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Far-infrared properties of superconducting $YBa_2Cu_3O_{7-\delta}$ films in high magnetic fields

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Abstract

We report the far-infrared properties of superconducting ab plane-oriented $YBa_2Cu_3O_{7-\delta}$ films in magnetic fields of up to 27 T. The applications of magnetic field (with H perpendicular to the ab plane and with unpolarized light) at low temperature produces no discernible field dependence. This observation differs from other previous far-infrared measurements in this temperature range. Only at fields and temperatures where the dc resistance is not zero – on account of dissipative flux motion – is a field-induced effect observed. © 1998 Elsevier Science Ltd. All rights reserved.

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1. Introduction

The electronic properties of the high-T_c superconductors are affected by the application of magnetic fields [1,2]. In the simplest picture, the sample in the mixed state is penetrated by an array of magnetic vortices, each of which contains a quantized amount of magnetic flux. If the vortices are at rest (pinned), the resistance will be effectively zero; if instead the vortices are moving, an electric field which is parallel to the current appears, leading to a finite electrical resistance. The behavior of vortex motion in the presence of viscous damping, pinning forces, and fluctuations either of thermal origin or as a result of the influence of defects in the sample, is complicated and is not fully understood.

Far-infrared spectroscopy has been applied several times to investigate the vortex dynamics in the high-T_c superconductors. In most – but not all – cases, evidence for field-induced absorption was observed. Brunel et al. [3] reported the reflectance of Bi2Sr2Ca-Cu2O8 (BSCCO) at several far-infrared frequencies as a function of temperature and field (up to 17 T). They found a field-induced drop in reflectance, which corresponded to the onset of a resistive state and thus only occurred at higher temperatures. In contrast, far-infrared transmittance measurements of thin $YBa_2Cu_3O_{7-\delta}$ (YBCO) films by Karrai et al. [4] showed an increase in transmission below $\backsim 125\ cm^{-1}$ with increasing field. This effect was attributed to dipole transitions associated with bound states in the vortex cores. Evidence for magneto-optical activity was also found, interpreted as cyclotron resonance in the mixed state. These effects occurred at

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Table 1
Sample characteristics

Sample	Thick (Å)	T_c (K)	ΔT_c (K)	ρ_{dc} (300 K) (cm)	λ_L (~ 20 K) (Å)
YBCO/200 Å PBCO/$YAlO_3$	300	83.5	3.5	550	2500 ± 100
YBCO/200 Å PBCO/$YAlO_3$	500	85.0	2.5	590	2300 ± 100
YBCO/MgO	400	83.0	3.0	400	2000 ± 200
YBCO/MgO	600	86.7	2.8	360	1900 ± 200
YBCO/$SrTiO_3$	5000	88.0	0.5	320	1750 ± 100

a temperature as low as 2.2 K and in magnetic fields between 2 and 15 T. The experiments prompted several theoretical calculations of the optical response of the vortex core states [5–7]. Shimamoto et al. [8] observed a large (20%) and temperature-independent change in far-infrared transmission in both YBCO and $Bi_2Sr_2Ca_2Cu_3O_x$ films at fields of up to 100 T; the behavior was explained by a flux-flow model. In contrast, Gerrits et al. [9] reported practically no influence of the magnetic field up to 15.5 T in the far-infrared reflectance of YBCO thin films at 1.2 K. Eldridge et al. [10] measured the reflectance of a YBCO film in magnetic fields up to 3.5 T at 4.2 K. A Kramers–Kronig analysis then, gave the field-dependent conductivity, which showed a broad resonance between 50 and 250 cm^{-1} whose shape, but not magnitude, could be fit by a theory involving vortex motion with pinning. Terahertz measurements over frequencies from 100 to 1000 GHz (3.3–33.3 cm^{-1}) found a non-linear dependence of the complex conductivity on the magnetic field strength, interpreted in terms of enhanced pair breaking, and corresponding superfluid depletion, because of nodes in a *d*-wave gap function [11].

