PHYSICAL REVIEW B **70**, 060501(R) (2004)

Experimental observation of the breakdown by a magnetic field of the superconducting fluctuations in the normal state

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(Received 7 April 2004; published 6 August 2004)

The effects induced on the normal state magnetization by fluctuating Cooper pairs have been measured in $Pb_{1-x}In_x$ alloys up to magnetic fields above $H_{C2}(0)$, the upper critical field extrapolated to $T=0$ K. Our results show that in dirty alloys these superconducting fluctuation effects are, in the entire $H-T$ phase diagram above $H_{C2}(T)$, independent of the amount of impurities and that they vanish when $H \sim 1.1 H_{C2}(0)$. These striking results seem to be consistent with the limits imposed by the uncertainty principle to the shrinkage of the superconducting wave function.

DOI: 10.1103/PhysRevB.70.060501 PACS number(s): 74.40.+k, 74.20.De, 74.25.Ha, 74.70.Ad

It is now well established that in addition to their intrinsic interest the fluctuating Cooper pairs created in the normal state by the unavoidable thermal agitation provide a useful tool in studying the superconducting transition.[1] As a consequence, the superconducting fluctuations (SCF) in the normal state have been extensively studied in low- and high-T_c superconductors, and at present many of their main aspects are well understood.[1–3] However, the behavior of the SCF in the normal state under strong magnetic fields, of the order of the upper critical field extrapolated to $T=0$ K, $H_{C2}(0)$, is still an open problem.[3,4]

Here we will present measurements of $\Delta M(T,H)$, the decrease induced by SCF on the magnetization in the normal state (the so-called fluctuation-induced magnetization),[5] in $Pb_{1-x}In_x$ alloys, with $0 \leqslant x \leqslant 0.45$, and up to fields well above $H_{C2}(0)$. Our experiments show that independently of the superconductors' dirtiness $\Delta M(T,H)$ vanishes when H becomes close to $1.1 H_{C2}(0)$. This striking behavior is not accounted for by the as-published expressions for $\Delta M(T,H)$ obtained on the grounds of the existing phenomenological[1–4,6–8] or microscopic[1–4,9–11] approaches. However our analysis suggest that, as was the case at high reduced temperatures but low fields [in low-T_c (Refs. 12–14) and high-T_c (Refs. 13–15) superconductors], at high fields $\Delta M(T,H)$ vanishes when the superconducting coherence length becomes of the order of the one at $T=0$ K (its minimum value compatible with the uncertainty principle), in spite of the fact that the magnetic field is an antisymmetric perturbation.[1]

Among the available low- and high-T_c superconductors, we choose the Pb-In alloys to study the high-field behavior of the fluctuation-induced magnetization in the normal state for four main reasons: i) Its entire $H-T$ phase diagram is, even for $H \gg H_{C2}(0)$, easily accessible with the existing high resolution, superconducting quantum interference device [SQUID] based, magnetometers. ii) By changing the In concentration, it is possible to cover both type I and type II superconductors and also the range from the clean to the dirty limits. This is a crucial advantage because it has allowed us to separate the "universal" magnetic field effects on the SCF from those associated with the dynamic and nonlocal electrodynamic effects, these last being strongly material-dependent.[1–4,9,13–16] iii) It is also possible to obtain alloys with high stoichiometric quality. This is another crucial advantage, because it minimizes the spurious magnetization roundings associated with T_c inhomogeneities that otherwise would be entangled with the intrinsic rounding due to the SCF. iv) The normal-state magnetic susceptibility of these samples is almost independent of T and H up to, at least, $5T_{c0}$ and $5H_{C2}(0)$. This allows a very reliable obtainment of the background magnetization around $T_c(H)$ by linear extrapolation of the as-measured $M(T)_H$ or $M(H)_T$ well above $T_c(H)$.

