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Vortex dynamics in single-crystal YBa$_2$Cu$_3$O$_7$ probed by ^{63}Cu nuclear spin echo measurements in the presence of transport current pulses

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We report ^{63}Cu NMR spin-echo experiments on a high-quality single crystal of YBa$_2$Cu$_3$O$_7$ ($T_c = 93$ K), to which we have bonded leads for both transport current pulse applications and four-point resistance measurements. For a 9 T field with H_0 parallel to c, the resistive transition onsets at 90 K, and R fully reaches zero at $T_{R=0} = 76$ K. The superconducting state NMR linewidth, however, does not exceed the normal-state value until $T < T_{R=0}$, where it is in agreement with predictions based on measured penetration depths. We discuss the possibility that a vortex liquid is present within the resistive transition, with vortex diffusion occurring at a rate fast enough to induce motional narrowing of the vortex lattice contribution to the NMR linewidth. We use the Einstein relation to show that this rapid vortex diffusion implies an upper bound for the correlation length for vortex motion. Inclusion of transport current pulses in the spin-echo pulse sequence is found to have no effect on the magnitude of the spin-echo signal for T both greater and less than $T_{R=0}$.

Nuclear magnetic resonance (NMR) has played a well-known role in the investigation of the microscopic mechanisms of superconductivity, providing one of the first important experimental confirmations of the Bardeen-Cooper-Schrieffer (BCS) theory,[1,2] and, more recently, guiding the development of a theory for high-T_c cuprate materials.[3] NMR, however, has also played a lesser known role in investigations of the magnetic properties of superconductor, in particular, the properties of the Abrikosov vortex lattice observed in the mixed state of type-II superconductors.[2]

High-T_c superconductor display highly unusual magnetic properties, which result from their anisotropic structure and high transition temperatures.[4,5] The magnetic properties of the various high-T_c compounds are quite varied—for example, the properties of the moderately anisotropic YBa$_2$Cu$_3$O$_7$ differ greatly from those of highly anisotropic Bi$_2$Sr$_2$CaCu$_2$O$_x$. There is, however, a common feature. All high-T_c compounds, for applied fields H such that $H_{c1} \ll H \ll H_{c2}$, display a high-temperature phase, called the "vortex-liquid" phase, in which vortex motion is thought to be relatively uninhibited. The vortex-liquid phases exists for $T_{R=0} < T < T_c(H)$, where $T_{R=0}$ is defined as the temperature at which sample resistance (in the low-current limit) reaches zero, and $T_c(H)$ is marked by the onset of superconductivity, as probed by either magnetic susceptibility or the onset of a dropoff in sample resistance. In the vortex-liquid phase the phenomenon of "flux flow" is observed: transport currents exert a Lorentz force per unit length $\mathbf{J} \times \mathbf{\Phi_0}$ upon the vortices which causes them to flow with velocity $\mathbf{v}$. The motion of the vortices results in an electric field $\mathbf{E} = -\mathbf{v} \times \mathbf{B}$, and hence resistivity $\rho = vB/J$. For $T < T_{R=0}$ vortices are frozen into either a vortex-lattice or "vortex-glass" state, and the resistivity (for small currents) is zero.

Although a quite sophisticated theoretical analysis of the vortex properties of high-T_c superconductors has been developed,[5] there remain very few experimental techniques that can probe vortex dynamics on a microscopic scale, especially for strong applied magnetic fields. NMR would appear to be a promising technique for obtaining various parameter values, such as correlation times for vortex motion. Indeed, a number of NMR investigations have appeared, notably several using ^{89}Y NMR to probe powder samples of YBa$_2$Cu$_3$O$_7$ ($T_c \sim 93$ K), with the magnetic field perpendicular to the CuO$_2$ planes. Brom and Alloul[6] reported the signs of "motional narrowing" of the ^{89}Y NMR line shape in the vortex-liquid regime (for 83 K $< T < T_c$ for their applied field of 7.5 T). Both Suh, Torgeson, and Borsa[7] and Corretta and Corti[8] provide ^{89}Y NMR evidence for vortex motion, but the characteristic correlation times [the characteristic time required for a vortex to move or diffuse approximately one intervortex spacing (~ 100 Å)] derived differ dramatically—Carretta and Corti obtain a time of 30 μs for 40 K $< T < T_c$, while Suh, Torgeson, and Borsa obtain a diffusion coefficient near T_c of (120 Å)^2/s, implying a much longer time of 100 ms.