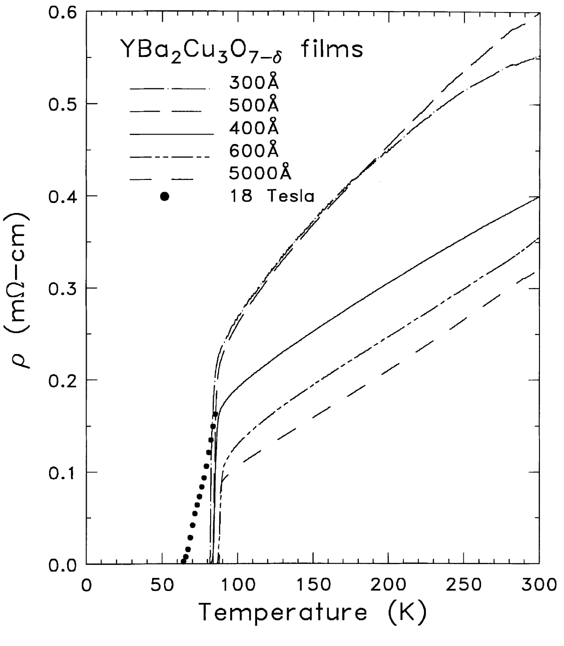

Fig. 1. The dc resistivity in the ab plane, as a function of temperature, for all the YBCO films. The magnetoresistance of a 400 Å film in 18 T is indicated by the symbols.

In this article, we report the results for the far-infrared reflectance ($\mathscr{R}$)and transmittance ($\mathscr{T}$) of superconducting YBCO films in magnetic fields of up to 27 T. We use $\mathscr{R}$ and $\mathscr{T}$ to extract the frequency-dependent optical conductivity. In addition, we carried out a detailed study of the zero-field $\mathscr{R}$ and $\mathscr{T}$, obtaining results in agreement with previous data [12]. The applications of magnetic field (with *H* perpendicular to the ab plane and with unpolarized light) at low temperatures produces no discernible field dependence. Only at fields and temperatures where the dc resistance is not zero – on account of dissipative flux motion – is a field-induced effect observed.

We studied three types of YBCO samples, all prepared by pulsed-laser ablation: (1) thin (300–500 Å) films on a $PrBa_2Cu_3O_{7-\delta}$ buffer layer on $YAlO_3$ substrates; (2) thin (400–600 Å) films on MgO substrates; and (3) a 5000 Å-thick film deposited on a SrTiO3 substrate. All the samples were structurally characterized by X-ray diffraction, which showed their *c*-axis orientation. The superconducting properties of the films were determined by dc resistivity or ac susceptibility measurements. The characteristics of all samples are listed in Table 1. Fig. 1 shows the dc resistivity. The 5000 Å-thick film has a slightly higher onset temperature and a sharper transition.

The films with $d < 1000$ Å were studied in both $\mathscr{R}$ and $\mathscr{T}$ whereas the 5000 Å-thick film on SrTiO3 was

