Low Temperature Fluctuations of Vortices in Layered Superconductors

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Data on the ^{205}Tl NMR frequency shift and linewidth due to the magnetic field variation in the mixed state of $Tl_2Ba_2Ca_2Cu_3O_{10}$ are presented. These data, which are commonly used to extract the London penetration depth, show a near linear temperature dependence at $T \ll T_c$. We offer an explanation of this effect as being caused by thermal fluctuations of vortices in weakly coupled layered systems. We argue that the fluctuations cannot be ignored in interpretation of NMR and muon-spin-resonance data for materials with large penetration depths even at low T.

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In weakly coupled layered superconductors, thermal fluctuations of vortices strongly influence the thermodynamics of the system in a magnetic field. This has been observed in "non-mean-field" dependence of the magnetization M on temperature T and magnetic field H in $Bi_2Sr_2CaCu_2O_8$ (Bi-2212) and $Bi_2Sr_2Ca_2Cu_3O_{10}$ (Bi-2223) well under the mean-field transition temperature T_{c0}. At a temperature T^*, a few kelvins below T_{c0}, M is field *independent* [1–3]. The effect has been explained quantitatively by the contribution of fluctuating vortices to the system entropy [4].

For high anisotropies and $H \ll H_{c2}$, this contribution was evaluated within the Lawrence-Doniach (LD) framework. The order parameter modulus of superconducting layers was set constant in space and time (at T's not too close to T_{c0}) while vortex positions (i.e., phases) were fluctuating [5]. As such the method is not restricted to high temperatures although, at first sight, there is no reason to test the effect of fluctuations at low T. Indeed, as follows from the data [1–3], the fluctuation contribution to the free energy is on the order of the mean-field free energy near T^*, which is not far from T_{c0}; with decreasing T, the unperturbed energy increases (in absolute value) while the fluctuation part diminishes.

This argument, however, does not necessarily hold if the measured quantity is related to the detail of the field distribution of the fluctuating vortices. The fluctuation amplitude depends on the vortex "stiffness," which is suppressed with increasing anisotropy and penetration depth λ; thus, anisotropic materials with large λ (Bi- and Tl-based compounds or some organic superconductors) are candidates for strong fluctuation effects. NMR, muon-spin-resonance (μSR), and small angle neutron scattering techniques are sensitive to the field variation within the flux-line lattice, and as such may be affected by fluctuations in vortex positions even at low T. In this Letter, we present results on the peak position of the ^{205}Tl NMR spectra and spectral width as functions of T for $Tl_2Ba_2Ca_2Cu_3O_{10}$ (Tl-2223). We find that the T dependence of the penetration depth extracted from these data, disregarding fluctuations, deviates from the standard BCS behavior. The deviations may well be understood in terms of fluctuations even at low T.

The powder sample was prepared by solid-state reaction following the procedure of Refs. [6, 7]. X-ray diffraction analysis indicated that the structure was $Tl_2Ba_2Ca_2Cu_3O_{10+\delta}$ with only traces of unreacted starting material. The transition temperature as determined by the diamagnetic shielding signal was $\simeq$ 120 K. The sample was ground thoroughly, mixed with an epoxy resin at a nominal filling fraction of 15% by volume. This superconductor-epoxy composite was cured in a 3.5 T field. The procedure results in a stable, predominantly c-axis aligned sample [8].

Pulsed NMR measurements were performed in the 3.5 T field with a heterodyne spectrometer. ^{205}Tl spectra were recorded with a spin-echo sequence and by shifting the spectrometer frequency through the resonance. Typical delay times for echo formation were $\tau_e \approx$ 40–60 μs.

We have studied the c-axis alignment by ^{205}Tl NMR in the normal state [6, 7]. From the analysis of the spectra for $\mathbf{H} \parallel \hat{\mathbf{c}}$, $\mathbf{H} \perp \hat{\mathbf{c}}$, and the random powder, we found that in this sample (48±2)% of the Tl-2223 crystallites were aligned and the rest randomly oriented. This information was used later to obtain the aligned spectrum

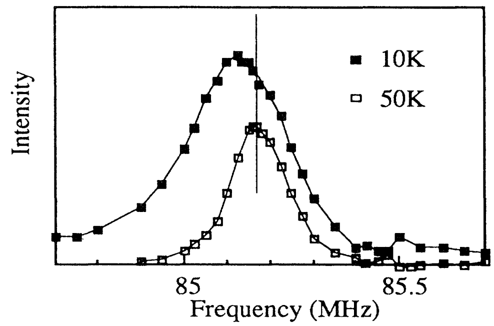

FIG. 1. ^{205}Tl spectra of the aligned crystallites at 10 K (filled squares) and 50 K (open squares) for $\mathbf{H} \parallel \hat{\mathbf{c}}$. The subtraction of the random powder contribution is described in the text. Note the relative shift in the peaks of the two spectra.

