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 reflectance ($\mathcal{R}$) and transmittance ($\mathcal{T}$) of superconducting *ab*-plane-oriented $YBa_2Cu_3O_{7-\delta}$ films in magnetic fields up to 30 Tesla. The frequency-dependent optical conductivity is determined directly from the $\mathcal{R}$ and $\mathcal{T}$ spectra. The application of magnetic field (with *H* perpendicular to the *ab* plane and With unpolarized light) at low temperatures 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.

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The electronic properties of high-T_c superconductors are affected by the application of magnetic fields.[1-3] 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. With an applied current density $\vec{J}^{ext}$ and average magnetic flux density $\vec{B}$, there will be a Lorentz force density $\vec{f} = \vec{J}^{ext} \times \vec{B}$ on the vortices. If the vortices are at rest (pinned), the resistance will be effectively zero whereas if the vortices are moving with a mean velocity $\vec{v}$, an electric field $\vec{E} = \vec{v} \times \vec{B}$, which is parallel to $\vec{J}^{ext}$, appears, leading to a finite electrical resistance. However, the complex behavior of vortex motion in the presence of viscous damping, pinning forces, and fluctuations either of thermal origin or due to the influence of defects in the sample is complicated and is not yet well understood.

Historically, microwave experiments have been widely used to study the vortex dynamics in type II superconductors.[4] When the vortices are pinned, their dynamics are invisible to dc transport. On the other hand, microwave measurements can sense the fluctuations about the pinning sites and give information about the effective pinning force constant and vortex viscosity. There have been numerous measurements of the effect of magnetic fields on the microwave response functions of the high-T_c superconductors.[5-8] It *is generally* agreed that the microwave results are affected by two mechanisms: vortex motion and superconducting condensate depletion. More recently, terahertz time domain spectroscopy spans the frequency from 100 to 1000 GHz (3.3-33.3 cm^{-1}).[9-11] The nonlinear dependence of the complex conductivity as a function of magnetic field has been interpreted in terms of enhanced pair breaking due to nodea in a d-wave gap function.

At higher frequencies, far-infrared spectroscopy has been applied *several* times to investigate the vortex dynamics in the high-T_c superconductors. In most cases, evidence for field-induced absorption is seen, although there are some differences among the measurements. Brunel *et al.*[12] *reported* the reflectance $\mathcal{R}$ of $Bi_2Sr_2CaCu_2O_8$ (BSCCO) at several far-infrared frequencies, w, as a function of temperature, *T*, and magnetic field *H* up to 17 Tesla. They inferred *a* value for the superconducting gap by setting $2\Delta(T,H) = \hbar\omega$ at the *T* and *H* where $\mathcal{R}$ first dropped below the low-T, zero-H value. The drop in far-infrared reflectance corresponded to the onset of a resistive state, and thus only occurred at the higher temperatures or fields. In contrast, the far-infrared transmittance measurements through a thin film of $YBa_2Cu_3O_{7-\delta}$ (YBCO) in magnetic field by Karrai *et al.*[13,14] showed an increase in transmission below ~ 125 cm-1 with increasing field. This 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 temperature as low M 2.2 K and in magnetic fields between 2 and 15 Tesla. The experiments prompted several theoretical calculations *of* the optical response *of* the vortex core **states**.[15–18] The theory of Hsu including the vortex motion[17,18] **describes** the **experimentally-observed chiral** response[14] very **well**, but the **agreement** for the **nonchiral response** at ~ 65 cm^{-1},[13] which the authors attributed to the vortex core resonance, **is only** partial. Shim-to *et al.*[19] **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 up to 100 **Tesla**; the behavior **was** explained by a flux-flow model. **Gerrits** *et al.*[20] reported practically no influence of the magnetic field up to 15.5 **Tesla** in the far-infrared reflectance of YBCO thin **films** at 1.2 K. **Eldridge** et *al.*[21] **measured** the ratio of normal incidence **reflectance** $\mathcal{R}$ of a YBCO **film** in a magnetic field to that in no field, in conjunction with a separate measurement of 5?(0), to obtain the absolute **values** of $\mathcal{R}(H=$ 0.7, 1.4, and 3.5 **Tesla**) 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}. They were able to fit the shape, but not the magnitude, of the peak by a **theory** involving vortex motion with pinning.

