Nonlocal electrodynamics and low-temperature magnetization of clean high-κ superconductors

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We show that magnetic properties of clean superconductors with a large Ginzburg-Landau parameter κ at low temperatures are affected by the nonlocality of the microscopic current-field relation and can be described by modified London equations. We argue that for clean materials at low temperatures, the standard London formula for the reversible magnetization in intermediate fields, $M\sim\ln(H_{c2}/B)$, should contain the field $H_0\sim\phi_0/\rho^2$ instead of $H_{c2}\sim\phi_0/\xi^2(T)$, with ρ being the nonlocality range on the order of ξ_0, the zero-T coherence length. Since ρ depends weakly on T, the magnetization should exhibit an approximate scaling $M(T,B)=X(T)Y(B)$ as observed in Bi- and Tl-based compounds in a broad temperature domain well below T_c. Our expression for the magnetization reduces to the standard London result near T_c and at any temperature for the dirty case. Implications of our results for interpretation of neutron scattering data and for procedures of extracting the penetration depth are discussed. [S0163-1829(96)06941-X]

I. INTRODUCTION

Magnetic properties of type-II superconductors near the critical temperature T_c are described by the Ginzburg-Landau (GL) theory. Outside the GL domain, no simple macroscopic description is available, and the full microscopic formalism should be employed to properly describe the cores of vortices; in fact, this can be done only numerically; see, e.g., Ref. 1. Fortunately, in materials with a large GL parameter $\kappa=\lambda/\xi$ (the ratio of the penetration depth to the coherence length), the core contribution to the total energy is small, and a very simple London approach suffices for many applications.[2–4] Within this approach, the magnetic field of a single vortex is given (in Fourier space) by $h(\mathbf{k})=\phi_0/(1+\lambda^2k^2)$, and the free energy density of a flux-line lattice is

$$F=B^2\sum_{\mathbf{G}} h(\mathbf{G})/8\pi\phi_0. \tag{1}$$

Here, the sum runs over the reciprocal lattice $\mathbf{G}$, ϕ_0 is the flux quantum, and B is the (average over many vortices) magnetic induction; for the anisotropic case, h should be replaced with the field component along the common direction of vortices.

For a dense vortex lattice in intermediate fields $H_{c1}\ll B\ll H_{c2}$ (a domain which exists only in materials with $\kappa\gg1$), one can replace the sum (1) over nonzero $\mathbf{G}$'s with an integral:[2]

$$\widetilde{F}=F-\frac{B^2}{8\pi}=\frac{B^2}{8\pi}\int\frac{2\pi G\,dG}{4\pi^2B/\phi_0}\,\frac{1}{1+\lambda^2G^2}. \tag{2}$$

The integral is extended from $G_{\min}=2\pi/a$ with the intervortex spacing $a=(2\phi_0/\sqrt{3}B)^{1/2}$ for a triangular lattice. With the upper limit of integration at ∞, the intergal is logarithmically divergent, and one has to introduce a cutoff at $G_{\max}$ on the order of $2\pi/\xi'$ where ξ' is an *effective* core size. The divergence and the cutoff are inherent shortcomings of the London approach which breaks down at distances $r\sim\xi$. The cutoff cannot be ''improved'' within the London theory; for this one should turn to a theory which is able to handle the core structure properly, e.g., to GL theory near T_c. This has been done by Hao and Clem[5] (who developed a variational procedure to avoid nonlinearities of the GL equations) and by Koshelev[6] (who utilized the circular cell approximation).

To keep the model simple, we add to the London energy an estimate for the core contribution,

$$\frac{H_c^2}{8\pi}\pi\xi^2\eta_1\frac{B}{\phi_0}=M_0B\eta_1, \tag{3}$$

where

$$M_0=\frac{\phi_0}{32\pi^2\lambda^2}, \tag{4}$$

H_c is the thermodynamic critical field, and the constant $\eta_1\sim 1$ is introduced to account for the uncertainty in defining the core size. Incorporating Eqs. (3) and (4) in Eq. (2) one obtains

$$\widetilde{F}=M_0B\left(\ln\frac{a^2}{\xi'^2}+\eta_1\right)=M_0B\left(\ln\frac{\eta_2H_{c2}}{B}+\eta_1\right), \tag{5}$$

where the constant η_2 accommodates the uncertainty in the cutoff ξ'. This yields the familiar London magnetization $M=B/4\pi-\partial F/\partial B=-\partial\widetilde{F}/\partial B$:

$$M=-M_0\ln(\eta H_{c2}/B), \tag{6}$$

where η absorbs all uncertainties mentioned. Experimental data show that close to T_c, $\eta\approx 1.2-1.5$;[7] the value 1.4 is obtained with the Hao-Clem method.[8]

It is worth stressing that the London model *per se* is only responsible for the prelogarithmic factor in Eq. (6) and for the linear dependence of M on $\ln B$. The term ηH_{c2} under the logarithm is, in fact, imposed by the cutoff (H_{c2}) and by the necessity to match the data (η). The appearance of H_{c2} in Eq. (6) is hardly accidental, since ξ is the only relevant length in the problem other than λ. Again, near T_c, this eclectic approach can be improved by employing the GL theory (the Hao-Clem procedure is an example). However, it is not obvious *a priori* that the same scheme will work at low temperatures. Still, there is a widespread opinion that the London equations along with their consequences and, in particular, Eq. (5) for the energy and Eq. (6) for the magnetization hold and are accurate at low T's as well. In the past, it was hard to verify whether or not this is true since the pinning is usually too strong at low T's, preventing reliable data on the *reversible* $M(T,B)$. With the arrival of low-pinning, high-κ, and high-T_c materials such as Bi- or Tl-based compounds, such data became available. By and large, they confirm the nearly linear dependence $M(\ln B)$. However, close examination shows that application of Eq. (6) to the low-temperature data is problematic (as discussed below).

The question of the validity of Eq. (6) at low T's became relevant in particular since it offers a simple method to extract the upper critical field from the magnetization data at temperatures down to reduced temperatures $t=T/T_c\approx 0.4$ (30–40 K, for Bi- and Tl-based compounds) where H_{c2} is too large for a direct measurement. While at high temperatures, Eq. (6) (corrected for vortex fluctuations) produced well-behaved $H_{c2}(T)$ linear in (T_c-T) for Tl-2223 and Hg-1201,[9] the method failed when applied to low T's. Extensive magnetization data on Bi-2212 by Cho *et al.*[10] analyzed with the help of either Eq. (6) or the Hao-Clem procedure have generated a puzzling result: H_{c2} so obtained was nearly T independent between 35 and 70 K, whereas the standard Helfand-Werthamer (HW) estimate predicts a change by a factor of 3.[11] Similar observations were reported by Triscone *et al.*[12] and Waldmann *et al.*[13] This contradiction motivated us to reconsider in detail the London approach at low temperatures.

In the following, we rederive the London equations from the microscopic theory, explicitly incorporating the basic nonlocality of the current-field relation in superconductors. We show that instead of $H_{c2}(T)\sim\phi_0/\xi^2$, the nonlocality sets a different scaling field $H_0\sim\phi_0/\rho^2$; here ρ is the nonlocality length that slowly decreases with temperature unlike $\xi(T)$. Having in mind high-T_c superconductors, we focus on layered strongly anisotropic materials which can be modeled with a cylindrical Fermi surface. We construct an expression for the magnetization and compare it with available data to show that our approach is in good agreement with the data for clean extremely anisotropic layered materials and is free of the contradiction described above.

The nonlocality of the Bardeen-Cooper-Schrieffer (BCS) electrodynamics has been studied extensively. Even before BCS, the nonlocal relation between the current density **j** and the vector potential **A** was suggested by Pippard[14,3] to explain data on the penetration depth; for the recent applications of Pippard's electrodynamics to the vortex structure see Ref. 15. The physics of nonlocality originates in a finite size, ξ_0 at $T=0$, of Cooper pairs: **j** at a given point is determined by the vector potential **A** within a domain $\sim\xi_0$ around this point. Instead of local relations between **j** and **A** of the GL or London theories, BCS provide an integral equation with a kernel $\hat{Q}$ extending to distances $\sim\xi_0$. In Fourier space this relation is of the form $\mathbf{j}(\mathbf{k})\propto\hat{Q}(\mathbf{k})\mathbf{A}(\mathbf{k})$ with the Fourier transform of $\hat{Q}$ explicitly depending on **k**. In the GL domain where $\xi(T)\gg\xi_0$, or far from the vortex cores in materials with the penetration depth $\lambda\gg\xi$, the nonlocal corrections vanish.