in the superconducting state. Prior to the alignment, we measured the ^{205}Tl NMR spectra in the superconducting state on the random powder sample at all temperatures. Since the percentage of the alignment is known, the random powder contribution can be subtracted from the spectrum of the aligned sample. This procedure was repeated for all temperatures. An example is given in Fig. 1 showing two subtracted spectra for $\mathbf{H} \parallel \hat{\mathbf{c}}$ at 10 K and 50 K. One sees that the peak of the 50 K spectrum is shifted to a higher frequency. It is worth noting that, in our analysis, no assumption about the line shape in the superconducting state for either the random powder or the aligned crystallites is made and the subtracted spectrum should be regarded as being solely from the aligned crystallites.

The inhomogeneous field, h, of the vortex lattice results in broadening of the NMR absorption line. In particular, the maximum of the NMR line corresponds to the saddle point value h_S of $h(x, y)$, which is shifted with respect to the average field B. It has been shown previously [7] that the shift, $h_S - B$, can be obtained as the difference between the Tl NMR frequency shifts at $\mathbf{H} \parallel \hat{\mathbf{c}}$ and $\mathbf{H} \perp \hat{\mathbf{c}}$. This is possible if the anisotropy parameter $\gamma = \lambda_c/\lambda_{ab}$ in the material of interest is large, and if the Tl spin shift is isotropic [7]. Unfortunately, there is still no reliable data on γ for Tl-2223 [9]. In this situation our approach is to study how the role of fluctuations varies with γ.

For $\mathbf{H} \parallel \hat{\mathbf{c}}$, in the *absence* of fluctuations, the shift is related to the penetration depth λ_{ab}: $B - h_S = 3.67 \times 10^{-2}\phi_0/\lambda_{ab}^2$ [7,11]. However, the observed shift *is affected* by fluctuations. We then introduce a new length Λ_{ab} which is related to the actual shift by the same formula:

$$B - h_S = 3.67 \times 10^{-2}\phi_0/\Lambda_{ab}^2\,, \tag{1}$$

where Λ_{ab} would coincide with λ_{ab} if there were no fluctuations. The length thus determined, $\Lambda_{ab}(T)$ is shown in Fig. 2; the data are normalized on the $\Lambda_{ab}(6\text{ K}) \simeq 1150$ Å

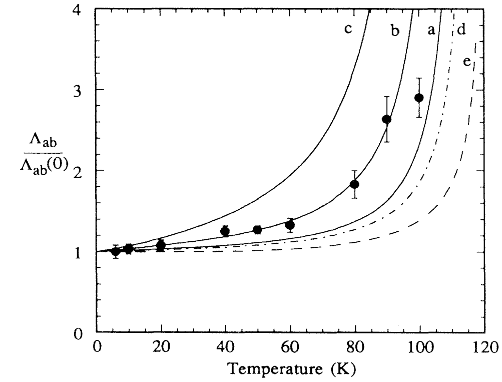

FIG. 2. Experimental data on $\Lambda_{ab}(T)$ extracted from the position of the NMR line-shape maximum using Eq. (1). The theoretical curves are evaluated for the following: (a) $\gamma = \infty$ and $\lambda_{ab}(0) = 1100$ Å, (b) $\gamma = \infty$ and $\lambda_{ab}(0) = 1500$ Å, (c) $\gamma = \infty$ and $\lambda_{ab}(0) = 1960$ Å, (d) $\gamma = 60$ and $\lambda_{ab}(0) = 1500$ Å, and (e) the "two-fluid" model, $\lambda_{ab}(T)/\lambda_{ab}(0) = (1 - t^4)^{-1/2}$.

(see footnote [12]); the near *linear* increase of Λ_{ab} with T at low temperatures is clearly seen, a considerable spread of data points notwithstanding.

From the subtracted spectra, we have also obtained the linewidth [full width at half maximum (FWHM)], as a function of T, shown in Fig. 3. Contribution from the normal state has been removed, and Fig. 3 shows the additional broadening due to the superconducting state. In principle, this linewidth may not be a good representation of the variance of the field distribution, due to distortions of the flux lattice and experimental difficulties in obtaining the complete spectrum [7]. However, for a static array of straight vortices, the field distribution is determined by $\lambda(T)$ and a similar temperature dependence for the FWHM and the variance might be

