 November 1999)

For the first time for a cuprate superconductor, measurements performed above T_c in high quality grain aligned $La_{1.9}Sr_{0.1}CuO_4$ samples have allowed the observation of the thermal fluctuation induced diamagnetism well inside the finite-magnetic-field fluctuation regime. These results may be explained in terms of the Gaussian Ginzburg-Landau approach for layered superconductors, but only if the finite field contributions are estimated by taking off the short-wavelength fluctuations.

PACS numbers: 74.40.+k, 74.20.De, 74.25.Ha, 74.72.Dn

Above any superconducting transition, Cooper pairs with a finite lifetime may be created by the always present thermal agitation energy [1]. One of the most interesting effects associated with these fluctuating Cooper pairs is the appearance, in some cases even well above the normal-superconducting transition temperature in the absence of a magnetic field (T_{c0}), of an appreciable decrease of the normal state magnetization [1,2]. In addition to its intrinsic interest, this effect [called fluctuation diamagnetism (FD)] may be used as a tool to probe the different descriptions of the superconducting transition. In particular, as stressed in Ref. [2] (p. 1062), FD is "useful for exploring the limits of the Ginzburg-Landau theory."

The first observation of the fluctuation induced diamagnetism above T_{c0} in a superconductor was reported by Gollub, Beasley, and Tinkham 30 years ago [3,4]. These measurements were done in different bulk isotropic metallic low-temperature superconductors (LTSC) and by using reduced magnetic fields $h \equiv H/H_{c2}(0)$ [where $H_{c2}(0)$ is the amplitude at $T = 0$ K of the upper critical magnetic field] within $10^{-3} \lesssim h \lesssim 1$. It was soon observed [2,4] that these data could not be explained, even when $h/\epsilon \ll 1$ [where $\epsilon \equiv (T - T_{c0})/T_{c0}$ is the reduced temperature], in terms of the Schmidt [5] approximation (henceforth called "Schmidt limit"), which corresponds to the Gaussian Ginzburg-Landau (GGL) theory in the zero magnetic-field limit. Furthermore, these data could not be explained in terms of the Prange [6] approximation (henceforth called "Prange regime"), which corresponds to the GGL description but by including the finite field contributions. Later, the failures of these approaches were explained in terms of nonlocal effects [7,8], which led to the suppression of the short-wavelength fluctuations. In fact, it is now well established that the nonobservation of the zero magnetic-field regime in LTSC [9], even when $h/\epsilon \ll 1$, is due to the importance of these cutoff effects in the LTSC [2,9].

In the so-called high-temperature cuprate superconductors (HTSC), FD was first observed qualitatively by Freitas, Tsuei, and Plaskett [10] and quantitatively by Lee, Klemm, and Johnston [11]. Because of the very high values of $H_{c2}(0)$ of most of the HTSC, typically of the order of 300 T or more (i.e., 2 orders of magnitude bigger than those of the LTSC), these data were obtained, as well as those measured at a quantitative level since then by various groups in different high quality HTSC samples [12,13], by using reduced magnetic fields within typically $10^{-3} \lesssim h \lesssim 10^{-2}$ (the existing high resolution, SQUID based, magnetometers working under magnetic fields amplitudes typically below 5 T) [14]. These FD results could be explained at a quantitative level in terms of the Schmidt limit by taking into account the layered structure of these materials [11,13,15,16]. This is just the result that one might expect, since for these values of h most of the experimentally accessible window in reduced temperature (typically $10^{-2} \lesssim \epsilon \lesssim 10^{-1}$) corresponds to the zero magnetic-field regime (i.e., $h/\epsilon \ll 1$). However, the comparisons of these results with those previously obtained in LTSC give rise to various crucial and interrelated questions eluded, at least at a quantitative level, until now: Why is the magnetic-field behavior of the FD, even for $h/\epsilon \ll 1$, so different in both types of superconductors? Is there any measurable finite magnetic-field effect on the FD for $h/\epsilon \gtrsim 1$ in HTSC? Is the FD in HTSC also affected by appreciable cutoff effects in the experimentally accessible h/ϵ window?

