Far-Infrared Magneto-Optical Activity in Type-II Superconductors

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Transmission measurements on superconducting $YBa_2Cu_3O_7$ films with circular polarized light show optical activity proportional to magnetic-field strength. The results are discussed within the context of cyclotron (diamagnetic) resonance of the superconducting ground state. Hole conduction is implied.

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Isotropic interacting electron systems in the presence of a homogeneous static magnetic field respond resonantly to a uniform electromagnetic field only at the bare cyclotron frequency. This result was first pointed out by Kohn nearly thirty years ago [1]. Its validity in a variety of two- and three-dimensional electron systems has been well established over the intervening years [2–5]. A question that does not appear to have been addressed is whether the Kohn theorem applies to electron systems in a superconducting state. On the face of it the conditions of the theorem seem to be violated because the Meissner effect produces an inhomogeneous static magnetic field. In extreme type-II superconductors, however, when the magnetic field B is much greater than the flux quantum Φ_0 divided by the square of the London penetration length λ_L, the magnetic field is highly uniform. In this case $\delta B/B \ll \Phi_0/B\lambda_L^2$ and for $YBa_2Cu_3O_7$ at 10 T, for example, $\Phi_0/B\lambda_L^2 \cong 10^{-2}$. Other violations of the conditions of the Kohn theorem may arise from anisotropy of the Fermi surface, and the presence of defects. In normal Fermi liquids, however, the cyclotron resonance is very robust, occurring in electron systems with anisotropic energy bands [2,3] and in the presence of both elastic and inelastic scattering [4]. Therefore we conclude that looking for evidence of cyclotron resonance in type-II superconductors would be fruitful.

In this Letter we report preliminary results on far-infrared transmission measurements on $YBa_2Cu_3O_7$ films in high magnetic fields to test the applicability of the Kohn theorem in type-II superconductors. In designing the experiment we have been guided by the predictions of the lossless free carrier (LFC) model in the spirit of the Kohn theorem. In this case the finite-frequency conductivity of the film at $B=0$ is given by $\sigma_F = n_0 e^2/im^*\omega$, where n_0 is the carrier density. The functional form of σ_F is identical to that derived from the London model in which n_0 is the carrier density of the superconducting condensate. From far-infrared studies this form for the imaginary part of the conductivity σ_2 has been found to be appropriate for superconducting $YBa_2Cu_3O_7$ at low frequencies [6,7]. Absorptivity measurements indicate that the real part of σ is small compared with σ_2 but nonzero even in single domain single crystals for light polarized in perpendicular to the chains [6]. In the presence of a magnetic field the conductivity function, within the LFC (hole) model, becomes $\sigma_F^{\pm} = n_0 e^2/im^*(\omega \pm \omega_c)$, where $\omega_c = eB/m^*c$ is the cyclotron frequency and the plus sign corresponds to the electron cyclotron resonance (ECR) active circular polarization and the minus to the hole cyclotron resonance (HCR) mode. The transmission of a film on a substrate in the limit where the film thickness d is small compared with λ_L is given by $T = 4n/|n+1+Z_0\sigma d|^2$, where n is the refractive index of the substrate and $Z_0 = 4\pi/c$ is the impedance of free space. The transmission of a film on a substrate relative to the substrate is then given by $T^{\pm} = (\omega \pm \omega_c)^2/[(\omega \pm \omega_c)^2 + \Omega^2]$, where $\Omega = 4\pi n_0 d e^2/m^* c(n+1)$. Therefore, the LFC model predicts that the film becomes optically active for $B \neq 0$.

