Angular Dependence of the Nonlinear Transverse Magnetic Moment of $YBa_2Cu_3O_{6.95}$ in the Meissner State

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The angular dependence of the nonlinear transverse magnetic moment of untwinned high-quality single crystals of $YBa_2Cu_3O_{6.95}$ has been studied at a temperature of 2.5 K using a low frequency ac technique. The absence of any signature at angular period $2\pi/4$ is analyzed in light of the numerical predictions of such a signal for a pure $d_{x^2-y^2}$ order parameter with line nodes. Implications of this null result for the existence of a nonzero gap at all angles on the Fermi surface are discussed. [S0031-9007(99)08921-8]

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Measurements of the low energy quasiparticle excitation spectrum of high temperature superconductors are crucial in elucidating the nature of the pairing state and gap function in these materials. For a $d_{x^2-y^2}$ order parameter, the gap function varies as $\Delta(\varphi) = |\Delta_0 \cos(2\varphi)|$, riding on a cylindrical Fermi surface in momentum space, with zeros or line nodes at $\varphi = (\pi/4 + n\pi/2)$. Here, Δ_0 is about 25 meV [1] for $YBa_2Cu_3O_{6.95}$ (YBCO), and φ is the angle measured from the crystallographic a or b directions. The density of states for quasiparticle excitations near these nodes on the Fermi surface increases linearly with energy. In a two fluid model, this results in the superconducting condensate density $n_s(T)$ decreasing linearly with increasing temperature from its zero temperature value $n_s(0)$ [2]. In conventional s-wave superconductors, there is a nonzero gap everywhere on the Fermi surface for quasiparticle excitations, and the depletion of the condensate is exponentially small at the lowest temperatures.

The temperature dependence of the in-plane London penetration depth [3] $\lambda(T)$ in these materials has been widely cited as evidence in support of the presence of line nodes in the gap function, though at the lowest temperatures there is some deviation from the expected linear behavior, even in the best samples [4]. Measurements of the magnetic field dependence of the low temperature specific heat have also been interpreted as evidence for the presence of nodes, but the analysis is subject to the limitations of a many-parameter fit [5]. Data from later experiments, taken over a wider range of temperature and field do not yield the same conclusions upon analysis [6]. Recent studies on the scaling of specific heat data has also been cited as evidence in support of nodes [7], but the scaling works poorly, and only over a limited temperature range. Among experiments that provide angular information, angle-resolved photoemission experiments can be interpreted as showing a minimum in the energy gap in the (110) direction in YBCO and BSCCO ($Bi_2Sr_2CaCu_2O_8$) [1,8], but have a resolution of only about 5 meV (1 meV = 11.62 K) and thus cannot resolve excitations or an underlying s-wave gap at the lowest energies. Inelastic neutron scattering shows evidence in support of gap anisotropy [9], but there, too, the resolution is not nearly enough to distinguish between a pure d-wave gap, and one with quasinodes at the level of a few meV or less. Thus, these experiments may at best set an *upper bound;* i.e., they *cannot* distinguish between nodes and quasinodes with a minimum gap at a level of 3% or less of the maximum gap Δ_0, and one can safely say that the overall picture is quite ambiguous regarding the possibility of an underlying gap below 1 meV.

In this Letter, we describe an experimental technique for distinguishing nodes from deep minima (quasinodes) in the order parameter that also provides information about the angular position of these nodes or quasinodes, by probing the existence of low lying excitations in response to an applied magnetic field in the Meissner regime. Here, the kinetic energy of the superflow of the screening currents provides the energy for quasiparticle excitations. Our null result for this probe rules out the existence of nodes, and allowing for quasinodes, sets a *lower bound* on the size of the underlying gap.

For a type-II superconductor in a magnetic field in the Meissner regime, screening supercurrents flow in a volume near the surface given approximately by the penetration depth λ. For the condensate participating in these currents, the quasiparticle excitation spectrum is modified by a semiclassical "Doppler shift" to $E(k) = \sqrt{(\Delta_k^2 + \epsilon_k^2)} + \hat{v}_s \cdot \hat{v}_F$ [10], where $\hat{v}_s$ is the superfluid flow field and $\hat{v}_F$ is the Fermi velocity. For a gap function with nodes, this leads to quasiparticle excitations even at zero temperature [11]. Because of the linear density of states near a line node, the depletion of the condensate due to quasiparticles is proportional to $(\hat{v}_s \cdot \hat{v}_F)^2$. These quasiparticles create a "backflow" which is then responsible for a nonlinear contribution to the magnetization that goes as $H^2\lambda^2$, where H is the applied magnetic field, and $v_s \sim H\lambda$. In the case of a $d_{x^2-y^2}$ order parameter, the gap function has a fourfold angular symmetry, and this gives rise to an intrinsic nonlinear *transverse* magnetization, superimposed on the nominal diamagnetic response [12]. Since this effect is felt only by a fraction of the condensate within a thickness of λ of the surface, the nonlinear magnetic moment is proportional to the surface area of the sample, and not the full sample volume. Also, since $\lambda \gg \xi$, where ξ is the in-plane coherence length, this is a bulk effect.