Early theoretical studies of nonlocal effects focused mainly on the vortex core region in materials with $\kappa\sim 1$. The treatment is difficult due to the spatial dependence of the order parameter $|\Delta|$; see, e.g., Refs. 1, 16, and 17. In this paper we argue that for *high-*κ materials at low T's, there exists a region around the vortex core in which the BCS *nonlocality* can be described by relatively simple corrections to London equations, while the spatial dependence of $|\Delta|$ due to supercurrents can be disregarded. We derive expressions for the free energy and indicate measurable quantities which can be affected by nonlocality: the field dependence of magnetization M, neutron scattering form factors, and the field variance in the mixed state [measured in muon spin resonance (μSR) and NMR experiments], to name a few. We show that nonlocality may well be responsible for a puzzling scaling $M(T,B)\approx X(T)Y(B)$ seen in Bi- and Tl-based high-T_c compounds[10,12,13] in broad temperature domains away of T_c.

The paper is organized as follows: In the next section we outline how the London equations are obtained from the microscopic theory. This approach has an added advantage of allowing us to obtain corrections to the London theory due to the nonlocality. We calculate the macroscopic free energy and discuss consequences of the correction terms. We then turn to the case of strongly anisotropic layered materials

modeled with a cylindrical Fermi surface. In Sec. III we discuss magnetization data on a few layered high-T_c superconductors for which the reversible data are available well below T_c. A short discussion follows.

II. MICROSCOPIC THEORY AND LONDON APPROACH

The main assumption in the derivation of London equations from the microscopic theory is that small currents do not suppress $|\Delta|$. Only the phase θ changes in space, a feature that preserves the flux quantization in the London approach. We begin with the quasiclassical Eilenberger version of the BCS theory:[18]

$$\mathbf{v}\cdot\mathbf{\Pi}f=2\Delta g/\hbar-2\omega f+(g\langle f\rangle-f\langle g\rangle)/\tau, \tag{7}$$

$$g^2=1-ff^+, \tag{8}$$

$$\frac{\Delta}{2\pi T}\ln\frac{T_c}{T}=\sum_{\omega>0}\left(\frac{\Delta}{\hbar\omega}-\langle f\rangle\right), \tag{9}$$

$$\mathbf{j}=-4\pi|e|N(0)T\ \mathrm{Im}\sum_{\omega>0}\langle\mathbf{v}g\rangle. \tag{10}$$

Here $\mathbf{v}$ is the Fermi velocity, $\mathbf{\Pi}=\nabla+2\pi i\mathbf{A}/\phi_0$, $\Delta(\mathbf{r})$ is the gap function (the order parameter), $f(\mathbf{r},\mathbf{v},\omega), f^+=f^*(\mathbf{r},-\mathbf{v},\omega)$, and g are Eilenberger Green's functions, $N(0)$ is the total density of states at the Fermi level per one spin, $\hbar\omega=\pi T(2n+1)$ with an integer n, $\langle\ldots\rangle$ stands for the average over the Fermi surface, and τ is the scattering time due to nonmagnetic impurities. The equation for f^+ is obtained from Eq. (7) by taking complex conjugate and replacing $\mathbf{v}\to-\mathbf{v}$. In the absence of currents, we have

$$f_0=\Delta_0/\beta_0,\quad g_0=\hbar\omega/\beta_0,\quad \beta_0^2=\Delta_0^2+\hbar^2\omega^2, \tag{11}$$

and Eq. (9) gives the uniform BCS gap $\Delta_0(T)$.

Let now a weak supercurrent flow in the system. We look for solutions of Eqs. (7)–(9) in the form:[19]

$$\Delta=\Delta_0 e^{i\theta},\quad f=(f_0+f_1)e^{i\theta},$$

$$f^+=(f_0+f_1^+)e^{-i\theta},\quad g=g_0+g_1, \tag{12}$$

where θ depends only on $\mathbf{r}$, and f_1, f_1^+, g_1 are small corrections. Substituting Eqs. (12) into Eq. (7) we obtain

$$\mathbf{v}\cdot(\nabla f_1+i\mathbf{P}f_1)=2\Delta' g_1/\hbar-2\omega' f_1-if_0\mathbf{v}\cdot\mathbf{P}. \tag{13}$$

Here $\Delta'=\Delta_0(1+\hbar/2\tau\beta_0)$ and $\omega'=\omega_0(1+\hbar/2\tau\beta_0)$; in the first approximation $\langle f_1\rangle=\langle g_1\rangle=0$, an assumption justified by the result. The ''supermomentum'' $\mathbf{P}=\nabla\theta+2\pi\mathbf{A}/\phi_0$ is assumed to be small, $P\ll\xi^{-1}$, so that $\mathbf{P}f_1$ can be neglected relative to ∇f_1. This implies that the current density is well under the depairing limit $j_d\approx c\phi_0/16\pi^2\xi\lambda^2$.

Equation (13) with those for f_1^+ and g_1 (which are not written down here) suffices to determine all corrections in Fourier space:

$$g_1(\mathbf{k})=\frac{i\hbar\Delta'^2}{2\beta'}\frac{\mathbf{v}\cdot\mathbf{P}(\mathbf{k})}{\beta'^2+\hbar^2(\mathbf{v}\cdot\mathbf{k})^2/4}, \tag{14}$$

where $\beta'=\beta_0+\hbar/2\tau$. Substituting g_1 in Eq. (10), we obtain the BCS current-field relation

$$j_i(\mathbf{k})=-[4\pi e^2N(0)T\Delta_0^2/c]a_l(\mathbf{k})\times\sum_{\omega>0}\frac{\beta'}{\beta_0^2}\left\langle\frac{v_iv_l}{\beta'^2+\hbar^2(\mathbf{v}\cdot\mathbf{k})^2/4}\right\rangle, \tag{15}$$

where $\mathbf{a}=\phi_0\mathbf{P}/2\pi=\mathbf{A}+\phi_0\nabla\theta/2\pi$ and the summation is implied over repeated subscripts. Note that Eq. (15) is explicitly gauge invariant and holds for any anisotropic Fermi surface. Finally, we substitute the solutions in Eq. (9) to obtain $\langle f_1(\mathbf{r},\mathbf{v})\rangle=0$, which is verified easily in Fourier space making use of $k_ij_i(\mathbf{k})=0$.

Since in the above derivation $|\Delta|$ is not altered by currents, Eq. (15) is a good starting point for observing how the London equations emerge from the microscopic theory. Clearly, for $\mathbf{k}=0$, Eq. (15) yields $\mathbf{j}\propto\mathbf{a}$ in both Fourier and real spaces; i.e., we have a *local* London relation between the current and the vector potential. Keeping the first correction in small $\mathbf{k}$'s we obtain expanding the denominator in powers of $(\mathbf{v}\cdot\mathbf{k})^2$:

$$\frac{4\pi}{c}j_i=-\frac{1}{\lambda^2}(m_{ij}^{-1}-\lambda^2n_{ijlm}k_lk_m)a_j. \tag{16}$$

Here

$$\frac{1}{\lambda^2}=\frac{16\pi^2e^2N(0)T\Delta_0^2\langle v^2\rangle}{3c^2}\sum_{\omega>0}\frac{1}{\beta_0^2\beta'},\quad m_{ij}^{-1}=\frac{3\langle v_iv_j\rangle}{\langle v^2\rangle},$$

$$n_{ijlm}=\frac{4\pi^2e^2\hbar^2N(0)T\Delta_0^2}{c^2}\langle v_iv_jv_lv_m\rangle\sum_{\omega>0}\frac{1}{\beta_0^2\beta'^3}. \tag{17}$$

We defined λ so as to have the correct isotropic limit and m_{ij} to have a convenient property $\det m_{ij}=1$. The tensor $\hat{n}$, symmetric with respect to all indices, can also be written as

$$\lambda^2n_{ijlm}=\frac{3\hbar^2\langle v_iv_jv_lv_m\rangle\Sigma\beta_0^{-2}(\beta')^{-3}}{4\langle v^2\rangle\quad\Sigma\beta_0^{-2}(\beta')^{-1}}. \tag{18}$$

This quantity is of the order $\hbar^2v^2/\Delta^2(0)\sim\xi_0^2$ in the clean case and of the order $v^2\tau^2$ for dirty materials. The first term on the right-hand side of Eq. (16) corresponds to the standard anisotropic London equation,[20] whereas the last term is due to nonlocality. We will discuss the general anisotropic case elsewhere; here we just note that being dependent on the shape of the Fermi surface, the fourth-rank tensor $\hat{n}$ couples supercurrents with the crystal lattice even in cubic materials which, within the local London theory, should behave isotropically.

