Role of Vortex Fluctuations in Determining Superconducting Parameters from Magnetization Data for Layered Superconductors

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The fluctuations of vortices in Josephson-coupled layered superconductors are shown to have a profound effect on values of the temperature-dependent penetration depth, Ginzburg-Landau parameter, and upper critical field extracted from reversible magnetization data below the critical temperature.

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Knowledge of the London penetration depth $\lambda(T)$ provides important information on the nature of the superconducting state. In the range of intermediate magnetic fields H with $\phi_0/\lambda^2 \ll H \ll \phi_0/\xi^2$, where ξ is the superconducting coherence length and ϕ_0 is the flux quantum, the reversible magnetization M of a superconductor with the Ginzburg-Landau parameter $\kappa = \lambda/\xi \gg 1$ is a linear function of $\ln H$ [1]. Apart from universal constants, the prefactor of $\ln H$ depends solely on λ. This behavior of $M(H)$ stems from the logarithmic interaction of straight vortices for intervortex distances smaller than λ and is well obeyed in both conventional and high-T_c superconductors [2–8].

The logarithmic dependence of M on H follows directly from the London model [1], which assumes that the normal vortex cores (of radius ξ) do not overlap; this condition is satisfied if $H \ll \phi_0/\xi^2$. The interaction of vortices is established through the overlapping fields and currents; if $H \gg \phi_0/\lambda^2$, the intervortex distance is smaller than λ and the overlap is strong. A variational model has been recently proposed by Hao and Clem to allow for the interaction between cores as well [9]. Since the core interaction is nonlogarithmic, M is only approximately logarithmic in H. Hao and Clem have developed a procedure for extracting λ, κ, and the upper critical magnetic field H_{c2} from the magnetization data; the procedure has been employed by several groups [10–14].

It is now accepted that superconducting fluctuations play an important role in high-T_c materials. Fluctuations are usually associated with high temperature and small ξ, the fluctuating quantity being the amplitude of the order parameter near the mean-field second-order phase transition at $H_{c2}(T) = \phi_0/2\pi\xi^2$ [15, 16]. However, as has been originally proposed by Nelson [17], even for $H \ll H_{c2}$, thermal fluctuations of vortices alter the thermodynamics of the system in the magnetic field in high-T_c superconductors. A striking manifestation of these fluctuations in Josephson-coupled layered superconductors [18] is the existence of a temperature T^*, a few degrees below the mean-field transition temperature T_{c0}, where the magnetization M is independent of H. If one plots M versus T for different fields, all curves cross at T^*, a feature observed in many experiments on high-T_c materials [6, 7, 11, 12].

In this Letter we describe a procedure for extracting λ from $M(H)$ data, which takes the vortex fluctuations into account. We demonstrate that the corrections due to these fluctuations can be substantial; for example, even at temperatures as low as 30 K the estimate of λ is decreased by about 10% in $Bi_2Sr_2CaCu_2O_8$, and by much larger amounts as T approaches T_{c0}. We show that the standard London and Hao-Clem procedures lead to a divergence of $\lambda(T)$ at T^* instead of T_{c0}, and also of $\kappa(T)$ near T^*. These unphysical divergences are removed by taking fluctuations into account.

Layered superconductors are aptly described by the Lawrence-Doniach model, within which the superconducting order parameter is defined only in conducting layers (*a-b* planes; see remark [19]) weakly coupled by Josephson tunneling. For the sake of simplicity, we model the variety of real materials by stacks of *equally* spaced layers, thus characterizing each compound with a single interlayer spacing s. We consider the fields $\phi_0/s^2\gamma^2 \ll H \ll H_{c2}$ ($\gamma^2 = m_c/m_{ab}$ is the superpair effective mass ratio) applied in the c direction; see remark [20]. Then, the intervortex distance is much larger than ξ_{ab}, and one can take the order parameter modulus as constant in space. Within such a scheme, a two-dimensional vortex (a "pancake") in a layer is characterized only by the phase which changes by 2π when one circles the vortex core. Continuous vortex *lines* of three-dimensional theories are replaced here with correlated stacks of pancakes [21, 22]. Whenever γ is large, the restoring force for a distorted stack becomes small, resulting in large vortex fluctuation effects.

