Effect of Vortex Fluctuations on ^{205}Tl Spin-Lattice Relaxation in the Mixed State of $Tl_2Ba_2CuO_6$

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The spin-lattice relaxation rate W on ^{205}Tl has been measured in the mixed state of a $Tl_2Ba_2CuO_6$ aligned powder sample with $T_c = 100$ K. At $T < 25$ K, $W \propto T$ and decreases with increasing magnetic field. This behavior is explained as caused by fluctuations of pancake vortices; the latter induce a time dependent random magnetic field which causes the relaxation of nuclear spins.

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The study of the nuclear spin-lattice relaxation rate, W, in high-T_c superconductors at temperatures below T_c is considered as a method providing useful information about the nature of their superconducting state [1]. In particular, a power law temperature dependence of planar ^{63}Cu and ^{17}O relaxation rates in YBCO has been suggested as an evidence for a non-s-wave pairing [2].

In drawing this conclusion, however, one has to be certain that only the nuclear spin relaxation via conduction electrons is involved at $T < T_c$, whereas other possible relaxation mechanisms play minor roles. In particular, for the NMR measurements, complications may arise due to the presence of vortices [3].

In this Letter we present the NMR data on the temperature and field dependencies of the ^{205}Tl relaxation rate in $Tl_2Ba_2CuO_6$ and show that vortex fluctuations constitute a dominant mechanism of the low temperature nuclear spin relaxation in this highly anisotropic superconductor. We observed, for the first time, that at temperatures well below T_c, *W varies linearly with temperature and decreases with field.* Then we show how to separate vortex and quasiparticle contributions to the relaxation rate in the superconducting state. We also show that W measurements provide information about the dynamics of the vortex lattice excitations analogous to phonons in the crystal lattice.

A magnetically oriented powder sample sealed in Stycast epoxy has been used in the measurements. The powder was prepared by grinding five $Tl_2Ba_2CuO_6$ single crystals selected from the same batch and having T_c near 100 K with the transition width of a few K. The method of crystal growth is described elsewhere [4]; a melt with no Ca oxide has been used. The linewidth of the aligned powder sample coincides with that of a single crystal within $\approx 10\%$. The data in fields of 3.58 and 7.05 T were obtained with a BRUKER MSL-300 NMR pulse spectrometer by using a saturation-recovery procedure. The NMR linewidth does not enter in our measurements. Its value, Δf, is field, temperature, and orientation dependent, being maximum, 149 kHz, at $T = 4.2$ K, $B = 7.05$ T at $\mathbf{B} \perp c$. The $\pi/2$ pulse length, τ, was 1.2 to 1.5 μsec and strength, H_1, calculated from $\gamma_n H_1 \tau = \pi/2$, about 70 G. One sees that we were able to provide $\tau\Delta f \ll 1$ condition. A one-exponential fit for the area of the spin-echo time dependence was employed. The in-field data were taken with $\mathbf{B}$ perpendicular and parallel to the ab crystal layers; the orientation has been fixed according to the minimum and maximum frequencies of NMR lines [4]. Note that even when $\mathbf{B}$ was "parallel" to ab, there was always a component $\mathbf{B} \perp ab$ due to a misalignment of crystallites within the sample.