In Fig. 1 we show a calculated transmission versus ω for the two circular polarizations. Also shown in the figure is the zero-field transmission of one of our films. In our experiment, where $B \leq 15$ T and $\omega \geq 40$ cm^{-1}, the cyclotron frequency $\omega_c \ll \omega \ll \Omega$ for m^* of the order of the free-electron mass m_0. Therefore for unpolarized or linearly polarized light the leading-order change in the transmission with magnetic field is $(\omega_c/\Omega)^2$, which is small and difficult to detect. For the case of circular polarized light this change is $\pm 2\omega\omega_c/\Omega^2$, which is larger and, moreover, sensitive to the sign of the carrier charge. Consequently, it is best to test the predictions of the Kohn theorem by measuring the transmission of a film with circularly polarized light. The experimental arrangement is shown schematically in Fig. 2. We have used a polarizer consisting of a linear polarizer whose polarization axis is oriented 45° from the optical axes of a 7-mm X-cut quartz crystal. Ideally this polarizer provides nearly ECR or HCR polarized light at frequencies $\nu \cong 11(4N \pm 1)$ cm^{-1} for integer N. At other frequencies the polarization state is elliptical and varies continuously with frequency. Because of the oscillations in the polarization state transmitted by the polarizer the transmission of the superconducting film is expected, within the LFC model, to oscillate between the ECR and HCR

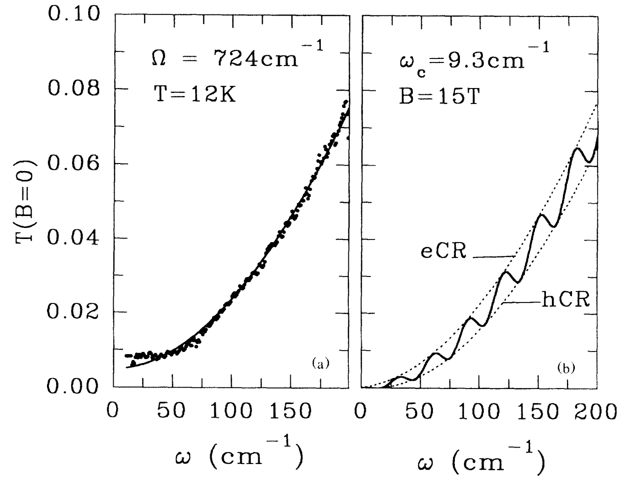

FIG. 1. (a) The observed far-infrared transmission (dots) is consistent with the London model (solid line) except for a 0.8% background which we attribute to radiation leakage around or through the film. Fitting the transmission data gives $\Omega=724$ cm^{-1}. The corresponding London length is consistent with the accepted value (1700 Å) in $YBa_2Cu_3O_7$ to within the uncertainty in the thickness of the film (20%). (b) The transmission is calculated in the LFC model for parameters of (a), $m^*=1.5m_0$ and $B=15$ T. The dotted curves are calculated for the two circular polarizations. The oscillating curve is calculated for the idealized polarizer arrangement used in this work.

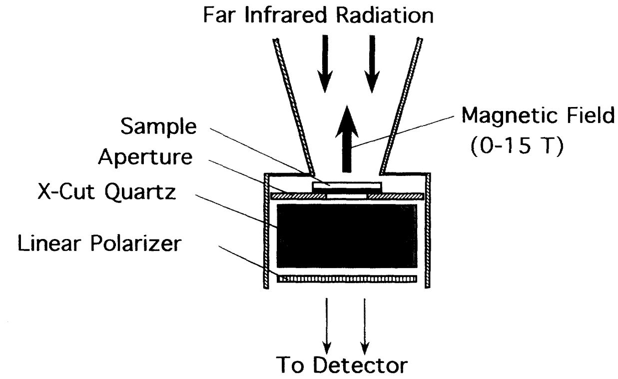

FIG. 2. Experimental arrangement for detection of far-infrared magneto-optical activity in transmission measurements.

transmission curves as shown in the right panel of Fig. 1. Therefore, this technique has the advantage that it provides an oscillatory signature of magneto-optical activity.

The films used in these studies were grown by pulsed laser ablation. In order to have transparency over a wide spectral range we have grown $YBa_2Cu_3O_7$ films on Si(100) substrates sandwiched between a yttria-stabilized-zirconia (YSZ) buffer layer and a YSZ cap layer [8,9]. The $YBa_2Cu_3O_7$ film thickness is $\cong 600$ Å as determined by calibrated time of growth. Electrical resistance measurements give T_c of 87 K with a transition width of ≤ 2 K. These films were described in more detail in earlier reports [8,9]. We have also made measurements on laser ablated $YBa_2Cu_3O_7$ films on $LaAlO_3$ substrates protected with $\cong 300$ Å $PrBa_2Cu_3O_7$ cap layers [10]. In these films $T_c \cong 90$ K with a ≤ 1 K width determined by shielding current measurements. The $B=0$ transmittance of these films was found to be governed by the London conductivity as illustrated for a $YBa_2Cu_3O_7$ film in Fig. 1.