Let us turn now to the case of a cylindrical Fermi surface, the situation reminiscent of the high-T_c superconductors. The Fermi velocity v_z along the cylinder axis $\hat{c}$ is zero, and for a circular cylinder $\langle\cdots\rangle=(2\pi)^{-1}\int_0^{2\pi}d\varphi\cdots$, where φ is the angle between $\mathbf{v}$ and $\mathbf{k}$. All indices in Eq. (16) now take only x,y values which we denote with Greek letters.

For a system isotropic in the layer planes, $n_{\alpha\beta\mu\nu}=n_0(\delta_{\alpha\beta}\delta_{\mu\nu}+\delta_{\alpha\mu}\delta_{\beta\nu}+\delta_{\alpha\nu}\delta_{\beta\mu})$. To keep the property $\det m_{\alpha\beta}=1$ we should replace the factor of 3 in definitions (17) of λ and $\hat{m}$ by 2. Then we obtain

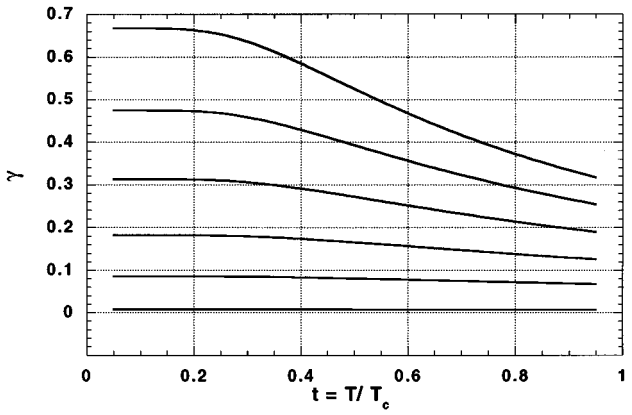

FIG. 1. Parameter γ vs reduced temperature which determines the temperature dependence of the nonlocality radius ρ according to Eqs. (19) and (20) for the scattering parameter $\xi_2/\ell=0$ (the upper curve), 0.2, 0.5, 1, 2, and 10 (the bottom curve).

$$n_0\lambda^2=\frac{\hbar^2v^2}{16\Delta_0^2(0)}\gamma(T)\equiv\rho^2(T), \tag{19}$$

where the distance ρ, on the order of the zero-T coherence length, will be called hereafter the nonlocality radius. The quantities λ, ξ, and ρ, appearing here and in the following analysis of the data for layered compounds, are commonly written with subscripts ab, which we omit hereafter for brevity. The temperature dependence of ρ is given by

$$\gamma(T)=\Delta_0^2(0)\sum\ \beta_0^{-2}(\beta')^{-3}\Big/\ \sum\ \beta_0^{-2}(\beta')^{-1}, \tag{20}$$

which can be evaluated numerically. Figure 1 shows γ vs $t=T/T_c$ for several values of the impurity parameter $\hbar/2\tau\Delta_0(0)=\xi_2/\ell$ where $\xi_2=\hbar v/2\Delta_0(0)$ is the two-dimensional zero-T coherence length, and ℓ is the mean free path. We see that the nonlocality radius is reduced by temperature and by impurities. In the clean limit ($\xi_2/\ell\ll1$) we have $\gamma(0)=2/3$ and $\gamma(T_c)\approx0.30$; see Appendix A. The nonlocality radius, being close to a constant below $t\approx0.2$, reaches at T_c about 0.7 of its value at $T=0$. The scattering suppresses ρ at all T's; also, the relative change of ρ with T is reduced. In the dirty limit $\beta'\to\hbar/2\tau$, and $\gamma\to\ell^2/\xi_2^2$; i.e., γ becomes T independent; the nonlocality range $\rho\to\ell/2$, which means that practically all nonlocal effects vanish.

We now apply the equations obtained to a vortex along the z axis. Equation (16) gives $4\pi j_\alpha/c=-(1-\rho^2k^2)a_\alpha/\lambda^2$. We isolate **a** and use the flux quantization condition $\mathrm{curl}\mathbf{a}=\mathbf{h}-\phi_0\hat{z}\delta(\mathbf{r})$ where **h** is the local magnetic field. Neglecting the terms $\sim\rho^4k^4$, we arrive at

$$h(k)=\frac{\phi_0}{1+\lambda^2k^2+\rho^2\lambda^2k^4}. \tag{21}$$

The implications of inverting this truncated Fourier transform into real space are discussed in Appendix B.

Let us turn now to vortex lattices in which the field is periodic: $h(\mathbf{r})=(B/\phi_0)\Sigma h(\mathbf{G})\exp(i\mathbf{G}\cdot\mathbf{r})$ where the sum runs over the reciprocal lattice vectors **G**. The energy density is given in Eq. (1) in which $h(\mathbf{G})$ of Eq. (21) should be substituted. As above, in fields $H_{c1}\ll B\ll H_{c2}$, we replace the sum over nonzero **G**'s with an integral from $G_{\min}=(2\pi^2\sqrt{3}B/\phi_0)^{1/2}$ (for a hexagonal lattice). Unlike the situation in the standard London model, the integral is convergent. However, to ensure that the G values corresponding to the vortex core are excluded, we set the upper limit of integration at $G_{\max}=2\pi/\xi'$. We then obtain neglecting terms $\sim O(\kappa^{-2})$:

$$\widetilde{F}=M_0B\left(\ln\frac{H_0/B+1}{H_0/\eta_2H_{c2}+1}+\eta_1\right), \tag{22}$$

$$H_0=\phi_0/2\pi^2\sqrt{3}\rho^2, \tag{23}$$

and M_0 is given in Eq. (4). Here again, the constants η_1 and η_2 accommodate uncertainties in the core energy and in the cutoff; in other words, this result has the same shortcomings as the standard London equation (5).

Still, there are important differences between Eqs. (5) and (22). In addition to H_{c2}, a characteristic field H_0 related to the nonlocality range enters the free energy. Equation (22) contains three characteristic lengths: $\lambda(T)$, $\xi(T)$, and the nonlocality range $\rho(T)$ (the Cooper pair size). This reflects the situation in the microscopic description where all three distances are implicitly present. Unlike H_{c2}, the field H_0 does not go to zero as $T\to T_c$; in fact, it increases, with T being inversely proportional to $\gamma(T)$; see Fig. 1. Therefore, near T_c, where $B\ll H_{c2}\ll H_0$, Eq. (22) reduces to the standard London energy (5). In the dirty limit for which $H_0\sim\phi_0/\ell^2\gg\phi_0/\xi_0\ell\sim H_{c2}(0)$, we again obtain the standard London result but now at all temperatures. Thus, the nonlocal corrections are noticeable at low T's for clean materials.

The magnetization $M=-\partial\widetilde{F}/\partial B$ now reads

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

$$\zeta=\eta_1-\ln\left(\frac{H_0}{\eta_2H_{c2}}+1\right). \tag{24}$$

The quantity ζ slowly decreases with temperature due to the second term. Note that the leading term in $M(B)$ of Eq. (24) is $\ln(H_0/B)$, corrected with terms on the order B/H_0, which are usually small.

For clean systems away from T_c the right-hand side (r.h.s.) of Eq. (24) is only weakly T dependent; in other words, we have an *approximate* property $M(T,B)\approx X(T)Y(B)$ where $X=M_0$ and the function Y, the rhs of Eq. (24), is nearly T independent. Experimental evidence for such a scaling behavior is given below.

