Low-temperature vortex dynamics in a high-temperature superconductor

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Magnetic-field gradients in the mixed state of a type-II superconductor are studied using ^{205}Tl nuclear magnetic resonance (NMR) on $Tl_2Ba_2Ca_2Cu_3O_{10+\delta}$. An anomalous peak was observed in the temperature dependence of the transverse relaxation rate at $T/T_c \approx 0.25$. We attribute this behavior to magnetic-field fluctuations from vortex dynamics. We interpret this behavior as a crossover of the principal time scale for vortex dynamics with that of the NMR experiment, approximately 100 μs. The temperature dependence of this time scale is discussed.

The dynamics of magnetic vortices in the mixed state of a high-temperature superconductor is intriguing for several reasons. The high transition temperature implies large thermal fluctuations with the possibility of various flux phases including melting of the flux lattice.[1] Second, the vortex-vortex interactions are substantially affected by anisotropy which is particularly strong in the Tl superconductors. Vortex dynamics affects the resistive transition in a magnetic field[2] and can be observed in mechanical vibration experiments.[3] Melting of the low-temperature flux lattice has been offered[2,3] as an explanation for some of these observations. It is generally assumed that at low temperature, the vortices settle into a solid lattice, the Abrikosov state (possibly with disorder), or a vortex glass[4] where they no longer contribute to dissipation processes detected by transport or torque measurements.

A spatially inhomogeneous magnetic field is intrinsic to type-II superconductors in their mixed state. The details of this field distribution, fixed by the superconducting penetration depth and the coherence length, as well as the applied magnetic field, can be directly determined from the NMR spectrum. In a vortex solid the NMR spectrum should be significantly broadened and shifted from that of the normal state. Vortex motion of sufficiently large amplitude and frequency will collapse and shift the field distribution back to its normal state value. Recently, Song *et al.*[5] have shown that the apparent penetration depth extracted from the spectrum shift in a vortex solid can acquire a linear temperature dependence at low temperatures as a result of thermal fluctuations of the vortices. In this application of the local vortex fluctuation model, estimates were made in the limit of short vortex correlation time.

However, the time scale for vortex dynamics can be expected to change significantly from low to high temperature. This can include thermal fluctuations of a pinned vortex about its equilibrium position, melting of the vortex solid, and vortex diffusion.[6] Since nuclear spin relaxation rates are sensitive to the spectral density of magnetic-field fluctuations, it is a natural tool for studying this aspect of vortex dynamics and is the subject of this paper.

Both spin-spin relaxation, $1/T_2$, and spin-lattice relaxation, $1/T_1$, in principal, can reflect vortex dynamics. The spin-lattice relaxation rate is determined by transverse field fluctuations at the Larmor frequency; for example, the ^{205}Tl resonance frequency is 85 MHz at 3.46 T. For large-amplitude vortex fluctuations, there is a significant probability of the core traversing many nuclei and this can be an effective channel for nuclear spin-lattice relaxation. Finally, $1/T_2$ is sensitive to the field fluctuations parallel to the applied field at low frequency. In this case, there can be a direct effect of vortex dynamics. The idea is the following. Each nucleus precesses at a rate determined by the local field. Fluctuations in the longitudinal component of the local field lead to incoherent dephasing among the nuclei and decay of the NMR spin-echo signal. In the superconducting state, the nuclear spin-spin interactions, giving rise to $1/T_2$ in the normal state, are quenched by the presence of magnetic-field gradients, thereby reducing $1/T_2$. In contrast vortex dynamics contributes directly to magnetic-field fluctuations at the nuclear sites, thus increasing $1/T_2$. In this paper we report observation of an anomalous peak in $1/T_2$ in the $Tl_2Ba_2Ca_2Cu_3O_{10+\delta}$ superconductor which we attribute to the vortex dynamics.

The powder sample was prepared by a solid-state reaction of a mixture of the appropriate metal oxides of high purity,[7,5] giving a high quality superconductor with a narrow diamagnetic transition at 120 K. The sample was ground repeatedly and mixed with an epoxy resin (Stycast 1266, Emerson and Cumming, Inc.) at a nominal filling fraction of 15% by volume. This superconductor-epoxy composite was cured in a 3.46 T magnetic field. This procedure produces stable, predominantly c-axis alignment.