We are interested in the nature of the gap function for supercurrents flowing within the Cu-O planes of the superconductor, which consists of layers of these planes. The YBCO crystals are flat with the c axis oriented perpendicular to the crystal plane which is the a-b plane. The magnetic field is applied parallel to the crystal plane and the transverse magnetic moment is measured in the crystal plane perpendicular to the applied field. As the crystal is rotated in the applied magnetic field about the c axis, the screening currents flow in different directions relative to the in-plane gap function, which is pinned to the crystal lattice. This leads to an angular modulation of the nonlinear transverse magnetic moment that provides *angle-resolved* information about the low lying excitations in YBCO.

Numerical calculations of the nonlinear transverse magnetic moment (m_T) of a sample with a finite disk shaped geometry have been carried out for pure d wave and mixed order parameters. For pure d wave, the calculation predicts the amplitude of the expected fourfold modulation of m_T with angular period $\pi/2$ [12], a consequence of the symmetry of the gap and angular position of the nodes. The presence of a small s-wave component as in $d + s$ changes the angular position of the line nodes, and introduces a component in the angular modulation of m_T with period π [13]. However, the angular period $\pi/2$ component is not adversely affected by small additions of s wave. Anisotropy of the Fermi surface and anisotropy in λ_a and λ_b have also been considered, and have very similar consequences. For experimentally determined values of such anisotropy [14], there is a small additional angular period π component, but the period $\pi/2$ component is essentially unchanged. On the other hand, for a d-wave-like order parameter, with varying levels of a nonzero quasinode, as in $d_{x^2-y^2} + is$ and $d_{x^2-y^2} + id_{xy}$ symmetries, the amplitude of the period $\pi/2$ component is suppressed. In addition, to prevent the nonlinear Meissner effect (NLME) from being thermally washed out, the temperature has to be such that $\frac{T}{\Delta_0} < \frac{H}{H_o}$, where $H_o = \frac{\phi_o}{\pi^2\lambda\xi}$, and ϕ_o is the flux quantum. For $\lambda_{ab} = 1400$ Å and $\xi_{ab} = 20$ Å, which are typical values, $H_o \approx 8000$ Oe. For measurements in a field of amplitude 300 Oe, this yields $\frac{H}{H_o} = 0.0375$, and requires that $T \lesssim 10$ K.

To measure m_T, an ac magnetic field is applied in the a-b plane of a mm size single crystal of YBCO. The field is modulated at 12 Hz, and the transverse magnetic moment at 36 Hz is detected by the transverse coil of a quantum design SQUID susceptometer. The signal is expected at the third harmonic because $m_T \sim \mathrm{sgn}(H)H^2$. The sample is maintained at 2.5 K with continuous cooling throughout the measurements. The superconducting magnet is operated with its persistent-current switch open, and the ac magnetic field is generated by driving a very low distortion (< -110 dB in higher harmonics) and low-noise ac current through the magnet coil, with a maximum amplitude of 300 Oe. The sample is placed in the center of the magnet coil and aligned optimally with the transverse flux detection coils. The analog output from the transverse SQUID is fed directly to a phase sensitive detection system. The large 12 Hz background from the fringing field of the magnet and geometric demagnetization fields from the sample are rejected by a high-pass filter section. The signal at 36 Hz is then detected with a phase sensitive detector locked to $3f$ of the current generator. The sample is rotated *in situ* between measurements in steps of 6° using a custom built sample holder that allows for *in situ* detection of the angular position of the sample at low temperatures. This system has been described in detail elsewhere [15]. It allows for precision in angular positioning of about 0.1° between steps and accuracy of better than 1° in an entire rotation. The sample holder and associated angular detection systems are designed to provide minimal magnetic background.