Additionally, Carretta[9] has performed ^{89}Y NMR experiments on unaligned, pressed pellets of YBa$_2$Cu$_3$O$_7$ in the presence of applied transport current, in order to monitor the induced motion of vortex lines—an experiment demonstrated by Delrieu[10] on Pb-Tl alloys. One expects that, in the YBa$_2$Cu$_3$O$_7$ system, with the applied field of 5.9 T used in the experiment, the sample resistivity would reach zero for $T < \sim 80$ K and that below that temperature it would be necessary to exceed the critical current density J_c in order to induce motion. Remarkably, Carretta and Corti, using current densities of only 1 A/cm^2, observe motional narrowing of the ^{89}Y line shape at all temperature values reported, down to 65 K.

For NMR investigation of high-T_c vortex dynamics, there are clearly advantages to the use of single-crystal

samples. One can, of course, obtain a well-defined orientation of the sample with respect to the magnetic field, but this can be achieved to a great extent by using powder samples aligned magnetically in an epoxy. A more important advantage is that the varied shapes of crystallites in powder samples result in an unknown spread in demagnetization factors,[11] which can affect the NMR line shape and linewidth. Magnetic fields produced by Meissner currents in neighboring crystallites in a powder sample also influence line shapes and linewidths. Finally, for NMR measurements with applied transport current, the use of a single crystal (with four-point probe monitoring of both current and voltage) enables one to obtain a well-defined current flow geometry and (presumably) eliminates grain boundary effects.

In this communication we report ^{63}Cu NMR measurements on the "planar" [Cu(2)] sits for a single crystal of YBa$_2$Cu$_3$O$_7$ with four attached leads for monitoring of the sample resistance and for application of transport current pulses during the NMR pulse sequence. Comparison of the temperature variation of the NMR linewidth and resistivity confirms aspects of the theory of vortex-lattice melting, and sets an upper limit on the volume of vortex bundles in the vortex-liquid state. We report spin-echo experiments accompanied by transport current pulses as large as 10 A/cm^2 (100 times greater than that of Ref. 9) but observe no effects on the echo amplitude or linewidth—although this must be regarded as a null result; we show that it also yields important information about vortex dynamics near and above the vortex melting temperature.

The single crystal of YBa$_2$Cu$_3$O$_7$ was prepared by the decanted flux technique of Liang et al.[12] The crystal dimensions are 2.4×0.9×0.1 mm^3. Superconducting quantum interference device (SQUID) susceptibility measurements indicated an onset T_c of 92.5 K and a width of 0.5 K (10–90% criterion) in a 10-G applied field.

Transport current and voltages were transmitted from the top of the cryostat to the crystal through Alpha 9450 coaxial cables. The crystal was glued (using GE No. 7031 varnish) to a narrow, machined, patterned piece of printed circuit (pc) board, which included four copper contact pads to which the coaxial cables were soldered. These contact pads were then attached to the voltage and current pads on the crystal as described below.

Using a mechanical mask, four silver pads of thickness 1 μm were evaporated onto the top surface of the crystal. The pads for the two current leads were deposited at opposite ends of the crystal. Two additional evaporations were used to extend the current lead pads to cover the sides of the crystal as well, in order to obtain a homogeneous current density throughout the depth of the crystal. Two voltage contacts (width ~0.25 mm) were deposited between the current pads. 0.0015-in.-diam. aluminum leads were bonded from the pc board contact pads to the silver pads on the crystal. It was observed that, upon applying current pulses larger than 1 A, leads often came detached from the crystal due to the Lorentz force on the wire (~0.027 N) in the 9 T field. Current carrying capacity may be increased to some extent by attaching more than one wire in parallel.