To answer the above addressed questions, we first present in this Letter detailed measurements of the in-plane magnetization above T_{c0} in a grain aligned $La_{1.9}Sr_{0.1}CuO_4$ (LaSCO) sample with mass as big as 48 mg but with the grains very well aligned and with a quite good stoichiometric homogeneity. The LaSCO system has a relatively low $H_{c2}(0)$, of the order of 45 T. Therefore, the magnetization measurements in this unique sample allow us to accurately determine the FD over almost three decades in h/ϵ, in the window $10^{-2} \lesssim h/\epsilon \lesssim 10$, which, in principle, covers, for the first time in any HTSC, both the zero and the

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finite magnetic-field regimes. Then, these data are analyzed on the grounds of the GGL approaches for layered superconductors.

The grain aligned LaSCO sample used in our present FD experiments was obtained by using the procedure described before [17]. The general aspects of this type of sample may be seen in Refs. [17] and [18]. The main difference of this sample from the other grain aligned LaSCO samples we have already used in other types of experiments [17,19] is its bigger mass, of the order of 50 mg, i.e., typically 2 orders of magnitude larger than most of the HTSC crystals used until now by different groups in other FD measurements [11–13]. However, in spite of its big mass, x-ray diffraction patterns of the sample we have chosen for the present experiments exhibit only the $(00l)$ peaks, which indicates excellent alignment of the grains. So, as stressed above, this type of grain-oriented LaSCO sample probably provides the best compromise for any HTSC to simultaneously have a low $H_{c2}(0)$ (and then a big h, of the order of 10^{-1} for $\mu_0 H = 5$ T), a quite good structural and stoichiometric homogeneity (and then a sharp transition, which allows us to experimentally penetrate as close to T_{c0} as $\epsilon = 10^{-2}$) and a big mass (which with this type of superconductor and for the fields used here may lead to magnetic moments as big as 10^{-7} A m^2, 10^3 to 10^4 times bigger than the instrumental resolution; see below). Also, the fact that the LaSCO is single layered has an additional advantage: The possible complications due to multilayering effects are avoided [11,13,15]. The magnetization measurements were made with a commercial SQUID magnetometer (Quantum Design, model MPMS). The magnetic moment resolution is 10^{-11} A m^2 for magnetic field amplitudes below 1 T and decreases until typically 10^{-10} A m^2 for higher fields. All the FD measurements have been done with the magnetic field applied perpendicularly to the CuO_2 superconducting layers (so, no particular notation will be used when referring to the different observables and parameters). A more detailed description of the experimental setups can be found in Refs. [12] and [17–19].

Some examples of the magnetization excess (normalized to their corresponding H amplitudes) versus temperature curves measured around T_{c0}, $\Delta M(T)_H/H$, are presented in Fig. 1. The data points have been corrected through the in-plane Meissner fraction for inhomogeneities at long length scales and for the possible small misalignment of some grains [17,19]. $T_{c0} = 27.1$ K has been determined within ± 0.2 K by the onset of the diamagnetic transition for H parallel to the CuO_2 layers, a field orientation for which FD is negligible [12,17]. The excess magnetization is defined by $\Delta M(T,H) \equiv M(T,H) - M_B(T,H)$, where $M(T,H)$ and $M_B(T,H)$ are, respectively, the as-measured magnetization and the background magnetization, the latter being associated with the normal contributions. $M_B(T,H)$ has been approximated by extrapolating through the transition the magnetization measured above $T = 36$ K, where the FD contributions are expected to be negligible. The main uncertainties of these data are associated with inhomogeneities (which mainly may affect the ΔM amplitude), with the background estimation (which affects the data mainly for $\epsilon \gtrsim 10^{-1}$) and with the T_{c0} determination (which affects the data mainly for $\epsilon \lesssim 10^{-2}$). In the window $10^{-2} \lesssim \epsilon \lesssim 10^{-1}$ the uncertainties on the absolute ΔM amplitudes are estimated to remain below 20%.