The detection setup has been calibrated and tested by "simulating" a real magnetic moment in an environment identical to that of the actual experiment. This is done by running an ac current through a copper coil mounted on the sample stage at a frequency $3f$, referred to the frequency $f = 12$ Hz of the oscillating magnetic field, which has an amplitude of 300 Oe. The ac moment of 1.5×10^{-8} emu rms amplitude at $3f$ is detected in the presence of the large background signal at frequency f due to the fringing field of the magnet. This calibration was done at 2.5 K, with just the bare sample holder. The measured amplitude is shown in Fig. 1 as a function of the angular orientation of the coil. The angular Fourier transform of these data (inset) shows the amplitude at angular period 2π. The "noise floor" in the Fourier transform of the calibration signal at higher angular frequencies corresponds to an amplitude less than 5×10^{-10} emu.

The angular dependence of the NLME has been measured in a number of untwinned single crystals of YBCO, including a very high-quality crystal (UWHC), grown in a YSZ crucible by the Argonne group. This crystal, which is 99.95% pure, has been cut into a disk shape with diameter 1.5 mm, and is 67 μm thick. The high quality of the crystal is borne out by a rocking curve with full width at half maximum of 0.08° for the 006 peak of a high resolution x-ray scan, and there were no twins observable in repeated area scans over different parts of the crystal. The superconducting transition of this crystal has an onset of 93 K and a width of less than 2.5 K as measured magnetically with a 10 Oe field applied in the *a-b* plane. The field of first flux entry was measured to be about 300 Oe in the *a-b* plane at 2.5 K.

Measurements of m_T are shown in Fig. 2 as a function of the angular orientation of the sample with respect to the applied magnetic field, with the "*a*" direction being initially oriented along the field. The sine Fourier transform of the data is shown in the inset. For a pure *d*-wave order parameter, we are interested in the angular period $2\pi/4$ component of m_T, the predicted value for which is 1.7×10^{-9} emu according to the calculations in Ref. [12]. This component is clearly in the noise of the data, below 5×10^{-10} emu. This "noise" is in part due to residual trapped flux, and also due to the noise floor of the measurement apparatus, as has been verified in repeated measurements. This imposes an upper bound on the size of the nonlinear Meissner effect, which, if there, is less than 30% of the predicted value. Identical measurements have also been carried out on a very high-quality rectangular crystal (UBCyca) grown by the UBC group in a BaSZ crucible [4]. It has the same surface area as our disk shaped crystal (UWHC) and the results were essentially the same.

This null result has to be examined in the light of measurements of the penetration depth. Scrutiny of data on the temperature dependence of the penetration depth $\lambda(T)$ indicates that there is always some curvature away from the pure *d*-wave result at the lowest temperatures. This is found in all published data on the cleanest bulk single crystals [3,4], and has been attributed to the presence of unitary scattering centers, that do not affect T_c [16,17]. However, even with the concentration of unitary scatterers needed to produce the required curvature, the nonlinear transverse magnetic moment effect may only be reduced to about 90% of the full value [11], and our data indicate a much stronger suppression. This same data for $\lambda(T)$ for UBCyca [18] can be fit at the lowest temperatures by a model which contains quasinodes, i.e., where the $\frac{\Delta_{\min}}{\Delta_o}$ is 2.5%, $\Delta_{\min}$ being the residual gap in the nodal direction. Such a fit is significantly better at the lowest temperatures than the fit for a pure *d*-wave order parameter. The suppression of the nonlinear transverse magnetic moment in all our measurements is consistent with $\frac{\Delta_{\min}}{\Delta_o}$ being *at least* 2%–3%. In the event that the $\lambda(T)$ and photoemission results can be explained by something other than a minimum in the gap, our experiment is also consistent with a $d + s$ order parameter where $\Delta_s > \Delta_d$.

Recently, other experiments that have attempted [19,20] to see the NLME in measurements of the field dependence of λ at low temperatures, but have not obtained conclusive results. According to theory [11], for a pure *d*-wave order parameter, λ should vary linearly with H at the lowest temperatures where $\frac{T}{\Delta_0} < \frac{H}{H_o}$. These most recent measurements were done at a very high level of sensitivity, about 100 times better than an earlier effort [21]. In one of these experiments done at UIUC [19], a significant field dependence in λ was observed, but this was attributed to trapped flux, as the field dependence was closer to $\sqrt{H}$, and the temperature dependence was not as predicted by theory. The experimental effort at UBC [20] observed a field dependence in λ, and also measured the effect as a function of the in-plane angular orientation of the applied field. However, they report that the field dependence

