Local vortex mobility below the irreversibility line of $Tl_2Ba_2CaCu_2O_8$: A ^{205}Tl NMR study of the transverse relaxation in single crystals

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We performed ^{205}Tl NMR on aligned single crystals of the 105-K superconductor $Tl_2Ba_2CaCu_2O_8$ in a field of 4.7 T as a function of angle θ between the field direction and the c axis. Irreversibility temperatures T_{irr} were determined by comparing the NMR linewidths after field cooling and zero-field cooling. The ^{205}Tl nuclear transverse relaxation rate T_2^{-1} shows a well-developed peak caused by vortex motion below T_{irr}. The peak shifts to higher temperatures with increasing θ. The data are analyzed analytically and by computer simulation and the obtained parameters of local vortex motion are discussed. [S0163-1829(97)50914-3]

A characteristic feature of the phase diagram of high-T_c superconductors as a function of magnetic-field B_0 and temperature T is the presence of an extensive vortex fluid phase, that starts just below T_c, and a vortex glass phase at low temperatures.[1,2] The transition from vortex fluid to vortex glass state has been studied intensively mainly by macroscopic methods (linear and nonlinear conductivity and magnetic measurements). Scaling theories for the critical slowing down of the vortex motion at this transition have been developed and applied to the results of these experiments.[1,2] Nuclear magnetic resonance (NMR) is a powerful tool to study local magnetic-field fluctuations caused by vortex motion — by the inherent short length scales, these fluctuations are not described by the renormalization theories. Various NMR properties are suited. For example, vortices give an inhomogeneous broadening of the nuclear resonance line, which effect is reduced by thermally activated vortex motion and the corresponding correlation time can be measured by NMR.[3–7] The fluctuating magnetic fields caused by vortex motion can also be studied by means of the nuclear relaxation.[7–13] The nuclear-spin-lattice relaxation T_1 is sensitive to transverse field fluctuations near the Larmor frequency $\omega_0 = \gamma_n B_0$. Recently a peak in $T_1(T)$ has been observed by ^{199}Hg NMR in $HgBa_2CuO_{4+\delta}$ (T_c = 96 K) at the irreversibility temperature, T_{irr}.[8] The transverse relaxation or spin-echo decay time T_2 is sensitive to motions that are typically a few orders of magnitude slower than seen in T_1, and the field fluctuations that cause the relaxation are along the B_0 direction. T_2^{-1} was studied in oriented powder of $YBa_2Cu_3O_{7-\delta}$ where the ^{89}Y transverse relaxation rate $T_2^{-1}(T)$ shows a peak below T_c.[9] The peak was analyzed in terms of fluctuations in the vortex field gradients. In oriented powder $Tl_2Ba_2Ca_2Cu_3O_{10}$ (T_c = 120 K) by NMR on the thallium nuclei located in the TlO layers, a similar peak was observed at 35 K and explained as the crossover of time scales with and without vortex motion.[12] In this paper we present data on aligned crystals of $Tl_2Ba_2CaCu_2O_8$ (T_c = 105 K), that give the precise location of the peak with respect to the irreversibility line and show its variation in location and height as a function of field along the c axis. The effect of vortex field fluctuations on the nuclear transverse relaxation is calculated.

All experiments have been performed on the same c-axis-aligned $Tl_2Ba_2CaCu_2O_8$ single crystals as used in a previous study on vortex motion in the vortex fluid state.[7] The compound has two types of thallium positions: the ''normal'' Tl(N) site in TlO double layers and the ''impurity'' Tl(I) site resulting from a partial replacement of Ca atoms by Tl (near 10%).[3,7,14] The average distance between Tl(I) and Tl(I) in the Ca layer is about 12 Å, compared to 3.8 Å between Tl(N) and Tl(N). Due to the large distance between Tl(I)'s, $T_2(I) > T_2(N)$ (see below). It makes the ''impurity'' site very convenient for the fluctuating field study. T_2 values were measured by means of the standard two pulse echo sequence at a field of 4.7 T. At high temperatures the Tl(N) and Tl(I) lines in this compound are well separated (>100 kHz) at all angles θ (the angle between the field direction and the crystallographic c axis), and the relaxation rates T_2 of the N and I line can be measured independently. Below 40 K for $\theta = 0$, the Tl(I) line shift is such that the two lines merge.[7] Because T_2(I) is much longer than T_2(N), separation of the two contributions remains possible.

