Fluctuation Induced Diamagnetism in the Zero Magnetic Field Limit in a Low Temperature Superconducting Alloy

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By using a Pb-18 at. % In alloy, the fluctuation induced diamagnetism was measured in the zero magnetic field limit, never observed until now in a low-T_C superconductor. This allows us to disentangle the dynamic and the nonlocal electrodynamic effects from the short-wavelength fluctuation effects. The latter may be explained on the grounds of the Gaussian-Ginzburg-Landau approach by introducing a total energy cutoff in the fluctuation spectrum, which strongly suggests the existence of a well-defined temperature in the normal state above which all fluctuating modes vanish. This conclusion may also have implications when describing the superconducting state formation of the high-T_C cuprates.

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The normal state behavior of a superconductor may be appreciably affected, mainly near the superconducting transition temperature, by the presence of fluctuating Cooper pairs created by the unavoidable thermal agitation energy. These thermal fluctuation effects were already predicted and observed more than 30 years ago in low temperature metallic or alloy superconductors (LTSC) [1]. In addition to their intrinsic interest, these fluctuation effects provided a useful tool to probe various general aspects of these superconductors and at present they are a textbook subject [2].

The discovery of the so-called high temperature cuprate superconductors (HTSC) has considerably enhanced the interest for the thermal fluctuation effects: As it is now well established [3,4], in HTSC the relative amplitude of these effects may be in some cases orders of magnitude larger than in LTSC. But, in addition, it was realized more recently that the presence of Cooper pairs well inside the normal region may directly concern some of the most central and still open aspects of the HTSC physics, including the formation of their superconducting state [5].

Although until now relatively unexplored at a quantitative level, a natural and promising way to better understand the creation and behavior of the fluctuating Cooper pairs in the normal state of the HTSC is to compare the thermal fluctuation effects in these superconductors with those arising in conventional (described by the BCS theory) LTSC. Probably, the best observable for this task is the so-called fluctuation induced diamagnetism (FD), i.e., the decrease of the normal state magnetization due to the presence of fluctuating Cooper pairs. In contrast with most of the other thermal fluctuation effects in superconductors, the FD is not only proportional to the density of fluctuating Cooper pairs but also to some extent to their dimension, i.e., to the superconducting coherence length [1,2]. As a consequence, the FD amplitude relative to the normal state magnetization is also quite important in bulk isotropic (3D) LTSC, due to their relatively large coherence length amplitudes (extrapolated to $T = 0$ K), $\xi(0)$. This is indeed an important and quite unique experimental advantage, which has already allowed a comparison between some of the earlier FD measurements of Gollub and co-workers in LTSC in the finite magnetic field (Prange) regime [6] and those recently obtained in a similar field regime in a HTSC [7]. One of the conclusions of such a comparison was that the HTSC are much less affected by dynamic and nonlocal electrodynamic effects than the LTSC studied until now. These striking differences, which may be in part attributed to the extremely type-II nature of the HTSC and to their reduced dimensionality [1], provide a first qualitative explanation of why, in contrast with the HTSC, the FD in LTSC had never been observed until now in the so-called zero magnetic field (or Schmidt and Schmid) limit. This limit is characterized, independently of the fluctuation dimensionality, by a linear dependence of the FD amplitude on H, the applied magnetic field [8].

The results summarized above enhance the interest for a possible experimental observation in LTSC of the FD in the zero magnetic field limit. In this Letter, we present detailed measurements of the fluctuation induced excess magnetization, $\Delta M(\epsilon, h)$, in a Pb-18 at. % In alloy. Here $h \equiv H/H_{c2}(0)$ and $\epsilon \equiv \ln(T/T_{C0})$ are, respectively, the reduced magnetic field and temperature, $H_{c2}(0)$ is the upper critical magnetic field amplitude (extrapolated to $T = 0$ K), and T_{C0} is the critical temperature at $H = 0$. These FD data will allow a separation of the dynamic and the nonlocal effects from the short-wavelength fluctuation effects. We will also see that the latter, which manifest mainly in the so-called high reduced temperature region, for $\epsilon \gtrsim 0.1$, may be taken into account by introducing a "total energy" cutoff in the Gaussian-Ginzburg-Landau (GGL) approach, the cutoff amplitude being similar, well within the combined experimental uncertainties, to the one already found in a HTSC ($YBa_2Cu_3O_{7-\delta}$, henceforth called Y-123) [9,10].