In **this** paper, we report the **far-infrared** reflectance ($\mathcal{R}$) and transmittance ($\mathcal{T}$) measurements of superconducting YBCO **films** in magnetic **fields** Up to 30 **Tesla**. We use $\mathcal{R}$ and 9 to extract the frequency-dependent optical conductivity **as** a function of temperature and applied magnetic field. **In** addition, we carried out a **detailed** study of the zero-field $\mathcal{R}$ and $\mathcal{T}$, obtaining results in agreement with previous data.[22,23] The application of magnetic field (with *H* perpendicular to the *ab* plane and with unpolarized light) **at** low **temperatures produces** no discernible field dependence. This observation differs from **several** far-infrared and millimeter measurements in this temperature **range**. Only at fields and temperature where the **dc** resistance is **not zero—on** account **of** dissipative flux motion—is a field-induced effect observed.

We have studied three types of YBCO samples: (1) thin (300-500 A) films made by *a* **KrF excimer laser** deposition on a $PrBa_2Cu_3O_{7-\delta}$ buffer **layer** on $YA10_3$ **substrates** at Jet Propulsion Laboratory; (2) thin (400-600 A) **films prepared** by pulsed-laser ablation at **Tsing** Hua University on (001)-oriented **MgO** substrates; and (3) a **5000-Å-thick film** deposited on a $SrTi0_3$ substrate by a **similar** method. All the **samples** have been structurally characterized by x-ray diffraction, which **has** clearly shown their **c-axis** orientation. The superconducting properties **of films have** been determined by dc resistivity or **ac** susceptibility **measurements**. The characteristics of **all samples** are listed in Table I. The **5000-Å-thick** film **gives** a higher onset temperature and sharper transition. On the other hand, **samples** prepared in similar conditions have shown close values *of* **resistivity** at room temperature,

The **films** with *d* <1000 A were studied in both **reflectance** ($\mathcal{R}$) and transmittance ($\mathcal{T}$) **whereas** the **5000-Å-thick** film on $SrTiO_3$ **was** studied only in $\mathcal{R}$. The far-infrared **studies** in magnetic field were carried **out** at the National High Magnetic Field Laboratory. Our measurements used a Bruker spectrometer and light-pipe optics to carry the far-infrared radiation through the magnet. The sample probes used in conjunction with a 20-Tesla superconducting magnet permit alternate sample and reference measurements (for both $\mathcal{R}$ and $\mathcal{T}$), allowing absolute measurements of these quantities. For the reflectance, an Au mirror was used as a references, while the transmittance was done relative to an empty diaphragm. The $\mathcal{R}$ and $\mathcal{T}$ were obtained at 4.2 K and applied fields Up to 17.5 Tesla. We also employed a 30- Tesla resistive magnet; in this magnet, only transmittance ratio is accessible. Thus, we report $[\mathcal{T}(H)/\mathcal{T}(0)]$ at temperature between 4.2 and T“. In all measurements, the unpolarized far-infrared radiation was incident nearly normal to the film, so that the electric field was in the ab plane. The magnetic field H was perpendicular to the ab plane. A detailed description of the experimental setup is given elsewhere.[24]

The complex dielectric fiction $\epsilon(\omega)$ or optical conductivity $\sigma(\omega)$ [where $\epsilon(\omega) = 1 + 4\pi i\sigma(\omega)/\omega$] were calculated **directly from the** measured $\mathcal{R}$ and $\mathcal{T}$ in the far-infrared region. To deal with dispersive and absorption effects in the substrate, the reflectance $\mathcal{R}_{sub}$ and transmittance $\mathcal{T}_{sub}$ of a bare $YAlO_3$ 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 analysis of the data for the films. These measurements turned out to be crucial in the case of the $YAlO_3$ substrata, where a doping with 4 at.% Nd gives field-dependent features in the transmittance.[25]

