Measurement of the Far Infrared Magneto-Conductivity Tensor of Superconducting YBa$_2$Cu$_3$O$_{7-\delta}$ Thin Films

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Abstract

We report measurements of the far infrared transmission of superconducting YBa$_2$Cu$_3$O$_{7-\delta}$ thin films from 5 cm$^{-1}$ to 200 cm$^{-1}$ in fields up to 14T. A Kramers-Kronig analysis of the magneto-transmission spectrum yields the magneto-conductivity tensor. The result shows that the magnetoconductivity of YBa$_2$Cu$_3$O$_{7-\delta}$ is dominated by three terms: a London term, a low frequency Lorentzian ($\omega_1 \approx 3$ cm$^{-1}$) of width $\Gamma_1 = 10$ cm$^{-1}$ and a finite frequency Lorentzian of width $\Gamma_2 = 17$ cm$^{-1}$ at $\omega_2 = 24$ cm$^{-1}$ in the hole cyclotron resonance active mode of circular polarization.

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The electrodynamic response of type II superconductors in the mixed state is strongly affected by vortex dynamics as extensive dc transport and microwave (µwave) frequency studies have shown for both conventional and high temperature superconductors [1,2]. For a pinned vortex lattice the results of the µwave experiments can be described in terms of a conductivity function consisting of two terms: a London term reduced from its zero field strength and a zero frequency oscillator with a width characterized by the “depinning frequency” \( \omega_d = \kappa/\eta \) where \( \kappa \) is the pinning force constant and \( \eta \) is the viscosity [1,3]. That this oscillator occurs at zero frequency has been attributed to the massless dynamics of the vortices. On the other hand a far richer phenomenology may be anticipated for the electrodynamics of the vortex system. The possibilities include: a pinning resonance associated with the inertial motion of the vortex in its pinning field [4], resonant excitation of quasiparticles to the quantized levels in the vortex core [5,6] and, in the clean limit, the collective cyclotron resonance of the electron system [7,8]. Moreover, quite generally, the response of an electron system in the presence of a magnetic field is expected to be chiral, so that \( \sigma_{xy}(\omega, H) \neq 0 \) [9]. Although these effects have not been observed at µwave frequencies, they have been reported from recent experiments in high \( T_c \) films at far infrared (FIR) frequencies [7,10,11]. These experiments were compared with a clean limit theory of vortex dynamics that predicts that these three resonances are hybridized leading to two finite frequency chiral resonances [11]. However, this theory does not predict the zero frequency resonance observed at µwave frequencies. The earlier FIR experiments were limited to \( \omega > 25 \text{ cm}^{-1} \) so that they did not observe the zero frequency oscillator nor did they completely resolve the pinning resonance [12]. Therefore there is a gap in our understanding of the electrodynamics of the vortex system that coincides with the frequency gap in the measurements between the µwave and FIR.

In this letter we present the magneto-transmission spectrum of YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) (YBCO) films over the frequency range from 5.26 cm\(^{-1}\) to 200 cm\(^{-1}\) by using the combination of broadband Fourier Transform Spectroscopy (FTS) and a FIR laser source. These measurements cover the entire range of frequencies relevant to the vortex system and provide a reconciliation
of the $\mu$wave and FIR phenomenologies. A Kramers-Kronig transformation (KKT) technique is used to obtain the real and imaginary parts of the magneto-conductivity tensor as a function of frequency. The results show that the electrodynamic response of the pinned vortex system is dominated by two major Lorentzian oscillators in addition to a London term. These results, together with evidence for the weaker vortex core resonances reported recently, suggest a vortex response that combines the features of both the massless vortex model of Gittleman-Rosenblum (G-R) and the clean limit model of Hsu which includes the vortex core structure and inertia.