The fitness of the Pb-In alloy used here to observe the FD in the zero-field limit is due to the fact that this LTSC is a dirty superconductor ($\xi_0/\ell \simeq 16$, where ξ_0 is the

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Pippard coherence length and ℓ is the mean-free path of the normal carriers) with a relatively high Ginzburg-Landau parameter, $\kappa = \lambda/\xi \simeq 3.8$ (λ is the magnetic field penetration length), which decreases the nonlocal effects [6]. But, in addition, this alloy has a still quite large coherence length $\xi(0) = 198$ Å, which increases the FD amplitude and contributes then to make possible the accurate measurement of the FD under quite low magnetic fields (which, in turn, again dramatically reduce the possible nonlocal and dynamic effects). In fact, by using a polycrystalline sample of 0.14 cm^3, it was feasible to determine $\Delta M(\epsilon, h)$ in the ranges $10^{-3} \lesssim h \lesssim 1$ and $10^{-2} \lesssim \epsilon \lesssim 0.6$. The big sample used here, which presents an excellent stoichiometric homogeneity, was grown following a standard procedure described elsewhere [11]. Let us just indicate that $T_{C0} = 6.95$ K was determined from resistivity measurements, in full agreement with the extrapolation to $H = 0$ of $T_C(H)$ determined from magnetization measurements, which also lead to $\mu_0 H_{c2}(0) = 0.83$ T. These magnetization measurements were performed by using a commercial SQUID magnetometer (Quantum Design, model MPMS).

An example of the as-measured magnetization as a function of the temperature at constant applied magnetic field, $M(T)_H$, is shown in Fig. 1(a). A detail around T_{C0} and T^C, the temperature where the fluctuation induced magnetization becomes zero, is shown in Fig. 1(b) (see below). Figure 1 also shows the background magnetization (dotted lines), $M_B(T)_H$, obtained by extrapolating through the transition the normal magnetization measured between $T/T_{C0} = 3.5$ and 5, a temperature region indicated by the two arrows on the right side of Fig. 1(a) and where the fluctuation effects are expected to be negligible. In fact, above $T/T_{C0} \simeq 1.7$ and up to the highest studied temperatures ($T/T_{C0} \simeq 6$), these $M(T)_H$ data do not show, at a quantitative level, any deviation from linear behavior as a function of T, which demonstrates then the robustness of such a background estimation. This is indeed another crucial experimental advantage of the Pb-In alloy studied here, mainly in the high reduced temperature region, when extracting the measured fluctuation induced excess magnetization, defined as $\Delta M(T)_H \equiv M(T)_H - M_B(T)_H$.

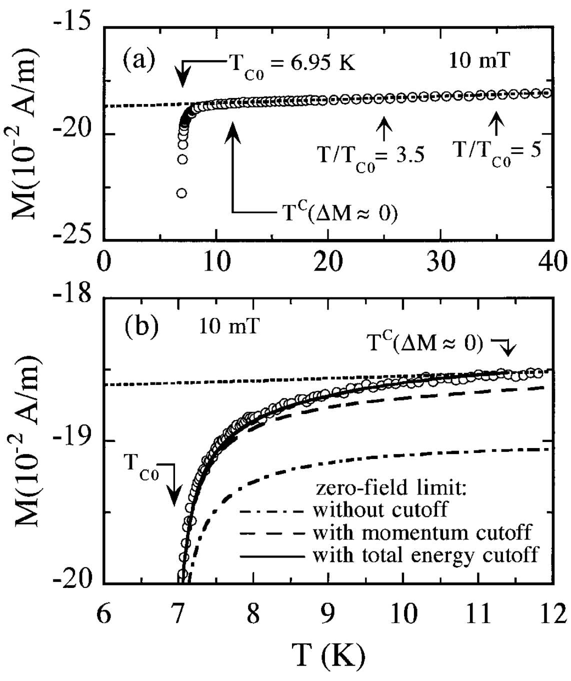

FIG. 1. (a) An example of the as-measured magnetization versus temperature at constant applied magnetic field. The background (dotted line) was extrapolated from the region $3.5 \leq T/T_{C0} \leq 5$. Above T^C the fluctuation effects vanish. (b) A detail around T_{C0} and the temperature where the fluctuation induced magnetization becomes zero, together with the predictions of the zero-field approach under different cutoff conditions. When compared to each other, these two figures provide evidence that the differences between both cutoff conditions cannot be absorbed by any realistic background.

