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Antiferromagnetism in the vortex cores of $YBa_2Cu_3O_{7-\delta}$

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We report spatially resolved nuclear-magnetic-resonance measurements on a high-temperature superconductor that indicate the presence of correlated antiferromagnetic fluctuations in the vortex core. The nuclear-spin-lattice relaxation rate $1/^{17}T_1$ of planar ^{17}O, in near-optimally doped $YB_2Cu_3O_{7-\delta}$, was measured. Outside of the core, $(^{17}T_1T)^{-1}$ is independent of temperature consistent with theoretical predictions for a *d*-wave superconductor. In the vortex core, $(^{17}T_1T)^{-1}$ increases with decreasing temperature following an antiferromagnetic Curie-Weiss law.

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High-temperature superconductors are commonly viewed as doped Mott insulators for which the parent compound is antiferromagnetic. For these materials competition between magnetism and superconductivity is unambiguously evident in the phase diagram. Varying the composition by increasing the oxygen content increases the electronic carrier density, suppressing the Néel phase and producing superconductivity. Even for those compositions that exhibit superconductivity, the presence of antiferromagnetic (AF) fluctuations is apparent in the normal state from inelastic neutron scattering[1] and copper nuclear-magnetic-resonance (NMR) relaxation.[2–4] Experiments have shown that antiferromagnetic fluctuations coexist with superconductivity even in optimally doped materials (highest T_c).[1] There have been predictions that magnetism can appear, possibly in a novel, spatially inhomogeneous form, coexisting with vortices.[7–12] Using spatially resolved, high-field, NMR experiments, we have found evidence for antiferromagnetism in the vortex cores of $YB_2Cu_3O_{7-\delta}$ (YBCO), lending support for these ideas.

In the mixed state, magnetic flux penetrates the sample in the form of quantized vortices, each with a core of radius that is the size of the superconducting coherence length $\xi_o = 16$ Å. According to theoretical models antiferromagnetism can appear in the vortex core, a region where superconductivity is suppressed. Zhang[7,10] has developed a theory that integrates antiferromagnetism and *d*-wave superconductivity based on SO(5) symmetry, predicting that the superconducting vortex can have an antiferromagnetic core. Arovas *et al.*[8] extended this work to consider the possible coexistence of superconducting vortices with antiferromagnetism as a function of doping. Demler *et al.*[9] have looked in the far-field region of the vortex finding that superconducting and coupled superconducting spin-density wave (SDW) phases can appear in the presence of vortices and that there are oscillations in charge density in good agreement with scanning tunneling microscopy experiment.[13] Charge- and spin-density wave structures near and inside the vortex have been explored theoretically by Zhu *et al.* and Chen and Ting.[14] They also find results consistent with experiment.[13] It has been shown by Herbut[11] that the *d*-wave superconductor is unstable to the formation of SDW owing to phase disordering in the core of a vortex. These theories have a common feature; antiferromagnetism can be associated with vortex cores.

Using elastic neutron scattering, Vaknin *et al.*[15] found evidence of static AF order in the mixed state of YBCO. They estimated that the upper limit of the average magnetic moment is $\mu \leq 0.004\mu_B$ per flux line in a layer. Katano *et al.*[16] found enhanced static AF correlations in the vortex state of $La_{2-x}Sr_xCuO_4$ $(x=0.12)$. Neutron inelastic-scattering experiments by Lake *et al.*[6] on the optimally doped $La_{2-x}Sr_xCuO_4$ $(x=0.163)$ show that the imaginary part of the magnetic susceptibility χ'' at low energy below the spin gap[1] and at an incommensurate AF wave vector, is strongly enhanced in a magnetic field $H_0 = 7.5$ T at temperatures below $T = 10$ K. They interpreted their results as evidence that the vortex cores are nearly ordered antiferromagnets that polarize the intervening medium. These measurements are close to NMR relaxation experiments since both are sensitive to the same component of the susceptibility. However, they do not have spatial resolution. This is possible with scanning tunneling microscopy where Hoffman *et al.*[13] have found checkerboard extended states that they associate with vortices although the method is not specifically sensitive to magnetism. All of these experiments consistently indicate unusual structure near the vortex core.

Following theoretical suggestions,[17,18] NMR measurements by Curro *et al.*,[22] Mitrović *et al.*,[5] and Kakuyanagi *et al.*,[19] as well as μSR line-shape measurements of Miller *et al.*,[20] show that it is possible to spatially resolve different regions of the vortex lattice by analyzing the internal field distribution of the corresponding resonance spectrum. However, resolution of the vortex core region is best achieved at relatively high applied fields since the fraction of the spectrum inside the core grows with increasing field. For a square vortex lattice, this fraction is $H_0\pi\xi_0^2/\phi_0$, where ϕ_0 is the flux quantum. In a field of 42 T, vortices are ~ 86 Å apart and vortex cores occupy $\sim 17\%$ of the total sample.

