Nonlinear Meissner Effect in CuO Superconductors

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Recent theories of the NMR in the CuO superconductors are based on a spin-singlet $d_{x^2-y^2}$ order parameter. Since this state has nodal lines on the Fermi surface, nonlinear effects associated with low-energy quasiparticles become important, particularly at low temperatures. We show that the field dependence of the supercurrent, below the nucleation field for vortices, can be used to locate the positions of the nodal lines of an unconventional gap in momentum space, and hence test the proposed $d_{x^2-y^2}$ state.

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Much of the interest in the CuO high-T_c superconductors is due to the belief by many theoreticians that these materials represent new, or at least novel, forms of superconductivity. Theories based on very different excitations (e.g., spinless fermions, anyons, and traditional charged fermions) have been proposed to explain the normal and superconducting properties of these materials. Superconductivity from anyons has received considerable attention. Similarly, several theories based on BCS-like pairing, either of novel excitations or of novel pair states of conventional charged fermions, have been proposed [1]. All of the theories of the superconducting state, except the conventional BCS pairing state, have the property that they break at least one additional symmetry of the normal state besides gauge symmetry. While there has been little compelling evidence to identify the order parameter of the high-T_c superconductors with any of these unconventional states, recent theories, based largely on interpretations of NMR in both the normal and superconducting states of $YBa_2Cu_3O_7$ (YBCO), have focused on the spin-singlet, $d_{x^2-y^2}$ state as the order parameter for these CuO superconductors [2, 3]. In this Letter we show that the field dependence of the supercurrent can be used to locate the positions of the nodal lines of the gap in momentum space; we suggest new experiments to resolve the gap structure of the $d_{x^2-y^2}$ state, or other unconventional BCS states.

Based on ideas from Hammel *et al.* and Shastry [4], a semiphenomenological model of a nearly antiferromagnetic (AFM) Fermi liquid that accounts for the anomalous NMR properties of the CuO superconductors has been proposed [5, 6]. Monthoux *et al.* [2] recently argued that the same AFM spin fluctuations that are responsible for the anomalous normal-state magnetic properties are also responsible for superconductivity in the layered CuO systems. These authors examine the weak-coupling gap equation with a pairing interaction given by the exchange of a single spin fluctuation, and find that the superconducting state with $d_{x^2-y^2}$ symmetry is the preferred solution.

Several authors [3] have extended the nearly AFM Fermi liquid into the superconducting state, and have concluded that the Knight shift and anisotropy of the spin-lattice relaxation rates in YBCO are most reasonably fit with a singlet, $d_{x^2-y^2}$ order parameter. Since the arguments put forward in favor of a $d_{x^2-y^2}$ order parameter are connected with the spin excitation spectrum, independent tests of the $d_{x^2-y^2}$ state based on the orbital structure of the gap or its excitation spectrum are needed to identify the order parameter.

The important feature of the quasiparticle excitation spectrum, which is required by symmetry for a clean superconductor with a $d_{x^2-y^2}$ order parameter, is that the excitation gap vanishes along four lines (nodal lines) located at the positions $|\hat{k}_x| = |\hat{k}_y|$ in the 2D plane and running along the length of the Fermi tube (see cross section in Fig. 1). These nodal lines imply low-energy excitations, at all temperatures, that give rise to power-law deviations of the penetration depth from its zero-temperature limit, i.e., $1/\lambda(0) - 1/\lambda(T) \sim (T/T_c)$ for $T \ll T_c$, and a gap with line nodes, while deviations proportional to higher powers are obtained from gaps with point nodes [7]. At present there is no consensus "for" or "against" a true gap in the spectrum from penetration depth measurements. Several measurements of the penetration depth in CuO superconductors are interpreted as supporting the predictions of BCS theory with a true gap, while the analysis by Annett *et al.* [8] of those

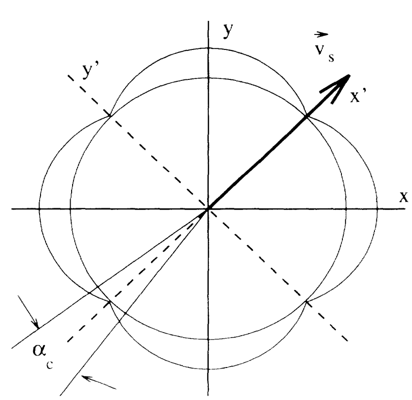

FIG. 1. Phase space for $\vec{v}_s \parallel \hat{x}'$.

same measurements (see also [8] for original references) suggests deviations from a true gap, but gives no indication of a linear temperature dependence that would be consistent with a $d_{x^2-y^2}$ state.