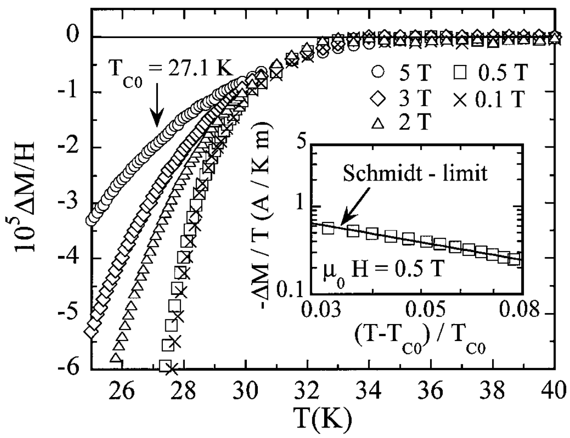

FIG. 1. Some examples of the $\Delta M/H$ versus temperature curves at constant magnetic field amplitudes. In the inset, the data for $\mu_0 H = 0.5$ T are compared with Eq. (1).

The $\Delta M(T)_H/H$ curves in Fig. 1 already illustrate some of the FD aspects that we are studying in this Letter. Note first that for $h/\epsilon \lesssim 1$ (which corresponds to $T \gtrsim 32$ K if $\mu_0 H = 5$ T and to temperatures as close to T_{c0} as $T \gtrsim 28$ K if $\mu_0 H \le 0.5$ T), all the $\Delta M(T)_H/H$ curves are H independent and, therefore, they agree with each other. This is just the FD behavior expected in the Schmidt region, which in single layered superconductors in the 2D limit, the case well adapted for the thermal fluctuations of Cooper pairs above T_{c0} in LaSCO, is given by [13,15,16]

$$\Delta M(\epsilon, h) = -\frac{k_B T}{6\phi_0 s}\,\frac{h}{\epsilon}\,, \tag{1}$$

where ϕ_0 is the magnetic flux quantum, k_B is the Boltzmann constant, and s is the superconducting layers' periodicity (which in this single layered compound is equal to 6.6 Å, one-half the unit cell length in the c direction). A quantitative comparison between the data for $\mu_0 H \le 0.5$ T in the region $3 \times 10^{-2} \lesssim \epsilon \lesssim 8 \times 10^{-2}$ and Eq. (1) is shown in the inset of Fig. 1. This leads to $\mu_0 H_{c2}(0) = 44.2 \pm 0.5$ T.

The results given in Fig. 1 also clearly illustrate the magnetic-field effects on the FD when $h/\epsilon \gtrsim 1$: For $T \lesssim 30$ K and for $\mu_0 H \gtrsim 0.5$ T, the $\Delta M(T)_H/H$ curves are progressively less depressed when H increases. Such a magnetic-field dependence can be much better observed in the example shown for $\epsilon = 3 \times 10^{-2}$ in Fig. 2, where the $\Delta M(H)_\epsilon/T$ data are presented as a function of $\mu_0 H$. In this figure, these data are also compared with the different

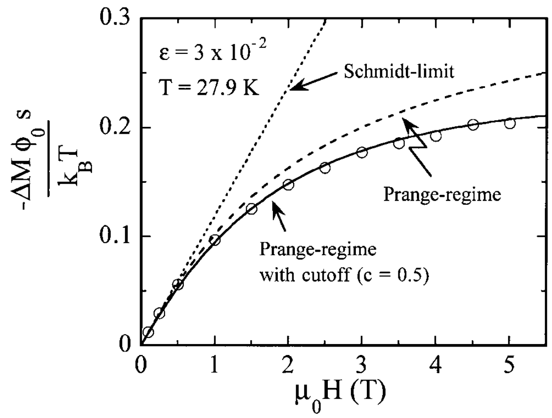

FIG. 2. An example of the $\Delta M/T$ versus H curves at constant reduced temperature and its comparison with the different GGL expressions.