In general, we found that the analysis of $\mathcal{R}$ and $\mathcal{T}$ gave more accurate results for the low-frequency (ω <100 cm^{-1}) dielectric response than optical reflectance of bulk single crystal or thick-film samples followed by Kramers-Kronig analysis, where extrapolation to zero and infinite frequencies were needed. A detailed discussion of the analysis for transmittance and reflectance has been given in previous work.[22,23]

In Fig. 1 we show the reflectance and transmittance of a 500Å YBCO/200Å PBCO/$YAlO_3$ film at 4.2 K and at several magnetic fields. We observed practically no influence of the magnetic field on the infrared response of the film. The band at 112 cm-l, which shifts to 130 cm-1 with field, was traced to the substrate, as mentioned above. The magnetic field and frequency-dependent conductivities σ_1 and σ_2 of the film are shown in Fig. 2. The spectra show no discernible field dependence at 4.2 K. This observation differs from previous *far-infrared* measurements in this temperature and frequency range.[13,19,21] Despite the lack *of* field dependence in the either σ_1 or σ_2 spectra, two interesting observations can be made. First, with the external field perpendicular to the ab plane of the superconducting YBCO film, no far-infrared magnetoresistance was detected at 4.2 K and in the high-field regime. Second, down to the low-frequency limit ($\sim$ 35 cm-1) of our measurements, the dielectric response does not change with magnetic field. Thus our results also differ from the data obtained in the terahertz region, where which reported a change in the σ_1 and σ_2 of YBCO and BSCCO films were reported over a broad range of field, frequency, and temperature.[9-11] Even t bough the terahertz measurements were done at frequencies below our low frequency limit, the changes in σ_1 and σ_2 that were seen would have extended well into our frequency range.

Figure 3 shows the temperature and field-dependent reflectance spectra of the 5000-Å-thick film. The low temperature reflectance is near unity at $\omega \rightarrow 0$. With increasing frequency it falls off slowly, showing a pronounced shoulder at ~ 450 cm-'. Increasing temperature decreases the reflectance in a characteristic way. The lower panel of Fig. 3 displays 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. In particular, the field independence of the 450 cm^{-1} edge favors the non-superconducting explanations of this feature.[26] Our results are in agreement with one earlier experiment,[20] but differ from several previous reports.[13,19,21]

Magneto-transmission measurements at several temperature and magnetic fields for both 400A and 600Å 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 to 27 Tesla. Typical noise variations of our measurements in a magnetic field are on the order of ± 1%. The low frequency limit of the measurement (about 35 cm-l) is due to 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-l transmission of these films is seen to change by more than 5% as field is increasing from 6 to 27 Tesla; changes at higher frequencies (ω >100 cm-l) are zero within our experimental error. Thus, large magnetic-field-induced increases in transmission occur only at temperature not too far below T,.

It is surprising that at low temperature 4.2-50 K, our spectra do not show any change with applied field. This result is in agreement with two early studies,[12,20] but appears to disagree with other infrared[13,19,21] and terahertz[9-11] experiments. To acquire a better understanding, we consider a simple picture at $T \ll T_c$. The vortex can be driven either by an ac electric field or by superflow. At high frequencies, the vortices oscillate within their pinning potential[27] and their natural motion in the presence of superfluid flow is of cyclotron type, i.e., adiabatically following the superconducting condensate.[28] The area within the vortex cores is in a "normal" state, that outside of them is in the superconducting state. The fraction area of the cores is $H/H_{c2}(T)$, where H_{c2} is the upper critical field. The superfluid density is expected to 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. The vortex response will 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 quasiparticle excitations inside the vortex cores. This depletion of the superfluid condensate has been used to explain terahertz impedance measurements in YBCO and BSCCO films, with pair-breaking playing a significant role.[9-11]

Why is there no such effect in our study? One possibility would be to assume 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 would reconcile the terahertz results[9-11] with our data. There are two problems with this explanation, First, our zero-field measurements show a normal-fluid component, with $1/\tau_D \approx$ 50 cm-l for $T < 50K$ and the viscous damping of the vortex motion should be of similar order. Second, in the terahertz measurements[9-11] there is about a 25% reduction in the superfluid density observed; in conduction with the change of σ_1 with field, this large effect suggests a similar frequency scale in these samples.