For $\gamma \gg 1$, it has been shown by Bulaevskii *et al.* that for $\mathbf{H} \parallel \hat{\mathbf{c}}$, the thermal distortions of the pancakes out of the straight stacks (arranged in a triangular lattice at $T = 0$) result in an extra contribution to the entropy [18]. The magnetization obtained from the total free energy is

$$-M = \frac{\phi_0}{32\pi^2\lambda_{ab}^2(T)} \ln\frac{\eta H_{c2}}{eH} - \frac{k_B T}{\phi_0 s} \ln\frac{16\pi k_B T\kappa^2}{\alpha\phi_0 s H\sqrt{e}}\,. \quad (1)$$

Here, λ_{ab} is the in-plane penetration depth, $\kappa = \lambda_{ab}/\xi_{ab}$,

$e = 2.718\ldots$, and η and α are constants of order unity. The first term on the right of Eq. (1) is the usual London result [1] for the dense system of straight unperturbed vortices, whereas the second term accounts for the fluctuations. The slope $\partial M/\partial \ln H$ is easily obtained:

$$\frac{\partial M}{\partial \ln H} = \frac{\phi_0}{32\pi^2\lambda_{ab}^2(T)}[1 - g(T)], \qquad (2)$$

where

$$g(T) = \frac{32\pi^2 k_B}{\phi_0^2 s} T\lambda_{ab}^2(T). \qquad (3)$$

In order to estimate the contribution of thermal fluctuations to $M(H)$, one ought to compare $g(T)$ with unity; $g(T) = 0$ corresponds to the standard London result. As is seen from Eq. (3), the fluctuations are enhanced by high T and large λ [the latter is due to the fact that the London part of M, the first term in Eq. (1), decreases as λ^{-2}, thus making the contribution of fluctuations relatively more important].

The conspicuous consequence of fluctuations follows from Eq. (2): At a temperature T^* defined by

$$g(T^*) = 1, \qquad (4)$$

M is *independent* of H. At T^*,

$$-M^* \equiv -M(T^*) = \frac{k_B T^*}{\phi_0 s} \ln \frac{\eta\alpha}{\sqrt{e}}. \qquad (5)$$

We note that within London theory one cannot evaluate the constants η and α. From the Hao-Clem variational approach one estimates $\eta \approx 1.4$ [23], the value we adopt in the following. The constant α enters the entropy of fluctuating pancakes through the "volume of particles" needed when the partition function is evaluated; this "volume" should be on the order of the core area and is taken as $\alpha\pi\xi_{ab}^2$ [18].

The theory of fluctuations for 2D systems ($\gamma = \infty$) developed recently by Tešanović *et al.* [16] for $H \sim H_{c2}$ yields Eq. (5) for M^*, but without the ln factor. Noting that γ does not enter either Eq. (1) or (5) for M, we expect $\ln(\eta\alpha/\sqrt{e}) = 1$ in our case as well, the difference in the H dependences of M in the domains $H \ll H_{c2}$ and $H \sim H_{c2}$ notwithstanding. In particular, this means that at T^*, the magnetization measured in the region $H \ll H_{c2}$ is the same as for $H \sim H_{c2}$, a feature clearly seen in the data [6, 11]. Thus we set $\alpha = e^{3/2}/\eta$. Although this choice of η and α affects the absolute values for H_{c2} and κ given below, our primary conclusion about the importance of vortex fluctuations remains unchanged.

To demonstrate how the fluctuations influence the determination of λ, we analyze $M(T, H)$ data with $\mathbf{H} \parallel \hat{\mathbf{c}}$ for a single crystal of $Bi_2Sr_2CaCu_2O_8$. The crystal used and the experimental details are described elsewhere [13]. The M vs $\ln H$ isotherms at temperatures of 72 to 83 K are plotted in Fig. 1. These data show that $|M^*| \approx 0.25$ G and $T^* \approx 80$ K; using Eq. (5), one obtains $s \approx 23$ Å.

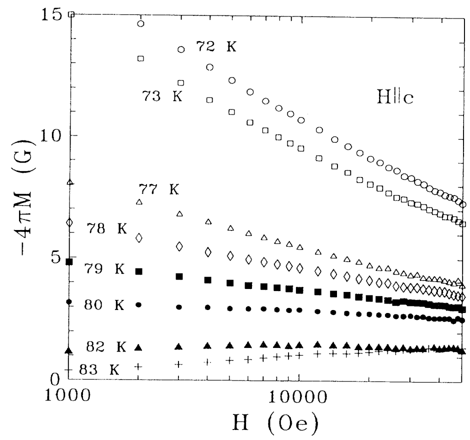

FIG. 1. Magnetization M vs ln$H$ ($\mathbf{H} \parallel \hat{\mathbf{c}}$ is the applied magnetic field) isotherms for a single crystal $Bi_2Sr_2CaCu_2O_8$; the temperatures are indicated. Similar data were obtained down to 30 K.

