Spin-Density Wave near the Vortex Cores in the High-Temperature Superconductor $Bi_2Sr_2CaCu_2O_{8+y}$

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Competition with magnetism is at the heart of high-temperature superconductivity, most intensely felt near a vortex core. To investigate vortex magnetism we have developed a spatially resolved probe based upon NMR spin-lattice-relaxation spectroscopy. With this approach we have found a spin-density wave associated with the vortex core in $Bi_2Sr_2CaCu_2O_{8+y}$, similar to checkerboard patterns in the local density of electronic states reported from scanning tunneling microscope experiments. We have determined both the spin-modulation amplitude and decay length from the vortex core in fields up to $H = 30$ T.

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Superconductivity and magnetism have been in the forefront of the study of high-temperature superconductivity, largely because their interplay is central to a wide range of physical properties [1]. An important manifestation of their competing orders has been suggested to exist near a vortex core where circulating supercurrents create a sufficiently large magnetic field as to suppress the superconducting state, thereby tipping the balance for stability in favor of magnetism [1,2]. A number of spin-sensitive, spatially resolved, probes have been employed to explore this vortex state, including muon spin resonance, small angle neutron scattering, and nuclear magnetic resonance (NMR). These experiments can give the spatial distribution of local magnetic fields, as in the example of the ^{17}O NMR shown in Fig. 1. This spectrum, as well as results of other methods, are a superposition of local fields from both magnetism and vortex supercurrents and their deconvolution can be quite difficult. In this Letter we report spatially resolved spin-lattice-relaxation (SLR) measurements which allow this separation to be performed.

Scanning tunneling microscopy (STM) experiments on $Bi_2Sr_2CaCu_2O_{8+y}$ (Bi2212) at moderate magnetic fields and low temperatures showed that vortices induce a checkerboard pattern of the local density of states with a spatial period of 15 Å $\approx 4a_0$ [3]. Further STM studies found such electronic ordering in $Bi_2Sr_2CuO_{6+\delta}$ (Bi2201) [4] and $Ca_{2-x}Na_xCuO_2Cl_2$ (NaCCOC) [5] persisting to nonsuperconducting dopings and above the superconducting transition temperature. Elastic neutron scattering experiments [6–8] on the cuprate superconductor, $La_2CuO_{4+\delta}$ (La214), produced magnetic Bragg reflections corresponding to a periodicity of 30 Å $\approx 8a_0$. Although spatially unresolved, this result was associated with the checkerboard pattern found from STM on Bi2212, since the period of a spin-density wave (SDW) is expected to be twice that found in the density of states.

Theoretical interpretation of these results includes competing SDW and superconducting order [9,10], a Wigner crystal of Cooper pairs or a pair density wave [11,12], striped ordering [13], and d-density wave checkerboard order [14]. More recently, a phenomenological octet model of quasiparticle scattering from the Fermi arcs [15] has gained popularity in interpreting STM and scanning tunneling spectroscopy (STS) results [16]. The octet model fits some STM, STS, and angle-resolved photoemission spectroscopy [17] experiments; however, this model does not account for the neutron scattering results [7] or STM on NaCCOC [5]. It has been suggested [18] that, by increasing the applied magnetic field, magnetic order might be "tuned" and further suppress superconducting order. Since NMR is a bulk, spin-sensitive probe, and can be

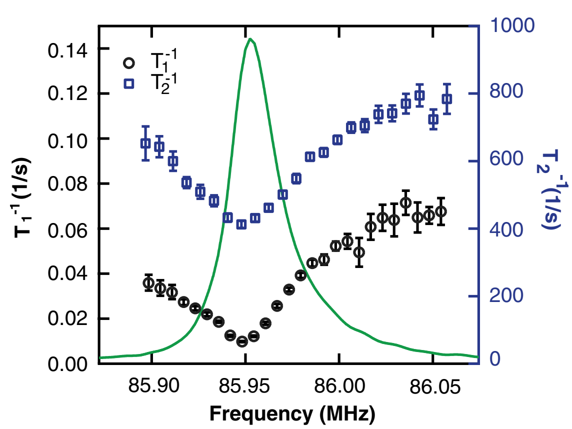

FIG. 1 (color online). The local field distribution and relaxation rate distribution at $H = 14.9$ T. The relaxation rates across the ^{17}O NMR spectrum (green curve) at $T = 4.2$ K for a Bi2212 single crystal. At low frequency there is an unexpected increase in the spin-lattice relaxation rate, T_1^{-1}, mirrored in spin-spin relaxation, T_2^{-1}.