The Pb-In alloys were prepared by first melting small pieces of the precursor Pb (Johnson Matthey, 99.9999% purity) and In (Alfa Aesar, 99.999% purity) in the appropriate proportions, and then annealing for 10 days at 3 to 5 K below the corresponding *solidus* temperature of each alloy. Other details of the samples preparation may be seen in Ref. 17. For the magnetic measurements we cut from the samples' inner part (more homogeneous) cylinders of typically ~6 mm diameter and ~6 mm height in order to optimize the available volume of our measurement system (a commercial SQUID magnetometer, Quantum Design, model MPMS2). Characterization by energy dispersive spectrometry showed that the In concentrations matched the nominal ones well within 10%. The relative changes in In concentration between different parts of each sample were less than 5%. Moreover, some of the samples have been electrochemically coated with Cu to eliminate the surface superconductivity between $H_{C2}(T)$ and $H_{C3}(T)$, which otherwise would complicate the analysis above $H_{C2}(T)$ for $T < T_{c0}$, the zero-field critical temperature.[16]

The normal-superconducting transition temperature in zero field, T_{c0}, was determined from measurements of the field-cooled (FC) magnetic susceptibility, χ^{FC}, versus temperature. Some examples are shown in the inset of Fig. 1. These measurements were performed with an external magnetic field of 0.5 mT which is much smaller than the corresponding lower critical magnetic field (at least for temperatures farther from T_{c0} than ~1 mK). The demagnetizing effects were estimated through the ellipsoidal approximation.

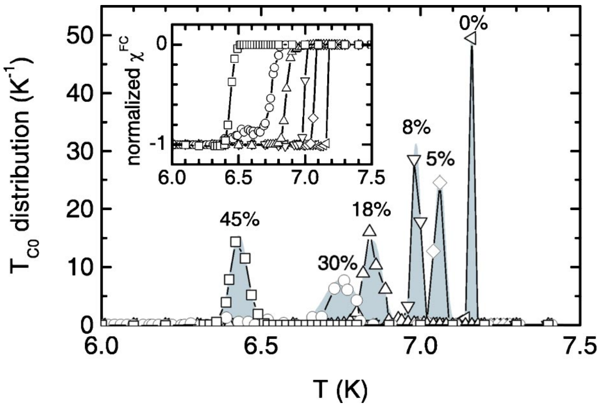

FIG. 1. T_{c0} distribution for each of the studied Pb-In alloys, as deduced from the $\chi^{FC}(T)$ curves normalized to -1 presented in the inset. The lines are guides for the eyes and the shaded areas represent the Gaussian distributions that best fit the data points (see the main text for details). Percentages indicate the In concentrations of each of the samples.

In the main Fig. 1, the T_{c0} distribution for each alloy is presented, obtained as the derivative of the corresponding $\chi^{FC}(T)$ curve. The shaded areas represent the best fits of Gaussian distributions $\exp[-(T-T_{c0})^2/\Delta T_{c0}{}^2]/\sqrt{\pi}\Delta T_{c0}$, with T_{c0} and ΔT_{c0} as free parameters. The resulting T_{c0} and ΔT_{c0} values are compiled in Table I. The other parameters summarized in Table I were obtained from measurements of the magnetization versus applied magnetic field at different constant temperatures below the transition and from the electrical resistivity versus temperature curves. For some In concentrations, there exist already detailed information of the general properties of the Pb-In alloys that were summarized in various earlier works addressing other phenomena in these materials.[17] We have checked that the data of Table I are in excellent agreement with these previous results.

The presence of fluctuating Cooper pairs above $T_c(H)$ produces a rounding of the as-measured susceptibility versus magnetic field curves, as illustrated in Fig. 2. This example also shows that this rounding is progressively reduced as the

TABLE I. Main parameters of the Pb-In alloys studied in this work. T_{c0} was determined from the field-cooled $M(T)_H$ curve at $\mu_0 H=0.5$mT. The Ginzburg-Landau parameter κ and $H_{C2}(0)$ were obtained from the reversible $M(T)_H$ curves in the mixed state. ξ_0^{Pb} follows from $\xi_0^{Pb}=1.35\xi^{Pb}(0)$, $\xi^{Pb}(0)$ from $\xi^2(0)=\phi_0/2\pi\mu_0 H_{C2}(0)$, and ℓ from measurements of the residual resistivity just above T_{c0}.