We observe that the field B enters M in the combination B/H_0 instead of the standard London or GL ratio B/H_{c2} (unless the material is dirty or T is close to T_c). This makes the procedure of extracting H_{c2} from the magnetization data at low T's more involved and difficult.

In deriving Eqs. (24) which should hold at all T's, we have ignored thermal fluctuations of vortices (phase fluctua-

tions). It is well established, however, that the latter significantly change M at high temperatures. Hence, we should add to M the fluctuation term

$$\delta M = \frac{k_B T}{\phi_0 s} \ln \frac{C \kappa^2 k_B T}{\phi_0 s B}, \tag{25}$$

where s is the interlayer spacing and the constant $C \approx 10.2$.[21,22] The ratio $k_B T/\phi_0 s M_0$ which determines the relative weight of the fluctuation term in M is about $0.5t$ at low T's. In other words, the fluctuation contribution cannot be neglected even at the lowest temperatures (30–40 K) available for measurement of reversible magnetization.[22] Note that, in principle, the term (25) violates the scaling $M(T,B) \approx X(T)Y(B)$. However, the comparison with data (the next section) shows that this violation is quantitatively significant only near T_c.

In the local London theory, $M(B)$ is linear in $\ln B$, a feature often used to extract $\lambda(T)$ from the magnetization data with the help of Eq. (6). The nonlocality modifies this to

$$S = \frac{dM}{d \ln B} = \frac{M_0}{(1 + B/H_0)^2} - \frac{k_B T}{\phi_0 s}. \tag{26}$$

This result does not depend on the uncertain η's. Near T_c and for dirty materials $B \ll H_{c2} \ll H_0$, and we have the standard London result. Again, the deviations from $S = M_0$ are expected at low T's for clean materials.

We now consider polycrystals of strongly anisotropic materials, for which the component of **M** along the layers can be disregarded. Clearly, for a grain with the $\hat{c}$ axis at an angle θ with the applied field $\mathbf{B} = B\hat{z}$, $M_z = M(B\cos\theta)\cos\theta$. We then have for a sample of randomly oriented grains $M_z = \int_0^{\pi/2} M(B\cos\theta)\cos\theta \sin\theta d\theta$.[23] With the help of Eqs. (24) and (25) we obtain

$$M = -\frac{M_0}{2}\left(\ln \frac{H_0 + B}{B} + \frac{H_0^2}{B^2} \ln \frac{H_0 + B}{H_0} - \frac{H_0}{B} + \zeta \right) + \frac{k_B T}{2\phi_0 s} \ln \frac{C' \kappa^2 k_B T}{\phi_0 s B}, \tag{27}$$

where $C' = C\sqrt{e} \approx 16.8$. In the dirty case or near T_c for any ℓ, this yields

$$M = -\frac{M_0}{2} \ln \frac{\eta \sqrt{e} H_{c2}}{B} + \frac{k_B T}{2\phi_0 s} \ln \frac{C' \kappa^2 k_B T}{\phi_0 s B}. \tag{28}$$

III. COMPARISON WITH DATA

In comparing the data on $M(B)$ at a given T with our results, Eq. (24) and Eq. (25) for crystals or Eq. (27) for random powders, we have to consider M_0, H_0, and ζ as fitting parameters. If a data set is sufficiently precise and extensive [say, $M(B)$ is known for a few T's], one can try to fit the above formulas to the data and see whether or not the temperature behaviors of so obtained $M_0(T)$, $H_0(T)$, and $\zeta(T)$ correspond to those theoretically expected. The requirements for the data quality are quite rigid because the numerical routines for ''best fits'' should deal with very shallow minima (H_0 enters M under the logarithm sign).

We start with the magnetization data for a single crystal

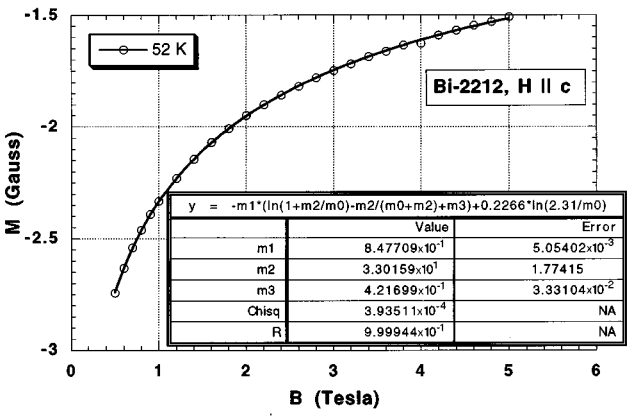

FIG. 2. A typical example of the magnetization data (for a Bi-2212 crystal in fields along $\hat{c}$ at 52 K). The solid line is calculated using Eqs. (24) and (25) with the best-fit parameters $M_0 \equiv m1$ (G), $H_0 \equiv m2$ (T), and $\zeta \equiv m3$. In the fitting function, $y \equiv M$ and $m_0 \equiv B$.

of Bi-2212 in fields parallel to the $\hat{c}$ axis ($T_c \approx 85$ K) for which the standard analysis with the help of either Eq. (6) or the Hao-Clem procedure leads to a conclusion that H_{c2} is T independent between 35 and 70 K.[10] Figure 2 shows a typical example in which we fit the data for $M(B)$ at $T = 52$ K to Eq. (24), with the contribution of fluctuations Eq. (25), included. In the latter we set $\kappa = 100$ and $s = c/2 = 15.3$ Å (the output fitting parameters are not sensitive to a precise value of κ since only $\ln\kappa$ enters δM; they are sensitive to s). We treat M_0, H_0, and ζ as fitting parameters (these are denoted in the fitting function seen in the inset of Fig. 2 as $m1, m2$, and $m3$, respectively). The quality of the fit and the accuracies with which the fitting parameters are extracted are also shown: In this case they are about 1%, 5%, and 8% for M_0, H_0, and ζ, respectively. For all data sets available, the accuracy deteriorates at the low-temperature edge of the T domains studied (presumably due to pinning) as well as at the high-T side (M becomes too small and noisy).

The whole data set for Bi-2212 and the theoretical curves [the solid lines obtained with the best-fit parameters $M_0(T), H_0(T)$, and $\zeta(T)$] are plotted in Fig. 3(a). Figure 3(b) shows that indeed the data can be *approximately* scaled on one curve, the feature that is related to the weak temperature dependence of H_0 as discussed above. We note that at first sight the fluctuation term (25) should have destroyed the model ability to describe this scaling; *quantitatively*, however, this does not happen since the data are well represented by our formulas which include the term (25).

Likewise, we have analyzed the data for a polycrystalline sample of Tl-2212 ($T_c \approx 103$ K; for the sample description see. Ref. 24) We consider the sample as being randomly oriented with $\kappa = 100$ and $s = 14.7$ Å and use Eq. (27) for the fit. The results are shown in Figs. 4(a) and 4(b).

The temperature dependences of H_0 for the two materials are shown in Fig. 5(a). Recall that the nonlocality radius decreases with T and therefore H_0 should increase with T. Taking the clean limit $\gamma(T)$ given in Fig. 1, we can check whether or not the relation $H_0(T) \propto 1/\gamma(T)$ holds as expected. Moreover, we can use $H_0(T)$ and the known clean

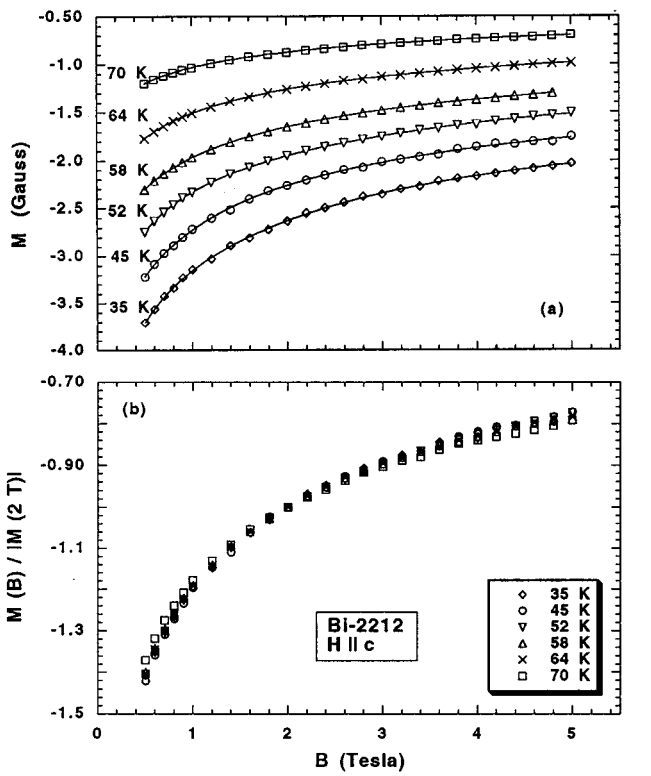

FIG. 3. (a) Magnetization data for a Bi-2212 crystal in fields along the $\hat{c}$ axis at the temperatures indicated. Solid lines are calculated using Eqs. (24) and (25) with fitting parameters M_0, H_0, and ζ shown in Fig. 5. (b) Approximate scaling of the data shown in (a).