The main idea of this paper is that the field dependence of the supercurrent may be used to locate the positions of the nodal lines (or points) of an unconventional gap in momentum space. This is a stronger test of the symmetry of the superconducting state than power-law behavior for the penetration length. This gap spectroscopy is possible at low temperatures, $T \ll T_c$, and is based on features which are intrinsic to nearly all possible unconventional BCS states in tetragonal or orthorhombic structures. The constitutive equation relating the supercurrent, $\vec{j}_s$, to the velocity, $\vec{v}_s = \frac{1}{2}(\vec{\partial}\phi + \frac{2e}{c}\vec{A})$ (this definition avoids the introduction of an arbitrary mass scale), is linear only for velocities small compared with the critical velocity, $v_c = \Delta/v_f$. Since the unconventional gaps of interest have nodal lines on the Fermi surface [9], nonlinear corrections to the constitutive equations become important, particularly at low temperatures. The low-energy states near the nodes of the gap lead to a quasiparticle contribution to the current that (i) persists at zero temperature, (ii) is a nonanalytic function of the velocity, and (iii) exhibits a large anisotropy at low temperature that is determined by the *positions* of the nodes on the Fermi surface. We discuss the theory of the nonlinear current-velocity relation at low temperature, and then show how the nonlinearity in the current leads to a novel basal-plane anisotropy of the screening length and magnetization energy (or magnetic torque), for the $d_{x^2-y^2}$ state when the field is parallel to the CuO planes.

Consider a superconductor-vacuum interface which is parallel to the magnetic field, and assume the penetration length is large compared to the in-plane coherence length, $\lambda \gg \xi$. The important spatial variations are then contained in the local value for the superfluid velocity, which is effectively uniform on the scale of ξ. The order parameter achieves its equilibrium value for the local value of the velocity, and the resulting current is [10]

$$\vec{j}_s = -eN_f \int d^2s\; n(s)\; \vec{v}_f(s) \left\{\sigma_v(s) + \int_0^\infty d\xi\, \left[f(E+\sigma_v(s)) - f(E-\sigma_v(s))\right]\right\} -eN_f \int d^2s\; n(s)\; \vec{v}_f(s)\; \pi T \sum_n \frac{\sigma_v(s) - i\epsilon_n}{\sqrt{[\epsilon_n + i\sigma_v(s)]^2 + |\Delta(s)|^2}}\,, \tag{1}$$

where $\sigma_v = \vec{v}_f(s)\cdot\vec{v}_s$ is the shift in the quasiparticle energy due to the superflow, N_f is the total density of states at the Fermi level, $n(s)$ is the angle-resolved density of states normalized to unity, $\vec{v}_f(s)$ is the quasiparticle velocity at the point s on the Fermi surface, and $E = \sqrt{\xi^2 + |\Delta(s)|^2}$. The first line of Eq. (1) is useful for calculating the current in the low-temperature limit. The first term represents the supercurrent obtained from an unperturbed condensate, i.e., $\vec{j}_s = -e\rho\vec{v}_s$, where ρ is the total density ($\rho = \frac{1}{2}N_f v_f^2$ for a cylindrical Fermi surface). The term involving the Fermi functions is the current from the equilibrium population of quasiparticles. This formula shows clearly that the quasiparticles do not contribute to the current at $T = 0$ for a superconductor with a true gap over the entire Fermi surface, provided the velocity is below the critical velocity, $\Delta_{\min}/v_f$. At finite temperature, the excitation term is responsible for the reduction in the superfluid density from ρ to $\rho_s(T)$ in the linear response limit. In addition, the quasiparticles give rise to a nonlinear reduction in the supercurrent.

In order to motivate the proposal for detecting the nodal structure of the gap in an unconventional superconductor consider the effect of a magnetic field on the penetration depth in a conventional, isotropic superconductor. For a field parallel to the surface, the screening current is proportional to the applied field, i.e., $j_s(0) \sim cH/\lambda$. This linear relation breaks down because of pair breaking induced by the velocity field,

$$\vec{j}_s = \rho_s(T)\; \vec{v}_s \left[1 - \alpha(T)\left(\frac{v_s}{v_c}\right)^2\right], \tag{2}$$