In at. %	T_{c0} (K)	ΔT_{c0} (K)	$\mu_0 H_{C2}(0)$ (T)	κ	ℓ (Å)	ξ_0^{Pb}/ℓ
0	7.18	0.01	0.070[a]	0.30[a]	$\sim 5\times 10^4$	$\sim$0.02
5	7.06	0.02	0.29	1.3	200	2.7
8	6.99	0.02	0.49	2.1	130	7.1
18	6.85	0.04	0.86	3.4	67	14
30	6.75	0.07	1.0	4.2	57	16
45	6.43	0.04	1.2	5.5	42	22

[a]Extrapolated from the values of the Pb-In alloys using the Gor'kov theory (Ref. 1).

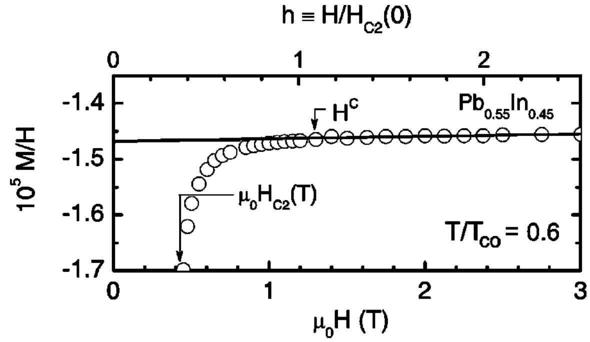

FIG. 2. An example, corresponding to the $Pb_{0.55}In_{0.45}$ alloy, of the as-measured magnetic susceptibility vs applied magnetic field at constant temperature below T_{c0}. The solid line is the background susceptibility, obtained by a linear fit in a region far from the superconducting transition, in this example from 2 to 3 T [i.e., from $\sim 5H_{C2}(T)$ to $\sim 7H_{C2}(T)$].

applied field increases and it vanishes when the reduced field, $h\equiv H/H_{C2}(0)$, becomes close to 1.1. These finite-field effects may be described quantitatively through the $\Delta M(h)_\varepsilon$ curves, as the one presented in Fig. 3, which corresponds to a temperature above T_{c0} [i.e., $\varepsilon>0$, where $\varepsilon\equiv\ln(T/T_{c0})$ is the reduced temperature]. These results show that when $h \gtrsim 0.2$, $\Delta M(h)_\varepsilon$ begins to decrease and for $h\gtrsim 1.1$ the fluctuation-induced magnetization vanishes. This behavior at high fields is confirmed by the experimental results at T_{c0} for all the samples studied in this work and summarized in Fig. 4: Independently of the superconductor dirtiness and also of their type-I or type-II character, $\Delta M(h)_{T_{c0}}$ vanishes when $h=1.1\pm 0.2$. Another central result shown in Fig. 4 is that when normalized by $H^{1/2}T_{c0}$ all the $\Delta M(h)_{T_{c0}}$ data for the different dirty alloys collapse on the same curve. In contrast, the data for pure Pb, which is in the clean limit, are appreciably lower. These last differences, that were already observed in other low-T_c alloys by Tinkham and co-workers in their pioneering measurements at low and intermediate fields (up to $h\simeq 0.6$), are due to the presence of appreciable nonlocal electrodynamic effects in the clean and low-κ Pb.[1–4,16]