In the present work, we report on the temperature dependence of spatially resolved measurements of the nuclear-spin-lattice relaxation rate $1/T_1$ in the mixed state. This re-

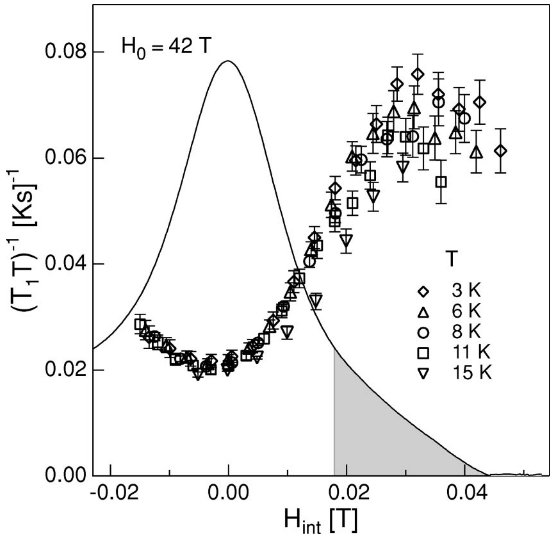

FIG. 1. Spin-lattice relaxation rate of planar ^{17}O divided by temperature as a function of internal magnetic field. The $(-1/2 - -3/2)$ satellite spectrum shown is measured at 37 T, same as at 42 T, and the shaded region corresponds to the fraction of the spectrum occupied by vortex cores at 42 T.

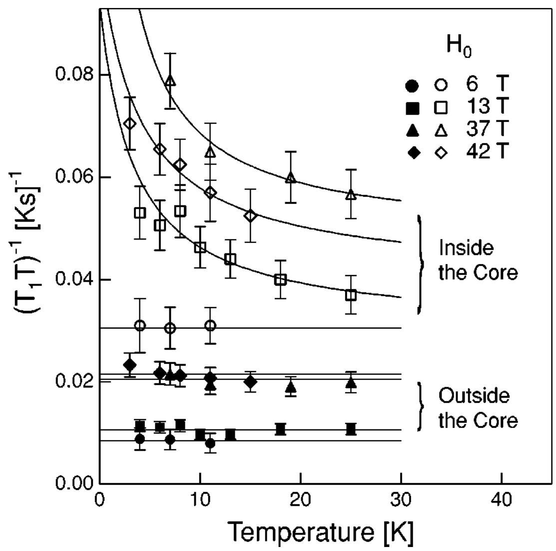

FIG. 2. Planar ^{17}O spin-lattice relaxation extracted from the saddle point (full symbols) and the vortex core regions of the spectrum (open symbols) versus temperature in 6, 13, 37, and 42 T applied fields. The solid lines are fits to the data as explained in the text.

laxation requires an electronic spin flip, and consequently, is particularly sensitive to magnetic fluctuations. Previously, we demonstrated[5] that we could resolve the NMR rate in the vortex core at high magnetic field, above 13 T. Applying this method, we have now measured the temperature dependence of $(^{17}T_1T)^{-1}$ and extended our range of field. We find that $(^{17}T_1T)^{-1}$ is independent of temperature outside the vortex cores, while in the vortex core region $(^{17}T_1T)^{-1}$ is enhanced, increasing with decreasing temperature following a Curie-Weiss (CW) law, reminiscent of the normal-state behavior of the copper relaxation, $(^{63}T_1T)^{-1}$. We associate this enhanced temperature dependence of $(^{17}T_1T)^{-1}$ in the vortex core region with correlated antiferromagnetic spin fluctuations such as are observed in the normal state.