We measured $1/T_2$ using a spin-echo sequence in the temperature range of 5–300 K, at fields of 3.46 T and 2.0 T. Examples of the measurements above and below T_c are

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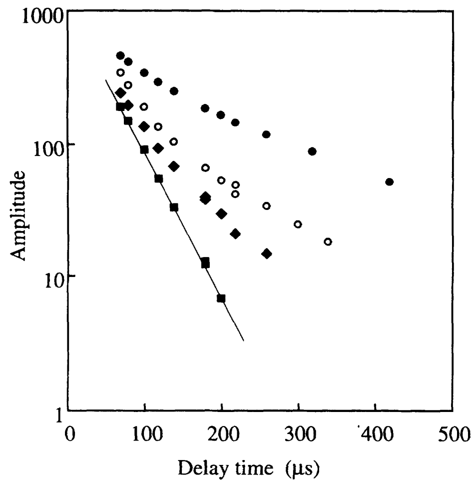

FIG. 1. Spin-echo amplitude (in arbitrary units) as a function of the delay time at 140 K (squares), 80 K (open circles), 30 K (solid diamonds), and 6 K (solid circles). The magnetic field is 3.46 T and parallel to the c axis.

shown in Fig. 1. The relaxation in the superconducting state is, in general, not a single exponential as is the case in the normal state. This nonsingle exponential behavior is likely due to a distribution of magnetic-field gradients. The relaxation rate determined from the data at short delay time gives an average of the relaxation rates and is defined to be $1/T_2$ in the following discussion. We have studied the magnetic field dependence of T_2 and the results are shown in Fig. 2(a). The temperature

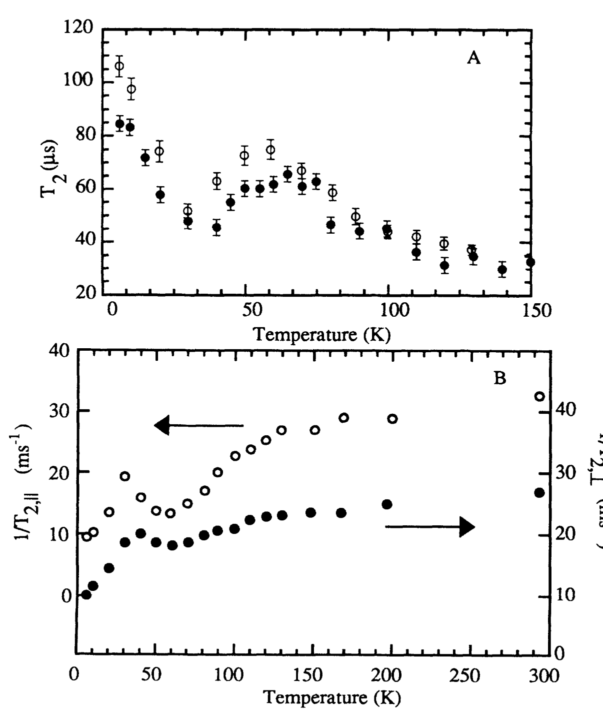

FIG. 2. (a) T_2 vs temperature at 3.46 T (open circles) and 2.0 T (solid circles) for $\mathbf{H}\|\, c$. (b) $1/T_2$ vs temperature at 3.46 T for $\mathbf{H}\|\, c$ (open circles) and $\mathbf{H}\perp c$ (solid circles).

dependence and the anisotropy are shown in Fig. 2(b). A peak in $1/T_2$ occurs at a relatively low temperature and the temperature dependence of $1/T_2$ is quite marked even at much lower temperatures. At a magnetic field of 3.46 T, the peak is strongest for $\mathbf{H}\|\, c$ although it is still clearly evident for $\mathbf{H}\perp c$. At 2.0 T, no peak in $1/T_2$ is observed for $\mathbf{H}\perp c$ (data not shown). The peak in $1/T_2$ at low temperature can be attributed to magnetic-field fluctuations.

In the flux lattice, the midpoint of two adjacent flux lines is a saddle point in the field distribution. Nuclear spins in its vicinity contribute to a peak in the NMR spectrum. The relaxation measurements were performed at the peak of the spectrum and thus are mostly sensitive to the field distribution and the fluctuations at the saddle points. The rate $1/T_2$ is generally smaller at frequencies away from this peak.