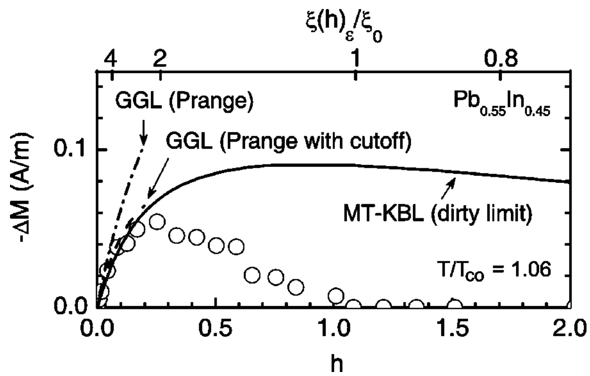

FIG. 3. An example, corresponding to the $Pb_{0.55}In_{0.45}$ alloy, of the reduced-magnetic-field dependence of the fluctuation-induced magnetization, at $T/T_{c0}=1.06$ (which corresponds to a reduced-temperature of $\varepsilon=6.2\times 10^{-2}$). The upper scale shows that $\Delta M(h)_\varepsilon$ vanishes around $\xi(h)_\varepsilon\simeq\xi_0$. The curves correspond to different theoretical approaches, as explained in the main text.

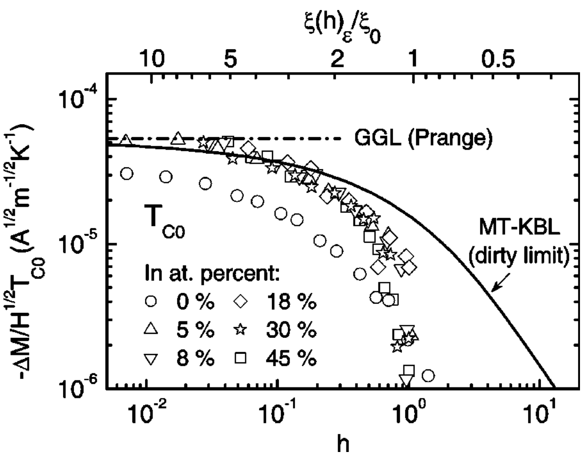

FIG. 4. $\Delta M(h)/H^{1/2}T_{c0}$ vs h at T_{c0} for all the compounds studied here. The upper scale illustrates that for all the compounds the fluctuation effects at T_{c0} vanish, sharply in this scale, when $\xi(h)_\varepsilon \simeq \xi_0$ (which corresponds to $h \simeq 1.1$). The data for pure Pb are strongly affected by non-local electrodynamic effects which, even at low fields, decrease the ΔM amplitude.

The results of Figs. 3 and 4 are particularly well adapted to be compared to the existing phenomenological[6–8] and microscopic calculations of $\Delta M(\varepsilon, h)$.[9–11] The dot-dashed curve in Fig. 3 corresponds to the Prange expression for $\Delta M(\varepsilon, h)$,[7] which is based on the Gaussian-Ginzburg-Landau (GGL) approach and that, therefore, is only applicable at low fields ($h \ll 1$). This curve was calculated with no free parameters, by using the values of Table I. As may be seen in Fig. 3, at low fields ($h \lesssim 0.05$) where this approach is applicable $\Delta M(h)_\varepsilon$ agrees at a quantitative level with the Prange predictions. This result provides then a further indication that in this dirty superconductor $\Delta M(h)_\varepsilon$ is not appreciably affected by non-local electrodynamic effects. As it is now well established,[1–4] the Prange approach may be extended up to intermediate fields ($h \simeq 0.1$) by introducing a momentum cutoff, which takes into account the short-wavelength fluctuations. The dashed line in Fig. 3 was obtained by using the values of Table I in the Prange expression with momentum cutoff calculated in Ref. 18. Although the agreement is extended now up to $h \simeq 0.2$, this approach still fails to explain the decrease of $\Delta M(h)_\varepsilon$ observed at higher fields.