The T dependences of T_2^{-1} for Tl(I) at four θ values are shown in Fig. 1. The peak in the relaxation rate at T_{peak} is seen to move to higher temperatures with increasing angle. For the Tl(N) site there is a similar peak near the same temperature although scatter in this case is larger due to the smaller absolute T_2 values and the larger influence of other relaxation processes. The inset shows this peak at $\theta = 45°$.

T_2 relaxation is usually caused by (in)direct dipolar coupling. Using appropriate published values for the Cu-relaxation rate[15,16] we find a maximum in the T_2 rate of 4 ms^{-1} around 150 K (for $\theta = 45°$) due to the Tl(I)–Cu dipolar interaction,[17] a value that is indeed observed experimentally; the next important interaction, Tl(I)–Tl(I), gives about 2.8 ms^{-1}. Below 150 K the Cu–Tl(I) T_2^{-1} is calculated

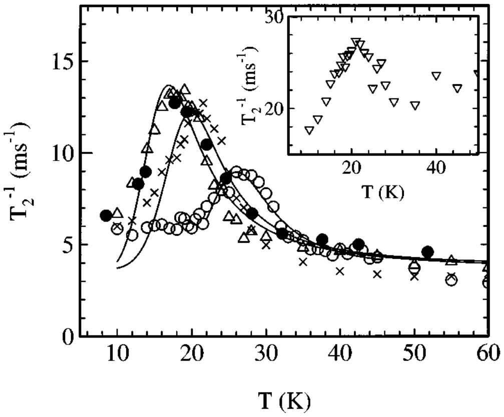

FIG. 1. T_2^{-1}(I) vs T at various θ. ●—0°, △—45°, ×—67°, ○—90°. The inset shows the $\theta=45°$ data for the normal line.

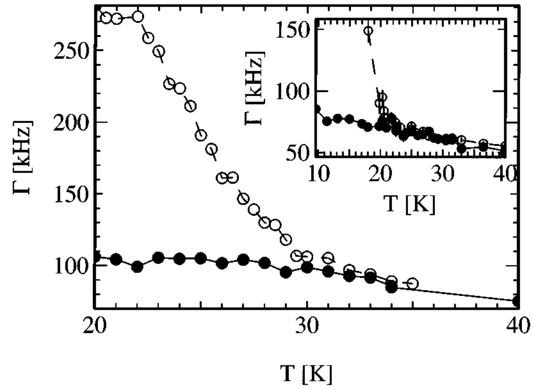

FIG. 2. The ^{205}Tl(N) linewidth Γ as a function of temperature for $\theta=90°$: ●—field cooled, and ○—zero-field cooled. The inset shows Tl(I) linewidth for $\theta=45°$.

to decrease smoothly with T to become of the order of a ms^{-1} or less below 50 K. It is clearly not the reason for the observed relaxation rate maxima. Also, the effect of the partial line merging of the Tl(N) and Tl(I) line at low temperatures, which in principle can give a maximum in T_2^{-1}, can be ruled out as line merging only occurs for $\theta\approx0°$ and the calculated effect is too small.