Sample temperature was controlled using an Oxford Instruments helium- or nitrogen-gas flow CF1200 cryostat. The time required for equilibration of the sample temperature was monitored using the resistance itself, and typically 2 or 3 h were required.

NMR line shapes were obtained by Fourier transform of the spin-echo signal. The line shapes observed were crudely Gaussian; to characterize linewidths we have extracted the full width at half maximum (FWHM) and then estimated the variance σ^2 and standard of deviation σ (plotted in the figure) according to the relation $\sigma = 0.4247$ (FWHM) for a Gaussian line shape. The amplitude of the pulsed rf magnetic field H_1 was typically 104 G ($^{63}\gamma H_1/2\pi = 117$ kHz), but in all cases it was sufficient to cover the full linewidth.

Sample resistance was monitored with a lock-in amplifier with current amplitude 120 μA and frequency 94 Hz. The sample resistance also served as an excellent monitor of the crystal temperature.

Figure 1 shows the planar ^{63}Cu NMR linewidth and sample resistivity, both taken at a 9 T field (H_0) applied parallel to the crystal c axis. Linewidths are given in angular velocity (ω) units, which can be converted to magnetic field units using the gyromagnetic ratio $^{63}\gamma$ of ^{63}Cu: $^{63}\gamma = 2\pi \times 1.1285$ kHz/G. The temperature dependence of the resistivity is quite similar to previous results. Rather than dropping abruptly to zero as T is lowered below T_c, the resistivity in the presence of the strong magnetic field makes a gradual descent—it steepens near 90 K±1 K, which we label as the onset T_c in the applied field (suppressed by only about 3 K), but it does not each zero until approximately 76 K.

We now discuss the temperature dependence of the NMR linewidth σ, and its relation with that of resistivity. The temperature-dependent NMR line broadening contribution due to a *static* Abrikosov vortex lattice is well known.[2,13,14] The line shape represents a profile of the magnetic-field distribution inside the sample, and, for a perfect triangular vortex lattice, includes singularities
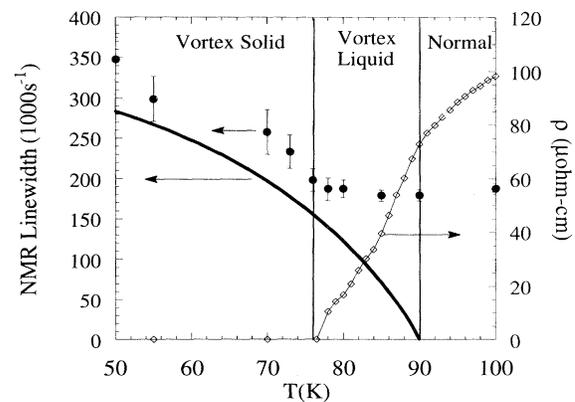

FIG. 1. Resistivity and ^{63}Cu NMR linewidth as a function of temperature for the twinned YBCO crystal in a 9 T field applied parallel to the crystalline c axis. No broadening of the ^{63}Cu line shape is seen down to tmeperatures below which the resistance becomes effectively zero.

associated with the magnetic-field extrema and saddle points. If pinning and lattice disorder are introduced, then the singularities are smeared, and often a Gaussian or near Gaussian results. The vortex-lattice contribution to the linewidth σ_{VL}, however, retains information about the extent of the magnetic-field distribution and, hence, the penetration depth λ:[15]

$$\sigma_{VL} \sim 0.0609 \frac{\phi_0}{\lambda^2} , \qquad (1)$$

where ϕ_0 is the magnetic flux quantum. Figure 1 includes this predicted contribution σ_{VL}, utilizing the penetration-depth measurements on similarly prepared high-quality single crystals by Hardy et al.[16]

