Optical Excitation of Quasiparticle Pairs in the Vortex Core of High-T_c Superconductors

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A far-infrared resonance has been observed in superconducting $YBa_2Cu_3O_7$ thin films in the presence of high magnetic fields. It corresponds to the quasiparticle pair creation process inside the vortex core. The resonance frequency is $\omega_0 = 1.3kT_c/\hbar$ with a linewidth $1/\tau \cong 0.6\omega$. This value for ω_0 implies, within BCS theory, a large energy gap in $YBa_2Cu_3O_7$.

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In type-II superconductors the magnetic field penetrates into the material in the form of a lattice of vortices each of which carries one quantum of flux Φ_0. Near the vortices the superconducting gap function $\Delta(r)$ is reduced and there exist low-lying quasiparticle states which can be thermally excited at temperatures much below the critical temperature T_c. The vortex lattice has been directly observed in both conventional [1] and high-T_c superconductors [2] and the quasiparticle density inside the vortex has been observed in scanning tunneling microscope (STM) measurements on conventional superconductors [3]. A discrete excitation spectrum was predicted by Caroli, de Gennes, and Matricon [4] and later investigated in greater detail by Bardeen *et al.* [5]. Spectroscopic measurements that probe the quasiparticle energy spectrum within the vortices have not been reported. The energy scale of the spectrum is set by the confinement energy $E_0 = \hbar^2/m\xi_0^2$ in the vortex core, where ξ_0 is the superconducting coherence length. For conventional superconductors $\xi_0 \cong 1000$ Å and the level spacing is too small to be measured by conventional techniques. For high-T_c materials, however, $\xi_0 \cong 20$ Å and $1 < m/m_e < 3$, so that $E_0 \cong 10$ meV and the vortex spectrum becomes accessible by far-infrared spectroscopy.

In this Letter we report the first spectroscopic observation of the quasiparticle pair creation process in the vortex core. The measurements are performed on epitaxial films of $YBa_2Cu_3O_7$ (YBCO). We find a vortex pair creation resonance at $\hbar\omega = 9.5$ meV and it has a linewidth of 5.5 meV. These quantities provide a measurement of the energy of the lowest quasiparticle state and its decay rate through an analysis based on the BCS theory.

In order to have transparency over a wide spectral range we used films grown by pulsed laser ablation on a Si(100) substrate sandwiched between a $\cong 800$ Å yttria-stabilized-zirconia (YSZ) buffer layer and a $\cong 400$ Å YSZ cap layer [6,7]. The YBCO film thickness $d \cong 600$ Å as estimated by calibrated growth time. Electrical resistance measurements give T_c of 87 K ($R=0$) with a transition width of ≤ 2 K. The patterned-line critical current densities of similarly grown films at 77 K are typically $J_c \cong 2\times10^5$ A/cm^2. X-ray rocking curve data demonstrate c-axis orientation of these films to within 0.6°.

We first measured the transmission of a thin film in zero magnetic field at 12 K using a fast-scan Fourier-transform Michelson interferometer with a resolution set to 1 cm^{-1}. Because of the very weak far-infrared signals transmitted through the YBCO film, 6000 spectra were taken and averaged in order to reduce the noise to an acceptable level. The transmission of the YBCO thin film on its substrate and that of a Si substrate (reference spectrum) with a buffer YSZ layer was measured using a sample holder designed to switch, *in situ*, the sample and reference in and out of the far-infrared beam. The ratio of these two transmissions gives the transmittance of the thin film which is shown in Fig. 1. The transmission coefficient of a film in the limit where the film thickness d is small compared with λ is given by $t = 2n/(n+1+Z_0\sigma d)$, where n is the refractive index of the substrate and $Z_0 = 4\pi/c$ is the impedance of free space. The low-frequency conductivity of a London superconductor in zero magnetic field is given by $\sigma_s = n_0e^2/m^*i\omega$, where n_0 is the particle density of the condensate. This form for the conductivity has been found to be appropriate for YBCO for frequencies below the gap frequency and at low temperatures from infrared reflectivity studies [8,9]. Absorptivity measurements indicate that the real part of the conductivity is small compared with σ_s but nonzero [10]. With σ_s only, the transmission of a film relative to the substrate is given by $T_0 = \omega^2/(\omega^2+\Omega^2)$, where $\Omega = 4\pi n_0 de^2[(n+1)m^*c]^{-1}$. This result is compared with a measurement of the transmission of a YBCO film in Fig. 1. The observed far-infrared transmission in Fig. 1 is consistent with the calculated transmission except for a 8×10^{-3} transmission background which we attribute to radiation leakage around the film for the configuration of