The nuclear spin-spin interactions arise from direct dipolar coupling and the indirect coupling through electronic polarization. They are primarily responsible for the transverse relaxation in the normal state.[8] It is important to note that these interactions are between the neighboring nuclear spins, regardless of their origin. When the resonance frequencies of the nearby nuclei are different, this flip-flop transition does not conserve the nuclear Zeeman energy and thus is forbidden.[9] In the mixed state of a superconductor there are large magnetic-field gradients associated with the static vortex structure. They increase in amplitude with decreasing temperature owing to the temperature dependence of the magnetic penetration depth. Their presence reduces the effective spin-spin interaction and in general $1/T_2$ becomes smaller in the superconducting state.

A similar phenomenon has been studied[10] in the context of the cross relaxation of nuclei with different gyromagnetic ratios γ, where the Larmor frequencies are not the same. Bloembergen *et al.* and others[10] studied quantitatively the cross relaxation process by considering the overlap of the effective local field distribution at adjacent nuclei i and j. In this case, the local field is due to the nuclear dipole-dipole interaction. Assuming a Gaussian shape for this distribution, the cross relaxation rate $1/T_{ij}$ is given by

$$\frac{1}{T_{ij}} = (\langle\Delta\omega^2\rangle)^{1/2} \exp\left[-\frac{\Delta h_{ij}^2}{2\langle\Delta\omega^2\rangle}\right], \tag{1}$$

where $\langle\Delta\omega^2\rangle$ is the second moment of the spin-spin coupling within each atomic species alone. Δh_{ij} is the static frequency difference at the adjacent nuclei, $\Delta h_{ij} = (\gamma_i - \gamma_j)H_{\text{ext}}$.

We begin by estimating this effect, in the absence of flux motion. We have calculated the field gradient using London theory. For an ideal flux lattice, the first derivatives of the field at the saddle points are necessarily zero. The second derivatives scale with the field:

$$\frac{\partial^2 h}{\partial {x_i}^2} \propto \frac{h}{\lambda_{ab}^2}, \tag{2}$$

where $x_i = x$ or y are the coordinates in the plane perpendicular to the applied field, and λ_{ab} is the inplane penetration depth. We have numerically calculated these derivatives for the isotropic case $\mathbf{H} \parallel c$. For example, taking $\lambda_{ab} = 1100$ Å, which is appropriate for $Tl_2Ba_2Ca_2Cu_3O_{10+\delta}$ at 5 K,[7] we found $\partial^2 h/\partial x^2 = 4.5 \times 10^{-2}$ G/Å^2, $\partial^2 h/\partial y^2 = -1.0 \times 10^{-2}$ G/Å^2. This gives a maximum field difference between adjacent Tl nuclei in the saddle point region of the magnetic-field distribution, $\Delta h = 0.7$ G, comparable to the Tl spin-spin coupling in the normal state. Consequently, one can expect such gradients to significantly reduce $1/T_2$ at low temperatures. From the field dependence of Eq. (2), one expects that T_2 is longer at higher field particularly at low temperatures when the fluctuation effect is smallest. This is confirmed by our measurement of the field dependence shown in Fig. 2(a). The results of the calculation of the temperature dependence of $1/T_2^{\text{static}}$ are shown in Fig. 3 as a heavy solid line constrained to match the experimental data at the lowest temperature where effects of flux motion are a minimum. A comparison of the experimental data with the calculation shows qualitatively the expected behavior at high temperatures, but with a dramatic departure near $T/T_c \approx 0.25$. We now turn to possible explanations of this behavior.[11]

One possible effect of vortex motion is its modification of the field gradients Δh_{ij} appearing in Eq. (1). It has been shown[12,5] that classical thermal fluctuations affect the field distribution through a "Debye-Waller" factor. The same factor reduces the field gradient. In Fig. 3, we show this reduction of $1/T_{2,\parallel}$ in the superconducting state as a thin solid line. This result is not significantly different from the static case at low temperatures shown as a thick solid line in this figure. More generally, one might argue that the field gradients which appear in Eq. (1) are averaged by vortex fluctuations of large amplitude, similar to motional narrowing, thereby restoring the nuclear spin-spin interactions characteristic of the normal state. $1/T_{2,\parallel}$ at the anomalous peak is almost as large as in the normal state so that this averaging would have to be about 80% efficient having a concomitant effect on the NMR spectrum itself. Neither the shift in the spectrum nor the linewidth shows evidence for this. Consequently, large-amplitude vortex fluctuations and vortex diffusion appear to be unlikely causes for the observed peak in $1/T_2$.