We then evaluate the experimental slopes $\partial M/\partial \ln H$ vs T, and determine $\lambda_{ab}(T)$ using Eqs. (2) and (3). The results are shown in Fig. 2(a) along with the $\lambda_{ab}(T)$ obtained neglecting the fluctuations [$g = 0$ in Eq. (2)]. At the lowest temperatures (≈ 30 K) the difference between the two results is small ($\approx 10\%$), although not negligible. The difference, however, increases rapidly with T. In particular, neglecting fluctuations results in an apparent divergence of $\lambda_{ab}(T)$ at T^* since at this temperature $\partial M/\partial \ln H = 0$. The correct $\lambda_{ab}(T^*)$ is finite.

The solid and dashed curves in Fig. 2 show the best fit of BCS $\lambda(T)$ [24] in the clean and dirty limits, respectively, to $\lambda_{ab}(T)$ obtained from the data taking fluctuations into account. Of course, one could do this fit for $\lambda_{ab}(T)$ extracted from the data neglecting fluctuations (the upper set of points in Fig. 2). Then, however, other quantities extracted from the same data [e.g., $H_{c2}(T)$] would display unphysical behavior (see below and Ref. [13]).

As we have mentioned, in the model used here [18] there is one infinitesimally thin conducting layer per unit cell. However, the unit cell of $Bi_2Sr_2CaCu_2O_8$ contains two pairs of closely spaced (≈ 3 Å) CuO_2 layers; the distance between the pairs is ≈ 12 Å. To apply our model to $Bi_2Sr_2CaCu_2O_8$ we assume that the two adjacent CuO_2 planes are strongly coupled so that we can treat them as a single superconducting layer; the average distance between the double layers is ≈ 15 Å. These double layers are weakly coupled [16] making our model applicable. Therefore, $s \approx 15$ Å is a proper value for $Bi_2Sr_2CaCu_2O_8$ described within our model. Further refinements of the theory are needed to explicitly include multiple interlayer separations actually present in various layered superconductors.

It should be mentioned that the data of Kes *et al.* [6]

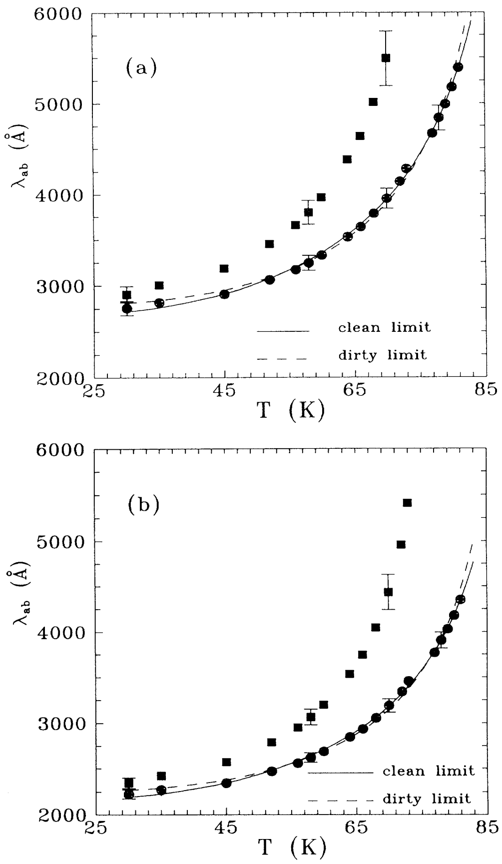

FIG. 2. (a) The in-plane penetration depth λ_{ab} vs temperature T extracted from $M(T,H)$ data as explained in the text (circles). The result fits the BCS clean limit better than the dirty one. Squares show $\lambda_{ab}(T)$ which is extracted from the same data neglecting fluctuations. The parameter $s = 23$ Å is obtained from the data using Eq. (5); the sample is assumed to be 100% superconducting. Representative error bars are shown. (b) Same as (a) except that $s = 15$ Å and the sample is assumed to be 65% superconducting.