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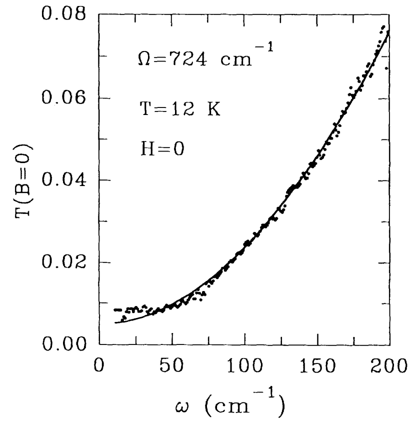

FIG. 1. Transmission of a thin YBCO film in absence of magnetic field ($H=0$) at $T=12$ K. The data (dots) are obtained by taking the ratio of the absolute transmission of the sample to a Si(100) substrate. The solid line is the calculated transmission for a thin superconducting film (as discussed in the text) using $\Omega=724$ cm^{-1}. The finite transmission at $\omega=0$ is due to a 0.8% light leakage around the sample for the geometry of this measurement.

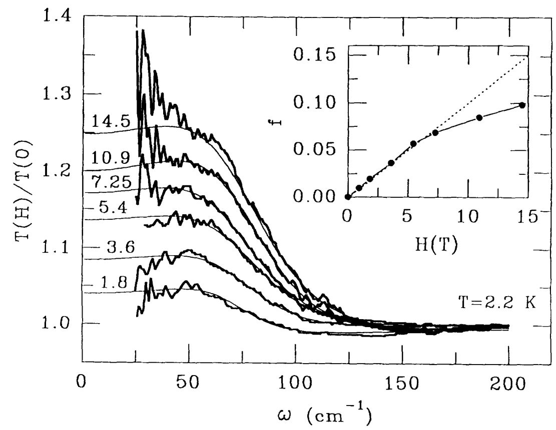

FIG. 2. Magnetotransmission measurements taken at 2.2 K and at several magnetic fields (H is expressed in tesla at the left of each spectrum). The data are obtained by taking the ratio of the transmission of the sample at H to the zero-magnetic-field transmission. The solid lines are given by a model calculation as discussed in the text and fitted to the experimental data using $\tau=8.5$ ps and $\omega_0 \cong 77$ cm^{-1}. The oscillator strengths f used in the model calculation to fit the data are shown in the inset as a function of H. The dashed line represents the fraction of vortex core area in the sample using $H_0=100$ T.

this measurement. Fitting the transmission data gives 724 cm^{-1} for Ω. The corresponding London length λ_L is consistent with the accepted value (1700 Å) in YBCO to within the uncertainty in the thickness of the film (20%).

We have performed far-infrared transmission measurements of this film at 2.2 K in magnetic fields up to 15 T. For these measurements the far-infrared radiation was guided to the sample using light pipe optics and the transmitted signal was detected by a 2.2 K bolometer placed below the sample and outside the magnetic field. In order to avoid the leakage signal that was observed in Fig. 1 we have mounted the sample on a 3-mm aperture which was much smaller than the geometric size of the sample. The sample substrate was mechanically wedged with a 3° angle in order to avoid optical interference effects. In Fig. 2 we present the results of these measurements in the form of the ratio of the transmission in magnetic field divided by the transmission at zero magnetic field $T(H)/T(0)$. It has proven difficult to obtain significant results for frequencies below 25 cm^{-1} because of the falloff of the Hg-vapor lamp source intensity and the ω^2 dependence of the transmission. $T(H)/T(0)$ displays an enhanced far-infrared transmission at low frequencies that increases with applied magnetic field. At high frequencies the transmission approaches the zero-field value to within our experimental uncertainty ($\pm 2\%$) due to instrumental drift. The enhanced transmission at low frequencies is qualitatively consistent with the reduction in the condensate density due to the presence of vortices. We interpret the broadened edgelike feature at 77 cm^{-1} as the *vortex quasiparticle pair excitation resonance*. To justify this interpretation we appeal to an analysis based on the BCS theory of the quasiparticle pair creation process in the vortex core.

To treat the optical properties of this system we take the 2D model (i.e., we take the c-axis mass m_c much greater than the in plane mass m_t) and examine the optical response of a single pinned vortex within the simple s-wave BCS theory. Therefore, we ignore the Abrikosov-lattice effects observed by Hess, Robinson, and Waszczak by STM measurements [11]. The bound quasiparticle states of the isolated vortex can be labeled by the angular momentum μ which takes on half-integer values [5]. The vortex bound quasiparticle spectrum is shown schematically in Fig. 3 where we have used the semiconductorlike representation. Within this picture the lowest excited states are the $\mu=\pm\frac{1}{2}$ states which occur