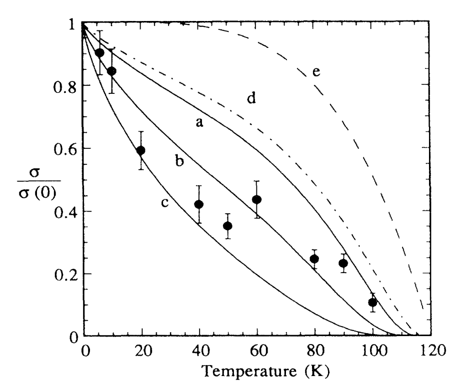

FIG. 3. Experimental linewidth (FWHM) vs temperature for $\mathbf{H} \parallel \hat{\mathbf{c}}$. The theoretical curves are evaluated for the following: (a) $\gamma = \infty$ and $\lambda_{ab}(0) = 1100$ Å; (b) $\gamma = \infty$ and $\lambda_{ab}(0) = 1500$ Å; (c) $\gamma = \infty$ and $\lambda_{ab}(0) = 1960$ Å, (d) $\gamma = 60$ and $\lambda_{ab}(0) = 1500$ Å, and (e) the "two-fluid" model, $\sigma/\sigma(0) = 1 - t^4$.

expected. This feature is commonly used to evaluate $\lambda(T)$. It is clear from Fig. 3 that there is a significant T dependence in the FWHM at low temperatures contrasting with the behavior expected for conventional s-wave superconductors. We show below that the fluctuations of vortices may well cause this temperature dependence.

For fluctuating vortices, the local field at nuclear sites varies in time. The NMR spectrum measures the time average of these fluctuating local fields over the time τ_e for echo formation. When the characteristic time τ_f of fluctuations is small relative to τ_e, the NMR linewidth will be reduced due to motional averaging; on the other hand, the line broadens for $\tau_f \gg \tau_e$ when the fluctuations are slow and averaging is not effective. We note that the position h_S of the line maximum is far less sensitive to τ_f than the FWHM. The time τ_f is estimated to be $\sim 10^{-10}$ s [13], smaller than τ_e; thus, motional averaging does take place. At very low temperature we might expect τ_f to be longer. For this reason, we do not attempt to derive λ_{ab} from FWHM data and we will only discuss its temperature dependence and compare it with the T dependence of the calculated field variance.

We now evaluate the distribution of the field averaged over the thermal motion within the LD model. One assumes that the superconducting order parameter is defined only in equidistant conducting planes x-y (a-b) with a period s in the z (c) direction. In the following we consider fields $H_{c1} \ll H \ll H_{c2}$ applied along c, so that the distance between vortices is large relative to the core size. The order parameter modulus can then be set constant, and only the phase variation is relevant. Continuous vortex *lines* of the 3D London description are replaced in the LD approach with stacks of 2D "pancakes" situated at $\mathbf{r}_{n\nu}$, where n and ν enumerate layers and vortices.

The pancakes interact magnetically and via Josephson tunneling. The relative strength of these two interactions depends on the ratio λ_J/λ_{ab}, where $\lambda_J = s\gamma$ characterizes the Josephson coupling: the weaker the coupling, the greater λ_J is. For Tl-2223 we should take $s \approx 18$ Å. In fact, there are two triple CuO_2 layers per each 36 Å of the unit cell; the three CuO_2 planes in each layer are in close proximity (≈ 3.2 Å) so that one can consider them as one superconducting layer.

Minimization of the LD free energy provides equations for the field and for the phase differences. One obtains (see appendix B of Ref. [14]) for arbitrarily positioned pancakes at $\mathbf{r}_{n\nu}$:

$$h_z = \frac{\phi_0 s}{\pi^3} \sum_{n\nu} \int_0^\infty dq \int_0^\infty \frac{dk_x dk_y}{1+\lambda_{ab}^2(k^2+Q^2)} \frac{k^2+Q^2}{k^2+q^2} \times \cos[\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}_{n\nu}) + q(z-ns)] \,. \quad (2)$$

Here $\mathbf{k} = (k_x, k_y)$, and $Q^2 = 2s^{-2}(1-\cos qs)$; we will not need other components of $\mathbf{h}$.

The profile $h_z(x,y)$ responsible for the outcome of the NMR experiments is the thermal average of the field (2) over all possible positions $\mathbf{r}_{n\nu}$. The equilibrium structure is a triangular lattice of straight z oriented pancake stacks: $\mathbf{r}_{n\nu} \equiv \mathbf{r}_\nu^{(e)}$. The pancakes are in thermal motion near their equilibrium positions, so that $\mathbf{r}_{n\nu} = \mathbf{r}_\nu^{(e)} + \mathbf{u}_{n\nu}$, with $\mathbf{u}_{n\nu}$ being the displacement of the pancake (n,ν). For $u^2 \ll \phi_0/H$, the free energy can be taken as quadratic in $\mathbf{u}$ [4,15]; then the thermal averaging is straightforward:

$$\langle h(\mathbf{r})\rangle = B \sum \frac{e^{-G^2\langle u^2\rangle/4}}{1+\lambda_{ab}^2 G^2} \exp(i\mathbf{G}\cdot\mathbf{r}) \,. \quad (3)$$

Here, $\langle u^2\rangle$ is the mean square displacement of pancakes from their equilibrium positions; the index z is omitted, and the sum is over $\mathbf{G} = (G_x, G_y)$ forming the reciprocal lattice. Equation (3) has been given by Brandt [16].