To analyze the results of Figs. 3 and 4 in terms of the microscopic approaches, we have chosen the expressions of $\Delta M(\varepsilon, h)$ proposed by Maki and Takayama[10] and by Klemm, Beasley and Luther[11] (MT-KBL theory) for the fluctuation-induced magnetization at finite fields and for the dirty limit, the one well adapted to the data in Fig. 3 and to most of the data in Fig. 4. The solid curves in these figures correspond to Eqs. (13) and (14) of Ref. 11. These last equations lead to results very similar to the pioneering MT calculations [Eqs. (16) and (17) of the second paper of Ref. 10]. As may be seen in Figs. 3 and 4, the agreement with the experimental data at low and moderate fields (up to $h \simeq 0.2$) is excellent. However, there is strong disagreement at high fields.

Another remarkable aspect [which may provide a hint for future extensions of the calculations of $\Delta M(\varepsilon, h)$ to the high field regime] of our experimental results is that, as illustrated in Figs. 3 and 4, for the field amplitudes where $\Delta M(h)_\varepsilon$ vanishes the GL coherence length, $\xi(h)_\varepsilon$, becomes of the order of ξ_0, the actual superconducting coherence length at $T=0$ K. The $\xi(h)_\varepsilon/\xi_0$ scale in Figs. 3 and 4 was obtained by using $\xi(h)_\varepsilon = \sqrt{2}\xi(0)h^{-1/2}$ and also $\xi(0) = 0.74\xi_0$, which is still a good approximation in the dirty limit.[1,2] When compared with our previous results at low fields but at high reduced temperatures,[12–14,18] this last finding already suggests that, in spite of the antisymmetric character of the magnetic field, the vanishing of $\Delta M(h)_\varepsilon$ may also be due to the quantum constraints to the shrinkage of the superconducting wave function:[13] Even above T_{c0}, the superconducting coherence length cannot be smaller than its minimum value, given by

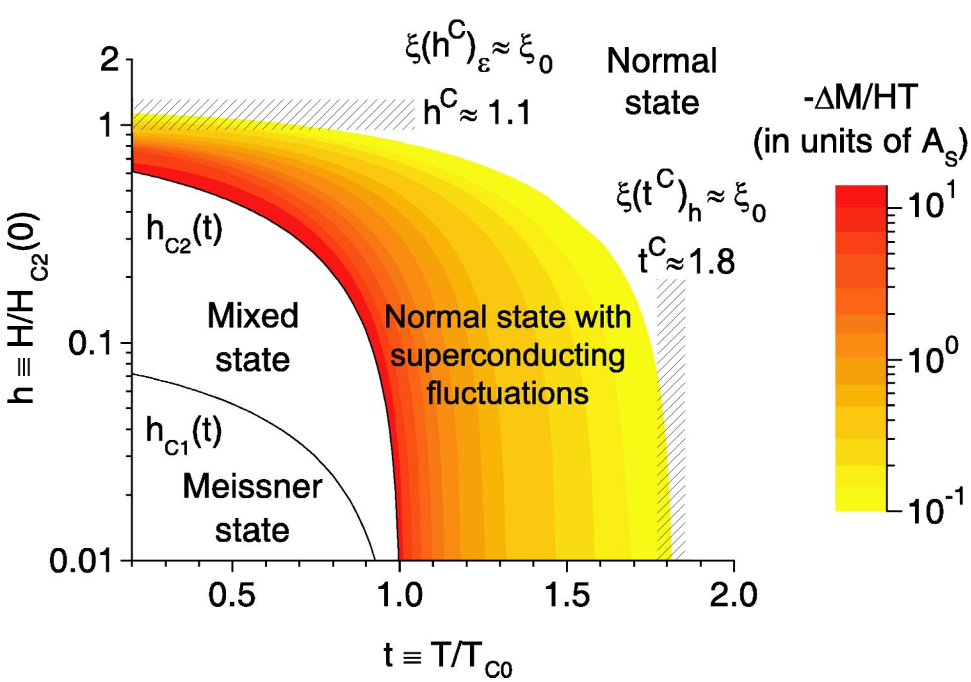

FIG. 5. (Color online) Measured $h-t$ phase diagram, including the SCF above $h_{C2}(t)$ for the $Pb_{0.55}In_{0.45}$ alloy. The color scale represents the fluctuation-induced magnetization (scaled by HT) in units of the Schmid amplitude, $A_S \equiv \pi\mu_0 k_B \xi(0)/6\phi_0^2$.

