Thermal Depinning of Flux Lines in $HgBa_2CuO_{4+\delta}$ from ^{199}Hg Spin-Lattice Relaxation

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^{199}Hg NMR spin-lattice relaxation rates in an aligned powder sample of the anisotropic high-T_c superconductor $HgBa_2CuO_{4+\delta}$ ($T_c = 96$ K) show anisotropy in the temperature interval 10–50 K, with an extra contribution present for the external magnetic field parallel to the c axis. The extra contribution shifts with magnetic field in a way related to the irreversibility temperature T_{irr}, indicating that the thermal motion of flux lines (FL's) is the cause of the extra relaxation. A simple model is presented to analyze the relaxation data and to extract values for the correlation time τ_c and the effective activation energy U of the FL motion.

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When a flux line (FL) in type II superconductors moves in the lattice, the surrounding nuclei are subject to a modulation of the local internal magnetic field which can affect the NMR parameters in various ways. While the applications of NMR to conventional type II superconductors have been limited [1,2], in high-T_c superconductors (HTSC's) NMR studies of FL dynamics appear more promising. The random thermal motion of the FL's has been detected through the anomalous narrowing of the inhomogeneously broadened NMR line [3–5] and from the recovery of the transverse nuclear magnetization in a spin-echo experiment [6,7]. Also the motion of vortices driven by an electric current or by a change in the applied dc magnetic field has been detected through linewidth [8] and spin-echo measurements [9,10].

Regarding the nuclear spin-lattice relaxation (NSLR, T_1^{-1} or R_1), unambiguous evidence for a direct contribution due to thermally fluctuating FL's has been obtained only in an organic superconductor [11]. For HTSC the evidence remains highly uncertain after a characteristic T and H dependence of the ^{205}Tl NSLR in $Tl_2Ba_2CuO_4$, initially attributed to FL motion [12], was shown to be present also above the superconducting transition temperature T_c [13].

The contribution to R_1 due to FL motion should be particularly prominent just above the irreversibility temperature T_{irr}, where the effective correlation time τ_c of the FL motion crosses, as a function of temperature, the value of the reciprocal Larmor frequency ω_L, i.e., $\tau_c\omega_L \approx 1$, for which a maximum in R_1 can be expected. The second condition to be met is that the NSLR of the nucleus investigated should not be dominated by other sources of relaxation.

In this Letter, we report ^{199}Hg nuclear spin-lattice relaxation rate (T_1^{-1}, R_1) studies in an oriented powder sample of $HgBa_2CuO_{4+\delta}$ ($T_c = 96$ K) for temperatures $10 \leq T \leq 300$ K and static magnetic fields $H = 5.9$, 8.2, and 9.4 T. We have observed a strongly field-dependent anisotropy of R_1 present only in a temperature interval of 10–15 K around the irreversibility line measured in the same sample used for NMR. The anisotropy can be attributed to the contribution to the NSLR from thermally fluctuating FL's present only when the external magnetic field $\boldsymbol{H}$ is perpendicular to the CuO_2 plane. We have fitted the data with an expression for R_1 derived for a simple model of FL pseudodiffusive motion under a restoring force. The effective correlation time τ_c of the excitation at the wavelength of the order of the intervortex distance obtained from the fit, is thermally activated with an effective activation energy, $U \propto 1/H$.

The measurements were performed in a single-phase powder sample with average grain size 12–20 μm. The grains were embedded in epoxy and oriented in a magnetic field to achieve the alignment of the grains along a common crystal c axis. X-ray diffraction and dc magnetization were measured to test the sample alignment, to determine the sample quality, and to measure $T_c = 96$ K. Details are given elsewhere [14]. The irreversibility temperature shown in Fig. 1 was measured as a function of the applied field up to 5.5 T using a SQUID magnetometer. T_{irr} was chosen as the temperature at which the