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performed up to high field, it is ideal for investigation of the magnetic vortex states of high-temperature superconductors. In the present work, we exploit these capabilities of NMR in addition to its potential for spatial resolution.

At low temperatures, vortices in Bi2212 form a solid [19]. The vortex lattice has a well-defined variation of local magnetic field with position in the vortex unit cell. Mitrović *et al.* [20] have shown that NMR relaxation in YBCO powders can be resolved spatially by measuring the relaxation at different positions in the NMR spectrum, thus in real space. The relaxation varies across the spectrum and, at low temperatures, it is dominated by the Doppler shift of nodal quasiparticles with the highest frequency portion of the NMR spectrum corresponding to the vortex core [20–22]. The Doppler contribution to the electronic excitations is proportional to the supercurrent momentum p_s and is constrained to each superconducting plane. Since SLR depends on p_s^2, it is strictly two dimensional, a decided benefit in comparison with measurements of the local field which is averaged in three dimensions. Most importantly, SLR has strong spatial contrast, increasing monotonically with the inverse square distance from the vortex core, $\sim r^{-2}$. Taking advantage of this spectroscopy, together with measurements of the NMR spectrum of local magnetic fields, we are able to characterize the magnetic SDW associated with vortices in our sample, consisting of a slightly overdoped, single crystal of the anisotropic high-temperature superconductor, Bi2212.

Our sample was postprocessed by isotope exchange in 1 bar of flowing ^{17}O ($\approx 90\%$ enriched) at 600 °C for 48 h followed by annealing for 150 h at 450 °C resulting in a superconducting transition temperature $T_c = 82$ K. Spatially resolved T_1 and T_2 measurements were taken by Fourier transforming progressive saturation and Hahn echo sequences, respectively, with 16-pulse, phase alternation, at $T = 4.2$ K over a wide range of applied magnetic field, $4 < H \leq 30$ T. Spectra from the central transition of the planar oxygen O(1) were acquired for various recovery times and then the frequency domain of each one was subdivided into 32 intervals. The recovery profiles in each interval were fit to a progressive saturation recovery [21] for T_1 and an exponential for T_2. Experiments were performed at the National High Magnetic Field Laboratory in Tallahassee, Florida, and at Northwestern University, Evanston, Illinois. Further experimental details can be found elsewhere [23,24].

In the case of an ideal, nonmagnetic, superconductor with straight vortex lines that form a two-dimensional lattice, the lowest frequencies of the NMR spectrum come from the resonant nuclei in the sample positioned farthest from the vortex cores; the highest frequency components are from the nuclei closest to the vortex cores where the field is highest; and the peak corresponds to a saddle point in the distribution of frequencies $\omega = \gamma H$, where γ is the nuclear gyromagnetic ratio. This relation between spatial positions in the vortex lattice unit cell and the NMR frequency is well established [25]. However, contributions to the local fields from magnetism associated with vortex cores must be accounted for separately.

