Fluxon Thermal Motion Detected by Nuclear Spin Echo Decay Measurements: ^{89}Y NMR in $YBa_2Cu_3O_7$

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A new NMR approach was devised to investigate thermal motion of vortices based on the observation of dephasing effects of the transverse nuclear magnetization below T_c. The effect was observed in ^{89}Y spin-spin relaxation time T_2 measurements of $YBa_2Cu_3O_7$ for two crystal orientations with respect to the applied magnetic field. The data were fitted with a simple model of diffusion of ^{89}Y nuclei with respect to the local magnetic field gradient generated by the moving vortices. Small values of a thermally activated effective diffusion constant of the vortices' motion well below T_c with reasonable activation energies were derived.

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In layered anisotropic high-T_c superconductors, thermal fluctuations of vortices strongly affect both thermodynamic and transport properties [1]. Different equilibrium dynamical regimes for the flux-line lattice are observed as functions of temperature and magnetic field: large amplitude thermal fluctuations, thermally activated depinning, and flux melting. NMR experiments have yielded information about microscopic fluxon dynamics in both conventional [2] and in high-T_c superconductors [3-5]. The main interest has been concentrated on NMR linewidth and line shape measurements. The line profile is determined by the local distribution of the magnetic field intensity in the flux lattice and is thus related to the London penetration depth λ [2,4]. The narrowing of the linewidth for increasing $T \rightarrow T_c$ can be an indication of averaging effects due to thermal motion of fluxons on a time scale of the NMR spectral linewidth (e.g., 10^{-5} to 10^{-3} sec) [3,5]. In this respect, NMR measurements yield information similar to the results of muon spin rotation (μSR) measurements wherein the main difference is the time "window" for the detection which is less than 10^{-6} sec in the μSR case [6].

In this paper, we propose a novel NMR approach to investigate fluxon dynamics based on the measurements of the decay of the echo signal amplitude in a two-pulse Hahn echo (TPHE) experiment. In a TPHE experiment one probes the irreversible dephasing of the transverse nuclear magnetization during the time interval τ between two radio frequency pulses [7]. If the vortex core moves even a few lattice spacings in the time interval between the two pulses, the nuclei will experience a dephasing effect due to the change of local magnetic field strength and thus of the Larmor precession frequency. This should result in an observable attenuation of the spin echo amplitude as an additional contribution to the nuclear magnetic relaxation time T_2. This effect is completely analogous to the one which occurs in the presence of atomic diffusion in a magnetic field gradient [8], except in our present case we look for the motion of the field gradient, fluxon motion, relative to the stationary nuclei.

The $I=\frac{1}{2}$, ^{89}Y nucleus appears to be an ideal probe since ^{89}Y has an intrinsically narrow NMR line, and no complications coming from nuclear electric quadrupole effects, plus a relatively long spin-spin relaxation time T_2. The ^{89}Y NMR data presented below for $YBa_2Cu_3O_7$ appear to confirm the predicted dephasing effect due to fluxon motion. The observed anomalous decay of the echo amplitude just below T_c is explained quantitatively in terms of an effective local diffusion constant which is indicative of very slow long wavelength modes which do not manifest themselves in other types of NMR measurements or in μSR measurements.

Two samples of $YBa_2Cu_3O_7$ with $T_c = 92 \pm 1$ K were prepared and the powder grains of an average diameter 25 to 38 μm were mixed with epoxy resin and oriented in an 8.2 T magnetic field. The estimated superconducting transition temperature at 8.2 T was $T_c = 87.5 \pm 1$ K for $H_0 \| c$ axis and $T_c = 89 \pm 1$ K for $H_0 \perp c$ axis, corresponding to the irreversibility transition temperature $T_{\rm irr} \approx 78.5 \pm 1$ K for $H_0 \| c$ axis and $T_{\rm irr} \approx 80 \pm 1$ K for $H_0 \perp c$ axis at 8.2 T, as measured from the detuning of the NMR probe. The NMR measurements were performed with a phase coherent pulse Fourier transform (FT) spectrometer [9] operating at 17.104 MHz in a field strength of 8.2 T. Typical $\pi/2$ pulse lengths for ^{89}Y were 20 to 25 μsec corresponding to an rf magnetic field strength of 60 G. The NMR linewidth ΔH was determined from the FT of half of the ^{89}Y echo signal. The echo decay rate was monitored with a Hahn sequence $(\pi/2)_x$-τ-$(\pi)_y$ with variable τ separation between pulses (x and y are the relative phases of the rf magnetic field pulses in the rotating frame). All measurements were taken in field cooled conditions with an applied magnetic field of 8.2 $T \gg H_{c1}$.

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Thus, the fluxon lattice should be viewed as a regular vortex lattice with a greater density of vortices than pinned vortex lines.