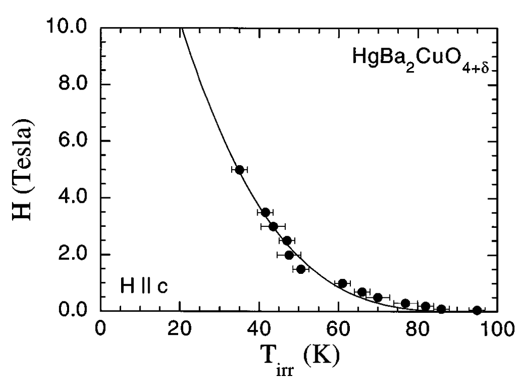

FIG. 1. The irreversibility line T_{irr} measured in oriented $HgBa_2CuO_{4+\delta}$ powder for $\boldsymbol{H} \parallel \boldsymbol{c}$. The solid curve is a fit by $H = 22[1 - T_{irr}(\mathrm{K})/96]^{3.3}$ T.

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field-cooled and zero-field-cooled magnetizations depart from each other.

The ^{199}Hg ($I = \frac{1}{2}$) NMR measurements were performed with Fourier transform (FT) spectrometers at 5.9, 8.2, and 9.4 T. Typical $\pi/2$ radio frequency pulses were 4 μsec, corresponding to $H_1 \cong 80$ G, sufficient to irradiate the whole NMR line with the exception of the lowest temperatures ($T < 20$ K). From an inversion recovery sequence, π-$\pi/2$, a single exponential recovery curve was obtained yielding R_1. Only at the lowest temperature investigated ($T \approx 8$ K) was a departure from single-exponential recovery observed at short times, which could be ascribed to incomplete saturation of the line and/or to diffusion-limited relaxation from the core of the fluxons. Some of the NMR measurements were performed with weaker radio frequency fields, $H_1 \sim 30$ G, using a long sequence of saturating pulses. The results of R_1 versus temperature at three different fields are shown in Figs. 2 and 3. Above T_c the relaxation rate is field and orientation independent, and R_1 follows a Korringa-type behavior: $T_1 T =$ const. Below T_c, R_1 remains isotropic except for the temperature interval 10–15 K where an extra contribution appears to be present for $\boldsymbol{H} \parallel \boldsymbol{c}$ (see the inset of Fig. 2).

The extra contribution has a maximum which shifts in temperature with a change of the applied magnetic field (see Fig. 3). The temperature- and field-dependent anisotropy of R_1 observed here are quite different from the ones observed in $YBa_2Cu_3O_7$ [15] and $YBa_2Cu_4O_8$ [16] for the R_1 due to the interaction with the Fermi liquid. Moreover, our results are inconsistent with a contribution due to fast relaxation within the vortex core nuclei in the superconducting region [1]. In fact, in the rapid diffusion limit one has [17] $R_{1s} = R_{1n} H \xi^2/\Phi_0$ where R_{1s} and R_{1n} are the NSLR in the superconducting and normal phases, respectively. With $\Phi_0 = 2.07 \times 10^{-7}$ G cm^2, $\xi_{ab} \cong 20$ Å, and $R_{1n} = 0.1T$ sec^{-1} (from Fig. 2) one has $R_{1s} \cong 2 \times 10^{-8} HT$ sec^{-1}. This estimate is of the wrong order of magnitude and predicts a H and T dependence which was not observed here. Also this contribution would give rise to a nonexponential recovery of the nuclear magnetization if the spin diffusion is not sufficiently rapid contrary to our observation of exponential recovery in the temperature range of interest ($T > 10$ K).

Instead, our results are quite consistent with a direct contribution to relaxation due to FL random thermal motion. The anisotropy is due to the fact, already observed in organic superconductors [11], that for $\boldsymbol{H} \perp \boldsymbol{c}$ and with a coherence length ξ_c of the order of the interplane spacing s in a strongly anisotropic superconductor, the FL's are self-trapped between the superconducting planes and thus do not contribute to the NMR relaxation. Furthermore, the FL contribution to $R_1(T_1^{-1})$ for $\boldsymbol{H} \parallel \boldsymbol{c}$ shifts with $\boldsymbol{H}$ in the same direction as the irreversibility temperature (see Figs. 1 and 3). It is noted that the maximum observed in $R_1(T)$ for $\boldsymbol{H} \parallel \boldsymbol{c}$ corresponds qualitatively to the conditions for which the motion of the FL's occurs with an effective frequency of the order of the nuclear Larmor frequency. When the magnetic field is increased, the T_{irr} decreases corresponding to higher mobility of the FL's, and thus the condition for the maximum in R_1 is met at lower temperature.