for $v_s/v_c \ll 1$, where $\alpha(T) \geq 0$ and $v_c = \Delta(T)/v_f$ is the critical velocity. One important point to note is that $\alpha(T) \to 0$ for $T \to 0$, since the quasiparticle occupation is zero for all quasiparticles states with velocity below the critical velocity. The importance of the nonlinear current-velocity constitutive equation to the penetration of magnetic fields into a superconductor is qualitatively clear. The reduction of the current by the pair-breaking effect reduces the effective superfluid density, and, therefore, increases the effective penetration length. Since the current is proportional to the field in linear order, the correction to the effective penetration length is quadratic in the surface field [11],

$$\lambda_{\text{eff}}(T,H)^{-1} = \lambda(T)^{-1} \left\{1 - \frac{1}{3}\alpha(T)\left[\frac{H}{H_o(T)}\right]^2\right\}, \tag{3}$$

where $H_o(T) = \frac{3}{4}cv_c(T)/e\lambda(T)$ is of order the thermodynamic critical field.

Now consider the $d_{x^2-y^2}$ order parameter, $\Delta(s) = \Delta_o(\hat{k}_x^2 - \hat{k}_y^2)$. The nodes in the gap imply that there is a quasiparticle contribution to the supercurrent even at $T = 0$, which is easily calculated in terms of the phase space of occupied quasiparticle states. Consider the case where the velocity is directed along the nodal line $k_x = k_y$; $\vec{v}_s = v_s\hat{x}'$ as shown in Fig. 1. For any nonzero v_s there is a wedge of occupied states near the node opposite to the flow velocity. For a cylindrical Fermi surface the quasiparticle current is

$$\vec{j}_{\rm qp} = -2eN_f\,(-2\Delta_o v_f)\int_{-\alpha_c}^{\alpha_c}\frac{d\alpha}{2\pi}\cos^2(\alpha)\sqrt{\sin^2(\alpha_c)-\sin^2(\alpha)}\,, \tag{4}$$

where α is the angle measured relative to the node at $-\hat{x}'$ and $\alpha_c = \sin^{-1}(\frac{v_f v_s}{2\Delta_o})$ is the maximum angle for which the quasiparticle states near the node are occupied. To leading order in $(\frac{v_f v_s}{2\Delta_o})$ we obtain a total supercurrent of

$$\vec{j}_s = \left(-\frac{e}{2}N_f v_f^2\right)\vec{v}_s\left\{1-\frac{|\vec{v}_s|}{2\Delta_o/v_f}\right\} \tag{5}$$

for $\vec{v}_s$ directed along any of the four nodes. Note that the current is parallel to the velocity, and that the occupied quasiparticle states reduce the supercurrent, as expected. Also the nonlinear correction is quadratic rather than cubic, as is obtained for the conventional gap, and with the characteristic scale determined by $v_o = 2\Delta_o/v_f$. Furthermore, the quasiparticle contribution to the current is a nonanalytic function of v_s [12].

Unlike the linear response current, the nonlinear quasiparticle current is anisotropic in the basal plane. A velocity field directed along the maximum direction of the gap, $\vec{v}_s = v_s\hat{x}$, produces two groups of occupied states, albeit with reduced populations because the projection of $\vec{v}_s$ along the nodal lines is reduced by $1/\sqrt{2}$. The resulting current is easily calculated to be

$$\vec{j}_s = \left(-\frac{e}{2}N_f v_f^2\right)\vec{v}_s\left\{1-\frac{1}{\sqrt{2}}\frac{|\vec{v}_s|}{2\Delta_o/v_f}\right\}, \tag{6}$$

which is again parallel to the velocity and has a quadratic nonlinear correction. However, the magnitude of the nonlinear term is reduced by $1/\sqrt{2}$. This anisotropy is due to the relative positions of the nodal lines and is insensitive to the anisotropy of the Fermi surface or Fermi velocity because the quasiparticle states that contribute to the current, for either orientation of the velocity, are located in a narrow angle, $\alpha \le \alpha_c \simeq (v_s/v_o) \ll 1$, near the nodal lines. Thus, the occupied quasiparticle states near the nodes have essentially the same Fermi velocity and density of states; only the relative occupation of the states is modified by changing the direction of the velocity.

The dependence of the supercurrent on the positions of the nodal lines in momentum space suggests that the anisotropy can be used to distinguish different unconventional gaps with nodes located in different directions in momentum space. For example, the d_{xy} state, $\Delta \sim \hat{k}_x\,\hat{k}_y$, would also exhibit a fourfold anisotropy, but the nodal lines are rotated by $\pi/4$ relative to those of the $d_{x^2-y^2}$ state, while the state $\Delta \sim \hat{k}_x\hat{k}_y(\hat{k}_x^2-\hat{k}_y^2)$ corresponding to the A_{2g} representation would exhibit an eightfold anisotropy.