Our sample is a near-optimally doped, $\sim 60\%$ ^{17}O-enriched $YBa_2Cu_3O_{7-\delta}$ powder sample, aligned with the crystal $\hat{c}$ axis parallel to the applied magnetic field. Low-field magnetization data show a sharp transition at $T_c(0) = 92.5$ K. Our measurements were made at temperatures from 3 to 25 K and magnetic fields from 6 to 42 T. We have independently determined from spin-spin and spin-lattice relaxation that the transition region in high fields near 30 T occurs at 80 K in our samples. The $(-1/2 - -3/2)$ $^{17}O(2,3)$ quadrupolar-split NMR satellite was used exclusively since it exhibits a sharp high-field edge in the normal state that broadens owing to the distribution of local fields from vortices.[5] The internal field variable in Fig. 1, is defined by $H_{int} = \omega/^{17}\gamma - H_0$, where $^{17}\gamma$ is the gyromagnetic ratio for oxygen and the spectrometer frequency ω is set to resonance at the peak of the spectrum, $H_{int} = 0$. This position corresponds to oxygen nuclei at the saddle point of the field distribution, a spatial location half way between neighboring vortices. Increasing H_{int} corresponds to spatial positions that approach the vortex core. We obtained precise spectra using a field-sweep technique[21] and we measured the relaxation rate using progressive saturation.[23] There is some overlap between the quadrupolar-split transitions. Their effect on the spectrum and on relaxation can be subtracted.[5] In Fig. 2, we report relaxation data for $H_{int} \leqslant 0.032$ T where the small correction for the $(-3/2 - -5/2)$ transition is less than 5%. However, the subtracted spectrum in the region near $H_{int} = 0.04$ T in Fig. 1, should be considered to be qualitative. In the present work, the main purpose of high applied field is to increase sensitivity to vortex cores since the corresponding portion of the NMR spectrum grows proportionately. This is shown for 42 T as the shaded region of the spectrum in Fig. 1.

The spin-lattice relaxation rate at 42 T, shown in Fig. 1, increases as a function of the internal field, i.e., on approaching the vortex cores. This is attributable[5] to a Doppler shift of the quasiparticle excitation energies, a consequence of the supercurrent momentum that increases toward the vortex core. Here, we will concentrate our discussion on the temperature dependence of the rate. Outside of the core region, we observed that $(T_1T)^{-1}$ is independent of temperature and at higher internal fields, in the vortex core, we observe a very different behavior.

In order to contrast the temperature dependence inside and outside the vortex cores in Fig. 1, we show $(T_1T)^{-1}$ in Fig. 2 for just these two regions of the spectrum in four magnetic fields, 6, 13, 37, and 42 T. The rates outside the cores were determined by evaluating an average in a narrow region of ± 0.002 T around $H_{int} = 0$. Inside the cores, our data are averaged over an interval of H_{int} from the point where $(T_1T)^{-1}$ is greatest to a point that is 0.01 T less than the peak position which occurs for $H_{int} \leqslant 0.032$. The clear distinction between the temperature dependences of the rate inside and outside the vortex cores holds for $H_0 \geqslant 13$ T. If the applied field is too low then the sensitivity of our NMR to the vortex core region is decreased and the enhancement in the temperature dependence of $(T_1T)^{-1}$ is not discernible, or possibly does not exist, as might be the case for the 6 T data in Fig. 2.

We first discuss $(T_1T)^{-1}$ outside the vortex core region and give a very simple interpretation of our observations. The rate can be viewed as an average of the product over all possible initial and final quasiparticle density of states.[5,18,24] At low temperature, the quasiparticle excitations outside of the vortex cores come exclusively from the four nodal regions. Since the density of states depends linearly on energy near the nodes of a *d*-wave superconductor, the rate can be expressed as the product of the initial and final quasiparticle excitation energies. The excitation energies are determined by temperature and, in a magnetic field, by two other variables: applied field through the Zeeman effect and superflow momentum $\mathbf{p}_s$ through the Doppler shift.[5] The latter are temperature independent and so, at sufficiently low temperatures, $T<20$ K the product of initial and final excitation energies is nonzero at the Fermi energy and temperature independent. It follows that $(T_1T)^{-1}$ is temperature independent[17,5,24] as we observe. Furthermore, the increase of the rate with applied field shown in Fig. 2, noted previously by Mitrović *et al.*,[5] is a consequence of node-to-node quasiparticle scattering indicating the presence of AF fluctuations.

Inside the core region, the temperature dependence of $(T_1T)^{-1}$ in Fig. 2 is quite different from outside the cores. Here, we find that $(T_1T)^{-1}$ increases with decreasing temperature. This temperature dependence resembles that of the relaxation at the planar copper site in the normal state, which is dominated by correlated AF fluctuations,[2–4] and for which the AF correlation length increases with decreasing temperature. Thus, we suggest that the temperature dependence of $(^{17}T_1T)^{-1}$ in the vortex core is also dominated by correlated AF fluctuations in a similar way. Furthermore, Moriya and Ueda's theory[25] shows that the temperature dependence of the imaginary part of the dynamic spin susceptibility, and thus $(T_1T)^{-1}$, for two-dimensional (2D) antiferromagnetic spin fluctuations of an itinerant electronic system follows a Curie-Weiss law. This behavior is central to the phenomenological model of Millis *et al.*[4] that successfully describes the normal state where the key parameter is the AF correlation length. Consequently, we proceed with the following analysis of the temperature dependence of $(T_1T)^{-1}$.

