

^{19}F NMR Study of Flux-Line Dynamics in Fluorinated $\text{YBa}_2\text{Cu}_4\text{O}_8$

Figen TABAK

*Department of Physics Engineering, Hacettepe University,
Beytepe, 06532, Ankara-TURKEY*

Mavrizio CORTÌ

*Department of Physics “A. Volta”, University of Pavia,
27100 Pavia-ITALY*

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Abstract

Measurements of the ^{19}F NMR linewidth $\Delta\nu$ and spin-lattice relaxation rate $1/T_1$ have been performed in an oriented powder sample of fluorinated $\text{YBa}_2\text{Cu}_4\text{O}_8$ at temperatures $4 < T < 300$ K and static magnetic fields $0.3 < H_0 < 9.4$ T for Holc. Thermal motion of flux lines was evidenced from the effect on NMR linewidth while $1/T_1$ showed the effects due to interaction with Fermi liquid.

Introduction

It is known that in high temperature superconductors (HTSC) an applied field H_0 greater than the lower critical field H_{c1} (≈ 100 Gauss) and less than the upper critical field H_{c2} ($\approx 10^6$ Gauss) penetrates in the bulk sample in the form of filaments of flux lines. A characteristic property of HTSC is their marked anisotropy resulting from their 2D structure. In $\text{YBa}_2\text{Cu}_4\text{O}_8$, with the NMR magnetic field along the c axis perpendicular to the CuO_2 planes, each flux line has a core radius of the order of the coherence length $\xi_{II} \approx 20$ Å. The flux lines form a triangular lattice (FLL) and for an external field H_0 in the range 5-10 T are at a distance $\ell_e = [2\phi_0 / \sqrt{3H_0}]^{1/2} \approx 100$ Å. Outside of the core of the filaments where the material is superconducting, the field decays in a manner determined by the London equation. In $\text{YBa}_2\text{Cu}_4\text{O}_8$, and for $T \ll T_c$, the London penetration depth is $\lambda_{II} \approx 1400$ Å. FLL can be considered an elastic continuum and shows interesting vibrational and fluctuational properties.

Current studies of HTSC concentrate on the dynamics of the flux lines. The understanding of the motion of the flux lines is an important problem both from a scientific as well as technological point of view, thermal motion of the vortices being related to the

critical depinning current. Above the so called irreversibility line $T_{irr}(H)$, which marks the onset of dissipative forces at low frequencies, the flux lines are mobile. Experimental approaches commonly used in the study of flux lines dynamics are the resistivity and magnetic type measurements of macroscopic character and reliable information is available. In recent years, NMR technique has proved to be a good tool for the study of the microscopic origin of spontaneous thermal fluctuations in HTSC for nuclei with no quadrupole moments ($I=1/2$) and with narrow intrinsic resonance lines. When a flux line moves in the lattice, the surrounding nuclei are subject to a modulation of the local internal magnetic field which can effect the NMR parameters in various ways. If the thermal fluctuations reach an amplitude which is a sizable fraction of the intervortex distance and characteristic frequency becomes faster than the inhomogeneously broadened linewidth ($10^3 - 10^5$ Hz), an anomalous narrowing is observed. On the other hand, thermal fluctuations drive the nuclear relaxation process and extra contributions to spin-lattice ($1/T_1$) and spin-spin ($1/T_2$) relaxation rates are expected. Characteristic frequency of the motions in the range $1-10^3$ Hz in T_2 measurements and $10^7 - 10^9$ Hz in T_1 measurements can be detected. From a combination of T_1 , T_2 and $\Delta\nu$ measurements one can derive microscopic insight, for example on the degree of correlation in the motion of 2D pancakes belonging to adjacent superconducting planes, on the barriers for the pseudo-diffusion of the flux lines and on their temperature and field dependences.