The only reasonable possibility left is to interpret the T_2^{-1} peak as a result of fluctuating fields caused by vortex motion. This interpretation is supported by the correlation of the field dependence of $T_{\rm peak}$ and the irreversibility temperature $T_{\rm irr}$. To determine $T_{\rm irr}$ for our sample we have measured the ^{205}TL line width at 4.7 T after zero field cooling (ZFC) and field cooling (FC). Figure 2 shows the result of this experiment.[18] At $T<T_{\rm irr}$ there is an additional broadening of the ZFC line,[19,20] because macroscopic field gradients exist inside the sample (the same mechanism causes the differences between ZFC and FC susceptibility data). At $T\geqslant T_{\rm irr}$, ZFC and FC line widths are equal since at these temperatures there is no pinning and hence no field gradient after ZFC. The obtained values of $T_{\rm irr}$ are 23±2 K at $\theta=45°$ and 33±2 K at θ near 90° and have to be compared with the T_2^{-1} peak positions at 18 and 28 K, respectively. According to magnetization measurements[21,22] on $Tl_2Ba_2CaCu_2O_8$ single crystals, the irreversibility temperature is near 20–25 K at 5 T ($\theta=0$) and increases as the field decreases. The increase of $T_{\rm irr}$ and $T_{\rm peak}$ with θ can analogously be explained, as it is the field component parallel to the c axis given by $B_0\cos\theta$, which is the most important in the formation of the vortex lattice in this strongly anisotropic system. The location of the T_2 peak at 90±5° is strongly influenced by the misalignment.

Intuitively it is clear that longitudinal field fluctuations may result in a peak in the spin-echo decay rate: very slow fluctuations have no effect on T_2 and rapid fluctuations also have no effect because in this case nuclear spins feel only a time-averaged field. In the intermediate region the dephasing effect of fluctuations on the echo formation should be maximal. Thus, if the correlation time of field fluctuations decreases with increasing temperature (e.g., with an Arrhenius law), it leads to a peak in $T_2^{-1}(T)$. What is the characteristic time or frequency connected with this peak? Let us consider the simple case of random-field fluctuations between $+b_z$ and $-b_z$ at every nuclear site with a correlation time τ_c. This means an exponential form $\exp(-t/\tau_c)$ for the time correlation function and hence a Lorentzian function for the fluctuations spectral density $f(\omega)\propto b_z^2\tau_c/(1+\omega^2\tau_c^2)$. For the explanation of the observed T_2^{-1} peak at 35 K in $Tl_2Ba_2Ca_2Cu_3O_{10}$, Song *et al.*[12] assumed that a spin-echo decay experiment probes this function near the frequency $\omega_{\rm peak}=T_{2\rm static}^{-1}$ where $T_{2\rm static}$ is the transverse relaxation time in the absence of field fluctuations. For two nonequivalent Tl nuclei with different $T_{2\rm static}$, the peak amplitude should then be proportional to the $\tau_{c\ \rm peak}=T_{2\rm static}$. As can be seen from Fig. 1 and its inset, the peak positions and amplitudes are the same for both thallium sites, while $T_{2\rm static}$ for the N line is much smaller than for the I line. It shows the proposed model to be incorrect.

In the following we will improve the analysis and present a numerical approach for independently or collectively moving vortices in a two-dimensional lattice. Before doing so, we consider the analytical result for a Gaussian frequency distribution of field fluctuations of width $\omega_p=\gamma_n b_z$,[24] which is a good approximation of the effects of fluctuation vortex fields on T_2. An evaluation of the magnetization decays shows a peak in the T_2 relaxation rate for $\gamma b_z\approx\tau_c^{-1}$, where τ_c is the time constant of the exponential decay of the correlation function of the fluctuations. The time dependence of the echo decay changes from single exponential at the high T side of the peak to $\exp(-t^3)$ at low temperatures. A similar expression is obtained, if the fluctuating field is supposed to take only two values: $+b_z$ and $-b_z$ with a correlation time τ_c.[23] The approach covers the same physics, but is more simple and leads to exponential decays of the magnetization in the short and long time limit. Between these limits the decay is not single exponential and the T_2 values (obtained by single exponential fits) are less accurate. The relaxation rates found in this way are well described by[23]

$$T_2^{-1}=\frac{\gamma_n^2 b_z^2\tau_c}{2+(\gamma_n b_z\tau_c)^2}+T_{2\rm static}^{-1}, \tag{1}$$

where we have added the parallel process $T_{2\rm static}^{-1}$. Equation (1) shows that fluctuations with a correlation time τ_c of the