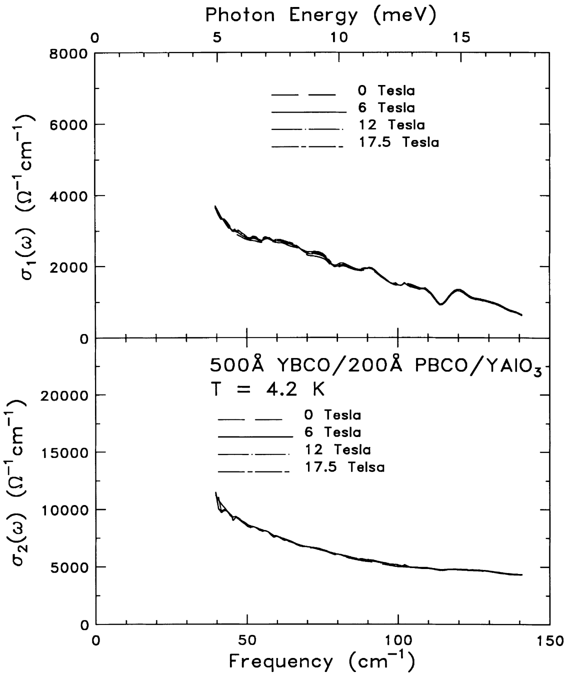

Fig. 2. The magnetic field and frequency-dependent conductivity of a 500 Å YBCO/200 Å PBCO/YAlO$_3$ film at 4.2 K.

studied only in $\mathscr{R}$. The far-infrared studies in magnetic field were carried out at the National High Magnetic Field Laboratory, using a Bruker 113v spectrometer, custom light-pipe optics to carry the far-infrared radiation through the magnet, and a 4.2 K helium-cooled bolometer detector [13]. Three sample probes were used. Two, used with a superconducting magnet, permit sequential measurements of sample and reference single-beam spectra, allowing absolute measurements of $\mathscr{R}$ and $\mathscr{T}$ up to 17.5 T. On account of the detector placement, these measurements are restricted to 4.2 K. We also employed a 27 T resistive magnet; in this magnet, the transmittance ratio, $[\mathscr{T}(H)/\mathscr{T}(0)]$, may be measured at temperatures between 4.2 and 100 K, but without the ability to replace sample with a reference. In all measurements, the unpolarized far-infrared radiation was incident nearly normal to the film, so that the electric field was in an ab plane and the magnetic field H was perpendicular to the ab plane.

To deal with the dispersion and absorption in the substrate, the reflectance $\mathscr{R}_{\mathrm{sub}}$ and transmittance $\mathscr{T}_{\mathrm{sub}}$ of YAlO$_3$ and MgO were also measured at each temperature and magnetic field where the film data were taken. The absorption coefficient $\alpha(\omega)$ and the index refraction $n(\omega)$ of the substrate were then used in the analysis of the data for films. These measurements turned out to be crucial in the case of the YAlO$_3$ substrates, which contained some 4% atomic weight Nd, giving field-dependent features in the transmittance. [14]

The 4.2 K frequency-dependent conductivities, σ_1 and σ_2, of a 500 Å YBCO/200 Å PBCO/YAlO$_3$ film at 4.2 K are shown in Fig. 2 at fields of up to 17.5 T, with σ_1 being smaller than σ_2. (Note the differing conductivity scales in the two panels). σ_1 is a measure of the absorption, while σ_2 is related (via the Kramers–Kronig relations) to the weight of the delta function at the origin, i.e., the superfluid density. Neither quantity shows any discernible field dependence.

Fig. 3 shows the temperature- and field-dependent reflectance spectra of the 5000 Å-thick film. The low-temperature relfectance is near unity at $\omega \rightarrow 0$. With increasing frequency it falls off slowly, showing a pronounced shoulder at $\sim 450\ \mathrm{cm}^{-1}$. Increasing temperature decreases the reflectance in a characteristic way (upper panel) [15]. The lower panel of Fig. 3 shows the far-infrared reflectance of the film at 4.2 K as a function of magnetic field. We find no field-induced effects in the spectra at the ± 0.25% level.

Magneto-transmission measurements at different temperature and magnetic fields for both 400 and 600 Å YBCO/MgO films are shown in Fig. 4. The data shown are the ratio of the transmission of the sample at H to the zero-field transmission. This ratio does not show any discernible field dependence from 4.2 to 50 K in fields of 27 T. Typical noise variations of our measurements in a magnetic field are of the order of ± 1%. The low frequency limit of the measurements (about 35 cm^{-1}) is caused by a combination of low transmitted intensity, decreasing source intensity, and reduced spectrometer efficiency at low frequencies.