The measured transmission of a film plus analyzer at $B=\pm 14.5$ T normalized to the zero-field transmission is shown in the lower panel of Fig. 3. The measured transmission exhibits oscillations and the phase of the oscillations reverses upon magnetic field reversal as predicted from the LFC model. Similar results were obtained for the $YBa_2Cu_3O_7$ film on $LaAlO_3$ but in this case the substrate transmits only up to 160 cm^{-1}. We have also measured the magnetotransmission of a sample consisting of a Si substrate with a YSZ layer but no $YBa_2Cu_3O_7$ film, which is also shown in Fig. 3. The absence of an oscillatory signal for this case demonstrates that the optical activity is coming from the $YBa_2Cu_3O_7$ film.

At frequencies below 100 cm^{-1} the oscillations in $T(B)/T(0)$ are superimposed on a rapidly rising background signal. This background signal was the subject of an earlier paper reporting the transmission ratio $T(B)/T(0)$ of $YBa_2Cu_3O_7$ on Si for unpolarized radiation [11].

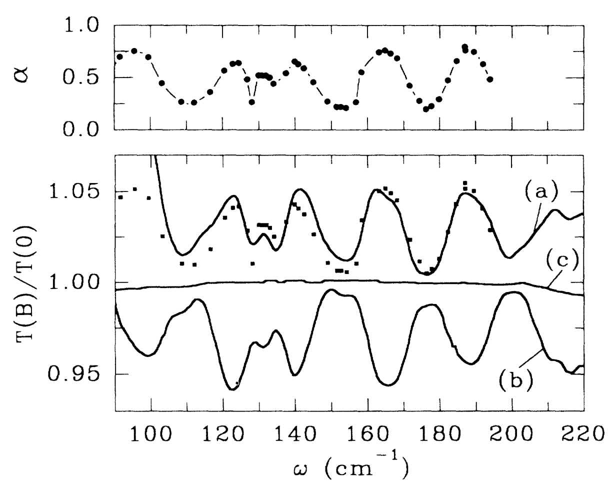

FIG. 3. Top panel: Measured fraction α of electron active circular polarization of the polarizer determined from measurements of cyclotron resonance on a high mobility two-dimensional electron gas in GaAs/GaAlAs heterostructure. The structure near 130 cm^{-1} is due to a quartz phonon. Bottom panel: $T(B)/T(0)$ for a $YBa_2Cu_3O_7$ film measured with the polarizer at $T=2.2$ K. The top and bottom solid curves are for $B=+14.5$ and -14.5 T, respectively. The middle solid curve is the transmission of a YSZ coated Si substrate at 14.5 T. The data points are a simulation of the +14.5-T data using the measured polarizer efficiency (top panel) and the lossless free carrier model with $m^*=3.1m_0$.

In Ref. [11] this effect was identified as a vortex resonance at $\omega_0 \cong 77\ \mathrm{cm}^{-1}$, associated with the pair creation excitation inside the vortex core [11]. An important issue for the work presented here is the optical activity of this vortex resonance. According to theory, for a rigidly pinned vortex, the vortex resonance is excited only in the hole active circular polarization [11]. In this case, the vortex excitation would produce electron active oscillations for $\omega < \omega_0$ and hole active oscillations for $\omega > \omega_0$. Consequently, the vortex excitation would be expected to reduce the cyclotron resonance optical activity for $\omega < \omega_0$ and enhance it for $\omega > \omega_0$. In recent theoretical work, the optical response of vortices with damped motion has been treated [12]. The prediction in this case is that the optical activity of the vortex excitation is reduced [12]. We find no evidence for optical activity of the vortex resonance in our measurements since the amplitude of the oscillations is found to remain nearly constant through the vortex resonance. Moreover, in the case of the $YBa_2Cu_3O_7$ film on $LaAlO_3$, the strength of the vortex resonance signal is reduced by about a factor of 4 but the amplitude of the optical activity remains the same, within the experimental error of the results on the $YBa_2Cu_3O_7$ film on Si. Therefore, we neglect the effects of the vortex resonance on the optical activity in our treatment of the polarization data presented here.