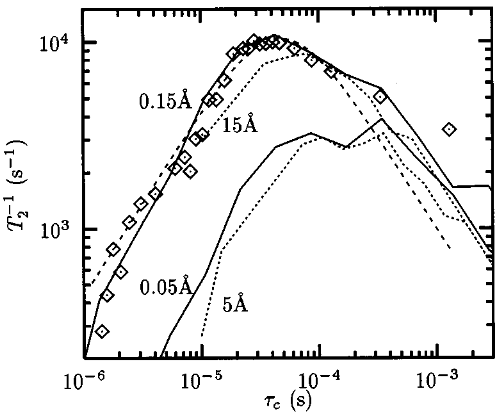

FIG. 3. T_2^{-1} vs τ_c for the independent (drawn) and collective (dotted) simulations. The diamonds are the experimental data points for $\theta=45°$ (see text), along with the fit based on Eq. (1).

order of the Larmor period $\gamma_n b_z$ (so that the nuclear spins make about one turn in the field b_z between two field hops) are the most effective.

The solid lines in Fig. 1 show the result of a fit to the data with Eq. (1) and the assumption of an Arrhenius law $\tau_c=\tau_0\exp(U/T)$ for the T dependence of τ_c with U an activation energy (in T units). In view of the simplicity of the model it is no surprise that the fit is not perfect, especially in the low temperature part of the peaks. The resulting values for $\theta\leqslant45°$ are $b_z=$ 0.2 mT and $U=80$ K. The amplitude of field fluctuations b_z is proportional to the peak amplitude and decreases at $\theta>45°$ while the activation energy U increases. At θ near 90°, $b_z=$ 0.1 mT and $U=180$ K. The prefactor τ_0 is of the order of 10^{-7}s at all θ. The correlation time τ_c at $T_{\mathrm{peak}}=18$ K equals 50 μs at $\theta\leqslant45°$.

The obtained values of b_z are much smaller than the average field inhomogeneity in the vortex state $\langle\Delta B^2\rangle^{1/2}=0.0609\Phi_0/\lambda_{ab}^2=9$ mT (λ_{ab} = 1250 Å). The average value of the field gradient in the vortex lattice may be evaluated as $G\approx\langle\Delta B^2\rangle^{1/2}/a_0=$ 0.5 G/Å ($a_0=$ 200 Å is the average intervortex spacing at 4.7 T). Thus, the amplitude of the corresponding displacement of the vortex lattice at T_{peak} is $b_z/G=4$ Å. This value will become a few times larger, if corrected for the much less effective displacements perpendicular to the field gradient and for the field gradient, which for the majority of the nuclei is overestimated.

For a more accurate calculation of the average displacement $\langle\Delta u^2\rangle^{1/2}$ of independently or collectively moving vortices from their equilibrium positions, we performed the following numerical analysis. Starting with a triangular lattice of vortices, we allow the vortex positions to fluctuate (independently or collectively) with a correlation time τ_c, and an average displacement $\langle\Delta u^2\rangle^{1/2}$. The thallium atoms, randomly distributed around the sample, feel a magnetic field $b(r)=\phi_0/\pi\lambda^2(\pi\lambda/2r)^{1/2}e^{-r/\lambda}$,[25] summed over the vortices. We now calculate the echo intensity after an echo pulse sequence for a series of times between pulses τ_c and average displacements, and fit the results with $m(t)=\exp(-t/T_2)$. The resulting relaxation rates are shown in Fig. 3. The experimental T_2 data [Tl(I) line, $\theta=45°$] are also shown after subtraction of $T_{2\mathrm{static}}^{-1}=3$ ms^{-1}. It is striking that we could use the same parameters as found from the fit in Fig. 1 $\tau_0=4.1\times10^{-7}$ s, $U=80.5$ K to convert the temperature scale to a τ_c scale on the x axis. As it is seen, within this model the average displacement of the independent vortices is about 0.2 Å. The value for the collective case is two orders larger, 15 Å [and similar to the prediction based on Eq.(1)]. These differences are as expected, because independently moving vortices produce higher fields as the single vortex field gradient is much larger than the average gradient.