We proceed now to calculate the FL contribution to relaxation with a simple model applicable to highly anisotropic HTSC. The relaxation rate due to the fluctuating local field is obtained from time-dependent perturbation theory as [18]

$$R_1 = \frac{1}{T_1} = \frac{1}{2} \gamma_N^2 \int_{-\infty}^{+\infty} \langle h_\perp(t) h_\perp(0) \rangle e^{-i\omega_L t} \, dt , \quad (1)$$

where γ_N is the nuclear gyromagnetic ratio, ω_L is the Larmor frequency, and $h_\perp$ is the local field perpendicular to the applied field $\boldsymbol{H}$. By referring to a model of a stack of superconducting planes of thickness d and the

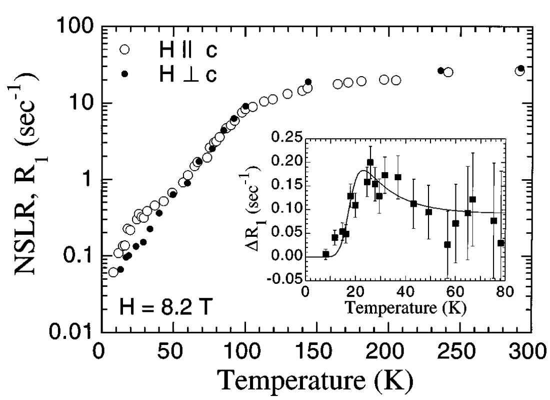

FIG. 2. Temperature dependence of ^{199}Hg NSLR, R_1, in oriented $HgBa_2CuO_{4+\delta}$ powder for $H = 8.2$ T. The inset shows the temperature dependence of the extra contribution to the R_1 for $\boldsymbol{H} \parallel \boldsymbol{c}$ obtained by subtracting the interpolated value of R_1 data for $\boldsymbol{H} \perp \boldsymbol{c}$ from the R_1 for $\boldsymbol{H} \parallel \boldsymbol{c}$. The solid curve is a fit of the extra R_1 data using Eq. (6) (see the text for details).

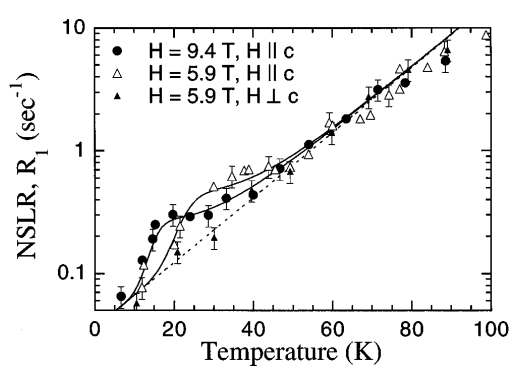

FIG. 3. Plot of ^{199}Hg R_1 vs T in oriented $HgBa_2CuO_{4+\delta}$ powder for two magnetic field intensities. Dashed line is the interpolation of the data of R_1 for $\boldsymbol{H} \perp \boldsymbol{c}$, yielding the background fitted as $R_1 = 0.036 \exp[0.061 T(\mathrm{K})]$ sec^{-1}. Solid curves are theoretical fits including the FL contribution (see the text for details).

interplane spacing s, the transverse field component for a single pancake vortex is, in cylindrical coordinates [19],

$$h_\perp(\rho,z) = \frac{\Phi_0}{4\pi\lambda^2}\frac{d}{\rho}\left[\frac{z}{|z|}\exp\left(-\frac{z}{\lambda_\parallel}\right) - \frac{z}{\sqrt{\rho^2+z_n^2}}\exp\left(-\frac{\sqrt{\rho^2+z_n^2}}{\lambda_\parallel}\right)\right] \cong \frac{\Phi_0}{4\pi\lambda^2}\frac{d}{\rho}\,, \quad (2)$$

where λ is the bulk London penetration length $\lambda = \sqrt{d/s}\,\lambda_\parallel$ and z_n is the z coordinate of the nth layer. $\lambda_\parallel$ is the penetration length for $\boldsymbol{H} \parallel \boldsymbol{c}$ and ρ is the radial distance from the center of the core. Since the average intervortex distance $l_e = (\sqrt{2}/\sqrt[4]{3})\sqrt{\Phi_0/B}$ is much smaller than $\lambda_\parallel$ for the B fields used here, we have simplified Eq. (2) by assuming $\rho, z \ll \lambda_\parallel$. We limit z_n to the two Cu planes above and below a given ^{199}Hg nucleus, i.e., $z_n = s/2 = d$. Equation (2) is valid only for $\rho > s, \xi$.