The samples are YBCO thin films grown by pulsed laser deposition on silicon substrates with yttria stabilized zirconia buffer layers and cap layers. The film thickness is typically $d = 400\,\text{Å}$ and typical critical temperatures are $T_c = 89\pm1\,\text{K}$ measured by ac susceptibility. Their growth and characterization is described in detail elsewhere. The broadband transmission of the films was measured from 30 cm$^{-1}$ to 200 cm$^{-1}$, using FTS with a 2.2 K bolometer detector. Magnetic fields up to 14T were applied perpendicular to the a-b plane of the YBCO thin films. The incident FIR radiation was elliptically polarized by a polarizer comprised of a metal grid linear polarizer and a x-cut quartz waveplate. The elliptically polarized transmission data was unfolded, using a calibration of the polarizer efficiency, to give the circularly polarized response, $T^\pm(\omega, H)$. For the low frequency measurements we use a CO$_2$ pumped FIR laser which has useful discrete FIR lines from 5.26 cm$^{-1}$ to 96 cm$^{-1}$. A more detailed description of the experiment and data manipulation are given elsewhere. Leakage of radiation around or through the sample is a potential problem at low frequencies where the transmission of the film is small (as low as 0.02% at 5 cm$^{-1}$). Leakage was eliminated by the use of Ecosorb and graphite to absorb stray light. The nearly quadratic $T(\omega, 0)$ observed down to our lowest frequencies attests to the low levels of radiation leakage in these experiments.

The experiments measured both the absolute transmission at zero field $T(\omega, 0)$ and the transmission ratio $T^\pm(\omega, H)/T(\omega, 0)$. The transmission ratio measurement is more accurate than the absolute transmission measurement since the sample is undisturbed, which makes
it possible to measure the change induced by vortex dynamics precisely. The transmission coefficient \( T^\pm(\omega, H) = |t^\pm(\omega, H)|^2 \) where the transmission amplitude \( t^\pm(\omega, H) \) is related to the conductivity by

\[
t^\pm(\omega, H) = (n_{Si} + 1) / \left( Z_0 \sigma^\pm(\omega, H) + n_{Si} + 1 \right)
\]

in which \( Z_0 \) is the impedance of free space, \( n_{Si} \) is the refractive index of the silicon substrate (nearly constant and real) and \( \sigma^\pm(\omega, H) \) is the conductivity in the two circular polarization modes. In order to eliminate multiple reflection effects in the substrate, we either averaged the spectrum with a resolution (4 cm\(^{-1}\)) lower than the spacing of the interference fringes (1 cm\(^{-1}\)) in the broadband measurement or use an anti-reflection coating on the back side of the substrate in the laser experiment [15].

Fig.1 shows \(|t^\pm(\omega, H)/t(\omega, 0)|\) as a function of frequency \( \omega \) at \( H=9T \) and 4 K. \(|t(\omega, 0)|\) at 4 K is also shown in the inset. The response \(|t^+(H)/t(0)|\) in the electron cyclotron resonance active polarization (eCP) mode is plotted for positive frequencies and the hole active polarization (hCP) response \(|t^-(H)/t(0)|\) is plotted for negative frequencies. This representation will be explained below (See Eq.(2).). The transmission ratios show a sharp rise for \(|\omega| < 30 \text{ cm}^{-1}\) corresponding to a peak centered between ±5 cm\(^{-1}\). This low frequency feature is asymmetrical around \( \omega = 0 \), i.e., \(|t^-(H)/t(0)| > |t^+(H)/t(0)|\). Its field dependence is slightly super linear. At high frequencies (\(|\omega| \geq 30 \text{ cm}^{-1}\)) the transmission ratio approaches unity in both modes such that \(|t^+(H)/t(0)| > 1 \) and \(|t^-(H)/t(0)| < 1\), with a minimum at -40 cm\(^{-1}\). This high frequency chiral response is found to scale linearly with magnetic field for \( H < 14T \ll H_{c2} \) [10] and has been interpreted in terms of the tail of a free hole-like cyclotron resonance response [7].

The overall shape of the transmission ratio spectrum in Fig.1 is simple which suggests that the underlying physics of vortex electrodynamics may be very simple and elegant. Therefore it is interesting to convert the transmission data into the conductivity function which is more useful for gaining insight into the phenomena. We do this by means of a Kramers-Kronig transformation technique. The most convenient quantity related to the
transmission experiment that satisfies the KKT conditions is \( \ln(t(\omega)) = \ln|t(\omega)| + i \arg[t(\omega)] \) in which \( \arg[t(\omega)] \) is the phase of the complex amplitude \( t(\omega) \).