T_{irr} is closely related to the melting and the depinning temperature. If one uses the Lindemann criterion for melting, only random hopping of independent vortices is considered. The vibration amplitude at T_{irr} obtained from this criterion is $\langle u^2\rangle^{1/2}\approx0.1a_0=20$ Å, much larger than in the corresponding simulation (0.2 Å). If we interpret T_{irr} as the depinning temperature, bundles instead of independent vortices may move over distances of the order of the correlation length $\xi=25$ Å. The 15 Å from the collective numerical analysis is a representation for this kind of motion. As the T_{peak} occurs just below T_{irr}, the expected vortex displacement ($\sim$20 Å) is in accordance with the collective model.

A feature of the simulation for the independent vortices, which is also seen in the experimental data, is the low slope at the low-temperature side of the peak, in comparison with the slope predicted by Eq. (1) (see also the long-dashed line in Fig. 3). This is related to a high-frequency tail of the fluctuation spectral density probed by the Tl nuclei. This spectrum can be thought of as consisting of a low- and high-frequency part. The low-frequency part arises from the nearby vortices and is characterized by a high b_z and an average time between hops of τ_c divided by the number of vortices within a radius of the order of λ. The high-frequency part is produced by many vortices further away, which produce a small high-frequency field at the nucleus. With only the nearest neighbors, we would have a relaxation behavior described by formula (1). The high-frequency (small τ_c) components produce additional relaxation in the low-temperature part, as can be seen in Fig. 3. Similar effects will occur in the case of flux bundles, if in the simulation the effects of independent bundles are incorporated.

As the echo decay at both high and low temperatures is dominated by another (dipolar) relaxation channel, we expect (and observe) an $\exp(t/T_2)^2$ time dependence. The decay shape around the peak will be determined by vortex fluctuations. Both simulation models predict that on the high-temperature side (small τ), the relaxation will be single exponential. In the low-temperature limit the collective hopping model shows again a single exponential behavior, while for the independent hopping model the low-temperature decay curves are better approximated by $\exp(t/T_2)^2$ (as for dipolar relaxation). At the peak the echo decays experimentally with an intermediate exponent as $\exp(t/T_2)^{1.4}$.

The evaluated activation energy U is comparable to values reported by Suh *et al.*[8] using the T_1 peak in $HgBa_2CuO_{4+\delta}$. The low limit of the typical U values obtained in magnetization relaxation measurements is also near 100 K,[26] though in these experiments a large current density j is present.[27] As NMR experiments deal with local-field fluctuations, thermally activated motion involves small jumping volumes and is determined by the short-range potential structure. In this sense our NMR experiments are indeed more analogous to large current transport or magnetic relaxation experiments where the jumping volume is also small.

In summary, the T_2^{-1} peak observed in high-T_c compound $Tl_2Ba_2CaCu_2O_8$ is accurately described by Eq. (1) and computer simulations. It shows the presence of field fluctuations by vortex bundles below T_{irr}. The observed vortex motion is relatively slow and short ranged and characterized by an activation energy of the order of 100 K. The computer simulation in addition shows that below T_{peak} the high-frequency part of the spectral density can have more weight than in the Lorentzian case, which explains the asymmetry seen in the relaxation peak.

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[18] The FC linewidth shown in Fig. 2 is smaller than the values in Ref. 7 because here the frequency sweep measurements were made without the readjustment of the resonance circuit. This procedure gives a more precise determination of relative changes in the linewidth, as wanted here, but is not suited for absolute values.

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