The extra contribution to relaxation rates due to flux line motion should be prominent just above the irreversibility line where characteristic peaks in $1/T_1$ and $1/T_2$ are expected when the effective correlation time of the flux line motion crosses as a function of temperature the value of the inverse Larmor frequency ω_L , (i.e. $\omega_L\tau_c \approx 1$). The second condition is that the spin-lattice relaxation of the nucleus should not be dominated by other sources of relaxation since thermally activated vortex motion is expected to be a very weak relaxation mechanism.

Unambiguous evidence for a direct contribution to relaxation due to thermally fluctuating flux lines has been obtained only in organic superconductors [1] [2]. Regarding HTSC, where controversial results have been obtained, the clear evidence comes from ^{199}Hg NMR spin-lattice relaxation rate measurements in $\text{HgBa}_2\text{CuO}_{4+\delta}$ [3].

In a recent study, ^{89}Y NMR linewidth $\Delta\nu$ and spin-lattice relaxation rate $1/T_1$ in $\text{YBa}_2\text{Cu}_4\text{O}_8$ were reported [4]. The flux line dynamics is evidenced by the extra narrowing of the NMR line. On the other hand, an anomalous enhancement of $1/T_1$ with a maximum above the irreversibility line is observed. The maximum is found to shift to higher temperatures for lower magnetic fields similar to the behavior of $T_{irr}(H)$.

In this paper we report ^{19}F NMR linewidth $\Delta\nu$ and spin-lattice relaxation rate $1/T_1$ measurements in an oriented powder sample of fluorinated $\text{YBa}_2\text{Cu}_4\text{O}_8$ for temperatures $4 < T < 300$ K and static magnetic fields $0.3 < H_0 < 9.4$ T parallel to the c axis. ^{19}F has nuclear spin $I=1/2$ and like ^{89}Y , is thus free from complications due to quadrupolar interactions, but having a large gyromagnetic ratio ($\gamma_F/\gamma_Y \approx 20$) makes it available as a suitable probe at low magnetic fields to study flux line dynamics.

Experimental Results and the Analysis of Data

A single-phase powder sample was prepared with average grain size 12-20 μm . After mixing with epoxy the grains were oriented in a magnetic field to achieve alignment along a common crystal c axis. The superconducting transition temperature estimated from the detuning of the NMR probe at zero field is $T_c=86\pm 1$ K.

Measurements of the ^{19}F NMR linewidths were obtained by Fourier transforming the second half of the spin-echo signal following a $(\pi/2)_x - \tau - (\pi/2)_y$ pulse sequence, with the exception of the test at $T=4.2$ K where an echo envelope measurement was performed. Typical $\pi/2$ radio frequency pulses were 2-3.5 μs , corresponding to $H_1 \approx 30$ Gauss, sufficient to irradiate the whole NMR line. The line shape is practically Gaussian at all temperatures and given by the sum of the second moment of intrinsic nature resulting from the dipole-dipole interaction and the second moment due to the distribution of the local magnetic field related to the flux line lattice. The $\Delta\nu$ (full-width at half maximum, FWHM) above T_c is around 45 kHz and practically temperature independent from 300 K down to 70 K. In the superconducting state the linewidth related to the FLL, $\Delta\nu_{FL}$ was obtained by subtracting the constant value ≈ 45 kHz from the experimental values. In Fig.1, resulting temperature dependence of $\Delta\nu_{FL}$ is shown for $H_0=9.4$ T. The linewidth related to flux lines is given as $\Delta\nu_{FL} = \sqrt{(M_2^{FL})^2} \approx 1.5 \times 10^{-2} \Phi_0 / \lambda_{II}^2(T)$, where M_2^{FL} is the second moment due to the modulation of the local field by the vortex configuration. In the absence of any motion of the flux lines the line narrows as $\Delta\nu(T)/\Delta\nu(0) = \lambda^2(0)/\lambda^2(T)$ for $T \rightarrow T_c$. The points (\bullet) in Fig.1 illustrates the behavior expected for $\Delta\nu_{FL}$ in the assumption $\Delta\nu_{FL}(0)=155$ kHz and $\lambda_{II}^2(T) = \lambda_{II}^2(0)[1-T/T_c]^{-1}$ (via the London two fluid model). The extra narrowing of the linewidth with respect to the rigid lattice value is evidence of the thermal motions of the flux lines. From the experimental results for $T \rightarrow 0$ an estimate of λ_{II} can be obtained as $\lambda_{II} \approx 2200$ \AA . This value is larger than the one in $YBa_2Cu_4O_8$.