In the zero field case, \( \arg[t(\omega, 0)] \) can be obtained through KKT by properly choosing an extrapolation function \( t_{\text{ext}}(\omega) \) for \( |t(\omega, 0)| \), which preserves time reversal symmetry (even function in \( \omega \)) and has correct asymptotic behavior at \( \omega \to 0 \) \( (t_{\text{ext}}(\omega) \propto \omega) \) and \( \infty \) \( (t_{\text{ext}}(\omega) \to 1) \). \( t(\omega, 0) \) can then be converted into \( \sigma(\omega, 0) \) since \( Z_0, d, \) and \( n_{\text{Si}} \) are known. Detailed analyses show that the film consists of \( f_{s0} = 0.61 \) of superfluid condensate with the London penetration depth \( \lambda_0 = 1850 \text{Å} \) \cite{[16]}.

When a magnetic field is applied to the system, time reversal symmetry is broken and it is then necessary to determine the proper response function for both positive and negative frequencies. In this case, the two circularly polarized modes are the canonical modes. We can extend the response function \( g^+(\omega, H) \) to the negative frequency range by

\[
g^+(\omega, H) \equiv \begin{cases} 
g^+(\omega, H), \text{ when } \omega > 0 \text{ (eCP)} \\
g^-(-\omega, H)^*, \text{ when } \omega < 0 \text{ (hCP)}
\end{cases}
\]

where \( g^\pm(\omega, H) \) are the physical quantities measured for positive frequencies in two circularly polarized modes. The KKT then is extended to the magnetic field case,

\[
\arg\left[\frac{t^+(\omega, H)}{t(\omega, 0)}\right] = \frac{1}{\pi} P \int_{-\infty}^{\infty} \frac{\ln|t^+(\omega', H)/t(\omega', 0)|}{\omega - \omega'} d\omega'.
\]

By combining this with the zero field data from which \( t(\omega, 0) \) is extracted, we can obtain \( \sigma^+(\omega, H) \) \((-\infty < \omega < \infty)\) and therefore \( \sigma^\pm(\omega, H) \) \((0 < \omega < \infty)\).

Fig.2 shows the magneto-conductivity \( \text{Re}[\sigma^+(\omega, H)] \) obtained by KKT on the transmission curves in Fig.1 with a \( \ln|t^+/t^-| \propto \omega^3 \) extrapolation scheme for \( |\omega| < 5 \text{ cm}^{-1} \), which will be described later in this letter. Apart from the residual metallic background which is present in \( \text{Re}[\sigma(\omega, 0)] \) \cite{[16]}, the conductivity function evolves from its zero field London form \( \left(\frac{ne^2}{m} f_{s0} i/\omega\right) \) into a form with a reduced London component and several finite frequency absorption bands. The resulting conductivity can be well represented as a finite sum of Lorentzian oscillators \( \sigma_H(\omega, H) = ne^2/m \sum_{i=1}^{M} f_i / (i(\omega - \omega_i) + \Gamma_i) \) where \( f_i \) represents the strength of the \( i \)th oscillator. Indeed, we found that \( \text{two} \) finite width oscillators \((M = 2)\) in
addition to a reduced strength London term ($\omega_0=0$, $\Gamma_0=0$) is sufficient to describe the main features induced by vortex dynamics. The best fit with $M=2$ in $\sigma_H(\omega, H)$ is shown as the dotted line in Fig.2. The first oscillator is at low frequency ($\omega_1=3.15$ cm$^{-1}$, in eCP mode) with $f_1=0.14$ (23% of $f_{s0}$) and a width $\Gamma_1=10$ cm$^{-1}$ [17] which is similar to the form of the G-R model [1]. The second oscillator is centered at $\omega_2=-24$ cm$^{-1}$ (in the hCP mode) with $f_2=0.11$ (18% of $f_{s0}$) and a width $\Gamma_2=17$ cm$^{-1}$, which produces the optical activity observed at higher frequencies [12]. The remaining oscillator strength gives $f_0=0.36$ (59% of $f_{s0}$) since $f_0 + f_1 + f_2 = f_{s0}$.