In Fig. 1(a) the relaxation rate W is presented as a

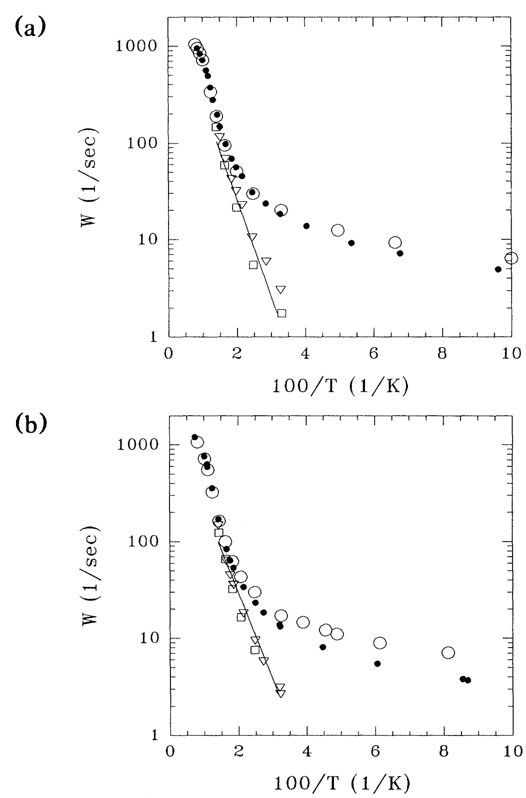

FIG. 1. The relaxation rate ^{205}W is shown as a function of $100/T$ for fields 3.58 T (open circles) and 7.05 T (black dots) for (a) $\mathbf{B} \perp ab$ and (b) $\mathbf{B} \parallel ab$. Open triangles (7.05 T) and squares (3.58 T) represent a quasiparticle contribution (see text).

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function of T for fields 7.05 and 3.58 T perpendicular to the layers. In a high temperature region, W is nearly field independent; a weak field dependence may be accounted for as arising from a T_c shift with the field B. At temperatures below 50 K, a stronger field dependence appears. As is seen from Fig. 1(b), the same W behavior is characteristic of the $\mathbf{B} \parallel ab$; the only difference is an almost nonshifted $T_c(B)$ and a stronger field effect at low temperatures.

The low temperature parts of W vs T dependencies are shown in Fig. 2 for fields perpendicular and parallel to layers. It is seen that below ≈ 25 K, $W = \alpha T$ with $\alpha_\perp = 0.49$ and 0.62, $\alpha_\parallel = 0.34$ and 0.56 $\text{sec}^{-1}\text{K}^{-1}$ for $B = 7.05$ and 3.58 T, respectively.

The observed field dependence excludes the spin diffusion mechanism of relaxation, i.e., the spin diffusion to the normal cores via cross relaxation. According to this mechanism the relaxation rate should increase with field due to increasing density of normal cores [3]. The relaxation due to quasiparticles is also expected to increase with field.

Vortices certainly present in the system and their thermally excited random deviations from the equilibrium positions provide some relaxation via induced alternating magnetic fields. In the following we use the term *vortons* for the excitations of the vortex lattice. Although similar to phonons, the vortons differ from the latter: they are overdamped oscillators [5] coupled to the magnetic field, whereas the phonons decay only via scattering processes and are coupled to the electric field [6]. In the following we show that the NMR relaxation due to alternating magnetic field induced by thermal vortons suffices to explain the value of the low temperature relaxation rate in $Tl_2Ba_2CuO_6$, as well as its T and B dependencies. Note that near T_c, in the liquid vortex phase, moving vortices can visit almost all nuclear sites and provide relaxation via normal electrons of their cores causing an increase of W. In the following we consider only the low temperature solid (vortex glass) phase, where diffusion of vortices is frozen.

We extrapolate the low temperature vortex contribution $W_v = \alpha T$ to higher temperature regions up to ≈ 50 K. The change in this power law in this temperature interval is unlikely because it is simply a consequence of the equipartition theorem. Subtracting this contribution from observed W values we obtain a quasiparticle contribution, W_q, shown in Figs. 1 and 2 by open triangles for $B = 3.58$ T and open squares for $B = 7.05$ T. We see that this contribution has a gaplike character $W_q \propto \exp(-\Delta/T)$ with $\Delta/T_c(H) \approx 3.6$ [7].