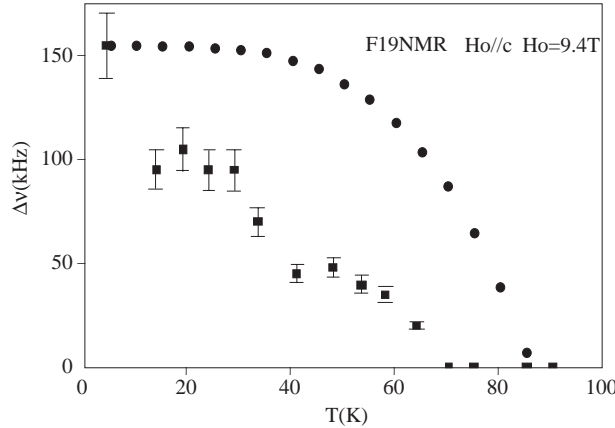


Figure 1. Contribution to the ^{19}F NMR linewidth from the vortices in fluorinated $YBa_2Cu_4O_8$ for $H_0=9.4$ T. The circles represents the behavior expected from the temperature dependence of the London penetration depth.

The ^{19}F spin-lattice relaxation rates $1/T_1$ were measured from the echo signal following the saturation of the line by a sequence of five $\pi/2$ pulses. Recovery is nonexponential and is fitted to a sum of two exponential functions,

$$[h(\alpha) - h(t)]/h(\alpha) = 0.45\exp(-t/T_{1\ell}) + 0.55\exp(-t/T_{1s}), \quad (1)$$

where $T_{1\ell}$ and T_{1s} are the long and short time constants, respectively.

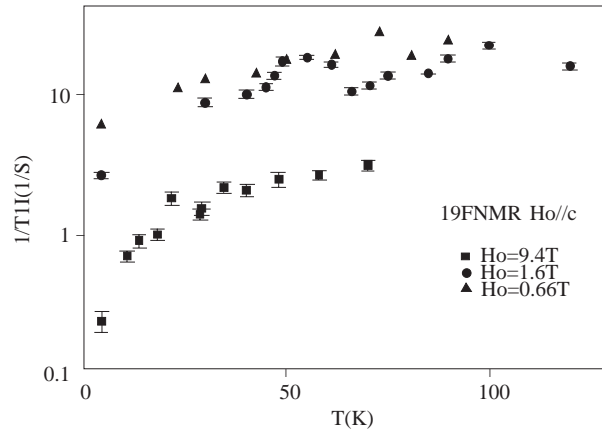


Figure 2. ^{19}F spin-lattice relaxation rate $1/T_{1\ell}$ in fluorinated $\text{YBa}_2\text{Cu}_4\text{O}_8$ for Ho//c ; (■) $H_0 = 9.4\text{T}$, (●) $H_0 = 1.6\text{T}$ and (▲) $H_0 = 0.66\text{T}$.