A sum rule on $t(\omega)$ follows from the superfluidity of the condensed state. For pinned type II superconductors the superfluid condensate response causes the low frequency conductivity to be dominated by the London screening, $\sigma(\omega \to 0) \sim i/\omega + \pi\delta(\omega)$. Therefore $t^+(0, H)/t(0, 0)$ is the ratio of the strength of two delta functions which is a real number (for $H < H_{c2}$). The left hand side of Eq.(3) is zero (when $\omega \to 0$) which leads to the relation

$$\int_0^\infty \ln |t^+(\omega', H)/t^-(\omega', H)| d\omega' = 0. \quad (4)$$

This sum rule provides a strong constraint on the magneto-transmission data which is useful for setting the extrapolations in the KKT analysis. It also allows inferences to be made on the electrodynamics of the vortex system outside the measurement range. Fig.3 shows $|t^+(\omega, H)/t^-(\omega, H)|$ as a function of $\ln(\omega)$ and the integral weight of Eq.(4) in the various regions. Assuming that free electron behavior eventually dominates the free carrier response at sufficiently high frequencies, then $\ln |t^+/t^-|/\omega \sim \omega_c/\omega^4\tau$ which indicates a strongly convergent (superconvergent) sum rule. Indeed, extrapolation of the broadband data by a simple cyclotron resonance model suggests that the frequency range beyond 190 cm$^{-1}$ contributes only about 10% to the integral.

As shown in Fig.3, we have confirmed the observation of a sign reversal of the optical activity ($T^+/T^- - 1$) in YBCO films (the same batch of samples as in this letter.) at about 30 cm$^{-1}$, reported by Choi et al. [11,12]. Remarkably, this feature is a natural consequence of Eq.(4) since the weight given by the hybridized hole cyclotron resonance [4,7,11] at higher
frequencies has to be balanced by an electron-like Hall effect at lower frequencies.

Even though several different extrapolation schemes with reasonable physical assumptions in the unmeasured regions ($|\omega| < 5.26 \text{ cm}^{-1}$ and $|\omega| > 200 \text{ cm}^{-1}$) give similar results and preserves the two oscillator picture of the conductivity, the use of Eq.(4) helps to refine the analysis. A simple cubic spline interpolation between $\pm 5.26 \text{ cm}^{-1}$ gives $\ln |t^+/t^-| \sim \omega$, which results in $\sim 30\%$ excess weight in the low frequency region compared to the high frequency region (double dotted dash line in Fig.3). (Note that $1/\omega$ in Eq.(4) dramatically increases the weight of the low frequency optical activity.) There are two possible resolutions of this discrepancy. The first possibility is that there are other (electron-like) chiral resonances in addition to the simple cyclotron resonance tail above $200 \text{ cm}^{-1}$ which balances the low frequency weight. Simulations show that an additional $7\%$ mid-IR chiral resonance at $\sim 500 \text{ cm}^{-1}$ will allow the sum rule to be satisfied. However, this resonance would cause the optical activity to change sign twice below $500 \text{ cm}^{-1}$ and there has been no evidence for this in our measurements up to $200 \text{ cm}^{-1}$ [11,12]. The second possibility is that $\ln |t^+/t^-|$ decreases faster than $\omega$ at low frequencies (below $5 \text{ cm}^{-1}$). By choosing an $\omega^3$ extrapolation for $\ln |t^+/t^-|$ (which is an odd function of $\omega$) we found that the sum rule can be satisfied (dashed line below $5 \text{ cm}^{-1}$). It turns out that the two-Lorentzian model mentioned above with a slightly electron-like low frequency oscillator satisfies the second criteria and is used as the extrapolation function for $|\omega| < 5.26 \text{ cm}^{-1}$ in Fig.3 (dotted line). This self-consistency further convinces us that the two-Lorentzian model properly represents the main response of the system.