theoretical expressions for $\Delta M(\epsilon, h)$ in single layered superconductors: The dotted line corresponds to the Schmidt limit [Eq. (1)], whereas the solid and dashed curves correspond to the Prange regime with and, respectively, without a cutoff in the energy spectrum. A calculation of $\Delta M(\epsilon, h)$ in the Prange regime without a cutoff was already attempted in Refs. [14] and [20] by using the Lawrence-Doniach-Yamaji (LDY) functional (which includes the terms associated with a nonzero magnetic field) [15,16,21], but these authors arrive at only approximate expressions. Such a calculation is, however, made easy if the cutoff in the kinetic energy spectrum of the fluctuating modes is already introduced, *from the beginning,* in the LDY functional. We will present the details of this procedure, which simplifies the sum over Landau levels of the LDY functional, elsewhere [22]. The final expression for the Prange regime with a cutoff in the 2D limit is [22]

$$\Delta M(\epsilon, h, c) = -\frac{k_B T}{\phi_0 s}\left\{-\left(\frac{\epsilon}{2h} + \frac{c}{2h}\right)\psi\left(\frac{1}{2} + \frac{\epsilon}{2h} + \frac{c}{2h}\right) - \ln\Gamma\left(\frac{1}{2} + \frac{\epsilon}{2h}\right) + \frac{\epsilon}{2h}\psi\left(\frac{1}{2} + \frac{\epsilon}{2h}\right) + \ln\Gamma\left(\frac{1}{2} + \frac{\epsilon}{2h} + \frac{c}{2h}\right) + \frac{c}{2h}\right\}, \tag{2}$$

where Γ and ψ are the gamma and, respectively, the digamma functions and c is a dimensionless energy cutoff amplitude. Such a cutoff accounts for the maximum in-plane kinetic energy of the Cooper pairs through $E_{\max} = c\hbar^2/[2m^*\xi^2(0)]$, m^* being their effective mass and $\xi(0)$ the in-plane coherence length at $T = 0$ K.

Equation (2) is general and includes the Prange regime in a 2D single layered superconductor *without a cutoff* as a particular case, which corresponds to $h \ll c$ and, simultaneously, $\epsilon \ll c$. Under these conditions, Eq. (2) leads to

$$\Delta M(\epsilon, h)_{c\to\infty} = -\frac{k_B T}{\phi_0 s}\left\{-\ln\Gamma\left(\frac{1}{2} + \frac{\epsilon}{2h}\right) + \ln\sqrt{2\pi} + \frac{\epsilon}{2h}\left[\psi\left(\frac{1}{2} + \frac{\epsilon}{2h}\right) - 1\right]\right\}. \tag{3}$$

(This equation includes, indeed, the approximate expression already obtained in Ref. [20].) It is also useful to comment here that two asymptotic cases may be obtained from Eq. (3): The Schmidt limit [Eq. (1)] is recovered by simply imposing $h/\epsilon \ll 1$; and the saturation value of the scaled magnetization at $h/\epsilon \gg 1$, which is found to be $\ln\sqrt{2}$ (the approximate value 0.346 was obtained in Refs. [20] and [23]).

The solid line in Fig. 2 corresponds to the fit of Eq. (2) to the data with the energy cutoff c as the only free parameter, $\mu_0 H_{c2}(0) = 44.2$ T being the value previously determined in the Schmidt region. As can be seen in this figure, the agreement with the data is excellent, and it leads to $c = 0.5$. Similar results, again with $c = 0.5$, were obtained for other reduced temperatures which cover all the experimentally accessible ϵ window ($2 \times 10^{-2} \leq \epsilon \leq 10^{-1}$). However, due to the uncertainties in the ΔM amplitudes, the dispersion of the absolute c values is estimated to be between 0.3 and 1. For completeness, in Fig. 2 we also show the Prange regime without a cutoff [Eq. (3) with again $\mu_0 H_{c2}(0) = 44.2$ T], which clearly does not agree with the data.