Figure 1 demonstrates that the NMR linewidth retains its normal-state width, $\sigma \sim 1.8 \times 10^5$ radians/s, until the temperature is reduced below 76 K, the temperature at which the resistance reaches zero $T_{R=0}$. Below that temperature the experimental line shapes are only slightly broader than predicted from Eq. (1); the slight excess broadening most likely results from convolution of the normal-state broadening mechanisms σ_{NS} with the vortex-lattice (VL) broadening σ_{VL}. For illustration we have also shown (Fig. 2) an estimate of the vortex-lattice broadening σ_{VL} alone by subtracting the normal-state variance σ_{NS}^2 from the measured total values: $\sigma_{VL}^2 = \sigma_{tot}^2 - \sigma_{NS}^2$. [Also shown, again, is the expected σ_{VL} for a static Abrikosov lattice, based on Eq. (1).] This subtraction procedure, though valid for a Gaussian, is crude and, in general, not justified (an exponential relaxation, for example, has a divergent σ^2)—at this point we will proceed with it tentatively. We subsequently report an experiment that confirms the major conclusions that are drawn.

The implication (from Fig. 2) is that the VL broadening σ_{VL} remains close to zero until the temperature is reduced below $T_{R=0}$ (~ 76 K), in contrast to the prediction based on the assumption of a static lattice. Only below
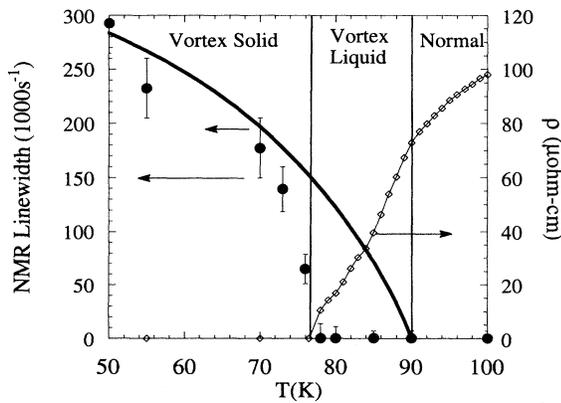

FIG. 2. Resistivity and vortex lattice contributioon to the second moment of the ^{63}Cu NMR line shape as a function of tmeperature. The 9 T applied field is parallel with the crystalline c axis. The solid line is the prediction based on a static vortex lattice using the penetration depth values from Hardy et al. and Eq. (1).

$T_{R=0}$ does the linewidth increase and match the prediction of (1). A reasonable explanation for such behavior is that for $T_{R=0} < T < T_c$ the broadening contribution from the vortex lattice is "motionally narrowed," suggesting a vortex-liquid state, with rapid diffusional vortex motion, for $T_{R=0} < T < T_c$. (A similar conclusion is drawn in Ref. 6 from ^{89}Y NMR on an aligned powder sample. Our results place an even faster lower limit on the time scale because the ^{63}Cu gyromagetic ratio is 5.4 times that of ^{89}Y.)

Suppose that an *instantaneous* magnetic field has a broadening (in magnetic field units) σ_i, which would result in an instantaneous frequency distribution $\Delta\omega_i \sim \gamma_n \sigma_i$, where γ_n is the nuclear gyromagnetic ratio. Motional narrowing of the NMR line shape will occur if in this situation the magnetic field fluctuates rapidly in time, so that a typical nuclear spin samples the full range of the field distribution within a correlation time τ with $\tau \ll 1/\Delta\omega_i$; then the observed NMR linewidth will be greatly narrowed from the value $\Delta\omega_i$ [to a value of order $(\Delta\omega_i)^2\tau$].

The assumption that the motional narrowing is the cause of the reduced linewidth for $T_{R=0} < T < T_c$ then implies a correlation time $\tau \ll 1/\Delta\omega_i \sim 1/(^{63}\gamma\sigma_{VL}) \sim 10$ μs for $T \sim 80$ K. Conversely, the *absence* of motional narrowing for $T < 76$ K$=T_{R=0}$ implies that $\tau > 10$ μs, in agreement with Suh, Torgeson, and Borsa[7], who estimate from T_2 measurements on ^{89}Y a correlation time of ~ 100 ms (for diffusing a distance approximately equal to one vortex lattice spacing). The combination of these results suggests that for magnetic fields of ~ 9 T, the correlation time as defined here increases by more than four orders of magnitude as T is lowered from approximately 78 to 73 K.