We also note that the FIR data can be used to infer information about the low frequency response of the system. $\ln |t^+/t^-| / \omega \propto Re[\sigma_{xy}]$ at low frequencies where $Z_0d\sigma \gg n_{Si} + 1$, $\sigma_{xx} \sim 1/i\omega$ and $\sigma_{xy} \ll \sigma_{xx}$. Therefore $\ln |t^+/t^-| \propto \omega$ implies that the low frequency limit of $Re[\sigma_{xy}]$ is a finite constant and $\rho_{xy} \propto \omega^2$. However if the second possibility holds true, an $\omega^3$ dependence of $\ln |t^+/t^-|$ would imply $Re[\sigma_{xy}] \propto \omega^2$ (dotted line in Fig. 3(b)) so that $\rho_{xy}$ would go to zero as $\omega^4$, corresponding to a very strong suppression of the Hall effect. Therefore $\mu$ wave studies of the Hall effect in type II superconductor may prove interesting.
Fig. 4(a) shows $\frac{Im[\sigma_{xx}(\omega, H)]}{Im[\sigma(\omega, 0)]}$ which describes the modification of the screening in the applied field. This ratio approaches 0.55 as $\omega \to 0$. In terms of the G-R model $\frac{Im[\sigma_{xx}(0, H)]}{Im[\sigma(0, 0)]} = \frac{\kappa}{\kappa + \frac{c^2}{4\pi\lambda_0^2} \frac{H \phi_0}{c^2}}$ which gives $\kappa \simeq 5.3 \times 10^5$ N/m². This value of $\kappa$ is consistent with diverse $\mu$wave measurements on films and single crystals [2,17,18], suggesting an intrinsic origin for $\kappa$.

There are other smaller structures (> 50 cm⁻¹) in the conductivity that correspond to only a few percent (or less) of the total oscillator strength. Many of these features are reproducible, scale in amplitude with $H$ and are found to correlate with the density of 45° misaligned grains in this sample. They have been discussed elsewhere in terms of vortex core excitations [10]. Also we note that $(T^+/T^- - 1)$ at low frequencies ($7 \sim 15$ cm⁻¹) decreases linearly with temperature and changes its sign at $\sim$20 K. This behavior is consistent with cyclotron resonance of thermally excited quasiparticles as has been reported by Spielman et al. [9]. This observation shows that the low temperature optical activity is dominated by vortex dynamics.

The nonzero center frequency of the low frequency oscillator is a consequence of the low frequency extrapolation as determined from the superconvergent sum rule. Within the G-R type models it implies a Magnus force, $n e (v_s - \beta v_L) \times \phi_0$ where $\beta \approx -0.08$ [19]. The negative $\beta$ is consistent with observation of a reversal of dc Hall effect in the flux flow regime [20,21]. No evidence for optical activity has been reported in the low temperature vortex response at $\mu$wave frequencies. This may be related to the strong suppression of the Hall resistance discussed above. In the G-R model optical activity requires a Magnus force, but the zero vortex mass keeps the resonance at low frequency. Hsu’s model contains a Magnus force, core excitations and, implicitly, a vortex mass and it produces two finite frequency chiral resonances. In the absence of pinning the Hsu model predicts that the core resonance is silent and the response of the superconductor is simple cyclotron resonance as expected from Kohn’s theorem [8]. Pinning hybridizes the cyclotron resonance and vortex core resonance with the pinning resonance. The result is a strong hybridized pinning resonance at a negative frequency, which gives optical activity and a weak hybridized vortex core resonance at a
positive frequency.

The observed response is seen to contain the features of both theories, but is inconsistent with either one alone. It appears that there is a massless response of the vortices that gives rise to the low frequency oscillator in addition to a finite vortex mass response that gives rise to the pinning resonance and the weak vortex core resonance in the eCP mode \([10,11]\). These observations suggest a model in which the vortex is considered as a composite object. The observed responses can then arise from the massless response of the vortex currents outside the core and the inertial response of the core. The observed universal value of \(\kappa\) then is a natural consequence of this model due to the pinning of the vortex current pattern to the vortex core. A paper describing this model will be presented elsewhere \([19]\).