Some examples of $\Delta M(T)_H$ curves, normalized to their corresponding H amplitudes, measured around T_{C0} are presented in Fig. 2(a). These curves already illustrate at a qualitative level some of the FD aspects that we are studying in this Letter. Note that the two curves obtained under the lowest fields agree with each other even very close to T_{C0}, i.e., they are H independent. This is the FD behavior expected in the zero-field limit, which in bulk isotropic (3D) superconductors is given by [8]

$$\Delta M(T, H) = -\frac{\pi k_B T}{6\phi_0^2} \xi(T) \mu_0 H , \tag{1}$$

where k_B is the Boltzmann constant, ϕ_0 is the magnetic flux quantum, and $\xi(T) = \xi(0)\epsilon^{-1/2}$. Such behavior for $\mu_0 H \leq 10$ mT indicates then the absence of appreciably nonlocal and dynamic effects which would introduce a

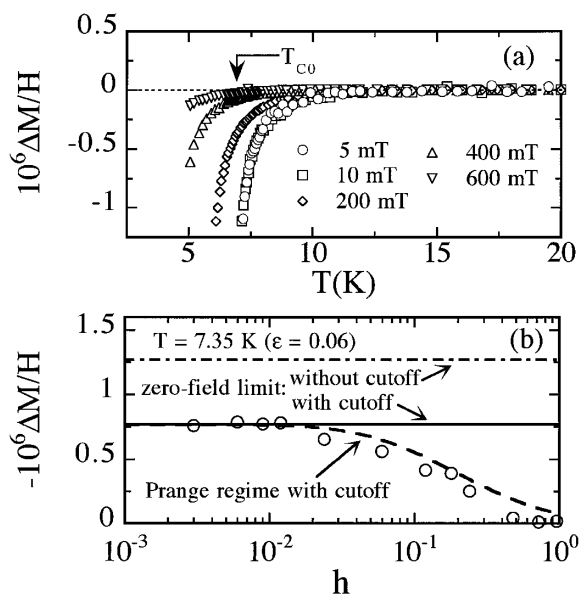

FIG. 2. (a) Some examples of the fluctuation induced magnetization versus temperature curves normalized to their corresponding magnetic field amplitudes. (b) Magnetic field dependence of the fluctuation induced magnetization at constant temperature.

strong h dependence. However, as can be seen in Fig. 2(b), the *amplitude* which may be obtained by using in this equation $\xi(0) = 198$ Å [determined from our magnetization measurements of $H_{c2}(T)$] clearly disagrees with the experimental data even in the zero-field limit ($h/\epsilon \lesssim 1$). This result already suggests the need for the regularization of the GGL approach through a cutoff to eliminate the short-wavelength fluctuations, which are always overestimated by the GGL theory [1,2]. These difficulties are much more important in 3D than in 2D or layered superconductors, just because the reduced dimensionality already introduces a cutoff [9,10]. So, we must calculate the FD in a 3D superconductor and in the zero-field limit under a cutoff. Let us, however, first briefly note that the results of Fig. 2(a) also illustrate the magnetic field effects on the FD when $h/\epsilon \gtrsim 1$: 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 $\epsilon \simeq 6 \times 10^{-2}$ in Fig. 2(b), where the $\Delta M(T)_H/H$ data are represented as a function of h.