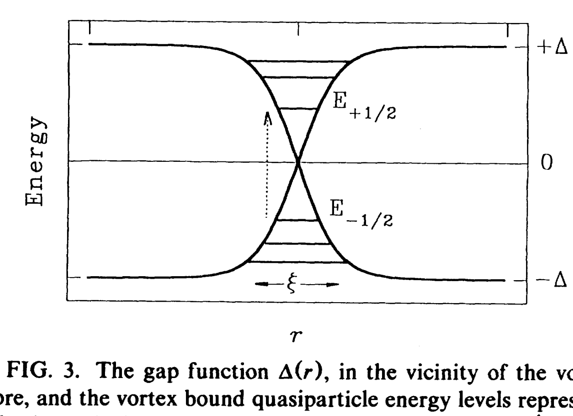

FIG. 3. The gap function $\Delta(r)$, in the vicinity of the vortex core, and the vortex bound quasiparticle energy levels represented schematically. The lowest excited states are $\mu=\pm\frac{1}{2}$ which occur at energies $\pm E_{1/2}$. The dotted vertical arrow represents the optical quasiparticle pair creation process. The quasiparticle states are confined on the scale of ξ_0.

at energies $\pm E_{1/2}$, where μ labels the angular momentum. Therefore the lowest energy for the creation of a pair of quasiparticles is $\hbar\omega = 2E_{1/2}$. From the theory we can write $E_{1/2} = K\Delta^2/E_F$, where Δ is the gap energy far from the vortex, E_F is the Fermi energy, and K is a dimensionless constant. Numerical calculations give $K \cong 0.75$ for the value of $E_F/\Delta \cong 7$ relevant for YBCO [12]. The value of K is relatively insensitive to E_F/Δ.

To treat the optical response we have calculated the conductivity using a Kubo formula approach. The calculated quantity is $\Sigma = \int \sigma d^3r$ for one isolated vortex. The result at finite frequencies in the lossless limit is $\Sigma = (\pi/\hbar\omega) N_0 \delta(\omega - 2E_{1/2}) |Q|^2$, where Q is the current matrix element for the transition and $N_0 = d/c_0$ is the density of states associated with the c-axis degree of freedom in the $m_c = \infty$ case, where c_0 is the unit-cell dimension. For finite m_c the density of states has a $(\omega - 2E_{1/2})^{-1/2}$ singularity due to k_c dispersion of the pair spectrum. The matrix element Q for the $\mu = -\frac{1}{2} \to \mu = \frac{1}{2}$ transition is nonzero only for the hole-cyclotron-resonance active mode of circularly polarized light (the carriers are assumed to be holes) [12]. Its magnitude is $|Q| = K_1 \hbar e k_F/2m_t$, where k_F is the Fermi momentum and K_1 is a dimensionless constant. We estimate $K_1 \cong 1.4$ from numerical calculations of the wave functions [12]. The only other transitions from the ground state allowed by angular momentum conservation involve the transitions to the continuum states above the gap [11].

Since the system is in the mixed state we must treat the optical response of an inhomogeneous system. We first define an average conductivity of the vortex, $\sigma_v = \Sigma/V_v$, where V_v is the vortex volume. The conductivity is spatially dependent, which leads to depolarization currents in the optical excitation process. This effect should be treated by a proper nonlocal calculation, such as the density-functional technique, which has been used for semiconductor heterostructures [13]. In this Letter we use a simpler technique which has proven adequate for heterostructures, giving results in good agreement with the density-functional approach and experiments [14]. In this approach we treat the single vortex as a cylinder of dielectric function ε_v embedded in the superconducting medium with dielectric function ε_s. The internal field in the vortex is then given by $E_v = E_s - LP$, where L is the depolarization factor and P is the polarization of the cylinder. For a cylinder, $L = 2\pi$ and we can express the internal field in terms of the field in the medium: $E_v = 2E_s(1 + \varepsilon_v/\varepsilon_s)^{-1}$.

For the superconducting medium $\varepsilon_s = 4\pi\sigma_s(i\omega)^{-1} = -(c/\omega\lambda_L)^2$, where λ_L is the London penetration depth [15]. For the vortex we take a lifetime-broadened Lorentzian centered at $\omega_0 = 2E_{1/2}$:

$$\varepsilon_v = \frac{4\pi}{i\omega}\sigma_v = \frac{4\pi n_v e^2/m_t}{\omega(\omega_0 - \omega + i/\tau)},$$