A clear indication that such contributions exist in Bi2212 is evident from the spectrum of the central transition for the O(1) planar site of ^{17}O NMR, Fig. 1, and in the linewidths, Fig. 2. We find the distribution of local magnetic fields to be much greater than that expected for vortex supercurrents alone; see the inset Fig. 2. Moreover, the linewidth increases systematically with applied magnetic field, rather than decreasing as would have been the case for supercurrents. Secondly, in Figs. 1 and 3, the SLR rate T_1^{-1} is strongly nonmonotonic across the spectrum contrary to predictions [26] for *d*-wave superconductors without magnetic vortices. Since SLR is from Doppler-shifted, nodal quasiparticles, it must increase smoothly and monotonically with decreasing distance approaching the vortex core. Thirdly, it is expected that the dominant interaction for spin-spin relaxation is from vortex vibrations which are also determined by vortex supercurrents [27]. Although there is not yet a quantitative theory for spin-spin relaxation, nonetheless it should mimic SLR. From Fig. 1, the strong similarity between the two rate profiles indicates that this is correct. As with SLR, we find that the spin-spin relaxation rate is not a monotonic function of local field and so we conclude that there must be both positive and negative local fields near the vortex core.

In order to model the measured local field distributions, we construct, analogous to STM observations in the charge channel, a spin checkerboard cosine wave decaying as a Gaussian centered on the vortex core,

$$B_{\rm SDW}(x, y) = A_{\rm SDW} \cos(2\pi x/\lambda) \cos(2\pi y/\lambda) e^{[-(x^2+y^2)/2\tau^2]}, \tag{1}$$

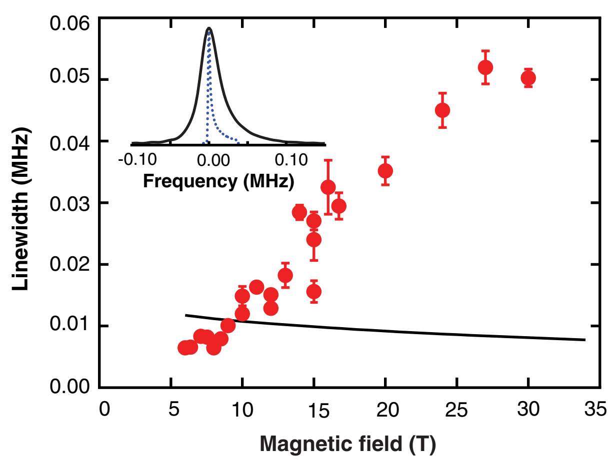

FIG. 2 (color online). The NMR linewidth, defined as the square root of the second moment of the spectrum, versus applied magnetic field at $T = 4.2$ K. The contribution from supercurrents was calculated from GL theory [28] (solid curve) and was subtracted from the data points. Inset: The experimental spectrum (solid, black curve) is much broader as compared to the calculated spectrum (dashed, blue line) for a perfect vortex lattice at $H_0 = 14.9$ T and $T = 4.2$ K.