To calculate the vortons' contribution to the relaxation rate, W_v, at low temperatures we use a simple model. We assume (a) the electromagnetic approximation for the pancakes' interaction within the vortex lattice [8] (Josephson interlayer coupling is neglected; see discussion below); (b) the harmonic approximation for vortex distortions; (c) the overdamped oscillator model to describe the dynamics of the pancake made vortex lattice [9]; and (d) a dense vortex lattice, $H_{c1,\perp} \ll B_\perp \ll H_{c2,\perp}$, where $B_\perp$ is the component of the field perpendicular to the layers, $B_\perp/\Phi_0$ being the area density of the pancakes. For the case $\mathbf{B} \parallel ab$ this implies that the misalignment is not too small (it should be $\approx 6°$–$10°$).

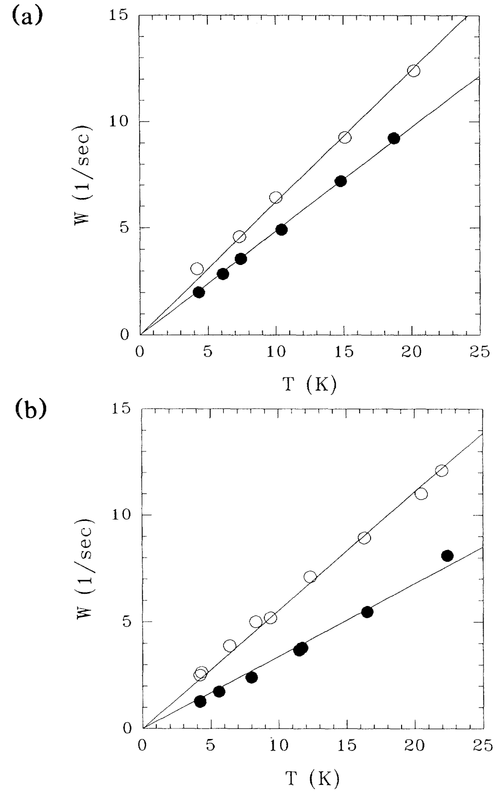

FIG. 2. Low temperature part of ^{205}W vs T dependence in fields 3.58 T (open circles) and 7.05 T (black dots) for (a) $\mathbf{B} \perp ab$ and (b) $\mathbf{B} \parallel ab$.

Let us consider first the case $\mathbf{B} \perp ab$ (along the z axis). In the absence of distortions, the pancakes are arranged in straight stacks along z, and an in-plane component of their field, $\mathbf{h} = (h_x, h_y)$, is absent. In the presence of thermal vortons, the time dependent component $\mathbf{h}(\mathbf{R}, t)$ gives rise to the relaxation of the longitudinal nuclear magnetization [here $\mathbf{R} = (\mathbf{r}, z)$, $\mathbf{r} = (x, y)$]. The relaxation rate for nuclear spin situated at $\mathbf{R}$ and precessing with frequency $\omega = \gamma_n B$ is given by the Fourier transform of the correlation function $K(\mathbf{R}, t)$:

$$W_v = \tfrac{1}{2}\gamma_n^2 K(\mathbf{R}, \omega), \tag{1}$$

$$K(\mathbf{R}, \omega) = \int_{-\infty}^{+\infty} dt \langle \mathbf{h}(\mathbf{R}, t) \cdot \mathbf{h}(\mathbf{R}, 0) \rangle e^{i\omega t},$$

where the brackets denote the thermal average. The coordinates of pancakes are $\mathbf{r}_{n\nu}(t) = [x_{n\nu}(t), y_{n\nu}(t)]$, where the integer n labels the layers and ν stands for pancake positions in a given layer. The vector potential for the field $\mathbf{h}(\mathbf{R}, t)$ is determined by

$$\text{curl curl } \mathbf{A} = -\frac{\Phi_0 s}{2\pi\lambda_{ab}^2}\sum_n \left(\nabla\phi_n + \frac{2\pi}{\Phi_0}\mathbf{A}\right)\delta(z - ns),$$
$$\phi_n = \sum_\nu \arctan\frac{x - x_{n\nu}}{y - y_{n\nu}}. \quad (2)$$

Here ϕ_n is the order parameter phase in the layer n and s is the interlayer spacing; for details see [10].