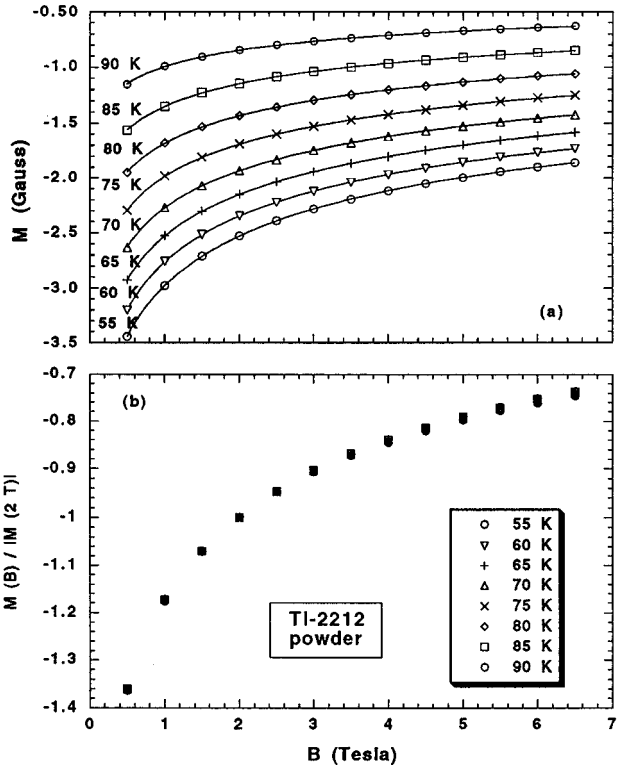

FIG. 4. (a) Magnetization data for the random polycrystal of Tl-2212 at temperatures indicated. Solid lines are calculated using Eq. (27) with fitting parameters M_0, H_0, and ζ shown in Fig. 5. (b) Approximate scaling of the data shown in (a).

limit $\gamma(T)$ in order to evaluate the two-dimensional (2D) zero-temperature coherence length ξ_2:

$$\xi_2^2=\frac{2\phi_0}{\pi^2\sqrt{3}H_0(T)\gamma(T)}; \tag{29}$$

Eqs. (19) and (23) were used. This quantity should come out the same for all temperatures. This, in fact, is the case: Figure 6 shows the product $H_0(T)\gamma(T)$ which can be considered as T independent over broad temperature domains for both materials given about 10% accuracy with which H_0 is extracted from fitting the data. We consider this constancy of ξ_2 as a major consistency test of our model.

Averaging the product γH_0, we obtain $\xi_2\approx 41$ Å for Bi-2212 and 44 Å for Tl-2212. Note that $\xi(0)$, which determines the upper critical field $H_{c2}(0)=\phi_0/2\pi\xi^2(0)$, differs from $\xi_2\equiv\hbar v/2\Delta_0(0)$. One can show that within a 2D version of the HW theory developed by Bulaevskii,[25] $\xi(0)=\hbar v/2.67\Delta_0(0)$, so that $\xi(0)\approx 0.75\xi_2$. This yields $\xi(0)\approx 31$ Å for Bi-2212 and 33 Å for Tl-2212. Perhaps, the best way to look at our numbers is to evaluate first the zero-T upper critical field for these two materials [$H_{c2}(0)\approx 35$ T and 30 T, respectively]; then, again with the help of 2D theory, calculate $H'_{c2}\equiv|dH_{c2}/dT|_{t=1}=H_{c2}(0)/0.59T_c$ (the factor 0.59 comes in 2D instead of 0.69, the 3D result of HW) and compare the results with the literature slopes. We obtain 0.7 T/K for Bi-2212 and 0.5 T/K for Tl-2212 to compare, e.g., with 0.9 and 1.0 T/K for these materials reported in Ref. 12. One should mention that literature values for H'_{c2} obtained with different techniques may be as high as 2.7 T/K for Bi-2212; see Ref. 12 and references therein.

Figure 5(b) shows the best-fit parameter $M_0=\phi_0/32\pi^2\lambda^2(T)$, which behaves qualitatively as expected. We will not analyze the T dependence of λ; it would take us too far from the main subject of this paper. We just mention that the magnitude of λ extracted from M_0 is quite reasonable: E.g., for $t\approx 0.5$ we obtain $\lambda_{ab}\approx 2600$ Å for Bi-2212 and ≈ 2000 Å for Tl-2212. Triscone *et al.* estimate the zero-T penetration depth for these two materials as 2770 Å and 1970 Å , respectively.[12]

The third fitting parameter $\zeta(t)$ is shown in Fig. 5(c); it decreases with T as Eq. (24) prescribes. We do not attempt to extract $H_{c2}(T)$ from the so-obtained $\zeta(T)$ since this would have required guesses for $\eta_{1,2}$.

Summarizing this part of the data analysis we can say that the nonlocal version of the London theory represents the data for the Bi-2212 and Tl-2212 compounds reasonably well, although the values of $\xi(0)$ extracted are larger than cited in the literature. Whereas the accuracy with which M_0, H_0, and ζ can be extracted from the magnetization data is not high, it is worth noting that our approach allows one to estimate the zero-T coherence length using the field dependence of magnetization at any T within a broad domain under T_c.

Our attempt to analyze in a similar manner the magnetization data for an aligned powder sample of Hg-1223 and for randomly oriented samples of Bi-2223 (with 25% of Bi re-

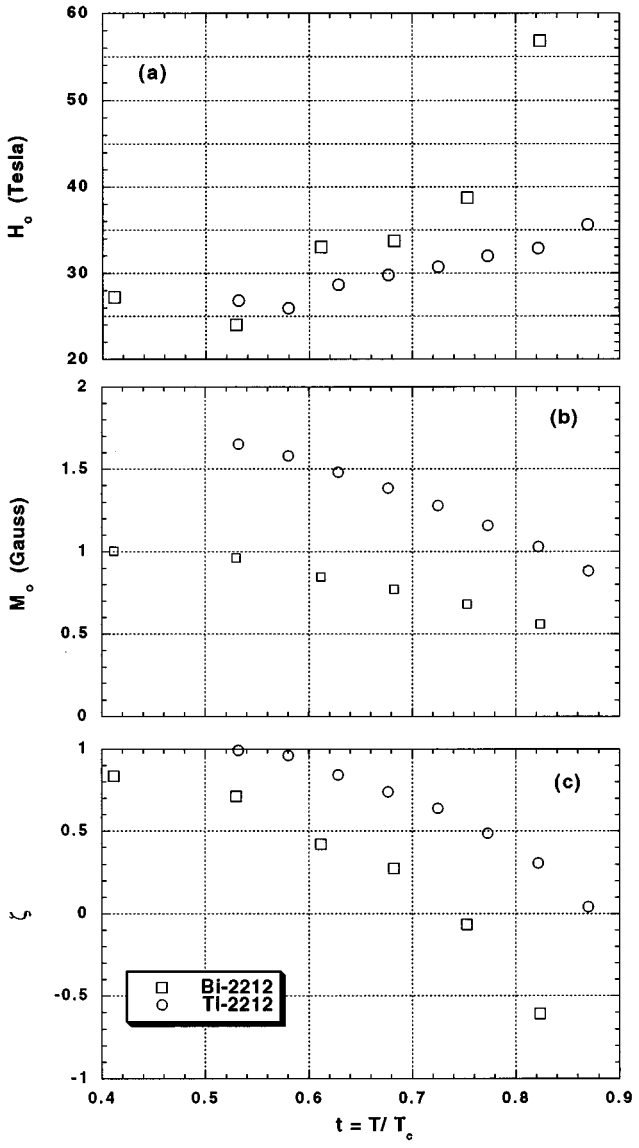

FIG. 5. Parameters H_0, M_0, and ζ used to fit the data $M(B,T)$ for Bi-2212 and for Tl-2212 [the data are shown in Figs. 3(a) and 4(a), respectively], vs reduced temperature t.