Thus, assuming that the thermal motion involves fluctuations of the equilibrium position of the FL which is a small fraction of the intervortex spacing l_e, we can write $h_\perp(\rho(t)) \cong h_\perp(\rho) + [\partial h_\perp(\rho)/\partial\rho]_\rho\,\delta\rho(t)$. From Eqs. (1) and (2), taking an ensemble average over ρ from ξ up to $l_m = (\sqrt[4]{3}/\sqrt{2\pi})l_e$ [20] and multiplying by 2 in order to account for the uncorrelated fluctuation of two pancake vortices in the plane above and below a given ^{199}Hg nucleus, one can write

$$R_1 \cong \frac{\gamma_N^2\Phi_0^2}{8\pi\sqrt{3}\,\lambda^4}\frac{d^2\langle u^2\rangle}{l_e^2\xi^2}\int_{-\infty}^{+\infty} g(t)\exp(-i\omega_L t)\,dt\,, \quad (3)$$

where the mean square fluctuation amplitude of the center of the FL core with respect to the equilibrium position can be expressed as [21]

$$\langle|\delta\rho(0)|^2\rangle = \langle u^2\rangle = \frac{16\pi\lambda_\parallel^2 k_B T}{s\Phi_0 B}\ln\left(\frac{\pi B\lambda_\parallel^2}{\Phi_0\ln(l_e/s)}\right). \quad (4)$$

For $s = 9.5$ Å, one has $\langle u^2\rangle/l_e^2 \cong 7\times10^6\lambda_\parallel^2 T$ with a negligible field dependence in the range $H = 5.9$–9.4 T. The correlation function (CF) $g(t) = \langle\delta\rho(t)\delta\rho(0)\rangle/\langle u^2\rangle$ contains the information on the FL dynamics. Above the irreversibility line the FL's should undergo thermally activated hopping. It is reasonable to assume a model of two-dimensional diffusivelike random motion under a restoring force. Thus we write the CF as a sum over q of collective components of the form $\exp(-Dq^2t)$ with D an effective diffusivity constant and Dq^2 the width of the central component in the spectrum of the FL excitations. The normalized spectral density becomes

$$j(\omega) = \int_{-\infty}^{+\infty} g(t)\exp(-i\omega t)\,dt = \frac{1}{N}\sum_q\frac{2Dq^2}{(Dq^2)^2+\omega^2} = \tau_D\ln\left(\frac{\tau_D^{-2}+\omega^2}{\omega^2}\right), \quad (5)$$

where we have integrated over $\vec{q}$ in two dimensions up to $q_m = (\sqrt{8\pi}/\sqrt[4]{3})\,(1/l_e)$ (in a triangular lattice) and set $\tau_D = (Dq_m^2)^{-1}$. Finally, from Eqs. (3) and (5) one has

$$R_1 \cong \frac{\gamma_N^2\Phi_0^2}{8\pi\sqrt{3}\,\lambda^4}\frac{d^2\langle u^2\rangle_{\rm irr}}{l_e^2\xi^2}\left(\frac{T}{T_{\rm irr}}\right)\tau_D\ln\left(\frac{\tau_D^{-2}+\omega_L^2}{\omega_L^2}\right), \quad (6)$$

where $\langle u^2\rangle_{\rm irr}$ is the value at $T = T_{\rm irr}$.