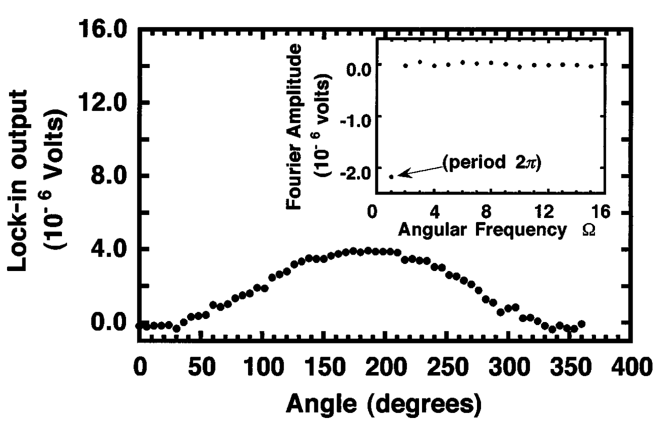

FIG. 1. Sensitivity of the measurement apparatus to a magnetic moment of 1.5×10^{-8} emu at $3f$. The amplitude at period 2π is shown in the cosine Fourier transform (inset) and is 2.2 μV.

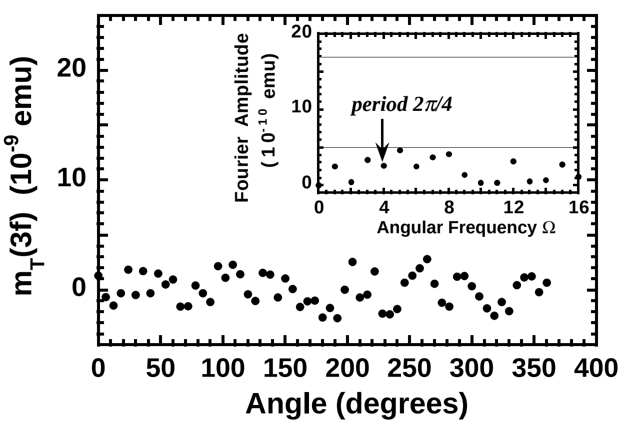

FIG. 2. $m_T(3f)$ as a function of angle with respect to applied magnetic field for UWHC YBCO sample. The initial orientation is with the "*a*" direction parallel to the applied magnetic field. The *absolute* value of the sine Fourier amplitudes are plotted in the inset. The expected amplitude at angular period $2\pi/4$ modulation for pure d wave is shown as a horizontal line at 17×10^{-10} emu. The level predicted for $\Delta_{\min}/\Delta_o = 2.5\%$ is shown by the horizontal line at 5×10^{-10} emu, near the noise floor.

measured at $\pm 45°$ to the crystal axes were not identical as would have been expected from theory, and the temperature and field dependence of the effect could not be understood within the current picture for the NLME [11].

Earlier attempts at measuring the angular dependence of the nonlinear transverse magnetic moment using a dc technique [22] were less sensitive by more than 2 orders of magnitude than the present work due to the very large linear background in a dc measurement, and the analysis overestimated the size of the effect significantly. In our experiments, trapped flux may produce a signal, but the angular dependence of this effect has the largest amplitude at angular period 2π, and is easily distinguished from the period $2\pi/4$ modulation for which we are searching. The angular modulation arising from the geometric demagnetization factor is linear in field below the field of first flux entry, and is minimized by looking at only the nonlinear component which is extracted directly in our work. Thus, artifacts due to trapped flux and geometry are both minimized by our technique.

There have been some calculations of the effects of nonlocal electrodynamics [23,24]. These are motivated by the idea that in a BCS-like picture, when the gap Δ_k goes to zero, then $\xi = \hbar v_F/\pi\Delta_k$ diverges, and one might no longer be in the local limit where $\xi < \lambda$. However, it turns out that these considerations are relevant only when $\hat{H} \| \hat{c}$. In our experiment, $\hat{H}$ is applied in the a-b plane, and the volume within a depth λ of the a-b plane crystal face that is responsible for the nonlinear Meissner effect is not affected by nonlocal effects. Even if we consider a "weakly 3D" case [24], the effect is at a field scale of about 20 Oe, far too small to suppress the NLME, whose characteristic field in our measurements is 300 Oe.

In summary, in view of the results of our measurements, there is a *minimum gap* of *at least* 2%–3% of Δ_0 everywhere on the Fermi surface of YBCO. However, in light of the evidence for a d-wave-like order parameter [25] from other experiments, and the constraints set by the data on $\lambda(T)$, it is possible that the order parameter in YBCO may still be d-wave-like, but have quasinodes instead of line nodes, with $\frac{\Delta_{\min}}{\Delta_o}$ between 2%–3%. Measurements of the penetration depth to lower temperatures or photoemission experiments at higher resolution may confirm this.

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