The upper bound on the correlation time $\tau \ll 10$ μs, which we have inferred, also has implications about the physics of the vortex-liquid state, including the effective volume of vortex "bundles" in the vortex-liquid regime. For the analysis we take a very crude model of vortex dynamics, which enables us to relate the observed rapid diffusional motion of the vortices with the measured electrical resistivity through the Einstein relation: we suppose that the vortex system can be viewed as a set of noninteracting particles (bundles) moving in a viscous medium, each having effective volume V. This volume V would be a kind of correlation volume throughout which the enclosed vortices (or "pancakes") move together in a highly correlated way. The vortices outside the correlation volume (at a distance greater than a correlation length $1 \sim V^{1/3}$) would move independently. In the presence of a transport current density J there is a force F on such a vortex bundle given by $F = JBV$ (using mks units), where B is the magnetic field. If the vortices, in response to this force, move with a drift velocity v then we may infer a vortex bundle mobility μ given by

$$\mu = \frac{v}{F} = \frac{v}{JBV} . \qquad (2)$$

Note, though, that the ratio v/JB can be obtained from the electrical resistivity, since $E = vB = J\rho$, and hence $v/J = \rho/B$. Thus from (2) we obtain

$$\mu = \frac{\rho}{B^2 V} . \qquad (3)$$

The mobility for a system of free, noninteracting particles can be related to the diffusion coefficient D through the Einstein relation

$$D = \mu kT . \qquad (4)$$

Combining relations (3) and (4), we obtain an expression for the effective bundle volume V:

$$V = \frac{\rho kT}{DB^2} . \qquad (5)$$

A lower limit on the diffusion coefficient D is known from NMR. Near 80 K vortices are known to diffuse a distance greater than order one lattice spacing (~ 100 Å) within a time 10 μs. The diffusion length x can be related to D according to

$$\langle x^2 \rangle \sim D\tau . \qquad (6)$$

Thus, $D > 1000$ Å^2/μs.

With the lower limit of $D > 1000$ Å^2/μs on the diffusion coefficient D, we now have, through Eq. (5), an *upper* limit on the effective volume V, which is proportional to resistivity ρ. ρ, in fact, reaches zero at 76 K, and the NMR line appears to remain motionally narrowed. We can obtain a quite *conservative* upper limit on V by using the value of ρ obtained at ~ 80 K, $\rho = 17$ $\mu\Omega$ cm. This yields $V = 2 \times 10^{11}$ Å^3, a volume that would contain $\sim 10^6$ vortex pancakes. The corresponding length L, given by $(2L)^3 = V$ is 4×10^3 Å, the same order of magnitude as the penetration depth, perhaps by coincidence. Of course, the possibility remains that the motion of vortices in the vortex-liquid state may be faster, and the bundle volume smaller, than we are able to verify. Additionally, our crude model of a vortex bundle with no features of pinning or anisotropy undoubtedly discards important physics. If, alternatively, we were to assume that individual vortices are restricted to point in straight lines throughout the sample thickness of 0.1 mm, then the volume $V = 2 \times 10^{11}$ Å^3 would imply a maximum correlation radius in the xy plane of 250 Å, a distance
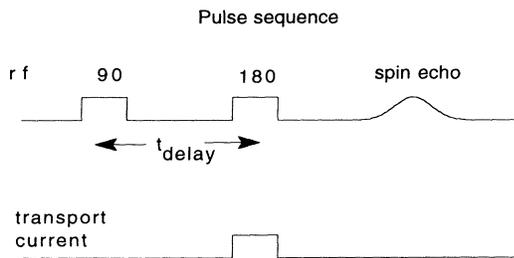
Pulse sequence

FIG. 3. The timing diagram for pulse sequences containing pulses of transport current. The transport current is pulsed on during the 180° RF pulse. For a static vortex lattice, current pulses exceeding the critical current density will displace the vortex lattice and rephasing after the 180 pulse should be diminished. For all current pulses investigated the spin-echo size is unaffected.

comparable to the intervortex spacing.