Since the FL contribution to R_1 is present only for $\boldsymbol{H} \parallel \boldsymbol{c}$, we assume as background contribution the experimental data for $\boldsymbol{H}\perp\boldsymbol{c}$. In the inset of Figs. 2 and 3 we show the theoretical curves obtained by adding the FL contribution from Eq. (6) to the background contribution obtained by interpolating the data of R_1 for $\boldsymbol{H}\perp\boldsymbol{c}$. To obtain a good fit of the data we assume in Eq. (6) $\tau_D = \tau_0\exp(U/T)$ and set $A = (\gamma_N^2\Phi_0^2/8\pi\sqrt{3}\,\lambda^4)\,d^2\langle u^2\rangle_{\rm irr}/\xi^2 l_e^2$ as a fitting parameter. We obtain $A \cong 10^8$ (rad/sec)^2 and $\tau_0 \cong 10^{-11}$ sec for all three curves while the activation energy U is found to be inversely proportional to H as shown in Fig. 4. Although the values of the parameters are only indicative rather than definitive due to the relatively small FL contribution to the total R_1 and the presence of three fitting parameters, it is encouraging that their order of magnitude is quite reasonable. From Eq. (4) one has $\langle u^2\rangle_{\rm irr}/l_e^2 \cong 5\times10^{-2}$ for $\lambda_\parallel(0) \cong 1500$ Å [22] and $T_{\rm irr} = 31$ K (at $H = 5.9$ T). Thus by taking $d = s/2 = 4.75$ Å, $\xi \cong 20$ Å one can estimate the theoretical value $A \cong 5\times10^9$ (rad/sec)^2. The calculation which leads to Eq. (6) overestimates the effect of fluctuations by considering the pancakes completely uncorrelated along the c direction. An even small degree of correlation between planes would reduce drastically the transverse field in Eq. (2) [19]. Thus the calculated value of A is not unreasonable.

Having established that the ^{199}Hg NSLR contains a measurable contribution from the FL's thermal motion for $\boldsymbol{H} \parallel \boldsymbol{c}$, we conclude by comparing the parameters obtained here with the ones extracted from magnetic transport measurements [23]. In doing so one should keep in mind that NMR probes the spontaneous fluctuations associated with the FL thermal hopping motion while other techniques

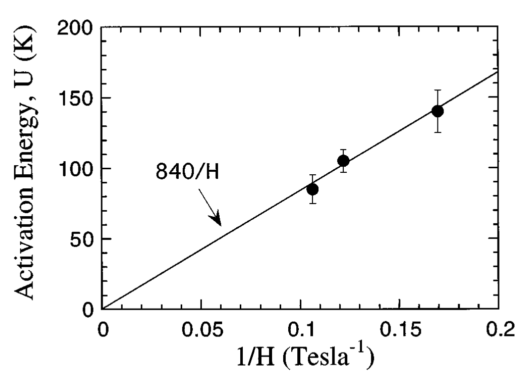

FIG. 4. Magnetic field dependence of the effective activation energy U of flux-line thermal motion obtained from ^{199}Hg NSLR in oriented $HgBa_2CuO_{4+\delta}$ powder. The line is a fit by $U = 840/H$ with H in tesla and U in kelvin.

probe dissipation effects related to the same FL dynamics. At the irreversibility line we find a smooth change in the correlation time consistent with continuous thermal depinning with a microscopic hopping time $\tau_c \cong 5 \times 10^{-8}$ sec. The prefactor τ_0 and the activation energy found here are of the same order of magnitude as found from dissipative flux motion in the thermally activated flux creep model for HTSC of similar anisotropy [24]. The simple field dependence $U \propto 1/H$ of the activation energy (Fig. 4) was derived on the ground of physical arguments [25] but experimentally one often finds more complicated H dependences [24,26]. For pancake vortices in anisotropic HTSC and for $\boldsymbol{H} \| \boldsymbol{c}$ it was shown [27] that the characteristic value of U depends strongly on the strength of the interlayer coupling with U becoming equal to the field-independent intrinsic pinning energy U_p only for the highly anisotropic HTSC. In the case where the Josephson interlayer coupling is not negligible one may have a field-dependent effective U for a flux bundle although the simple $1/H$ form is difficult to justify. We may thus conclude that the parameters derived near $T_{\rm irr}$ from the present NSLR experiment describe a thermally activated hopping motion of bundles of pancake vortices with non-negligible interlayer coupling.

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