The typical decay of the transverse nuclear magnetization in a TPHE experiment is shown in Fig. 1 for two temperatures, above and below T_c, and for two orientations of the c axis with respect to the magnetic field direction. The decay rate is orientation dependent even at room temperature. Part of the anisotropy can be explained in terms of ^{89}Y-^{89}Y dipolar interactions while a small contribution may be related to the anisotropy of Cu spin susceptibility [3]. The temperature dependence of the decay is of prime interest here. Above T_c the temperature dependence is negligible while below T_c the decay of the nuclear magnetization becomes faster and deviates strongly from exponential (see Fig. 1). The data were fit with the equation

$$M(t)=M(0)\exp(-t/T_2)\exp(-t/T_{2\mathrm{eff}})^3 . \qquad (1)$$

Note that the data could be fit equally well by replacing the $\exp(-t/T_{2\mathrm{eff}})^3$ term with a Gaussian term. Unfortunately, the signal to noise ratio was not sufficient to resolve the weight of the two terms, even with considerable signal averaging. However, above T_c the Gaussian form yields a slightly better fit while the $\exp(-t/T_{2\mathrm{eff}})^3$ term yields a better fit below T_c, indicating the probable presence of both terms. More relevant for the purpose of the present experiment is a comparison between the TPHE decay and the decay obtained by a Carr-Purcell-Meiboom-Gill (CPMG) sequence [8,10]. In fact, in the CPMG sequence given by $(\pi/2)_x$-τ-$[(\pi_y)$-2τ- $]_n$, any dephasing of the nuclear magnetization $M(t)$ due to a change of the local magnetic field at the nuclear site during the time interval τ is not cumulative and thus can be minimized by choosing τ shorter than the intrinsic spin-spin relaxation time T_2. In order to avoid spin-locking effects normally present in the CPMG sequence for short τ values [11], we have devised a modified sequence with alternating phases of the y pulses given by $(\pi/2)_x$-τ-$[(\pi)_y$-2τ-$(\pi)_y$-2τ-$(\pi)_{-y}$-2τ-$(\pi)_{-y}$-2τ- $]_n$, which will be called the AP-CPMG sequence. With the above AP-CPMG sequence, the results for the echo decay rate for $T>T_c$ are nearly the same as the results for the TPHE as expected in the absence of fluxon motion, confirming that the phase alternation does indeed eliminate spin-locking effects. On the other hand, just below T_c one observes the onset of an extra contribution to the TPHE decay, a contribution which becomes negligible in the AP-CPMG decay as shown in Fig. 2. We are thus led to the conclusion that the additional contribution to the decay of the Hahn echo signal is to be attributed to a change of local magnetic field due to the time dependence of the magnetic field gradient associated with fluxon motion.

We turn now to the temperature dependence of the echo decay. By referring to the fit of Eq. (1) we find a temperature independent exponential term yielding $T_2=13.5$ msec for $H_0 \parallel c$ and $T_2=7.4$ msec for $H_0 \perp c$ and a temperature dependent contribution $(T_{2\mathrm{eff}})^{-1}$. $(T_{2\mathrm{eff}})^{-1}$ is small and almost T independent above T_c, while just below T_c, $(T_{2\mathrm{eff}})^{-1}$ displays an enhancement followed by

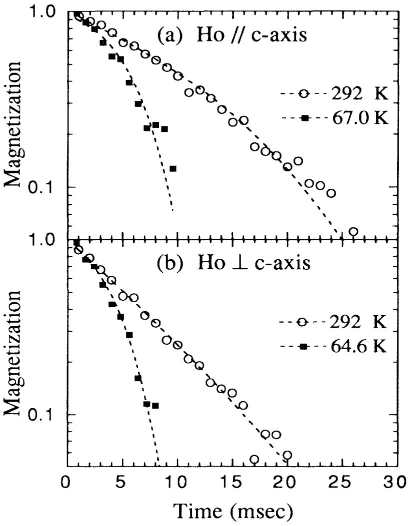

FIG. 1. Typical decay of the nuclear magnetization vs $t \equiv 2\tau$ in a TPHE experiment for ^{89}Y in $\mathrm{YBa_2Cu_3O_7}$. The dashed lines are a fit of Eq. (1) with (a) magnetic field $H_0 \parallel c$ axis: $T_2=13.5$ msec and $T_{2\mathrm{eff}}=23.9$ msec (at $T=292$ K); $T_{2\mathrm{eff}}=7.8$ msec (at $T=67$ K) and (b) $H_0 \perp c$ axis: $T_2=7.4$ msec; $T_{2\mathrm{eff}}=30.8$ msec (at $T=292$ K); $T_{2\mathrm{eff}}=6.8$ msec (at $T=64.6$ K).