In the vortex core, we find that $(T_1T)^{-1}$ consists of two parts, a temperature-independent contribution R_0 and a temperature-dependent part, with the form of a Curie-Weiss law, χ_{core}, giving $(T_1T)^{-1}=R_0+\chi_{core}=R_0+C/(T-\theta_{CW})$. In Fig. 3, we plot the inverse of χ_{core} as a function of temperature where the lines in the figure are linear fits. The inverse of the slope of these lines and their zero temperature intercept have a phenomenological interpretation as a Curie constant C and Curie-Weiss temperature θ_{CW}. Within the accuracy of these fits the Curie-Weiss temperature is negative lying in the range -1 to -4 K for magnetic fields from 13 to 42 T and the Curie constant is *field independent*; consequently, magnetic fields above 13 T do not affect the low-energy AF fluctuations. The dependence of antiferromagnetic fluctuations on doping of a 2D antiferromagnetic has been shown theoretically to have a quantum critical point[26] that is observed in normal-state measurements $(^{63}T_1)^{-1}$ in LSCO.[27] Similarly, the enhanced temperature dependence of $(^{17}T_1T)^{-1}$, moving toward the vortex core, can be interpreted as an approach to a quantum critical point where the amplitude of the superconducting order parameter plays the role of doping. It follows from the theory that a negative θ_{CW} indicates a spin gap in the vortex core, while a positive value of θ_{CW} would have suggested AF ordering with gapless spin-wave excitations.

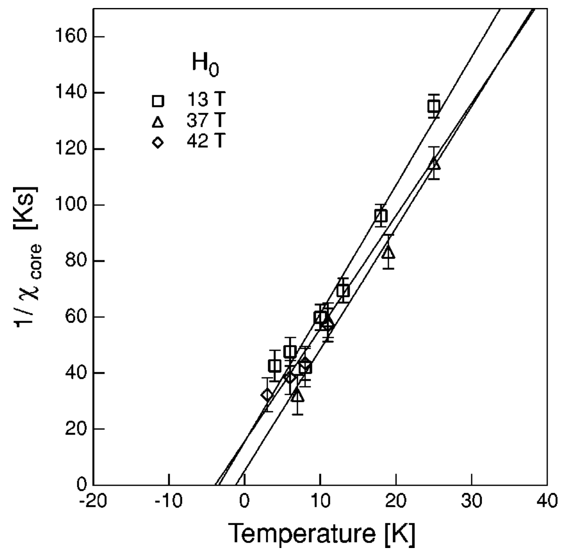

FIG. 3. The inverse of χ_{core} versus temperature, defined in the text. The solid lines are fits to the Curie-Weiss temperature dependence.

In contrast to χ_{core}, the term R_0 depends on the applied magnetic field. A natural extension of the latter term to the region outside the cores suggests that it describes relaxation that depends on thermal quasiparticles through the Zeeman and Doppler effects which are field dependent, but not temperature dependent. We note that at our highest field, 42 T, it appears that R_0 decreases, possibly from additional suppression of the superconducting order parameter at high field, reducing the Doppler term.

It might seem surprising that $^{17}T_1$ is sensitive to antiferromagnetic spin fluctuations at low temperatures in the superconducting state since in the normal state a geometric form factor screens the oxygen nucleus from these fluctuations.[2,28] There are two reasons for this.[24] At low temperatures, the superconducting energy gap constrains possible scattering processes to the nodal regions. This severely limits the available phase space and then screening of oxygen by form factors is much less effective. Second, incommensurability of the susceptibility in the superconducting state[1] increases oxygen sensitivity to antiferromagnetic fluctuations.

Our measurements show that the oxygen NMR relaxation rate, in the form $(^{17}T_1T)^{-1}$, is enhanced in the vortex core. This indicates the existence in the core of correlated antiferromagnetic fluctuations near a quantum critical point with a small spin gap. Outside the core the relaxation rate is proportional to the temperature at low temperatures, and can be attributed to the nodal quasiparticles of a *d*-wave superconductor.