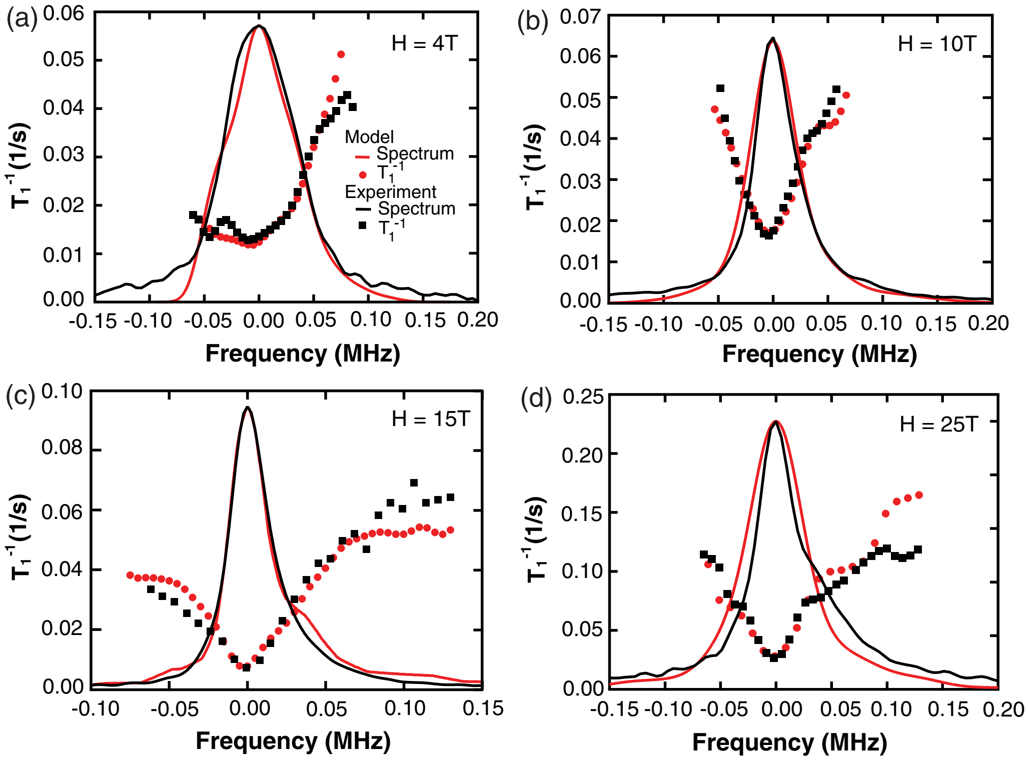

FIG. 3 (color online). The calculated [gray (red)] and experimental (black) results for the spectra (curves) and relaxation rates (dots, model; squares, experiment) for various fields, (a)–(d) for ($H = 4, 10, 15, 25$ T). At low magnetic field there is a near monotonic relationship for T_1^{-1} as a function of frequency (local field) as expected for the case $A_{\rm SDW} \le A_{\rm dia}$. At high magnetic fields the SDW dominates producing a nonmonotonic distribution of relaxation rates. The spectra are normalized to peak height and the calculated T_1^{-1} is offset vertically and scaled to compare with experiment.

where $A_{\rm SDW}$ is the amplitude of the magnetization at the vortex core, τ is its decay length, and λ is the period of modulation. An example is given in the inset of Fig. 4.

We fit the NMR spectrum of local fields, $B(x, y)$, including the contribution from diamagnetism of the vortex supercurrents, $A_{\rm dia}(x, y)$, for each position in the vortex unit cell: $B(x, y) = B_{\rm SDW} + A_{\rm dia}$. The supercurrent diamagnetism $A_{\rm dia}$ was calculated from Ginzburg-Landau (GL) theory for an ideal vortex lattice [28]. Holding λ constant at 30 Å, as suggested by STM [3] and neutron scattering [6–8], we obtained $A_{\rm SDW}$ and τ from a fit of this model to the spectrum for all magnetic fields, Fig. 4. With increasing applied field H there is an increase in $A_{\rm SDW}$ and a nearly constant τ, approximately twice the superconducting coherence length, $\tau \approx 2\xi_o \approx 22$ Å.

Next we calculate the SLR rate at each position in the vortex unit cell. The Doppler term [20], proportional to p_s^2, is determined from GL theory [28]. Then we associate the relaxation rate at each position in the unit cell with the corresponding local magnetic field from the NMR spectrum. Multiple values of the calculated SLR rate, within each of 32 contiguous intervals of local field, were averaged and then presented as a single value to be compared with the SLR data in Fig. 3, offset vertically to coincide with the measured rate at zero frequency. At high magnetic fields, $H > 20$ T, where the SDW dominates vortex diamagnetism, $A_{\rm SDW} > A_{\rm dia}$, we found $\lambda = 30 \pm 5$ Å for our SLR data by fixing $A_{\rm SDW}$ and τ to the values in Fig. 4 and allowing λ to be a free parameter when fitting both SLR and spectra. With our model we capture an excellent