When the temperature is raised above 60 K, the 35 cm^{-1} transmission of these films is observed to change by more than 5% as field increases from 6 to 27 T; changes at high frequencies ($\omega > 100\ \mathrm{cm}^{-1}$) are zero within experimental error. These large field-induced increases in transmission occur only at temperature not too far below T_{c}. Dc transport on the 400 Å film (shown in Fig. 1) demonstrated that the

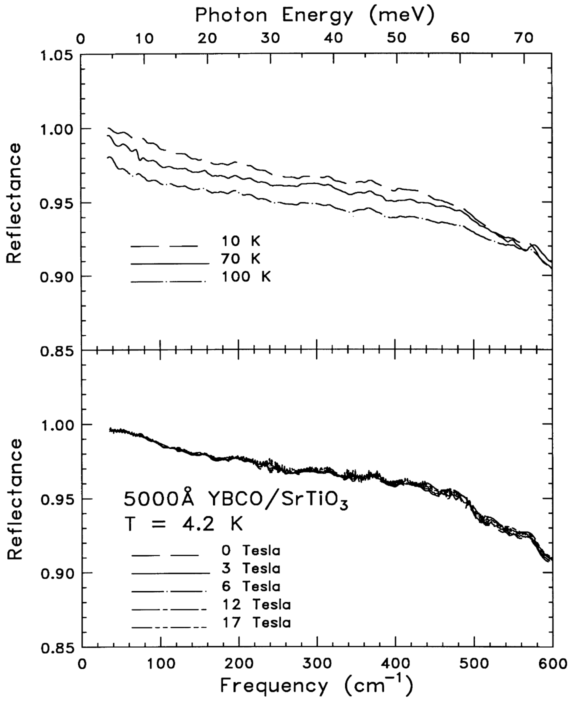

Fig. 3. The measured zero-field reflectance of a 5000 Å YBCO/SrTiO3 film at 10, 70 and 100 K (upper panel). The reflectance at 4.2 K as a function of magnetic field (lower panel).

magneto-optical effects correlated well with the onset of finite resistance.

It is surprising that at low temperatures, 4.2–50 K, our spectra show no change with applied field. We found this result in films from two laboratories and on three different substrates. It is in agreement with two earlier studies, [3,9] but appears to disagree with other infrared [4,8,10] and terahertz [11] experiments. To acquire a better understanding, we consider several effects at low temperatures. The applied field threads vortices through the film, and one expects these vortices to have several effects on the electromagnetic response of the film. First, the cores of the vortex contain normal (unpaired) charge carriers and these carriers should interact with the infrared electromagnetic field, giving rise to dissipation. Second, the infrared field drives a supercurrent that interacts with moving vortices causing a flux-flow loss. Third, the carriers in the vortex cores are no longer in the superconducting condensate, so the average superfluid density is decreased. We consider each of these effects in turn.

There were many studies of the quasi-particle states inside a vortex core [16–21]. The physics of the vortex core is usually described by the Bardeen–Stephen model, [17] a dirty-limit description ($l < \xi$) where the motion of the quasi-particles get randomized within the core. (Here, l is the mean free path, ξ the coherence length.) In the clean limit, [18] where ($l > \xi$) and for $H \ll H_{c2}$ in the case of s-wave symmetry, there is a quasi-continuum of bound states in the vortex core and a very small energy of the lowest bound state (minigap) of $E_0 \approx \hbar^2/m\xi^2 \approx \pi^2\Delta_0/2E_F$, where E_F is the Fermi energy. However, for the high-T_c superconductors the situation is quite different and ξ is about two orders of magnitude smaller than in classical clean superconductors. Hence, only a few bound states occur in the vortex core. Dipole transitions may occur among the $E_{\pm 1/2}$ quasi-particle levels in the vortex core; the field-induced absorption, which were reported earlier, was attributed to these transitions [4].