With the help of Eqs. (2) we obtain the field Fourier transform:

$$\mathbf{h}(\mathbf{p}, p_z) = -\sum_{n,\nu} \frac{\Phi_0 s \mathbf{p} p_z \exp[-i\mathbf{p}\cdot\mathbf{r}_{n\nu} - ip_z ns]}{[(p^2 + Q^2)^{-1} + \lambda_{ab}^2](p^2 + p_z^2)p^2}, \quad (3)$$

where the momentum $\mathbf{p}$ is in the x, y plane, p_z is along z, and $Q^2 = 2(1 - \cos p_z s)/s^2$. Assuming the distortions of vortices, $\mathbf{u}_{n\nu}(t) = \mathbf{r}_{n\nu}(t) - \mathbf{r}_\nu^0$, from the equilibrium positions $\mathbf{r}_\nu^0$ to be small at low T's, we obtain the correlation function averaged over the vortex lattice unit cell:

$$K(t) = \frac{B_\perp^2}{s^2\Phi_0^2}\sum_{\mathbf{G},\mathbf{k},q} \frac{q^2}{\{[(\mathbf{k}+\mathbf{G})^2 + Q^2]^{-2} + \lambda_{ab}^2\}^2[(\mathbf{k}+\mathbf{G})^2 + q^2]^2} \times \left\{\frac{[\mathbf{G}\times\mathbf{k}]^2}{(\mathbf{k}+\mathbf{G})^2 k^2}\langle u_{\rm tr}(\mathbf{k},q,t)u_{\rm tr}(\mathbf{k},q,0)\rangle + \langle u_l(\mathbf{k},q,t)u_l(\mathbf{k},q,0)\rangle\right\}. \quad (4)$$

Here, $\mathbf{G}$ form the reciprocal lattice, the summation over momentum $\mathbf{k}$ (in the ab plane) and q (along the c axis) is restricted by the first Brillouin zone, $u_{\rm tr}$ and u_l are the transverse and longitudinal amplitudes of the distortions $\mathbf{u}$, and

$$\mathbf{u}(\mathbf{k}, q, t) = \frac{s\Phi_0}{B}\sum_{n,\nu}\mathbf{u}_{n\nu}(t)\exp(i\mathbf{k}\cdot\mathbf{r}_\nu + iqn). \quad (5)$$

The function $K(t)$ determines the relaxation rate $\overline{W}_v$ of Tl nuclei averaged over their positions in a vortex unit cell. $\overline{W}_v$ reproduces correctly the relaxation rate at least for the initial decay of nuclei magnetization.

Within the harmonic approach, the free energy functional for distortions reads

$$\mathcal{F}\{\mathbf{u}(\mathbf{k}, q)\} = \frac{B_\perp}{2s\Phi_0}\sum_{\mathbf{k},q}[\epsilon_l(k,q)|u_l(k,q)|^2 + \epsilon_{\rm tr}(k,q)|u_{\rm tr}(k,q)|^2], \quad (6)$$
$$\epsilon_l(k,q) = c_{11}k^2 + c_{44}Q^2, \qquad \epsilon_{\rm tr}(k,q) = c_{66}k^2 + c_{44}Q^2,$$

where c_{66}, c_{11}, and c_{44} are the flux lattice shear, compression, and tilt moduli. In the electromagnetic model, the moduli are given by [11]

$$c_{66} = \frac{B_\perp\Phi_0}{64\pi^2\lambda_{ab}^2}, \quad c_{11} = \frac{B_\perp^2(k^2 + Q^2)}{4\pi k^2[1 + \lambda_{ab}^2(k^2 + Q^2)]},$$
$$c_{44} = \frac{B_\perp\Phi_0}{32\pi^2\lambda_{ab}^4 Q^2}\ln\left(1 + \frac{\Phi_0 Q^2}{4\pi B_\perp}\right). \quad (7)$$