This anisotropy in the current implies a similar anisotropy in the field dependence of the penetration length, which can be calculated from Eqs. (5) and (6) and Maxwell's equation; in the gauge $\vec{\partial}\cdot\vec{v}_s = 0$, and for surface fields parallel to a nodal line, the equations reduce to

$$-\frac{\partial^2 v_s}{\partial z^2} = \frac{4\pi e^2}{c^2}\,j_s[\vec{v}_s] = -\frac{v_s}{\lambda^2}\left\{1-\frac{|v_s|}{v_o}\right\}. \tag{7}$$

The magnetic field in the superconductor, $\vec{b} = (c/e)\vec{\partial}\times\vec{v}_s$, is also parallel to a node, and has magnitude $b = (c/e)(\partial v_s/\partial z)$ with $b(0) = H$. For field penetration into a bulk superconductor we define the effective screening length in terms of the surface impedance, $1/\lambda_{\rm eff} = -(1/H)(\partial b/\partial z)|_{z=0}$. We obtain

$$\frac{1}{\lambda_{\rm eff}} = \frac{1}{\lambda}\left(1-\frac{2}{3}\frac{H}{H_o}\right), \quad \vec{H}\parallel \text{node}\,,$$
$$\frac{1}{\lambda_{\rm eff}} = \frac{1}{\lambda}\left(1-\frac{1}{\sqrt{2}}\,\frac{2}{3}\frac{H}{H_o}\right), \quad \vec{H}\parallel \text{antinode}\,, \tag{8}$$

to leading order in H/H_o, where $H_o = (v_o/\lambda)(c/e) \sim \phi_o/(\lambda\xi) \sim H_c$. Observation of this anisotropy and linear field dependence would be a strong indication of a $d_{x^2-y^2}$ order parameter.

Sridhar *et al.* [13] have measured the field dependence of the penetration length for $\vec{H}$ parallel to the a and b axes of single crystals of YBCO. These authors report an in-plane penetration depth which obeys $\lambda(H,T) = \lambda(T) + \kappa(T)H^2$. At intermediate temperatures, $T \simeq 80$ K, they measure $\kappa(80) \simeq 1$ Å/G^2, which drops by 3 orders of magnitude by $T \simeq 10$ K, $\kappa(4) \simeq 10^{-3}$ Å/G^2. The simplest interpretation is that the data are in good agreement with a full gap on the Fermi surface, in which case the $d_{x^2-y^2}$ state is not the order parameter of YBCO. However, we can ask if the data, so far, force this conclusion. First of all, the $d_{x^2-y^2}$ state will also show a quadratic field correction to the effective penetration depth at intermediate and high temperatures, with a coefficient that is similar in magnitude to that of the conventional BCS prediction, and also drops rapidly with temperature. The question is, "At what temperatures and fields does the effective penetration depth exhibit a linear field dependence?" The linear field regime is inferred from Eq. (1). The linear behavior arises from the nonanalytic dependence of $\vec{j}_s$ on $\vec{v}_s$ at $T = 0$ for the nodal directions where $\Delta(s) = 0$. Nonzero temperature removes the singularity. For sufficiently small fields an expansion in v_s is valid; however, the crossover field is determined when the smallest Matsubara frequency becomes comparable to the superflow kinetic energy shift per particle, i.e., $\pi T \simeq v_f v_s \simeq 2\Delta_o(H/H_o)$. This implies a crossover field, $H_x \simeq H_o(T/T_c)$. Thus, to obtain a significant linear field regime down to $H \simeq H_{c1}/10 \simeq 25$ G requires temperatures $T \le 2$ K. Thus, the experiments of Ref. [13], which are reported down to 10 K, are apparently at the high end of the temperature regime where a linear term might become observable.