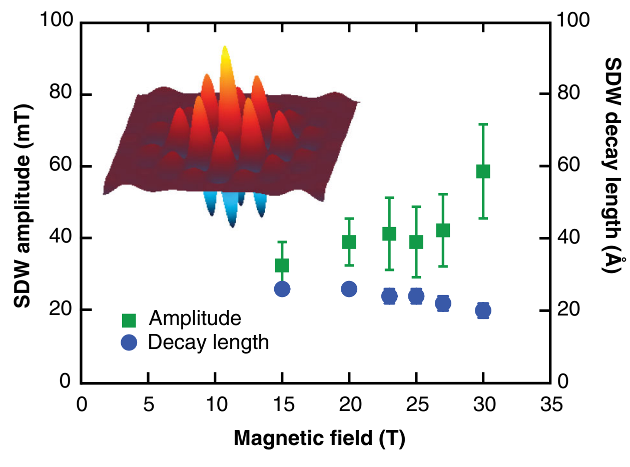

FIG. 4 (color online). The amplitude $A_{\rm SDW}$ (squares) and decay length τ (circles) for the best fit from Eq. (1) to our measured spectra. There is an increase in $A_{\rm SDW}$ with increasing magnetic field while τ is nearly constant. Inset: The spatial distribution of the local magnetic fields near a vortex within a CuO_2 plane calculated from Eq. (1) for a spin-density wave with these parameters at $H = 15$ T.

representation of both the field distribution and the unusual nonmonotonic behavior of the SLR rate as a function of frequency, i.e., local field.

There are three ranges of applied magnetic field which characterize different profiles of T_1^{-1} across the NMR spectrum, depending on the relative contributions of the diamagnetic local fields $A_{\rm dia}$ and the vortex-induced magnetic contribution $A_{\rm SDW}$. At low magnetic fields, 4–6 T, where $A_{\rm SDW} < A_{\rm dia}$, T_1^{-1} resembles the near monotonic behavior expected for a d-wave superconductor without magnetic vortices [26]. The spectrum width is larger in this range owing to a vortex instability, possibly associated with vortex charge [23]. Upon increasing the external magnetic field to 6–12 T there is a sharp increase of T_1^{-1} on the low frequency side of the spectrum where $A_{\rm SDW} \gtrsim A_{\rm dia}$. Finally, at high magnetic fields, 15–30 T, there is a significant increase of T_1^{-1} on both low and high frequency sides of the spectrum corresponding to $A_{\rm SDW} \gg A_{\rm dia}$. Measurements on YBCO [21,29] have revealed a small upturn in T_1^{-1} at low frequency, but much less pronounced than what we report here for Bi2212. In the YBCO work, contamination on the low frequency side of the spectrum from quadrupolar transitions precluded an unobstructed view of the magnetic field distributions over the same wide range as is possible [19] with Bi2212. It is likely that vortices in YBCO are accompanied by a SDW pattern which was not resolved in these earlier NMR experiments.

It is instructive to compare the linewidth for ^{17}O NMR reported here with that measured in YBCO in the normal state for various cations (Zn, Ni, and Li) substituted for copper in the CuO_2 plane producing an impurity local moment that generates an oscillatory local magnetic field [30]. The ^{17}O NMR broadening we observe in Bi2212 is similar in magnitude to that for Zn in YBCO scaled to the same field and temperature. In this sense vortices can be viewed as an impurity in a d-wave superconductor. In fact, a weak impurity effect has been reported for pure Bi2212 [31], and was identified with interstitial oxygen, disappearing in the superconducting state as does the Knight shift.

In summary we have shown that the local field distribution in the mixed state of Bi2212 is not that of an ideal superconductor and has substantial magnetic contributions. Furthermore, the well-defined correlation between NMR relaxation and position in the spectrum requires that these additional contributions be closely associated with vortices. In fact, the nonmonotonic behavior of the relaxation rate profiles can only be understood if the vortex local fields are oscillatory. We find that a spin-density wave model for vortex magnetism can completely describe both field distribution (spectrum) and supercurrent distribution (SLR) and thereby characterize the vortex, spin-density wave state in Bi2212. Our results establish a link between previously reported checkerboard STM patterns [3] and spin-dependent elastic neutron scattering in a cuprate superconductor [7].

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