Using the Langevin equations for the overdamped motion of pancakes, we find the distortions correlation function:

$$\langle u_{{\rm tr},l}(\mathbf{k},q,t)u_{{\rm tr},l}(\mathbf{k},q,0)\rangle = \frac{Ts\Phi_0^2}{B_\perp^2\Gamma_{{\rm tr},l}(k,q)\eta}\exp[-\Gamma_{{\rm tr},l}(k,q)t], \quad (8)$$

where $\Gamma_{{\rm tr},l}(k,q) = \Phi_0\epsilon_{{\rm tr},l}(k,q)/B_\perp\eta$ are vorton frequencies. Commonly, η is estimated using Bardeen-Stephen's $\eta = \Phi_0^2\sigma_n/2\pi\xi_{ab}^2c^2$, where σ_n is the normal state conductivity. We note, however, that in the high-T_c superconductors at low T this may not be the case due to the very small radius of vortex core ξ_{ab}.

The characteristic vorton frequencies in the problem are the maximum frequency of the shear vortons ω_s (at $q = 0$ and $k = K_0$, where $K_0^2 \approx 4\pi B_\perp/\Phi_0$) and the maximum frequency of the tilt vortons ω_t (at $k = 0$ and $q = \pi/s$):

$$\omega_s = \frac{\Phi_0 c_{66}K_0^2}{B\eta} \approx \frac{B_\perp\Phi_0}{16\pi\lambda_{ab}^2\eta}, \quad (9)$$

$$\omega_t = \frac{\Phi_0 c_{44}Q^2}{B\eta} \approx \frac{\Phi_0^2}{32\pi\lambda_{ab}^4\eta}. \quad (10)$$

In the highly anisotropic superconductors we have $c_{44} \ll c_{66}$, and $\omega_t \ll \omega_s$ for large k, q. The main contribution to $\overline{W}_v$ at $\omega \ll \omega_s$ comes from transverse distortions because $c_{66} \ll c_{11}$ [only the small k's and large q's are relevant in the summation over $\mathbf{k}, q$ in Eq. (4)]. We substitute Eqs. (6)–(8) in Eq. (4) and replace the sum over $\mathbf{G}$ by an integral with the lower limit $\approx K_0$ and the upper one $\approx 1/\xi_{ab}$, where the correlation length ξ_{ab} is slightly larger than s. We obtain

$$\overline{W}_v \approx \frac{\gamma_n^2 T}{2\xi_{ab}\lambda_{ab}^2\omega}\tan^{-1}\frac{32\pi^2\lambda_{ab}^4\omega\eta}{\Phi_0^2\ln(\Phi_0/\xi_{ab}^2B_\perp)}, \quad \omega \ll \omega_s, \quad (11)$$

$$\overline{W}_v \approx \frac{\gamma_n^2 T\Phi_0 B_\perp}{16\pi\xi_{ab}\lambda_{ab}^4\omega^2\eta}, \quad \omega \gg \omega_s. \quad (12)$$

Note that $\omega = \gamma_n B_\perp$ for $\mathbf{B} \perp ab$. Thus, at low frequencies, $\omega \ll \omega_t$, in the three-dimensional (3D) regime of pancake coupling, $\overline{W}_v$ is nearly field independent (the B dependence is logarithmic). The crossover 2D–3D occurs when ω passes through ω_t. In the 2D regime $\overline{W}_v$ decreases as $1/B$, with different coefficients for $\omega \ll \omega_s$ and for $\omega \gg \omega_s$. The frequency dependence of the relaxation rate at a given $B_\perp$ reproduces the vortons density of states weighted with a factor describing their interaction with the transverse magnetic field. This interaction is strong for the field components $\mathbf{h}(\mathbf{p}, p_z)$ with large p_z.