We also consider the effect of impurity scattering on the linear field dependence of the effective penetration length. Quasiparticle scattering from impurities plays a similar role to that of temperature; the impurity lifetime removes the singular behavior of the current at sufficiently low velocities. The crossover field due to impurity scattering can be obtained by including the impurity scattering self-energy. The resulting current is given by Eq. (1) with the replacement of $\epsilon_n \rightarrow \tilde{\epsilon}_n = \epsilon_n + (\tilde{\epsilon}_n/2\tau)\langle 1/\sqrt{\tilde{\epsilon}_n^2 + |\Delta(s)|^2}\rangle$ where τ is the s-wave scattering rate in the Born approximation, and the average is over the Fermi surface. At zero temperature, impurity scattering leads to a new low-energy scale [14], $\tilde{\epsilon}_n = 4\Delta_o e^{-\pi\tau\Delta_o}$, which smooths out the linear behavior of the effective penetration length below the crossover field, $H_\tau = 2H_o e^{-\pi\tau\Delta_o}$. This condition is considerably less restrictive than that for the temperature. For YBCO with a mean-free path of $l/\xi = \pi\tau\Delta_o \simeq 10$ the crossover field is of order $2H_o e^{-10} \ll H_{c1}$. Thus, there should be no difficulty from impurity scattering in observing the linear behavior in the effective penetration length for moderately clean YBCO crystals.

Another test of the presence of nodal lines associated with a $d_{x^2-y^2}$ order parameter would be to measure the magnetic anisotropy energy, or magnetization torque, for in-plane fields. Consider a velocity field $\vec{v}_s = v_{x'}\hat{x}' + v_{y'}\hat{y}'$. The projections of the velocity along the nodal lines $\hat{x}'$ and $\hat{y}'$ imply two occupations of quasiparticle states of differing magnitudes, which generate the current

$$\vec{j}_{\rm qp} = \left(-\frac{e}{2}N_f v_f^2\right)\left[\left(\frac{v_{x'}^2}{v_o}\right)(-\hat{x}') + \left(\frac{v_{y'}^2}{v_o}\right)(-\hat{y}')\right]. \tag{9}$$

The important feature is that the current is not parallel to the velocity field, except for the special directions along the nodes or antinodes; neither is the magnetic field parallel to the applied field, $\vec{H}$. This implies that the magnetic energy, $U = -\frac{1}{8\pi}\int \vec{b}\cdot\vec{H}d^3x$, is anisotropic in the a-b plane. For a film in a parallel field, $\vec{H} = H(-\sin\vartheta\hat{x}' + \cos\vartheta\hat{y}')$, the magnetic anisotropy energy to leading order in H/H_o is [11]

$$U_{\rm an}(\vartheta) = -\frac{H^2}{2\pi}\left(\frac{H}{H_o}\right)\lambda A\,\Phi\,[\sin^3\vartheta + \cos^3\vartheta], \quad 0 \le \vartheta \le \pi/2\,, \tag{10}$$

where A is the surface area of the film and $\Phi = \frac{8}{3}[1 + \frac{1}{2}\cosh(\frac{d}{2\lambda})]\sinh^4(\frac{d}{4\lambda})/\cosh^3(\frac{d}{2\lambda})$, which is constant (equal to $\sim 1/3$) for $d \ge \lambda$, and varies as $\Phi \simeq \frac{1}{4}(d/2\lambda)^4$ for very thin films. The anisotropy energy is minimized for field directions along the nodal lines, and is maximum for fields along the antinodes. In order to suppress vortex nucleation, thin films with dimensions $d \le \lambda$ are desirable; the lower critical field for vortex nucleation is increased by roughly (λ/d) in a thin film. The optimum geometry might be a superlattice of superconducting-normal (SN) layers with an S-layer thickness $\xi \ll d < \lambda$. In this case the field at each SN interface is essentially the external field, and the anisotropy energy is enhanced by the number of S layers.

Torque magnetometry has proven extremely useful for measuring the anisotropy of the penetration lengths for current flow along the c axis compared to the basal plane [15]. In the linear response limit we do not expect anisotropy of the penetration depth for current flow in different directions in the basal plane; however, in-plane anisotropy of the current arising from nonlinear field corrections should be observable at low temperatures, if the superconducting state has an unconventional order parameter. For comparison, the maximum magnetic torque from Eq. (11) is $\tau \simeq (1/6\sqrt{3}\pi)H^2(H/H_o)A\lambda \sim 10^{-3}$ dyn cm/rad, for $H = H_{c1} = 250$ G, $A = (2000\ \mu\text{m})^2$, and $\lambda = 1400$ Å, which is small, but comparable to the smallest values of torque reported in [15].

In summary, we suggest that measurements of the field dependence of the penetration depth for fields parallel to the CuO layers at temperatures well below 4 K should provide an answer to the question of whether or not the gap in the CuO superconductors has a line of nodes. If so, torque magnetometry and/or the anisotropy of the low temperature penetration length could be used to locate the nodal lines on the Fermi surface, thus providing direct evidence for or against a $d_{x^2-y^2}$ order parameter for the CuO superconductors.

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