The remaining possibility is that flux motion produces magnetic-field fluctuations, thereby contributing directly to the incoherent phase accumulation and thus $1/T_2$. Only fluctuations in the component of local field along the direction of the applied field are effective in producing dephasing. Furthermore, such field fluctuations contribute significantly to $1/T_2$ only when the fluctuation rate is close to $1/T_2^{\text{static}}$. If the fluctuations are too rapid, the nuclear spins merely experience the average of the local field and the fluctuations are ineffective for relaxation. If they are too slow, they contribute only to the static inhomogeneous field distribution but not to relaxation. If the characteristic vortex fluctuation time increases over a wide enough range with decreasing temperature, it will produce a peak in $1/T_2$. Below we discuss a simple model for vortex fluctuations which contains this essential character.

In order to estimate this effect we assume that the longitudinal field fluctuations have an exponential time correlation function with τ being the correlation time. The spectral density is then a Lorenztian function $f(\omega) = \overline{h^2}\tau/[1 + (\omega\tau)^2]$, where $\overline{h^2}$ is the mean square amplitude of the field fluctuations. It is reasonable to take the difference between the measured $1/T_2$ and $1/T_2^{\text{static}}$ to be given by $f(\omega \equiv 1/T_2^{\text{static}})$. The τ dependence of such a function has a peak at $1/\tau = \omega$. When the fluctuation time scale sweeps through T_2^{static}, a peak in the measured $1/T_2$ should be observed. For purposes of illustration we have calculated τ from our data holding $\overline{h^2}$ to be a constant and show these results in Fig. 4. The correlation time τ inferred from this procedure is a smoothly decreasing function of temperature from 6 to 60 K. With this interpretation there is no evidence of an abrupt change that might be associated with a first order thermodynamic flux phase transition in this material. However, this model for the field fluctuation spectrum is only qualitative. In particular, the τ values

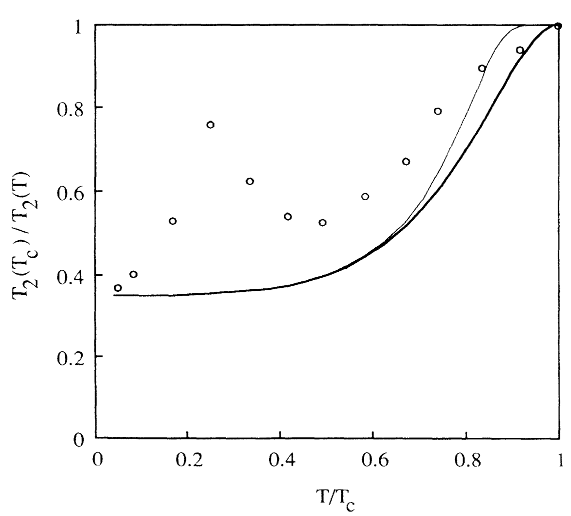

FIG. 3. Reduction of $1/T_{2,\parallel}$ in the superconducting state at field 3.46 T. The lines are calculations using Eq. (1) with (thin line) and without (thick line) fluctuation effects. Parameters are $\lambda_{ab}(0) = 1100$ Å and $T_c = 120$ K.

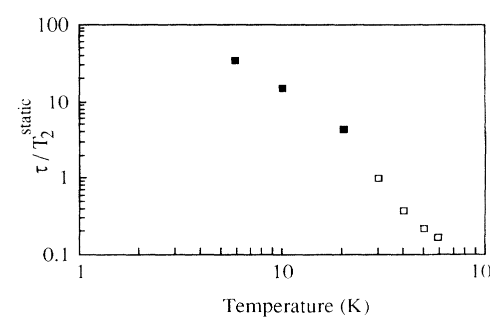

FIG. 4. Correlation time τ as a function of temperature derived from $1/T_2$ measurements. Definition of τ is discussed in text.

obtained for temperatures much higher than 30 K are less reliable partly due to the subtraction procedure in extracting the contribution from vortex dynamics where the bare spin-spin interaction is assumed to remain constant below T_c. If these factors are considered, it is likely that τ might be much smaller above 30 K. It was pointed out recently by Moonen, Reefman, and Brom[13] that at high temperature, vortex diffusion might dominate the vortex dynamics and the local vortex fluctuation model[5] might not be applicable.