the uncertainty principle, the one at $T=0$ K (which, in fact, is the characteristic length of the Cooper pairs[1]). When the shrinkage of the superconducting wave function is due to a magnetic field, this condition may be written as $\xi(h)_\varepsilon \gtrsim \xi_0$, where ξ_0 for each alloy is related to the one of pure Pb, ξ_0^{Pb}, by[1] $\xi_0 \simeq (\xi_0^{\mathrm{Pb}}\ell)^{1/2}$, ℓ being the electronic mean free path. Such inequality directly leads to a critical reduced-field, h^C, given by $h^C=2(\xi(0)/\xi_0)^2$, above which all the SCF vanish. By using again $\xi(0)=0.74\xi_0$, we obtain $h^C\simeq 1.1$, in excellent agreement with the results of Figs. 2 and 4. As $\xi(0)/\xi_0$ is almost material-independent,[1] the relationship $\xi(h)_\varepsilon \gtrsim \xi_0$ predicts that the above value of h^C will be "universal," in strikingly good agreement with the experimental results at T_{c0} for all the samples studied in this work and summarized in Fig. 4.

Although our present paper is centered on the measurements of $\Delta M(\varepsilon,h)$ at high reduced fields, we have also measured in detail its reduced-temperature dependence in all the samples studied here. These last experiments confirm our previous findings obtained at low fields in other low- [12–14] and high-T_c (Refs. 13 and 15) superconductors: At low fields, $\Delta M(\varepsilon)_h$ vanishes when $\varepsilon \simeq 0.6$, which corresponds well to the condition $\xi(\varepsilon)_h \simeq \xi_0$. An example of our results on the dependence of $\Delta M(t,h)$ on t and h is presented in Fig. 5, with $t\equiv T/T_{c0}$. This example provides an overview of the *measured* SCF effects on the normal state magnetization above the superconducting transition in a low-T_c alloy.

In conclusion, the experimental results summarized here provide for the first time experimental information on the behavior of the superconducting fluctuation effects on the magnetization at high reduced fields above any superconducting transition. These results show an unexpected behavior of these fluctuation effects, which is not accounted for by the expressions for the fluctuation-induced magnetization existing in the literature. Moreover, they suggest the relevance in the SCF of the quantum effects associated with the shrinkage of the superconducting wave function at high fields. We note also that, although our experiments have been done in low-temperature superconductors, the overview for the SCF given in Fig. 5 could apply in principle to cuprate superconductors, which are extremely type II superconductors also unaffected by non-local effects. However, this last suggestion obviously needs further experimental verification, in particular in view of the recent observation of anomalous thermomagnetic effects well above $T_c(H)$ in some underdoped cuprates.[19] Other open questions are the SCF in presence of magnetic order (our present results suggest the robustness of the SCF against this antisymmetric perturbation) or the relationships between the vanishing of the SCF when $\xi(h)_\varepsilon \simeq \xi_0$ and the MT microscopic approach[10] if the zero-point fluctuations are taken into account.

We thank Professor J. B. Goodenough and Professor K. Maki for their helpful remarks. We acknowledge the financial support from the ESF "Vortex" Program, the CICYT, Spain, under Grant Nos. MAT2001-3272 and MAT2001-3053, the Xunta de Galicia under Grant Nos. PGIDT01PXI20609PR, and Unión Fenosa under Grant No. 220/0085-2002.

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[5] The fluctuation-induced magnetization is defined as $\Delta M(T,H) \equiv M(T,H)-M_B(T,H)$, where $M(T,H)$ is the as-measured magnetization and $M_B(T,H)$ the so-called background or bare magnetization, which may be approximated by extrapolating the magnetization measured either at $T \gg T_{c0}$ or $H \gg H_{C2}(0)$.

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