To regularize the GGL approach we empirically introduce a *total energy* cutoff given by (in units of $\hbar^2/2m^*$, where $\hbar$ is the reduced Planck constant and m^* is the effective mass of the Cooper pairs)

$$k^2 + \xi^{-2}(\epsilon) < c\xi^{-2}(0), \tag{2}$$

where k is the modulus of the wave vector of the fluctuation modes and $c \leq 1$ is the cutoff amplitude which, in the absence of nonlocal and dynamic effects, may be approximated as a constant (see below). Equation (2) eliminates the most energetic fluctuation modes and not only those with short wavelengths [1,2]. This total energy cutoff is then obtained by adding to the kinetic energy of the fluctuating modes (proportional to k^2) a contribution arising from the "spatial smallness" of the fluctuations. This last contribution may be seen as due to the "localization energy" associated with the shrinkage, when the temperature increases, of the superconducting wave function. An analysis of this localization energy in terms of the uncertainty principle which directly leads, below *but also above* T_{C0}, to the condition $\xi(T) \gtrsim \xi_0$ (where ξ_0 is the actual coherence length at $T = 0$ K) will be published elsewhere [12]. The adequacy of such a total energy cutoff is being suggested by our recent results in a HTSC [9,10]. To probe its adequacy also in a LTSC is another aim of this Letter. Note also here that, by using $\xi(\epsilon) = \xi(0)\epsilon^{-1/2}$, Eq. (2) may be rewritten as $k^2 < (c - \epsilon)\xi^{-2}(0)$. Therefore, for $\epsilon \ll c$ the total energy cutoff reduces to the conventional momentum cutoff. However, in contrast with all the regularization conditions proposed until now, Eq. (2) *suppresses all the fluctuation modes* above a well-defined temperature, ϵ^C, defined by $\epsilon^C = c$ [which will correspond to $\epsilon^C \equiv \ln(T^C/T_{C0})$, with T^C defined by $\xi(T^C) = \xi_0$ [12]].

The easiest way to introduce the above cutoff condition in the FD calculations is to extend to the 3D case our recent GGL results in layered superconductors [7]. This may be straightforwardly done by following the procedure proposed by Hikami and Larkin in the case of the magnetoconductivity without any cutoff [13]. In the Prange regime this leads to

$$\Delta M(\epsilon, h, c, c_z)_E = -\frac{k_B T}{\pi \phi_0 \xi(0)} \int_0^{\sqrt{c_z - \epsilon}} d\tilde{k}_z \Big[-\frac{c + \tilde{k}_z^2}{2h} \psi\Big(\frac{h + c + \tilde{k}_z^2}{2h}\Big) - \ln\Gamma\Big(\frac{\epsilon + h + \tilde{k}_z^2}{2h}\Big) + \ln\Gamma\Big(\frac{h + c + \tilde{k}_z^2}{2h}\Big) + \frac{\epsilon + \tilde{k}_z^2}{2h} \psi\Big(\frac{\epsilon + h + \tilde{k}_z^2}{2h}\Big) + \frac{c - \epsilon}{2h}\Big], \tag{3}$$

where Γ and ψ are, respectively, the gamma and digamma functions, $\tilde{k}_z \equiv k_z \xi(0)$ and c_z are, respectively, the dimensionless momentum of the fluctuations and the cutoff in the z direction (parallel to H), and c is the cutoff in the directions perpendicular to the applied magnetic field. Note that the z spectrum of the fluctuations is not sensible to the applied magnetic field and, therefore, $c \to c_z$ in the absence of appreciable nonlocal or dynamic effects. This further simplifies the ΔM expression in the zero-field limit under a total energy cutoff, which may be easily obtained from Eq. (3) by just imposing $c_z = c$ and $h \ll \epsilon, c$:

$$\Delta M(\epsilon, h, c)_E = -\frac{k_B T}{6\pi \phi_0 \xi(0)} h \Big(\frac{\arctan\sqrt{(c - \epsilon)/\epsilon}}{\sqrt{\epsilon}} - \frac{\arctan\sqrt{(c - \epsilon)/c}}{\sqrt{c}} \Big), \tag{4}$$

where the cutoff strength, c, is a constant (independent of ϵ and h) close to 1 [14].

The FD expressions in a 3D superconductor under a momentum cutoff in the zero-field limit and in the Prange regime may be easily obtained by just changing c and c_z by, respectively, $c + \epsilon$ and $c_z + \epsilon$ in Eqs. (3) and (4). Note also that these expressions also include the ones without cutoff as a limiting case, which corresponds to $\epsilon, h \ll c$.