We consider now the case $\mathbf{B} \parallel \mathbf{a}$ taking into account the misalignment of crystallites. We use the same Eqs. (11) and (12) for $\overline{W}_v$, but now $B_\perp$ depends on the misalignment angle, $B_\perp \ll B$, while ω is determined by the total field, $\omega = \gamma_n B$. The components $h_y(t)$ (parallel to the b axis) and $h_z(t)$ give rise to the relaxation (they are transverse to $\mathbf{B}$). The effect of h_z is approximately the same as that of $h_y(t)$. As a result, for $\omega \ll \omega_s$, the relaxation rates for $\mathbf{B} \parallel \mathbf{a}$ and for $\mathbf{B} \perp ab$ are approximately the same because $\overline{W}_v$ depends very weakly on $B_\perp$. For $\omega \gg \omega_s$, these two become different due to the explicit dependence of $\overline{W}_v$ on $B_\perp$.

One can understand the data if we assume that ω lies in the interval $\omega_t \ll \omega \ll \omega_s$. This implies that $\eta > 10^{-6}$ g/cm^2 sec, i.e., $\sigma_n > 10^4$ ohm^{-1} cm^{-1} if we use the Bardeen-Stephen η. Then $\overline{W}_v \propto 1/B$ for any orientation of the applied field. The data for $\mathbf{B} \parallel ab$ are in accordance with this dependence. There are deviations from $\overline{W}_v \propto 1/B$ for $\mathbf{B} \perp ab$ (see the α values above). Taking $\lambda_{ab} = 1700$ Å [12], $\xi_{ab} \approx 20$ Å, and $s = 11.7$ Å (the distance between CuO_2 planes for $Tl_2Ba_2CuO_6$), we obtain $\alpha_\perp = \overline{W}_v/T \approx 0.44$ sec^{-1}K^{-1} in the field 7.05 T in agreement with the experimental value 0.49 sec^{-1}K^{-1}.

In the above calculations, we have ignored the Josephson interlayer coupling and pinning.

The model without the Josephson coupling is valid under the condition that the anisotropy parameter $\gamma \geq \lambda_{ab}/s$ [see [11(b)]], which may be fulfilled in Bi- and Tl-based superconductors [13]. As γ decreases, c_{44} and ω_t increase. Then the interval of 3D behavior at low ω, where $\overline{W}_v$ depends weakly on ω, becomes broader. The large anisotropy is a necessary condition for the high relaxation rate W_v as well as for the existence of the 2D region where $\overline{W}_v \propto 1/\omega$. This could be the reason why W increases with B in more three-dimensional YBCO [2], opposite to what is observed in our case.

Pinning can be accounted for by taking $1/L$ as the lower limit of integration over k in Eq. (4), where L is the correlation length of pancake ordering in the ab plane. For $\overline{W}_v(\omega)$ pinning is important if pancakes are disordered on distances smaller than $L \approx \Phi_0/8\pi\lambda_{ab}(\omega\eta)^{1/2} \approx 2000$ Å.

In conclusion, we have measured the relaxation rate W in $Tl_2Ba_2CuO_6$ and explained the low temperature results within the model of fluctuating vortices. The model correctly reproduces the temperature dependence of the relaxation rate, its value, and, qualitatively, its field dependence.

We note also that the fluctuation mechanism of relaxation should be taken into account before and when extracting the contribution of quasiparticles to the relaxation rate from the NMR data in highly anisotropic superconductors. We have shown that the account of vortex fluctuations in extracting quasiparticle $W_q(T)$ at $T < 50$ K from experimental data in $Tl_2Ba_2CuO_6$ changes the result from a nonexponential dependence $W_q(T)$ to the exponential gaplike one. The importance of vortex fluctuations for the interpretation of the NMR experiments has recently been discussed by Song *et al.* [14].

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