These transitions are not evident in our high-field measurements. We suggest that anisotropic pairing (or gap) effects in the high-temperature superconductors might modify significantly the states inside a vortex core. In particular, the quasi-particle levels in the vortex cores of d-wave superconductors differed significantly from the s-wave case. In addition to the set of localized levels similar to that found in s-wave superconductors there are also continuum levels outside the core that are associated with the s-wave admixture induced by the vortex [22–24].

The flux-flow loss can be understood in a simple way. The applied infrared field drives a supercurrent $\vec{j}_s$in the film, and this supercurrent imposes a force $\phi_0\vec{j}_s \times \hat{z}$ on the vortices. (Here, ϕ_0 is the flux quantum and $\hat{z}$ the direction of the external magnetic field, assumed to be perpendicular to the film surface and the applied infrared electric field.) In the ideal case (no pinning) but with a phenomenological viscous damping force on the moving vortex, the vortices have a steady-state drift at right angles to the supercurrent. If the damping force is written as $\eta\vec{v}_L$, where $\vec{v}_L$ is the vortex lattice drift velocity, then in the steady-state the two forces balanced and $\vec{v}_L = \phi_0\vec{j}_s \times \hat{z}/\eta$. The moving flux induces an electric field $\vec{E} = \vec{B}_0 \times \vec{v}_L$ that is parallel to the supercurrent and the time average $\langle \vec{E}\cdot\vec{j}_s \rangle$ gives a resistive loss.

A number of additional effects must be added to the simple picture described earlier. To be complete, one

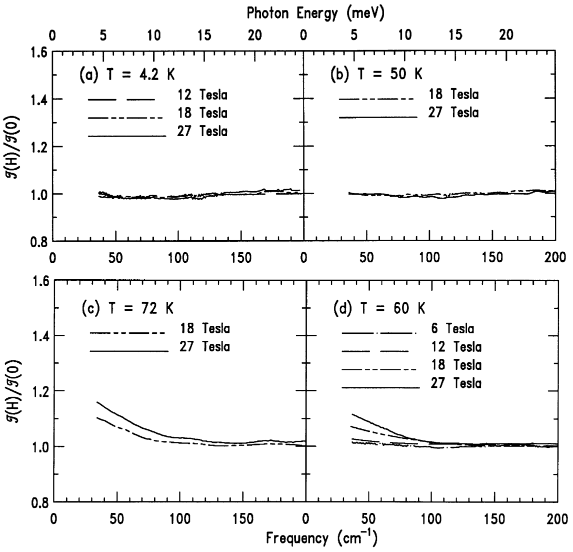

Fig. 4. The magneto-transmittance at several temperatures and magnetic fields for YBCO/MgO films (a)– (c) 400 Å, (d) 600 Å.

may need to include the effect of pinning, to add the Magnus (hydrodynamic) force, to consider the inertial mass of the vortex, and to allow for cyclotron motion [1,5,6,25]. In particular, pinning is necessary to have zero resistance in the field, and at low temperatures the flux lattice in the high-T_c materials is pinned. At high frequencies, the vortices oscillate within their pinning potential [26] and their natural motion in the presence of superfluid flow is of cyclotron type, i.e., adiabatically following the superconducting condensate [27]. The cyclotron gives a chiral response, but no effect in unpolarized light [5,6].

We now discuss the change of superfluid density. The area within the vortex cores is in a ''normal'' state; and that outside in the superconducting state. The fractional area of the cores is $H/H_{c2}(T)$, where H_{c2} is the upper critical field. The superfluid density is be decreased by this factor, so $\omega_{ps}^2 \rightarrow \omega_{ps}^2[1 - H/H_{c2}]$, where ω_{ps} is the superfluid plasma frequency. This loss of spectral weight from the superfluid condenstate appears in the vortex response. The vortex response will thus be proportional to H/H_{c2} and the change in the dielectric response can be attributed to the pair-breaking effect of the field and to quasi-particle excitations inside the vortex cores. This depletion of the superfluid condensate was used to explain terahertz impedance measurements in YBCO and BSCCO films, with pair breaking playing a significant role [11]. In the terahertz measurements [11] a 25%–50% reduction in the superfluid density with 8 T applied field was reported.