For layered Josephson coupled systems, $\langle u^2\rangle$ has been evaluated by Glazman and Koshelev [15]. In fields $B \gg \phi_0/\lambda_J^2$, their result is

$$\langle u^2\rangle = \frac{8\pi\lambda_{ab}^2 T}{s\phi_0 B} \ln\frac{B\lambda_J^2}{\phi_0}\,, \quad \lambda_J = s\gamma \ll \lambda_{ab} \,. \quad (4)$$

Similar calculation gives in the limit of vanishing Josephson coupling:

$$\langle u^2\rangle = \frac{16\pi\lambda_{ab}^2 T}{s\phi_0 B} \ln\frac{\pi B\lambda_{ab}^2}{\phi_0 \ln(1/k_0 s)}\,, \quad \lambda_J \gg \lambda_{ab} \,, \quad (5)$$

where $k_0^2 = 4\pi B/\phi_0$. To demonstrate the relevance of the Debye-Waller factor in Eq. (3), we calculate the value h_S at the saddle point and the variance $\sigma^2 = (B/\phi_0)\int d^2r[\langle h(\mathbf{r})\rangle - B]^2$.

Since the Debye-Waller factor does not alter the vortex lattice symmetry, the saddle point positions within the cell are unchanged by fluctuations: they are in the middle of lines connecting equilibrium positions of the nearest neighbors. One then obtains for h_S in a triangular lattice:

$$\langle h_S\rangle - B = \frac{\phi_0\sqrt{3}}{8\pi^2\lambda_{ab}^2} \sum_{m,n}{}' \frac{(-1)^n}{m^2-mn+n^2} \exp\left[-\frac{2\pi^2 B}{\sqrt{3}\phi_0}\langle u^2\rangle(m^2-mn+n^2)\right] ; \quad (6)$$

the term $m=n=0$ is omitted in the sum $\sum'$.

To compare this with the data of Fig. 2, we use the "two-fluid" approximation $\lambda_{ab}^2(T)/\lambda_{ab}^2(0) = (1-t^4)^{-1}$ with $t = T/T_{c0}$, $T_{c0} = 120$ K, and evaluate Λ_{ab} with the help of Eqs. (6) and (1). The solid curves of Fig. 2 are obtained using $\langle u^2\rangle$ of Eq. (5) where the Josephson coupling is disregarded ($\gamma = \infty$). The dash-dot-dashed curve shows the same for the anisotropy parameter $\gamma = 60$ and $\lambda_{ab}(0) = 1500$ Å (see footnote [17]). As can be seen, the fluctuation effect is stronger for higher anisotropies and larger λ_{ab}; physically, this is due to the reduced "stiffness" of the vortex system with respect to tilt deformations.

Similarly, we have evaluated the variance $\sigma^2 = \sum' \langle h(\mathbf{G})\rangle^2$, where $\langle h(\mathbf{G})\rangle$ is the Fourier transform of $\langle h(\mathbf{r})\rangle$; see Eq. (3). Figure 3 shows $\sigma(T)/\sigma(0)$ with the same parameters as those of Fig. 2. Both figures indicate that fluctuations of vortices can explain deviations of experimental results for $\Lambda_{ab}(T)$ and $\sigma(T)$ from the standard T dependences: $\lambda_{ab}(T)$ and $\propto \lambda_{ab}^{-2}(T)$, respectively.

It should be noted that our approach disregards pinning. However, within the collective pinning model (many vortices per pinning center), most pancakes fluctuate freely independent of the pinning potentials. Hence, one expects that the NMR line shape, being dependent on properties on the scale of the vortex lattice cell, is unaffected by weak pinning.

In conclusion, we have shown that fluctuations of vortices, ever present in layered Josephson coupled superconductors, strongly affect the outcome of NMR experiments in the mixed state even at low temperatures. These fluctuations should be considered when the NMR data are used for determining $\lambda_{ab}(T)$ before resorting to more sophisticated possibilities such as unconventional pairing. Behavior similar to that described in this paper has been seen in μSR data on Bi-2212 [18], on polycrystalline Tl-2212 [19], and on organic material (BEDT-TTF)$_2$Cu(NCS)$_2$ [20].

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