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[16] $\sigma(\omega, 0)$ can be represented very well by a sum of three components $\frac{n e^2}{m} f_{s0} i/\omega + f_{m1}\sigma_{m1} + f_{m2}\sigma_{m2}$: (1) superfluid condensate, $f_{s0} = 0.61$; (2) normal metal, $f_{m1} = 0.28$ and $\sigma_{m1} = \frac{n e^2}{m}/(-i\omega + 1/\tau)$ with $1/\tau = 150$ cm$^{-1}$; and (3) a nearly frequency independent normal component (over the measurement range), $f_{m2} = 0.11$ and $\sigma_{m2} = \frac{n e^2}{m}/(-i\omega + 1/\tau)$ with
$1/\tau \approx 600 \text{ cm}^{-1}$. The total oscillator strength is given by $4\pi ne^2/m = c^2/\lambda_0^2$ which corresponds to $\lambda_0 = 1850\text{ Å}$. The strength of the Drude components are found to be sample dependent. The second term is believed to represent superconducting dead layers at the interfaces of the superconducting films and the third term is related to either the mid-IR contribution [22] or the chain conductivity in YBCO. Both terms are believed to be field independent.

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FIGURES

FIG. 1. The transmission amplitude ratio \( |t^+/-(\omega, H)/t(\omega, 0)| \) as a function of frequency \( \omega \) for a YBCO thin film at \( H=9T \) and \( 4 \) K for the circularly polarized light. The solid curve represents the FTS data from 30 cm\(^{-1}\) to 200 cm\(^{-1}\) and the triangular points represent the data from the FIR laser lines. The region between 125~140 cm\(^{-1}\) corresponds to a quartz phonon and the half waveplate condition of the polarizer. The dotted line is the \( \ln |t^+/t^-| \sim \omega^3 \) extrapolation described in text. Inset: Zero field transmission amplitude \( |t(\omega, 0)| \) vs. frequency \( \omega \) at 4\(^\circ\)K (the solid line). The dotted line is the extrapolation function \( t_{\text{ext}}(\omega) \).

FIG. 2. The magneto-conductivity \( \text{Re}[\sigma^+(\omega, H)] \) obtained from the Kramers-Kronig analysis. The change induced by the magnetic field can be described as the sum (dotted line) of a low frequency oscillator of width \( \Gamma_1 = 10 \) cm\(^{-1}\) at \( \omega_1 = +3.15 \) cm\(^{-1}\) (single dotted dash line) and an oscillator of width \( \Gamma_2 = 17 \) cm\(^{-1}\) at \( \omega_2 = -24 \) cm\(^{-1}\) (double dotted dash line). The small dotted line is \( \text{Re}[\sigma(\omega, 0)] \).

FIG. 3. \( |t^+(\omega, H)/t^-(\omega, H)| \) is plotted as a function of \( \ln (\omega) \). The spectral weights of different frequency regions to the sum rule of Eq.\(^{(4)}\) are indicated. The dashed line between laser data points is a guide for the eye. The double dotted dash line is the simple cubic spline extrapolation which behaves as \( \ln |t^+/t^-| \sim \omega \) below 5 cm\(^{-1}\). The dashed line is an \( \omega^3 \) extrapolation for \( \ln |t^+/t^-| \) which satisfies the sum rule.

FIG. 4. The magneto-conductivity shown in the Cartesian coordinates, \( \sigma_{xx} \) and \( \sigma_{xy} \). Dotted lines are the two-Lorentzian model fits described in Fig.\(^{3}\). (a) \( \text{Im}[\sigma_{xx}(\omega, H)]/\text{Im}[\sigma(\omega, 0)] \) represents the reduction of screening in the applied field. (b) \( \text{Re}[\sigma_{xy}(\omega, H)] \) and (c) \( \text{Im}[\sigma_{xy}(\omega, H)] \) show a resonance at 24 cm\(^{-1}\).
Fig.

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\[ \text{Re}[\sigma_{xy}(\omega, H)] \quad \text{Im}[\sigma_{xy}(\omega, H)] \quad \text{Im}[\sigma_{xx}(\omega, H)]/\text{Im}[\sigma(\omega, 0)] \]

\[ (x10^{15} \text{ sec}^{-1}) \]

\[ \omega (\text{cm}^{-1}) \]

\[ (\text{m}\Omega\cdot\text{cm})^{-1} \]

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