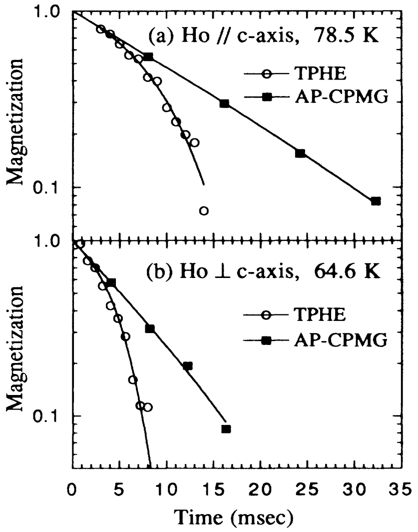

FIG. 2. Comparison of the nuclear magnetization vs time for ^{89}Y in $\mathrm{YBa_2Cu_3O_7}$ for the two pulse sequence TPHE and for the multiple pulse sequence AP-CPMG described in the text. The full lines are fits with Eq. (1) with (a) $H_0 \parallel c$ axis: $T_2=13.5$ msec and $T_{2\mathrm{eff}}=13.0$ msec (TPHE); $T_{2\mathrm{eff}}=66.7$ msec (AP-CPMG) and (b) $H_0 \perp c$ axis: $T_2=7.4$ msec and $T_{2\mathrm{eff}}=6.8$ msec (TPHE); $T_{2\mathrm{eff}}=29.0$ msec (AP-CPMG).

a decrease back to the same value as above T_c (Fig. 3). We propose the maximum of $(T_{2\mathrm{eff}})^{-1}$ below T_c can be understood in terms of thermal motions of the vortices. To this aim, we fit the data with an equation valid for atomic diffusion in a uniform magnetic field gradient whose average value is G [8]. In fact, around a given flux core there exists a radial magnetic field gradient whose average value G is of order $\Delta H/d$, where ΔH is the NMR inhomogeneous linewidth and d is the intervortex spacing. Large amplitude diffusional or vibrational motion of the vortex core would produce a dephasing of the nearby nuclei in a manner analogous to nuclei diffusing in a steady magnetic field gradient. Although the gradient G will not be uniform, particularly near the saddle points of the vortex lattice (where G is zero), in order to be able to analyze the data quantitatively, we make the simplifying assumption that a majority of nuclei sit in the same average gradient. Furthermore, we consider the case in which the vortex cores have moved on the average only a fraction of d in the measuring time τ. Under these two conditions we can borrow from the theory of the effect of atomic diffusion on the decay of the nuclear magnetization, and write [8]

$$M(t)=M(0)\exp(-t/T_2)\exp\left[-\frac{D\gamma_N^2G^2t^3}{12}\right], \qquad (2)$$

where T_2 is related to nuclear dipole-dipole interactions.

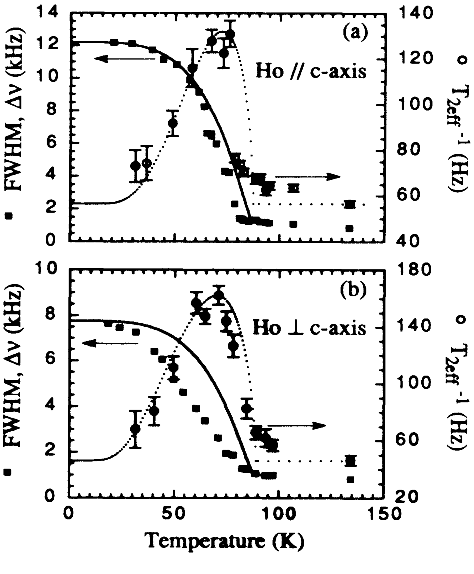

FIG. 3. (■) Full width at half intensity (FWHM) of the ^{89}Y NMR line in YBa$_2$Cu$_3$O$_7$ vs temperature. The full line is the behavior predicted by the T dependence of the penetration length without motional narrowing effects (see text). (o) Effective decay rate $(T_{2\mathrm{eff}})^{-1}$ obtained from the fit of Eq. (1) plotted vs temperature. The dotted line is the theoretical fit from the expression for $(T_{2\mathrm{eff}})^{-1}$ in Eq. (2) with D given by the values in Eq. (3) and a constant term given by $(T_{2\mathrm{eff}})_0^{-1}=56.5$ Hz for $H_0\parallel c$ axis and $(T_{2\mathrm{eff}})_0^{-1}=46$ Hz for $H_0\perp c$ axis.