We now discuss an experiment that verifies our earlier inference, from Fig. 2, that the vortex lattice contribution to the NMR linewidth σ_{VL} within the temperature range 76 K $< T <$ 90 K, is quite small. Let is first assume the contrary, that at temperature 80 K the vortex-lattice contribution σ_{VL} to the linewidth is the value obtained using the assumption of a static lattice, or 1.2×10^5 rad/s. In that event, we could easily detect the presence of σ_{VL} using a "spin-echo" NMR experiment, accompanied by applied transport current pulses to displace the vortex lattice.

Figure 3 gives a timing diagram for the spin-echo experiment with an applied transport current pulse.[17] A 90° pulse is applied to the spin system initially at thermal equilibrium, followed after a time t_{delay} by a 180° pulse. Let us suppose that a typical spin has frequency ω following the 90° pulse and ω' following the 180° pulse. Then, if $\omega = \omega'$ the NMR signal will grow to a maximum at a time t_{delay} following the 180° pulse.

In our experiment, however, the current pulse is expected to displace the vortex lattice, so that a typical spin should see a quite different magnetic field before and after the 180° pulse, and hence $\omega \neq \omega'$. The size of the echo should then be severely diminished if $(\omega - \omega')t_{delay} \gg 1$. We have seen no such diminution in any of our experiments, confirming our assumption of motional narrowing. A wide range of current magnitudes, pulse lengths, and temperatures have been used, but here we present a few representative examples.

At 80 K we used $t_{delay} = 40$ μs and a current pulse of magnitude 20 mA ($J = 21$ A/cm^2) and duration 5 μs. Using the measured resistivity of 17 $\mu\Omega$ cm, we infer an electric field during the pulse of 3.6×10^{-2} N/C, and a vortex drift velocity v given by $E = vB$, where B is 9 T. This gives $v = 40$ Å/μs, so that a 5-μs pulse gives a 200-Å average displacement of the vortex system—larger than the intervortex spacing for a triangular lattice of 160 Å. The typical frequency jump $(\omega - \omega')$ should be $\sim \sigma_{VL}$ at 80 K, or $\sim 10^5$ s^{-1}, and $(\omega - \omega')t_{delay} \sim 4 \gg 1$, yet the echo amplitude was unaffected by the current pulse.

We also attempted the same experiment at temperatures below 76 K, where R reaches zero, hoping to cause a vortex displacement by exceeding the critical current J_c. J_c is not known precisely for this crystal, but Senoussi *et al.*[18] find $J_c \sim 1000$ A/cm^2 for single crystal YBa$_2$Cu$_3$O$_7$ at 70 K in a 1-T field, with a strong field dependence (typically an order of magnitude decrease in J_c for each 1-T increase in field, over the measured range). At 73 K, where σ_{VL} is clearly nonzero, we applied a 1-μs current pulse with current density $J = 320$ A/cm^2, and with $t_{delay} = 40$ μs. Again the current pulse resulted in no diminution of the echo amplitude. We infer from this result that either we are not exceeding the critical current or that we are exceeding it but that even for this high current the differential resistivity and the resulting electric fields in the sample are low.

In conclusion, our experiment gives clear microscopic evidence for presence of the vortex-liquid state with rapid vortex diffusional motion of distances greater than the in-

tervortex spacing within times less than 10 μs. The NMR experiment, coupled with measurements of resistivity, imply that, in the vortex-liquid state, vortex motion is highly correlated only over distances of at most ~ 3000 Å. At temperatures below $T_{R=0}$ (76 K in our 9-T field) the NMR linewidth is in agreement with the predictions based on the assumption of a static vortex lattice, but also not inconsistent with the slower diffusional motion (~ 100 ms for an intervortex spacing) obtained by Suh, Torgeson, and Borsa,[7] suggesting a jump of at least four

orders of magnitude in the correlation time as T is lowered below $T_{R=0}$.

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