An overview of the FD behavior as a function of h/ϵ is shown in Fig. 3, together with the theoretical predictions [Eqs. (1) to (3), always with $\mu_0 H_{c2}(0) = 44.2$ T and $c = 0.5$]. The circles and squares data points correspond to $\mu_0 H = 0.5$ T and, respectively, $\mu_0 H = 5$ T in all the

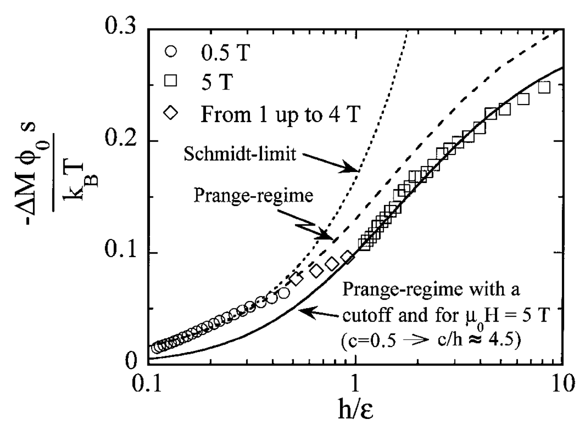

FIG. 3. An example of the h/ϵ dependence of the dimensionless fluctuation induced diamagnetism showing the different behavior for $h/\epsilon < 1$ and $h/\epsilon > 1$. The dependence on the ratio c/h can also be clearly observed (see main text).

experimentally accessible ϵ window. For completeness, in this figure we have also represented with rhombi some data for $\mu_0 H$ between 1 and 4 T. As can be observed in this figure, for $\mu_0 H = 5$ T the data show an excellent agreement with the Prange regime with the cutoff corresponding to $c/h \simeq 4.5$ (solid line). The data points from $\mu_0 H = 4$ T down to 1 T illustrate how the cutoff effects become less important when the ratio c/h increases (see above), being negligible for $\mu_0 H \lesssim 0.5$ T. These results lead to another important aspect of the FD behavior, already observed in LTSC [4,9], but indeed never observed until now in a HTSC compound: ΔM not only depends on h/ϵ but also on c/h. Let us stress also that the sample independence of the $\Delta M(\epsilon, h)$ behavior has been checked by measurements in the other grain-oriented LaSCO sample.

Let us finally note that by simply equating at $\epsilon = 0$ Eq. (2) with one-half the value predicted by Eq. (3), c can be easily related to the scaling field H_s, used to analyze the FD in LTSC [4,9]. For $c = 0.5$, this leads to $H_s/H_{c2}(0) \simeq 0.8$, i.e., a ratio 20 times higher than in clean LTSC. In other words, the present results also demonstrate experimentally another important aspect until now only suggested [11,15]: The kinetic energy cutoff of the fluctuation modes affects much less the FD in LaSCO than in LTSC in the clean limit (this last limit being the one well adapted to compare with the HTSC). This striking result, associated at least in part with the lower dimensionality behavior of FD in HTSC (a dimensionality effect predicted for the LTSC [23], but not observed until now), explains why the Schmidt regime, which was never observed in LTSC, may be easily observed in HTSC.

In conclusion, the fluctuation diamagnetism well inside both the zero and the finite-magnetic-field regimes has been observed for the first time in a cuprate superconductor. When compared with the GGL theoretical expressions, the results presented here provide unambiguous answers to the questions addressed in the introduction of this Letter. Indeed, they have also opened new interesting theoretical and experimental questions as, for instance, those that concern the amplitude of the cutoff in the energy spectrum of the Cooper pairs in other HTSC or the implications of such a cutoff on the physics of these superconductors.

This work has been supported by the CICYT, Spain, under Grant No. MAT98-0371, by the CICYT-FEDER Funds, Spain, under Grant No. 1FD97-0146, and by Unión Fenosa under Grant No. 0666-98. C. C. acknowledges financial support from Fundación Ramón Areces (Spain).

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