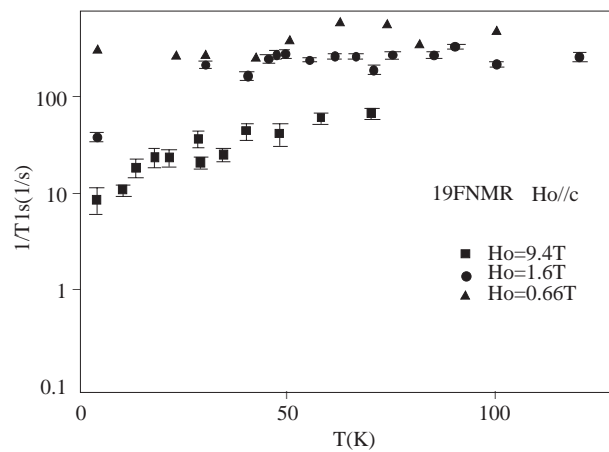


Figure 3. ^{19}F spin-lattice relaxation rate $1/T_{1s}$ in fluorinated $\text{YBa}_2\text{Cu}_4\text{O}_8$ for Ho//c ; (■) $H_0 = 9.4\text{T}$, (●) $H_0 = 1.6\text{T}$ and (▲) $H_0 = 0.66\text{T}$.

In Fig. 2 and Fig. 3 the behavior of $T_{1\ell}$ and T_{1s} is reported for $H_0=9.4$ T, $H_0=1.66$ T and $H_0=0.66$ T as a function of temperature. We analyze the data in terms of a simple model of two dimensional diffusive-like random motion of 2D pancake vortices under a restoring force developed in references [3] and [4]. From the time dependent perturbation theory, the relaxation rate due to the fluctuations in local field due to the time dependence of the FL position is given by [5]

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

h_{\perp} being the local field perpendicular to the applied field and for a single 2D pancake vortex, is given as [6] [7],

$$h_{\perp} = \frac{\Phi_0}{4\pi\lambda_{II}^2} \frac{s}{\rho} \left[\frac{z}{|z|} \exp(-z/\lambda_{II}) - \frac{z}{\sqrt{\rho^2 + z^2}} \exp[(-\sqrt{\rho^2 + z^2})/\lambda_{II}] \right], \quad (3)$$

width s the distance between superconducting planes and ρ the radial distance from the center of the core.

Thus by considering that the motions around the equilibrium positions involve a small fraction of the equilibrium intervortex distance ℓ_e , one can expand $h_{\perp}(\rho, t)$ in terms of $\rho(t)$ and by an ensemble average from the vortex core ξ up to $\ell_m = \ell_0(\sqrt{3}/2\pi)^{1/2}$ arrives at [4]

$$1/T_1 = (\gamma^2/2) h_e^2(T) J(\omega_L), \quad (4)$$

where $h_e(T) = [(2s/\xi^2 \lambda_{II}^2) k_B T \ln(2\lambda_{II}^2/\sqrt{3}\ell_e^2)]^{1/2}$ is a temperature dependent effective field and $J(\omega_L) = \tau_d \ln[(\tau_d^{-2} + \omega_L^2)/\omega_L^2]$ is the spectral density with $\tau_d \approx D_{\perp}^2/\ell_e^2$ having the meaning of an effective correlation time describing the pseudo-diffusion motion of the 2D pancake vortices with a pseudo-diffusion constant D_{\perp} .

According to this theoretical expression one expects a maximum in the relaxation rate at the temperature T_m where $\tau_d = 0.86/\omega_L$. In $YBa_2Cu_4O_8$ a maximum in the relaxation rate is observed at $T_m=35$ K for $H_0=9.4$ T which shifted to $T_m=53$ K for $H_0=5.9$ T, consistent with the theoretical value. Fitting the data according to Eq.(4), assuming for τ_d the thermally activated temperature dependence $\tau_d = \tau_0 \exp(U/T)$, one extracts the barrier U for the thermal depinning of the flux lines as $U=450 \pm 50$ K for $H_0=9.4$ T and $U=700 \pm 50$ K for $H_0=5.9$ T with $\tau_0 = 1.0 \times 10^{-14}$ s.