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[1] P. Dai, H.A. Mook, R.D. Hunt, and F. Dogan, Phys. Rev. B **63**, 054525 (2001); P. Dai *et al.*, Science **284**, 1344 (1999); J. Rossat-Mignod, L. P. Regnaud, P. Bourges, C. Vettier, P. Burlet, and J. Y. Henry, in *Frontiers in Solid State Science*, edited by L. C. Gupta and S. Murani (World Scientific, Singapore, 1993), p. 365.

[2] M. Takigawa, P.C. Hammel, R.H. Heffner, Z. Fisk, J.D. Thompson, and M. Maley, Physica C **162-164**, 175 (1989).

[3] C. Berthier, M.J. Julien, M. Horvatic, and Y. Berthier, J. Phys. I **6**, 2205 (1996).

[4] A.J. Millis, H. Monien, and D. Pines, Phys. Rev. B **42**, 167 (1990).

[5] V.F. Mitrović, E.E. Sigmund, H.N. Bachman, M. Eschrig, W.P. Halperin, A.P. Reyes, P. Kuhns, and W.G. Moulton, Nature (London) **413**, 505 (2001).

[6] B. Lake *et al.*, Science **291**, 1759 (2001); B. Lake *et al.*, Nature (London) **415**, 299 (2002).

[7] S.C. Zhang, Science **275**, 1089 (1997).

[8] D.P. Arovas, A.J. Berlinsky, C. Kallin, and S.C. Zhang, Phys. Rev. Lett. **79**, 2871 (1997).

[9] E. Demler, S. Sachdev, and Y. Zhang, Phys. Rev. Lett. **87**, 067202 (2001).

[10] J.P. Hu and S.C. Zhang, cond-mat/0108273 (unpublished).

[11] I.F. Herbut, Phys. Rev. Lett. **88**, 047006 (2002).

[12] M. Franz, Z. Tešanović, and O. Vafek, Phys. Rev. B **66**, 054535 (2002).

[13] J.E. Hoffman, E.W. Hudson, K.M. Lang, V. Madhavan, S.H. Pan, H. Eisaki, S. Uchida, and J.C. Davis, Science **295**, 452 (2002).

[14] J.X. Zhu, I. Martin, and A.R. Bishop, Phys. Rev. Lett. **89**, 067003 (2002); Y. Chen and C.S. Ting, Phys. Rev. B **65**, 180513 (2002).

[15] D. Vaknin, J.L. Zarestky, and L.L. Miller, Physica C **329**, 109 (2000).

[16] S. Katano, M. Sato, K. Yamada, T. Suzuki, and T. Fukase, Phys. Rev. B **62**, R14 677 (2000).

[17] M. Takigawa, M. Ichioka, and K. Machida, Phys. Rev. Lett. **83**, 3057 (1999).

[18] R. Wortis, A.J. Berlinsky, and C. Kallin, Phys. Rev. B **61**, 12 342 (2000).

[19] K. Kakuyanagi, K.I. Kumagai, and Y. Matsuda, Phys. Rev. B **65**, 060503 (2002); cond-mat/0206362 (unpublished).

[20] R.I. Miller *et al.*, Phys. Rev. Lett. **88**, 137002 (2002).

[21] It has been suggested that a field sweep can distort the NMR spectra and current distributions and thereby affect the relaxation rates (Ref. 19). We have compared field-sweep spectra, obtained as described in Ref. 5 where the sample was cooled to 10 K in zero-field prior to starting the field sweep, to spectra obtained after the sample was annealed at 100 K before cooling back to 10 K at each field step. We found that the zero-field-cooled spectra are broader for $H_{int}<0$, but the rates extracted from spectra for $0<H_{int}<0.032$ T are identical regardless of the data acquisition procedure.

[22] N.J. Curro, C. Milling, J. Haase, and C.P. Slichter, Phys. Rev. B **62**, 3473 (2000).

[23] V.F. Mitrović, E.E. Sigmund, and W.P. Halperin, Phys. Rev. B **64**, 024520 (2001).

[24] V. F. Mitrović, Ph.D. thesis, Northwestern University, 2001.

[25] T. Moriya and K. Ueda, Adv. Phys. **49**, 555 (2000); T. Moriya, Y. Takahashi, and K. Ueda, J. Phys. Soc. Jpn. **59**, 2905 (1990).

[26] A.V. Chubukov, S. Sachdev, and J. Ye, Phys. Rev. B **49**, 11 919 (1994).

[27] T. Imai, C.P. Slichter, K. Yoshimura, and K. Kosuge, Phys. Rev. Lett. **70**, 1002 (1993).

[28] V.F. Mitrović *et al.*, Phys. Rev. B **66**, 014511 (2002).