In order to analyze the optical activity data in terms of the LFC model it is necessary to calibrate the polarizer. We have characterized the polarizer by using it to measure the magnetotransmission of the high mobility two-dimensional electron gas (2DEG) in a GaAs heterostructure. The narrow line width ($1.6\ \mathrm{cm}^{-1}$) of the 2DEG cyclotron resonance allows a calibration of the polarization state transmitted by the quartz polarizer at each frequency by varying the applied magnetic field. In the top panel of Fig. 3 we show the measured polarization state α of the polarizer, defined as the fraction of ECR polarization intensity transmitted by the polarizer.

In order to test the consistency of the $YBa_2Cu_3O_7$ transmission data with the LFC model we have simulated our measured curves with the free-carrier model and the measured polarization of the polarizer using only the charge sign and carrier mass m^* as fitting parameters. The results of the fitting are shown as data points in the lower panel of Fig. 3 for a cyclotron mass of $3.1m_0$. It is clear from the comparison of Figs. 1(b) and 3 that the data are only consistent with *hole conduction.* An equally good fit to the data on the $YBa_2Cu_3O_7$ film on the $LaAlO_3$ substrate was found with the same mass and charge sign. These fits support the interpretation of these data in terms of cyclotron resonance of the superconducting state.

We have also studied the magnetic field dependence of these spectra. In order to enhance the amplitude of the oscillations we have plotted the transmission ratios $T(B)/T(-B)$ for different values of the magnetic field in Fig. 4(a). For the conditions of this experiment,

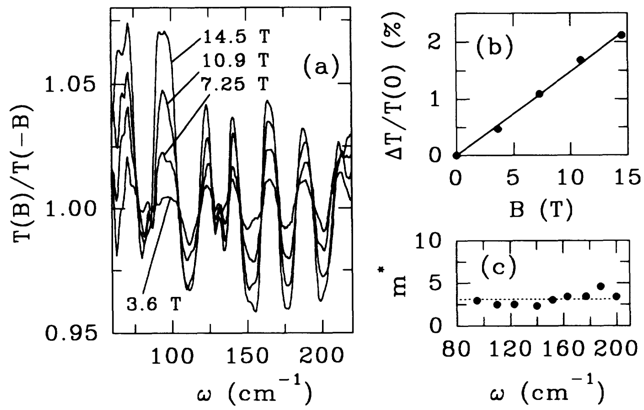

FIG. 4. (a) The transmission ratio $T(B)/T(-B)$ at $T=2.2$ K for the $YBa_2Cu_3O_7$ film with a polarizer for different magnetic fields. (b) The magnetic field dependence of the oscillation amplitude $\Delta T/T(0) = |T(B) - T(0)|/T(0)$ at $\omega = 175\ \mathrm{cm}^{-1}$. The straight line is the result predicted from the lossless free carrier model with $m^* = 3.1m_0$. (c) m^* deduced from the magnetic-field dependence of the transmission at the frequencies where the polarizer has its extrema of circular polarization.