It is worthwhile intercomparing the temperature dependence of the spectrum linewidth,[5,14] shift,[5] and $1/T_2$ data on the same material near 40 K. The shift data show a smooth temperature dependence. However, a change in the temperature dependence of the linewidth is observed at low temperature for both our powder sample[14] at 1.12 T and 3.46 T and the aligned sample[5] at 3.46 T. At low temperature, it was found that the linewidth grows approximately linearly as the temperature decreases for all cases. The temperature at which such changes in the temperature dependence occur is 30 K for the powder sample at 3.46 T, and 40 K for the aligned sample at 3.46 T,[5] 10 K higher than that of the $1/T_2$ peak. The local vortex fluctuation model was shown[5] to qualitatively account for this temperature dependence; however, at low temperature when the vortex correlation time is long, there will be additional broadening due to incomplete motional averaging.

A measurement of the magnetic irreversibility, defined to be the temperature when the departure of the field-cool and zero-field-cool magnetization occurs, was reported[15] and the irreversibility temperature was found to be around 20 K at 3.5 T. The irreversibility line is most often interpreted as the temperature below which flux pinning becomes effective. More generally, it signifies a slowing down of the vortex dynamics and the growth of the correlation time. This is more so when a smooth frequency dependence of the irreversibility line is observed. Our analysis of $1/T_2$ also shows a gradual growth of the vortex correlation time over the same temperature region and provides a picture consistent with earlier measurements of the irreversibility line. It is natural that the peak in $1/T_2$ occurs at a higher temperature than the irreversibility line[15] since our $1/T_2$ experiments are sensitive to dynamics at the higher frequency of order 10 kHz, in comparison with the low-frequency magnetization measurements.

In summary, the spin-spin relaxation experiment is unique in its sensitivity to the magnetic-field gradients on a microscopic length scale, and to the magnetic-field fluctuations arising from vortex dynamics. The experiments are conducted in thermal equilibrium in contrast to transport or torque experiments where the response to a driving force is measured. This spin-spin relaxation technique is specifically sensitive to the field fluctuation frequency which provides information related to the vortex-vortex coupling and the dissipation processes in the vortex core.

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[1] D. J. Bishop, P. L. Gammel, D. A. Huse, and C. A. Murry, Science **255**, 165 (1992).

[2] H. Safer *et al.*, Phys. Rev. Lett. **69**, 824 (1992); W. K. Kwok *et al.*, *ibid.* **69**, 3370 (1992).

[3] D. E. Farrell, J. P. Rice, and D. M. Ginzburg, Phys. Rev. Lett. **67**, 1165 (1991).

[4] M. P. A. Fisher, Phys. Rev. Lett. **62**, 1415 (1989); D. S. Fisher, M. P. A. Fisher, and D. A. Huse, Phys. Rev. B **43**, 130 (1991).

[5] Y.-Q. Song *et al.*, Phys. Rev. Lett. **70**, 3127 (1993).

[6] B. J. Suh, D. R. Torgeson, and F. Borsa, Phys. Rev. Lett. **71**, 3011 (1993); L. N. Bulaevskii, N. N. Kolesnikov, I. F. Schegolev, and O. M. Vyaselev, *ibid.* **71**, 1891 (1993).

[7] Y.-Q. Song *et al.*, Phys. Rev. B **45**, 4945 (1992); Y.-Q. Song *et al.*, *ibid.* **44**, 914 (1991).

[8] The contribution to $1/T_2$ from the spin-lattice relaxation is less than 10% in the normal state and much less in the superconducting state.

[9] C. P. Slichter, *Principles of Magnetic Resonance*, 3rd ed. (Springer-Verlag, New York, 1989).

[10] N. Bloembergen, S. Shapiro, P. S. Pershan, and J. O. Artman, Phys. Rev. **114**, 445 (1959); A. Abragam and W. G. Proctor, *ibid.* **109**, 1441 (1958); P. S. Pershan, *ibid.* **117**, 109 (1960); B. N. Provotorov, Sov. Phys. JETP **42**, 882 (1962).

[11] It is also possible that the spin-spin interactions are modified in the superconducting state; however, it is unlikely that it would have a nonmonotonic temperature dependence.

[12] E. H. Brandt, Phys. Rev. Lett. **66**, 3213 (1991).

[13] J. T. Moonen, D. Reefman, and H. B. Brom, Phys. Rev. Lett. **72**, 176 (1993); D. Reefman and H. B. Brom, Physica C **213**, 229 (1993).

[14] M. Lee *et al.*, Phys. Rev. B **40**, 817 (1989).

[15] V. K. Chan and S. H. Liou, Phys. Rev. B **45**, 5547 (1992).