placed by Pb) and Hg-1201 did not produce ''well-behaved'' $H_0(T)$. The failure may have been caused by variety of factors: misalignment, residual pinning, a hard to control procedure of subtracting the contribution of the normal phase from the measured M; for whatever reason noisy data sometimes take the numerical routine far from ''reasonable'' values of fitting parameters. However, the most likely reason for the failure of Eqs. (24) or (27) which incorporate nonlocality might simply be that the nonlocality is irrelevant in these samples. Although Eqs. (24) and (27) apply formally for any mean free path ℓ, for short ℓ ($H_0 \gg H_{c2}$), H_0 practically drops off all expressions for M. In this situation, the standard London equation complemented with the fluctuation term (25) should provide a proper description of the data.

Figures 7(a) and 7(b) show the results of fitting magnetization data for an aligned sample of Hg-1223 ($T_c \approx 133$ K; for sample characteristics see Ref. 26) to Eqs. (6) and (25) and for powder samples of Bi-2223 and Hg-1201 (the

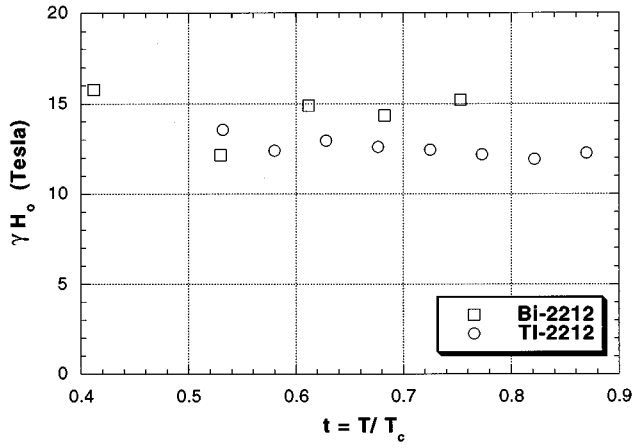

FIG. 6. The product of $H_0(T)$ and $\gamma(T)$ vs reduced temperature $t = T/T_c$ for Bi-2212 and Tl-2212. For our model to be consistent, this product should be T independent.

samples are described in Refs. 23 and 27) to Eq. (28). The two quantities M_0 and $\eta' H_{c2}$ ($\eta' = \eta$ for the aligned sample and $\eta' = \sqrt{e}\,\eta$ for powders) were used as fitting parameters. Figure 7(a) gives $\eta' H_{c2}(t)$ for these three materials. While plotting $\eta' H_{c2}$ and M_0 against the temperature we have observed that extrapolations of these quantities to zero in all cases yield T_c's a few K in excess of reported critical temperatures. We associate this with the effect described by the theory of vortex fluctuations: the ''mean-field'' T_{c0} exceeds the temperature where the resistivity drops. Also, we observed that for all materials, the parameter $\eta' H_{c2}$ shows a better pronounced linear behavior at high temperatures than M_0; we therefore defined T_c's by extrapolating $\eta' H_{c2}$ to zero and obtained 140, 112, and 100 K for Hg-1223, Bi-2223, and Hg-1201, respectively. The T-dependent λ^{-2}, obtained as the second parameter from the same fits, is shown in Fig. 7(b). We will not make quantitative conclusions from these; we note, however, qualitatively correct temperature behavior of the two parameters for all three materials. Finally, we note that an attempt to scale the data, in the manner shown in Figs. 3(b) and 4(b) for Bi- and Tl-2212, for the three latter materials fails since the T dependence of H_{c2} is much stronger than that of H_0.

IV. DISCUSSION

The method we suggest for analyzing the data on reversible magnetization in clean systems is, in fact, quite simple; it is based on explicit analytic expressions (24), (25), for crystals or (27) for random polycrystals. The three fitting parameters we have used reflect actual physical complexity: Magnetization does depend on the penetration depth $\lambda(T)$ (via M_0), zero-T coherence length $\xi(0)$ (or ξ_2 in 2D, via H_0), and on uncertain constants of the order unity $\eta_{1,2}$ [via $\zeta(T)$] inescapable within any version of a London approach which avoids complexity of the core structure. Having all weaknesses of a London technique, the method is perhaps the only one practically available for temperatures out of the narrow GL domain. As we have pointed out, the variational procedure of Hao and Clem, being free of London shortcomings near T_c, fails when applied to clean systems at low

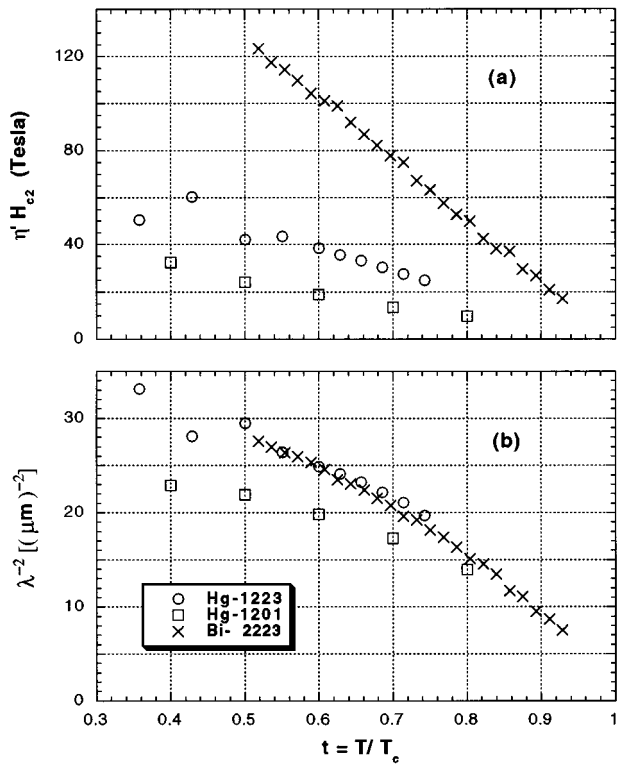

FIG. 7. (a) Parameter $\eta' H_{c2}(t)$ ($\eta' = \eta$ for aligned sample of Hg-1223 and $\eta' = \sqrt{e}\,\eta$ for randomly oriented polycrystalline samples of Bi-2223 and Hg-1201) vs reduced temperature obtained by fitting the magnetization data to Eqs. (6) and (25) for Hg-1223 and to Eq. (28) for Bi-2223 and Hg-1201. The fits are not sensitive to the value of κ; we use $\kappa = 100$ in the fluctuation term for all materials. As explained in the text, T_c's are chosen by extrapolating $\eta' H_{c2}(T)$ to zero. (b) Parameter $\lambda^{-2}(t) = 32\pi^2 M_0/\phi_0$ obtained in the same fitting procedure.

temperatures. Also, our method preserves the approximately linear in ln$B$ dependence of M of the standard London approach; corrections to the logarithmic term are small. The major difference for clean systems at low T's is in the value and physical meaning of the scaling field H_0 which takes the role of H_{c2} in the standard approach: The magnetization depends on B/H_0 instead of the standard B/H_{c2}.