where n_v is the effective carrier density in the vortex. n_v is given by the optical strength of the $\mu = -\frac{1}{2} \to \mu = \frac{1}{2}$ process which is proportional to $|Q|^2$. The f-sum rule states that $\int \Sigma d\omega = \pi n e^2 V_v/2m_t$, where V_v is the effective volume of the vortex. To estimate the strength of the pair creation process we can integrate Σ to find n_v [12]. Using the BCS expression for the coherence length, $\xi_0 = \hbar v_F/\pi\Delta$, we have $n_v = \pi^2 K_1^2 d (k_F\xi_0)^2/8Kc_0V_v$. We can write $V_v = \pi(\alpha\xi_0)^2 d$, with $\alpha\xi_0$ as the effective radius for appreciable oscillator strength of the pair creation process. If we assume that the optical process exhausts the sum rule, we have $n_v = n_0$, and we obtain $\alpha \cong 4$. This shows that the effective volume of the vortex is considerably larger than the vortex volume $\pi\xi_0^2 d$. To obtain a more precise calculation of the optical strength it will be necessary to treat the nonlocal conductivity more carefully. The effective conductivity in the vortex is then

$$\sigma_{ve} = \frac{2\sigma_v}{1 + \varepsilon_v/\varepsilon_s} = \frac{n_0 e^2/m_t}{\beta/\tau + i(\omega - \beta\omega_0)},$$

where $\beta = n_0/(n_0 + n_v)$. We see that the resonance condition becomes $\omega_r = \beta\omega_0$. For what follows we assume that $n_v = n_0$, so that $\beta = 0.5$ and within these approximations the position of the resonance is $\omega_r = E_{1/2}$ which is the quasiparticle energy.

The effective average conductivity of the system is then $\sigma_{\text{eff}} = (1 - f)\sigma_s + f\sigma_{ve}$, where f is the fraction of the volume occupied by the vortices, and $f \propto H$. Using this form of σ_{eff} we have fitted the transmission data as shown as the solid lines in Fig. 2. The fitting parameters were ω_0, $1/\tau$, and f. The model is seen to provide an excellent description of the experiments. There are small systematic deviations at low frequencies which appear to be outside of the noise in the measurements. The downward deviation at low fields we understand in terms of the leakage signal T_L. Including the leakage effect in the fitting we find that $T_L \le 5 \times 10^{-4}$. The low-frequency positive deviations at high magnetic fields are not understood at this time. We have also used a Maxwell Garnett [16] effective medium theory (modified for cylindrical inclusions) to analyze these data. The calculated line shapes remain the same but the f values are a factor of 2 smaller. From the fits we find that $\beta\omega_0 = 77 \pm 3$ cm^{-1} and $\beta/\tau = 45 \pm 6$ cm^{-1}, nearly independent of H.

The inset in Fig. 2 shows the resulting f vs H. The dashed curve is based on $f = H/H_0$ with $H_0 = 100$ T which compares reasonably with estimates of H_{c2}. As the field is raised above 7 T it appears that the oscillator strength saturates. The theory presented here considers a completely pinned vortex model. Recently, Hsu has treated the case of damped vortex motion [17]. In this case the vortex motion must also be included in the optical response. This model also predicts a resonance for $\omega \cong \Delta^2/E_F$ but the selection rule for circular polarization is relaxed [17]. Moreover, Hsu finds that the oscillator strength depends on τ, the damping parameter, and vanishes in the absence of damping. Therefore the observed saturation of the oscillator strength may depend on the nature and distribution of the pinning centers in the film. We have also attempted to check the angular momentum selection rule as predicted by the pinned vortex model by performing transmission measurements with circular polarized light. The result is not yet conclusive, however, since the expected signal is found to be dominated by the magneto-optical activity of the condensate which will be reported elsewhere [18]. We conclude that further experimental and theoretical work will be required to establish the proper description of the vortex optical response.

The general shape of the curves in Fig. 2 are to be understood in terms of the modifications of the London screening due to the vortex excitation resonance since the transmission is dominated by $\mathrm{Im}(\sigma_{\mathrm{eff}})$. Therefore the line shape has an antiresonance rather than a resonance character. The observed quasiparticle relaxation rate, $1/\tau \cong 6$ meV, is only somewhat smaller than $E_{1/2}$. One possible mechanism for this large relaxation rate is scattering into the chain states.

We can use these results for ω_0 to obtain an estimate of Δ. From the equation for $E_{1/2}$ we get $\Delta^2 = E_{1/2}\hbar^2 k_F^2/2m_t K$. Taking 0.25 hole per unit cell [8] we can estimate $k_F \cong 3\times10^7$ cm^{-1} in a free-carrier model. Taking $m_t \cong m_e$ we find $\Delta \cong 65$ meV. We note that this implies, within BCS theory, a coherence length $\xi_0 \cong 13$ Å which is within the range of accepted values. The value of Δ, however, is somewhat larger than that estimated from other experiments [8,19]. Taking larger masses reduces Δ but then makes ξ_0 smaller than expected. These results should be considered as an indication that the simple BCS theory does not provide an adequate description for the high-T_c superconductors. However, the large excitation energy $E_{1/2}$ does suggest a large energy scale in these materials.

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