Regarding our NMR $1/T_1$ data in fluorinated $YBa_2Cu_4O_8$, no peaks which can be attributed to vortex motion seems to be observed. By using the theoretical expression (4) we estimated the temperatures where the maxima in $1/T_1$ are expected by scaling our ^{19}F NMR $1/T_1$ data with ^{89}Y NMR data in $YBa_2Cu_4O_8$ assuming that the inverse field dependence $U=4200/H_0$ for the activation energy found in $YBa_2Cu_4O_8$ holds also for fluorinated $YBa_2Cu_4O_8$. Thus by taking $\tau_0 = 1.0 \times 10^{-14}$ s, $s = 10 \text{ \AA}$, $\lambda_{II} = 1400 \text{ \AA}$, $\xi = 20 \text{ \AA}$, and $\ell_e = 100 \text{ \AA}$ the temperatures for the expected peaks are calculated to be $T=42$ K for $H_0=9.4$ T and $T=63$ K for $H_0=5.9$ T, which is not the case as observed here. Also

the estimated relaxation rates turn out to be small compared with high relaxation rates observed. One estimate is $1/T_1=0.18 \text{ s}^{-1}$ at $T=42 \text{ K}$ for $H_0=9.4 \text{ T}$ while the experimental values are $1/T_1=25 \text{ s}^{-1}$ and $1/T_1=50 \text{ s}^{-1}$ for the long and short components, respectively (see Fig.2 and Fig.3). The absence of vortex peaks can be attributed to the masking caused by the high relaxation rates due to the interaction of fluorine nuclei with the Cu conduction electrons. This leads to the conclusion that while linewidth senses and gives information about vortices, T_1 shows the effects due to a Fermi liquid.

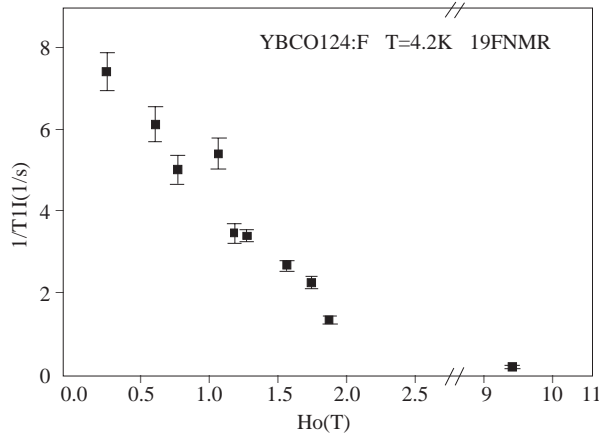


Figure 4. Field dependence of the ^{19}F spin-lattice relaxation rate $1/T_{1l}$ in fluorinated $\text{YBa}_2\text{Cu}_4\text{O}_8$ at $T=4.2 \text{ K}$ for the long T_1 component.

An important feature of these results is the sizable field dependence of the relaxation rates. In Fig.4 we report this dependence for $T=4.2 \text{ K}$. Field dependent relaxation rates in the mixed state in type II superconductors have been previously reported [8]. For HTCS the only characteristic field dependent relaxation rates which have been claimed to be due to the relaxation by vortices are reported in highly anisotropic $Tl_2Ba_2CuO_4$ [9] where $1/T_1$ decreases with the increasing field, as we observed here. However the similar field dependence also observed above T_c both for $Tl_2Ba_2CuO_4$ [10] and in our case rules out this possibility. Moreover, our results are also inconsistent due to rapid relaxation of the nuclei in the vortex core where the metal is in its normal state followed by spin diffusion to more distant nuclei in the superconducting region. In fact, in the rapid diffusion limit, one has $R_{1s} = R_{1n}H\xi^2\Phi_0$ where, R_{1s} and R_{1n} are the relaxation rates in the superconducting and normal phases [11]. This equation predicts an H dependence which was not observed here. It seems that the field dependence is neither related to flux lines thermal motion nor superconductivity. At the moment the origin of the field dependent relaxation rates is an open question.

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