Simple examination of expressions (24) and (27) for M shows that the upper critical field cannot be extracted from the data on $M(B)$ with the help of our method (unless T is close to T_c or the sample is dirty for which ηH_{c2} can be extracted). The method, however, allows one to extract $\xi(0)$ and $H_{c2}(0)$; this, in turn, gives $H_{c2}(T)$ with the help of the theoretically known T dependence of H_{c2}.[25]

Among the magnetization data sets examined, we see a profound difference in the behavior of Bi- and Tl-2212, on the one hand, and of Bi-2223, Hg-1223, and Hg-1201, on the other: The former show clearly the nonlocal effects while the latter do not. We tentatively ascribe the difference to a shorter mean free path in the samples of the second group, a speculation still to be verified.

The magnetization is not the only quantity which may be affected at low temperatures by the nonlocality of the current-field relation. We now discuss briefly other examples starting with the Fourier transform of the vortex field, which is measured as a form factor $F(k) = h(k)/\phi_0$ in neutron scattering. To our knowledge, no data on $F(k)$ are published for high-κ materials to compare with Eq. (21). The data for polycrystalline NbTa with $\kappa \approx 4$ taken in relatively low fields[28] lie considerably lower than London's $F(k) = (1+\lambda^2 k^2)^{-1}$. This reduction can be well described by Eq. (21) within values of relevant parameters provided in Ref. 28. However, given the low value of κ and the fact that the material was dirty rather than clean, the agreement between the data and our model might be fortuitous. Nonetheless, we note that the nonlocal terms in $F(k)$ clearly correct the London theory in the right direction.

Another quantity affected by nonlocality is the field variance $\sigma^2 = \overline{[h(\mathbf{r}) - \mathbf{B}]^2} = \mathrm{B}^2 \Sigma_{\mathbf{G}\neq 0} [h(\mathbf{G})/\phi_0]^2$ within the flux lattice (the overbar stands for the average over the vortex lattice cell). In intermediate fields, the convergent sum can be calculated directly.[29] We approximate it by an integral and obtain for $\kappa \gg 1$ making use of Eq. (21)

$$\sigma^2 = \sigma_0^2 \frac{H_0 + 2B}{H_0 + B}, \tag{30}$$

where $\sigma_0^2 = \phi_0^2/8\pi^3\sqrt{3}\lambda^4$ corresponds to the nonlocal effects neglected [the direct summation yields $\sigma_0 = 0.061\phi_0/\lambda^2$ (Ref. 29)]. Again, near T_c, $B \ll H_{c2} \ll H_0$, and the nonlocal correction can be disregarded; the same happens in the dirty case at all T's. Hence, in clean systems at low T's, the variance is enhanced by nonlocality. The penetration depth $\lambda(T)$ extracted from the measured variance is, therefore, underestimated if nonlocality is disregarded. If, for instance, the μSR experiment is done with Bi-2212 in the field $B = 5$ T ($H_0 \approx 30$ T), disregarding nonlocality may cause an error in σ^2 about $B/H_0 \approx 15\%$.

Another deviation from the local London theory is the decrease of the slope $S(B) = dM/d\ln B$ with B seen in high-T_c materials (see, e.g., Refs. 30 and 31) and ascribed to quantum fluctuations of vortices.[32] Our data on Bi- and Tl-2212 are reasonably well described by Eq. (26). We must note, however, that nonlocality does not predict a plateau in $S(B)$ observed for $B > 7$ T,[31] an effect explained by quantum fluctuations. This implies that the nonlocality is not the only cause of the observed suppression of $S(B)$.

A question arises whether or not it would be worthwhile to continue the expansion of the kernel Q in Eq. (15), $4\pi j_i(\mathbf{k})/c = -Q_{il}(\mathbf{k}) a_l(\mathbf{k})$, in powers of $\rho^2 k^2$ in order to obtain a better representation of the vortex field, or to work, if possible, with the exact $Q(\mathbf{k})$ [see Appendix C where $Q(k)$ is given for the clean limit at $T = 0$]. The problem, however, is that even the exact Q describes the current under the assumption $|\Delta| = \text{const}$, which implies that the core region is inaccessible. best - Nevertheless, already the first nonlocal corrections to Q reveal that at low T's of clean materials, the scaling field in the expression for the free energy and the magnetization should be $H_0 \propto \phi_0/\rho^2$, and not the upper critical field $H_{c2} \propto \phi_0/\xi^2$; this is in fact our main result. Adding more terms to Q will not change this result, making, however, the model forbiddingly cumbersome. An exact $Q(k)$ is only available for the clean limit at $T = 0$; on the other hand, using the truncated Q, we are able to evaluate the nonlocal corrections to M at any temperature and for

arbitrary impurity concentration. One can say that the scheme we propose is not a microscopic theory, but a refinement of the London approach which has been proved to be the only tool practically available for interpreting data on the low-temperature magnetic behavior of high-κ superconductors. Still, our model, the usefulness of which has been demonstrated here on two high-T_c compounds, is subject to further experimental tests.

Finally we comment on the possible effect of the nonconventional symmetry of the order parameter on our results, a question which calls for a separate and careful study. We do not expect these effects to be strong because all quantities we discuss here are obtained by averaging over the Fermi surface. Still, it remains to be seen whether or not the nonlocal corrections to the London equations may contain interesting information regarding the possible *d*-wave symmetry.

In conclusion, we have used a gauge-invariant BCS current-field relation valid for any Fermi surface to derive the anisotropic London equations describing nonlocal effects for clean high-κ materials at low T's. The equations well represent the magnetization data for Bi- and Tl-2212 down to the reduced temperature $t\approx 0.4$ where the data are still reversible. We show that one can extract the zero-T coherence length from these data taken at any T in a broad domain away from T_c. We explain why $M(T,B)$ can be approximated by a product of $M_0(T)$ and of a function of B/H_0 with the scaling field H_0 being related to the nonlocality radius which is only weakly T dependent. The nonlocal terms are shown to be responsible for significant deviations of the neutron scattering form factors from standard London predictions. Nonlocal effects should be taken into account in extracting the penetration depth from the μSR and NMR data. Our model reduces to the standard London approach near T_c and for the dirty case at all temperatures.

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APPENDIX A

The quantity $\gamma(T,\alpha)$, with $\alpha=\xi_2/\ell$ being the impurity parameter, can be evaluated analytically at $T=0$ and $T=T_c$. In the first case, the sums in Eq. (20) are replaced with integrals according to $2\pi T\Sigma_\omega\to\int_0^\infty \hbar\, d\omega$. Then, $\gamma(0,\alpha)=I_3/I_1$ where

$$I_1(\alpha)=\int_0^\infty \frac{dx}{(1+x^2)(\sqrt{1+x^2}+\alpha)}$$

and $I_3=I_1''(\alpha)/2$. The integral depends on $\mathrm{sgn}(\alpha-1)$. Since $\alpha=0$ in the clean limit, we are interested mostly in $\alpha<1$, for which

$$I_1=\frac{\pi}{2\alpha}-\frac{2}{\alpha\sqrt{1-\alpha^2}}\tan^{-1}\sqrt{\frac{1-\alpha}{1+\alpha}}. \tag{A1}$$

One can expand this in powers of α and obtain

$$\gamma(0,\alpha)=\tfrac{2}{3}(1-1.865\alpha+3.471\alpha^2-\cdots). \tag{A2}$$

At $T=T_c$, one has $\gamma(T_c,\alpha)=0.315S_3/S_1$ where the BCS ratio $\Delta(0)/T_c=1.764$ has been used. The sum $S_3=\Sigma(2n+1)^{-2}(2n+1+\alpha_1)^{-3}=S_1''(\alpha_1)/2$ where $\alpha_1=\alpha/0.567$ and

$$S_1(\alpha_1)=\sum_{n=0}^{\infty}\frac{1}{(2n+1)^2(2n+1+\alpha_1)}$$
$$=\frac{\pi^2}{8\alpha_1}+\frac{1}{2\alpha_1^2}\left[\psi\left(\frac{1}{2}\right)-\psi\left(\frac{1+\alpha_1}{2}\right)\right]; \tag{A3}$$

ψ is the digamma function. Expanding this in powers of $\alpha_1/2$ we obtain

$$S_1=\sum_{m=0}^{\infty}\frac{(2^{m+3}-1)\zeta(m+3)}{2^{m+3}}(-\alpha_1)^m, \tag{A4}$$

where ζ is the Riemann zeta function. This yields

$$\gamma(T_c,\alpha)=0.301(1-2.026\alpha_1+2.135\alpha_1^2-\cdots). \tag{A5}$$

Note, however, a slow convergence of both series (A2) and (A5) which are given here just to show that $\gamma(\alpha)$ is regular at $\alpha=0$.