$\omega_c \ll \omega \ll \Omega$, the LFC model predicts a linear dependence of $T(B)/T(-B)$ with magnetic field with the slope varying inversely with m^*. A plot of the magnetic-field dependence of $T(B)/T(-B)$ is shown in Fig. 4(b). The mass values determined from these data, after correcting the amplitudes for the polarization of the polarizer, are plotted in Fig. 4(c) for the frequencies corresponding to the extrema in α. From this plot we find $m^* = (3.1 \pm 0.5)m_0$. The scatter in the mass is consistent with the noise in the amplitude data. We emphasize that this is an inferred mass since our frequency range does not include the cyclotron frequency and we have not directly observed the cyclotron resonance. Nevertheless, this inferred mass is in satisfactory agreement with other experiments and the results from band structure calculations. Optical measurements, which are sensitive only to n_0/m^*, report a band mass close to m_0 [6]. In de Haas-van Alphen measurements recently reported on YBa_2Cu_3O, several small pieces of Fermi surfaces were identified from the data and the corresponding masses were all estimated to be around $7m_0$ [13]. The authors interpreted these masses as (enhanced) dressed masses. Band-structure calculations find several pieces of Fermi surface [14,15]. The largest pieces associated with the CuO_2 planes, the so-called barrels, which are expected to dominate the *a-b* plane excited optical properties, have masses that are reported to range from $1.3m_0$ to $1.9m_0$ [14]. We note that, within the context of the Kohn theorem, the mass measured in our experiment is the phonon dressed band mass. However, this is complicated by the anisotropy of the band structure and the multiple connectedness expected for the Fermi surface in these materials. For the off resonance conditions of our experiment ($\omega \gg \omega_c$) the mass that we are measuring is presumably a specific average over the Fermi surface. Also, our inferred mass should be regarded as an upper limit at this point since any effect that reduces the magnetic field dependence of the oscillation amplitude will increase the measured mass.

Additional evidence for the cyclotron resonance interpretation is found in the $T(B)/T(0)$ data for unpolarized light that was reported earlier [11]. For $\omega < 30\ \mathrm{cm}^{-1}$, the transmission is found to increase at high magnetic fields so that $T(B)/T(0)$ rises more quickly than predicted by the vortex excitation models [11]. This rise can be understood in terms of the magnetic-field dependence of the condensate response. The effective conductivity at low frequency is $\sigma_{\mathrm{eff}}^{\pm} = (1-f)\sigma_F^{\pm}$, where f is the vortex volume fraction and the vortex contribution to σ_{eff} is negligible for $\omega \ll \omega_0$. For unpolarized light the transmission ratio $T(B)/T(0) \cong (1-f)^2(\omega^2+\omega_c^2)/\omega^2$ when ω_c and $\omega \ll \Omega$. This expression diverges at zero frequency and can account for the low-frequency rise observed in the data.

How are we to understand these magneto-optical activity results? We note that both the LFC model and the London superconductor model are lossless at all frequencies. In these models the optical properties are controlled by the diamagnetic (screening) response of the conducting system. The content of the Kohn theorem is that the *diamagnetic response frequency is shifted to* ω_c and that it corresponds to the collective cyclotron motion of the condensate. In real systems there are loss mechanisms and violations of the conditions of the Kohn theorem which can lead to finite σ_1. The diamagnetic response dominates in these thin-film transmission measurements, however, since σ_2 is very large near the resonance. Therefore, the losses can be undetected.

In conclusion, we have observed magneto-optical activity of superconducting $YBa_2Cu_3O_7$. The observed signal is found to be consistent with cyclotron resonance of the superconducting condensate. Additional measurements at frequencies closer to the cyclotron frequency are being carried out to clarify these results. There are other interesting implications of this experiment. For example, helicon propagation should be possible in bulk crystals of extreme type-II superconductors at frequencies sufficiently low that losses due to the vortex response are small. We believe that these are general effects not confined to $YBa_2Cu_3O_7$ or high-T_c superconductors but intrinsic to all extreme type-II superconductors.

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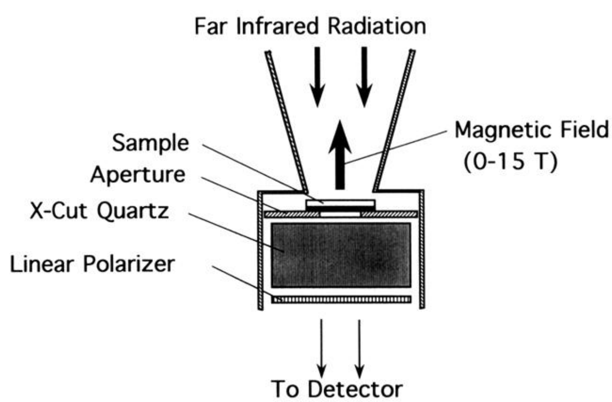

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