The solid line in Fig. 2(b) was obtained from Eq. (4) with again $\xi(0) = 198$ Å [and, then, $\mu_0 H_{c2}(0) = 0.83$ T] and with c as the only free parameter. This leads to $c = 0.5$. So, the central task now is to probe whether this cutoff amplitude also explains the ϵ behavior of $\Delta M(\epsilon)_h$ in the zero-field limit, mainly in the high reduced temperature region, where the short-wavelength effects are expected to be very important. But before that, let us just note here that by using this cutoff amplitude in Eq. (3), together with $c_z = c$, we obtain the dashed line in Fig. 2(b). Although the h behavior of this Prange approximation with cutoff agrees qualitatively with the measurements, the amplitude

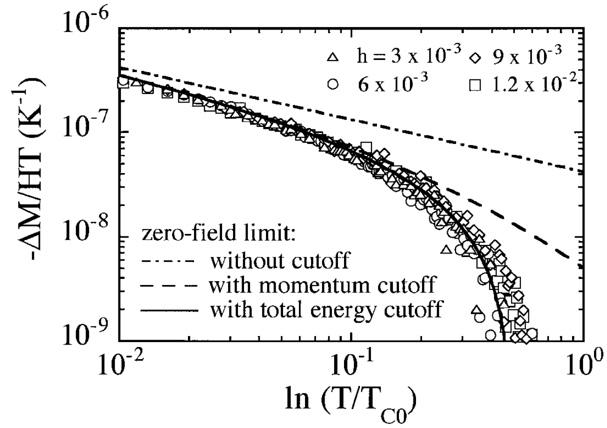

FIG. 3. Reduced temperature dependence of the fluctuation induced magnetization over HT measured under weak reduced fields. The curves correspond to the zero-field approximation under different cutoff conditions.

differences, which are quite important, are due to the presence at these relatively high h values of nonlocal and dynamic effects.

An overview of the FD data for $h \lesssim 10^{-2}$ for all the experimentally accessible reduced temperatures is presented in Fig. 3, together with a comparison with the GGL approach in the zero-field limit under different cutoff conditions, always with $\xi(0) = 198$ Å and $c = 0.5$. The dispersion between these data points represents well all the experimental uncertainties, including those associated with the background magnetization estimations, and it leads to an uncertainty on the c value of less than 15%. These FD results penetrate, to our knowledge for the first time in a LTSC and under any magnetic field, well inside the high reduced temperature region (for $\epsilon \gtrsim 0.1$), where the short-wavelength effects are expected to dominate the fluctuation spectrum. In addition, the data for different reduced magnetic fields agree with each other. This confirms that they correspond well to the FD in the zero-field limit, to our knowledge never observed before in LTSC at any reduced temperature. When compared with the GGL approach under different cutoff conditions, these results provide the first experimental demonstration of the adequacy of the total energy cutoff condition given by Eq. (2) to explain the FD in the zero magnetic field limit in a LTSC in all the experimentally accessible reduced temperatures, including the high ϵ region. Some aspects of these results are also shown in Fig. 1(b), now in linear scales.

In conclusion, the comparison of our present results on $\Delta M(\epsilon)_h$ in a LTSC with those that we have recently obtained in a HTSC (Y-123) [9,10] suggests that, independently of the absolute values of $\xi(0)$, *coherent* Cooper pairs cannot exist in the normal state for $\epsilon \gtrsim c$ ($\lesssim$1) (i.e., *also above* T_{C0}, the smallest possible size of the superconducting coherence is of the order of ξ_0 [12,14]). In addition to their interest in the understanding of the thermal fluctuations above T_{C0} in superconductors, these findings may have implications in the superconducting state formation in HTSC. For instance, in the scenarios where the local pairing is supposed to occur at a different temperature (T^*) than the long range phase order (T_{C0}) [5], our results suggest that $\ln(T^*/T_{C0}) \leq c$ [12]. It would be, however, of crucial importance to probe the universality of these ideas by studying other LTSC and HTSC compounds (in this last case with different dopings) and other observables. From a theoretical point of view, it will be useful to further work on the physical meaning of the total energy cutoff condition on the grounds of both the microscopic and the GGL approaches (in this last case by introducing higher order terms in the Ginzburg-Landau functional).

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