APPENDIX B

It is of interest to examine the field structure of a vortex in real space in this approximation. Note that for this one has to formally extend the Fourier transform (21) to arbitrary k's. Then, inversion of Eq. (21) for $\lambda\gg\rho$ yields

$$h(r)=\frac{\phi_0}{2\pi\lambda^2}\left[K_0\left(\frac{r}{\lambda}\right)-K_0\left(\frac{r}{\rho}\right)\right], \tag{B1}$$

an expression similar to that obtained by Kramer[33] and Brandt[34] in the GL domain; it is done here for any T. Clearly, for $r\gg\rho$, the second term is negligible, and Eq. (B1) coincides with the standard London field. However, at shorter distances, the field (B1) is ''well behaved,'' unlike the divergent London solution. As $r\to 0$, h goes to a finite limit $h(0)=(\phi_0/2\pi\lambda^2)\ln(\lambda/\rho)$. For comparison, recall Abrikosov's result $h(0)\approx 2H_{c1}=(\phi_0\ln\kappa)/2\pi\lambda^2$ near T_c.[4] We note also that the nonlocal electrodynamics at Josephson contacts results in a finite $h(0)$ for the vortex situated at the junction.[35]

In real space Eq. (21) takes the form

$$h-\lambda^2\nabla^2 h+\rho^2\lambda^2\nabla^4 h=\phi_0\delta(\mathbf{r}). \tag{B2}$$

This equation (without the explicitly specified T- and ℓ-dependent coefficients and without the source term on the right) was proposed by Koppe prior to BCS theory.[36] It can be obtained by minimizing a functional

$$8\pi\mathcal{F}=\int dV[h^2+\lambda^2(\mathrm{curl}\mathbf{h})^2+\rho^2\lambda^2(\nabla^2 h)^2]. \tag{B3}$$

APPENDIX C

In the discussion of nonlocal effects we assumed that out of the vortex core $\Delta=$ const and that the quadratic expansion of $Q(k)$ in the small parameter $k\xi$ suffices for our purpose. In the immediate vicinity of the core, these assumptions are bound to be violated. Physically, this happens due to two major reasons: (a) The gap is suppressed by high currents and (b) the influence of the full k dependence of Q becomes important.

The GL theory provides the following formula for the suppression of the order parameter by current: $\Delta^2=\Delta_0^2[1-(j/2j_d)^2]$, where Δ_0 is the gap at $j=0$ and $j_d=c\phi_0/16\pi^2\lambda^2\xi$ is on the order of the depairing current density.[4] Using the London expression $j(r)=c\phi_0/8\pi^2\lambda^2 r$ for $j(r)$ near the vortex core, we obtain $\Delta^2(r)=\Delta_0^2(1-\xi^2/r^2)$. At $T\approx T_c$ and $r\sim\xi$, this results in the long-range power-law corrections to the London theory which mask the short-range nonlocal effects [see Eq. (B1)].

In clean superconductors the situation changes qualitatively at $T\ll T_c$: It has been shown by Maki[37] that within the BCS theory, a uniform current at $T=0$ does not suppress $|\Delta|$ as long as $j<j_d$; at $j=j_d$, $|\Delta|$ drops discontinuously to zero. This argument can be extended to currents varying slowly over distances $\sim\xi$ by expanding Δ^2 in series of scalar combinations of the vector $\mathbf{j}$ and its spatial derivatives. The first current corrections to Δ_0 can only be proportional to j^2, $\mathbf{j}\mathrm{curl}\mathbf{j}$, and $|\mathrm{curl}\mathbf{j}|^2$, since $\mathrm{div}\mathbf{j}=0$. At $T=0$ the term j^2 does not contribute to Δ (the Maki effect). The term $\mathbf{j}\mathrm{curl}\mathbf{j}$ is a pseudoscalar which vanishes in crystals invariant under inversion. Hence, $\Delta^2=\Delta_0^2(1-C\lambda^2|\mathrm{curl}\mathbf{j}|^2/j_d^2)$, with $C\sim 1$. However, in high-κ materials the second term in the square brackets is of order $\kappa^{-2}\ln^2\kappa\ll 1$ and thus can be neglected. This follows from the London equation $\mathrm{curl}\mathbf{j}=-c\mathbf{h}/4\pi\lambda^2$ in which $h\sim\phi_0\ln\kappa/2\pi\lambda^2$ is the field on the vortex axis. Therefore, in clean superconductors at low T's, the effect of current on $|\Delta|$ outside the core is suppressed.

The influence of higher-order terms in k can be demonstrated for an exactly solvable case of the cylindrical Fermi surface in the clean limit at $T=0$, for which the BCS $Q(k)$ can be calculated analytically. To this end, it is convenient to take local $\mathbf{j}$ along the x axis, then $\mathbf{k}\|\hat{y}$ (due to the current continuity $\mathbf{k}\cdot\mathbf{j}=0$). Then $4\pi j_x/c=-Qa_x$ and Eq. (15) yields

$$Q=\frac{8\pi e^2N(0)T\Delta_0^2v^2}{c^2}\sum_{\omega>0}\frac{1}{\beta_0}\int_0^{2\pi}\frac{\cos^2\varphi d\varphi}{\beta_0^2+(\hbar v k\sin\varphi)^2/4}. \tag{C1}$$

At $T=0$, we replace $2\pi T\Sigma_\omega$ with $\int_0^\infty\hbar d\omega$ to obtain

$$Q(k)=[2q\tan^{-1}q-\ln(1+q^2)]/q^2\lambda^2, \tag{C2}$$

where $q=k\xi_2$, and $\lambda^2=c^2/4\pi e^2N(0)v^2$. Hence $Q=1/\lambda^2$ for $q\ll 1$ and $Q=\pi/q\lambda^2$ if $q\gg 1$.

For a vortex we have $\mathrm{curl}\mathbf{a}=\mathbf{h}-\phi_0\hat{z}\delta(\mathbf{r})$ and $i\mathbf{k}\times\mathbf{h}(k)=-Q(k)\mathbf{a}(k)$, which gives

$$h(k)=\frac{\phi_0 Q(k)}{k^2+Q(k)}. \tag{C3}$$

Substituting this in Eq. (2), we obtain the free energy by replacing the sum over the reciprocal lattice with an integral from $G_{\min}=2\pi/a$ to $G_{\max}=2\pi/\xi'$ k dependence (the convergence of the integral notwithstanding, we have to cut the integration domain at $G_{\max}$ to avoid the core). For $H\gg H_{c1}$ the Q term in the denominator of Eq. (C3) can be neglected, and we obtain

$$\widetilde{F}=M_0B\left(\ln\frac{1+b}{b}-\frac{\ln(1+b)}{b}+\frac{4\tan^{-1}\sqrt{b}}{\sqrt{b}}+\zeta\right), \tag{C4}$$

where $b=6B/H_0$. It is worth noting that the cutoff at $2\pi/\xi'$ results in renormalization of the core correction η_1, which is uncertain within our scheme anyway. We have now the magnetization M and $S=dM/d\ln B$ in the form

$$-\frac{M}{M_0}=\ln\left(1+\frac{1}{b}\right)+\frac{2\tan^{-1}\sqrt{b}}{\sqrt{b}}+\zeta, \tag{C5}$$

$$S=M_0\frac{\tan^{-1}\sqrt{b}}{\sqrt{b}}\approx M_0\left(1-\frac{b}{3}+\frac{b^2}{5}-\cdots\right). \tag{C6}$$

These formulas describe $M(B)$ behaving qualitatively similar to the given in Eqs. (24) and (26) based on the quadratic expansion of $Q(k)$. One can check that for the usual situation of $B/H_0\ll 1$, the full $Q(k)$ results in the same field dependences of M as that of Eq. (24) if the terms $\sim B^2/H_0^2$ are neglected. On the other hand, the approach of the main text based on the truncated Q has the advantage of incorporating both finite temperatures and the impurity scattering.

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