The effect of fluxon motion is described by the effective rate $(T_{2\mathrm{eff}})^{-1}=(D\gamma_N^2G^2/12)^{1/3}$. Here γ_N is the ^{89}Y nuclear gyromagnetic ratio and D is an effective local diffusion constant describing the vortex dynamics. We assume $\gamma_N G=\gamma_N\Delta H(T)/d$, with $d=(2/\sqrt{\pi})\sqrt{\phi_0/H}=1.8\times10^{-6}$ cm, $H=8.2$ T, and

$$\frac{\Delta H_{\parallel,\perp}(T)}{\Delta H_{\parallel,\perp}(0)}=\frac{\lambda^2_{\parallel,\perp}(0)}{\lambda^2_{\parallel,\perp}(T)}=1-\left[\frac{T}{T_{c,\parallel,\perp}}\right]^4,$$

where $\lambda(T)$ is the penetration depth. The NMR linewidth has been measured in our samples and is also shown in Fig. 3. Our results are in reasonable agreement with previous measurements [3], except the deviation of $\Delta H(T)$ from the theoretical curve describing the behavior of $\lambda(T)$ in the two-fluid model is less pronounced in our data particularly for $H_0\parallel c$. The deviation from $\lambda(T)$ is ascribed to motional narrowing effects due to fluxon thermal fluctuations [3]. Thus the discrepancy could be due to a different concentration of pinning centers in the different samples investigated. By using $\gamma_N\Delta H_\parallel(0)=12.2$ kHz and $\gamma_N\Delta H_\perp(0)=7.7$ kHz from Fig. 3, one finds a theoretical curve for $(T_{2\mathrm{eff}})^{-1}$ which fits the data in Fig. 3 for the following choice of

$$\begin{aligned}D(H_0\parallel c)&=4\times10^{-10}\exp(-500/T),\\ D(H_0\perp c)&=4\times10^{-10}\exp(-350/T).\end{aligned} \qquad (3)$$

D has units of cm^2/sec. The general behavior of the observed maximum is reproduced remarkably well by the theoretical curve, except close to T_c where flux-lattice melting may produce a sudden increase in D thus invalidating Eq. (2). The $D_0=4\times10^{-10}$ cm^2/sec and $E_a=500$ and 350 K values deduced for $D(T)$ are only indicative of fluxon motion in view of the simplicity of the model and of the uncertainty in the estimate of the average magnetic field gradient G. If one interprets D as describing the diffusional displacement of the core of a vortex line in a given CuO$_2$ plane, one has $\langle a^2\rangle=4D\tau$, which implies an rms displacement of order of 10 Å in a time $\tau=1$ msec for T close to T_c and in a time of about 1 sec at $T=40$ K. A comparison with the NMR linewidth narrowing is in order. The departure of $\Delta H(T)$ from the T dependence of $\lambda(T)$ has been ascribed to the onset of about 40 K (in our case this would be at a higher T) of vortex motion with correlation time $\tau_c<10^{-4}$ sec [3], a much faster correlation time than the diffusional correlation time estimated from the echo decay. A possible explanation for the difference is that the linewidth narrowing is due to a short wavelength segmental motion of vortex lines or to depinning effects while $(T_{2\mathrm{eff}})^{-1}$ measurements probe the long wavelength segmental motion of the vortex cores. It should be emphasized that the huge magnitude of the magnetic field gradient which exists around a given vortex line allows one to observe very slow local diffusional displacements not accessible to NMR linewidth and μSR measurements. Note that there is no contradiction with the zero average displacement of pinned vortices if only a fraction of the large total segmental displacement is detected in the time interval separating two radio frequency pulses. The situation is reminiscent of the dynamics of entangled polymers in a gel where a description in terms of an effective diffusion constant is commonly adopted [12].

Alternative possible explanations for the effects we observed are considered below and shown to be inconsistent with the data. An enhancement of the ^{89}Y NMR echo decay rate below T_c was reported before [13] and attributed to the spin-flip suppression of unlike ^{63,65}Cu nuclear moments with a resultant increase of Y-Cu nuclear dipolar coupling. This interpretation predicts a monotonic increase of $(T_{2\text{eff}})^{-1}$ below T_c rather than the maximum observed experimentally in Fig. 3. A ^{89}Y-^{89}Y pseudodipolar interaction via the charge carriers could be effective above T_c, similar to what was reported for ^{63,65}Cu [14]. This indirect coupling could indeed contribute to the T_2 anisotropy above T_c. However, it could not explain the data below T_c since the opening of a superconducting gap could result in a decrease of $(T_{2\text{eff}})^{-1}$, but not an increase.

In conclusion, we have suggested a novel NMR method to investigate fluxon thermal motion. We have presented convincing experimental evidence that the effect is indeed observable in ^{89}Y echo decay rates in YBCO. From a quantitative fit of the data, an effective diffusion constant was derived which corresponds to very slow components of the microscopic vortex-line dynamics. This opens up the interesting possibility of comparison of the experimental results with hydrodynamic models in terms of statistical motion of polymerlike assemblies of parallel lines [12] and/or of collective low frequency, long wavelength modes of coupled vortices [1].

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