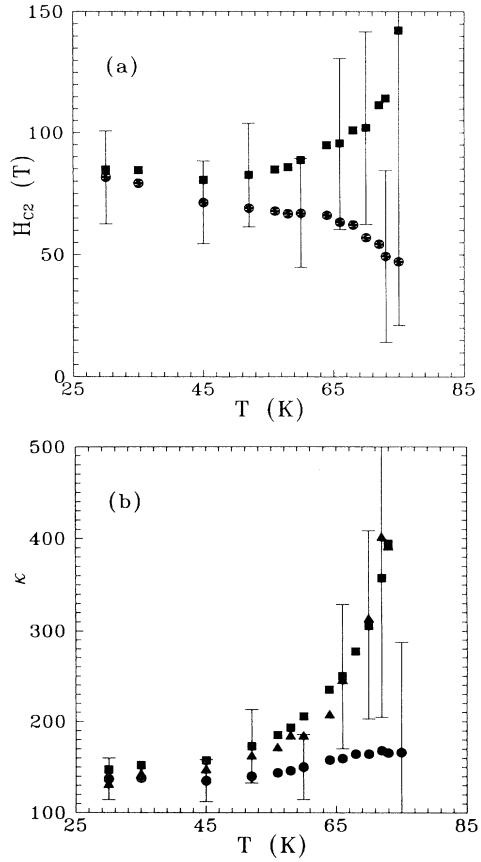

FIG. 3. (a) The upper critical field H_{c2} vs temperature T (circles) obtained from the $M(T,H)$ data using Eq. (6) for the sample parameters as in Fig. 2(a). Squares show the results for H_{c2} with fluctuations neglected. (b) Ginzburg-Landau parameter κ vs temperature T (circles) obtained from the $M(T,H)$ data using Eq. (6) and the sample parameters as in Fig. 2(a). Squares are obtained neglecting the fluctuations. The triangles show $\kappa(T)$ obtained using the Hao-Clem procedure [13].

for the same compound give $s = 20.6$ Å, while those of Kadowaki [7] yield $s = 16.5$ Å. A possible cause for this variation may be sample variations. From Eq. (5), $s \propto 1/M^*$. If only a fraction of a sample is superconducting, the value of M^* associated with the *superconducting* volume is underestimated. Thus an overestimate of s may occur. To correct for the inconsistency between $s = 23$ Å extracted from the raw data and $s = 15$ Å dictated by the structure, we assume that only $15/23 = 65\%$ of our crystal is superconducting. We should then rescale the slopes $\partial M/\partial \ln H$ by a factor of 0.65, and recalculate $\lambda_{ab}(T)$ with $s = 15$ Å. The result is shown in Fig. 2(b). Thus, Fig. 2 shows that, independent of a particular choice of s or of the actual superconducting volume fraction, our conclusion that the vortex fluctuations have a strong influence on the derived $\lambda_{ab}(T)$ remains unchanged.

The $M(H)$ data can also be used to determine $H_{c2}(T)$ (and κ) with the help of Eq. (1). To this end we rewrite Eq. (1) in the form

$$\ln\frac{\eta H_{c2}}{eH} = \left(-\frac{32\pi^2\lambda_{ab}^2}{\phi_0}M + g\ln\frac{g}{e}\right)\frac{1}{1-g}\,. \qquad (6)$$

By plotting the expression on the right of Eq. (6) versus $\ln H$ for different temperatures and extrapolating the straight lines so obtained to the $\ln H$ axis, one obtains $\eta H_{c2}(T)/e$. The result of this procedure is shown in Fig. 3(a). The same method implemented by setting $g = 0$ in Eq. (6), i.e., disregarding fluctuations, yields the upper set of points in Fig. 3(a). Given H_{c2}, one estimates $\xi_{ab} = \sqrt{\phi_0/2\pi H_{c2}}$ and $\kappa = \lambda_{ab}/\xi_{ab}$, as shown in Fig.

3(b). Clearly, neglecting fluctuations leads to an unphysical increase of the derived H_{c2} and κ with temperature. This is true for the standard London model [1] as well as for the Hao-Clem procedure [9, 13]. The main source of errors is in extracting the slopes $\partial M/\partial \ln H$ from the data; near T^*, $|\partial M/\partial \ln H|$ is small, so that the relative error increases.

We note that the contribution of vortex fluctuations to the magnetization is not an exclusive property of high-T_c superconductors. It should be taken into account whenever the anisotropy and the penetration depth are sufficiently large. As an example, we mention the organic superconductor κ-(BEDT-TTF)$_2$Cu(NCS)$_2$; strong fluctuations near H_{c2} of this compound have already been noticed [25] [although T_{c0} is only about 10 K, estimates of $\lambda_{ab}(0)$ range from 6800 Å [26] to $\sim 10^4$ Å [27]].

In conclusion, we have demonstrated that fluctuations of vortices can by no means be neglected in the determination of the London penetration depth from reversible magnetization data, when dealing with Josephson-coupled layered materials. In particular, the fluctuations must explicitly be taken into account when only the high-temperature data are available [4, 5, 14].

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[19] The notation we adopt here is not universal; e.g., the conducting planes of the organic superconductor κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ are labeled as *bc* (see, e.g., Ref. [25]).
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