If the superfluid density is decreased by a factor of 2, $\sigma_2(\omega)$ will be reduced by a similar amount; in turn the transmittance will increase, with the amount of increase determined by the spectrum of the absorption. Yet, we see no decrease of $\sigma_2(\omega)$ and no increase in transmittance when fields of up to 27 T are applied at low temperatures. One possible explanation is that in our samples the frequency scale for the vortex loss, γ_v, is small. If $\omega \gg \gamma_v$, then the dielectric response is little different from that of the superconductor. (A narrow Drude peak, for example, looks very much like a delta function.) This explanation might reconcile the terahertz results [11] with our data. However, our zero-field measurements show a normal-fluid component, with $1/\tau_D \approx 50\ \mathrm{cm}^{-1}$ for $T < 50$ K and the viscous damping of the vortex motion should be of similar order. In addition, the effects of a transfer of spectral weight out of the superfluid may be measured to quite high frequencies.

Thus, we find no fundamental reason as to why our data differ from those of earlier experiments [4,8,10,11]. The difference may be because of differences among the samples or some other cause. We can say from our measurements (Figs. 1 and 2 and Ref. [14]) that the films we investigated are quite similar to high-quality films studied earlier [12,15].

The contrast between the high and low temperature data in Fig. 4 is striking. Above 60 K at 27 T, our films have a finite dc resistance, which implies that the system has entered the flux flow regime. In this regime, we observe a definite field-induced increase in transmission at low frequencies. At these elevated temperatures, one can expect that vortices become more mobile and thermally activated (Brownian) motion becomes possible. The electromagnetic interaction of the induced currents with the vortex lattice was treated explicitly by many workers [1,17,28,29]. In these models, the interaction with the vortices is treated phenomenologically by introducing an effective vortex mass M, pinning force constant κ_p, and vortex viscosity η. The latter two lead to a vortex relaxation time $\tau_v = \eta/\kappa_p$.

Unfortunately, it is difficult to relate our experimental data to the model described earlier because the case of a clean limit in our films must be distinguished from the Bardeen–Stephen [17] model, which is valid in the dirty limit. Further, we are presently unaware of any calculations for the vortex dynamics in the flux flow regime taking into account the influence of a clean limit and possibly anisotropic symmetry of the order parameter and can give the following qualitative discussion. The total transmittance through a thin film of thickness $d \ll \lambda$ is [30] $\mathscr{T} = 4n/[(y_1 + n + 1)^2 + y_2^2]$, where $y_1 + \mathrm{i}y_2 = (4\pi/c)d(\sigma_1 + \mathrm{i}\sigma_2)$ is the film admittance. At temperatures where the dc resistance is not zero – on account of dissipative flux motion – the London screening and the imaginary part of the conductivity σ_2 is significantly reduced. Then, the transmittance is increased at low frequencies.

Finally, we can compare the magnetic phase diagram for YBCO films from our measurements with data from dc transport and I–V measurement [31]. The magnetic phase diagram for high-T_c superconductors is not simple [32] and we cannot expect to use the far-infrared measurements simply to study the phase boundaries in the H–T diagram. At least in principle, however, there is a relationship between far-infrared properties in a magnetic filed and two distinct parts (the vortex solid and the vortex liquid) on the H–T phase diagram. At low temperatures the vortices are not easily moved because of pinning effects, i.e., the vortex lattice is most likely a solid and the magnetic field has no effect on our spectra. However, when temperature is increased into the vortex liquid state and the flux pinning is overcome, vortex motion is driven by optical current and there is a corresponding change in the far-infrared properties. Thus, a magnetic-field-induced enhancement of the transmittance can be explained as the ac analog of flux-flow resistance.

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