There have been many studies of the quasiparticle local density of states inside a vortex core.[29-35] The physics of the vortex core for a type II superconductor is usually described by the Bardeen-Stephen model,[31] a dirty-limit description ($l < \xi$) where the motion of the “ quasiparticles gets randomized within the core. In the clean limit,[32,33] where ($l > \xi$), and for $H < H_{c2}$ *in* the case of s-wave symmetry, there is *a* quasicontinuum of bound states in the vortex core and a very small energy of the lowest bound state (minigap). However, for the high-T_c superconductors the situation is quite different, since A is larger and E_F is smaller than in the classical superconductors. (ξ^2 is about four orders of magnitude smaller than in classical clean superconductors). Hence, only a few bound states occur in the vortex core. Recent evidence for a large core spacing has been found in scanning tunneling spectroscopy (STS) on YBCO single crystals.[36]

Dipole transitions among the quasiparticle levels in the vortex core, which have been reported earlier,[13] 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. The quasiparticle levels in the vortex cores of *d*-wave superconductors differ 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 s-wave admixture induced by the vortex.[37-39] Kopnin and Volovik[40] pointed out in a clean d-wave superconductor, gap nodes cause the electronic density of states associated with a vortex to diverge at low energies: $N_{vortex}(E) \sim 1/|E|$. This divergence will presumably be cut off in a dirty *d*-wave case. At higher fields the distance between the *cores* become small and the electronic structure of the vortex core will be influenced by the competition between electrodynamic effects and spatial variations of the superconducting order parameter near the vortex core. Thus, it seems likely that the excitations from the continuum levels might remove spectral weight from the $E_{\pm 1/2}$ state making its oscillator strength too small to be observed. For our YBCO films, we may approach already the limit where the core region of the vortex is empty of low-energy excitations.

As can be seen in Fig. 4. the contrast between the high and low temperature data is striking. Above 60 K where the system enters the flux flow *regime,* we observe a definite field-induced increase in transmission at low frequencies. At these elevated temperatures, one may expect that vortices become more mobile and thermally activated (Brownian) motion becomes possible. The electromagnetic interaction of the induced currents with the vortex lattice has been treated explicitly by many workers.[31,41–43] 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 above model because the case of a clean limit in our films must be distinguished from the Bardeen-Stephen[31] model which is valid in the dirty limit. Furthermore, 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. We can give the following qualitative discussion. The total transmittance through a thin film of thickness $d \ll \lambda$ is[44] $\mathcal{T} = 4n/[(y_1 + n + 1)^2 + y_2^2]$ where $y_1 + iy_2 = (4\pi/c)d(\sigma_1 + i\sigma_2)$ is the film admittance. At temperature 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 measurements.[45,46] The magnetic phase diagram for high-T. superconductors is not simple[47] and we cannot expect simply to use far-infrared measurements to study the phase boundaries in the H - T diagram. At least in principle, however, there is relationship between far-infrared properties in a magnetic field 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 due to pinning effects, i.e., the vortex lattice is moat 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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Table I. Sample characteristics.

Sample	Thick	T_c	ΔT_c	ρ_{dc}(300 K)	λ_L(~ 20 K)
	A	K	K	$\mu\Omega$-cm	A
YBCO/200Å PBCO/$YAlO_3$	300	83.5	3.5	550	2500±100
YBCO/200Å PBCO/$YAlO_3$	500	85.0	2.5	590	230W100
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

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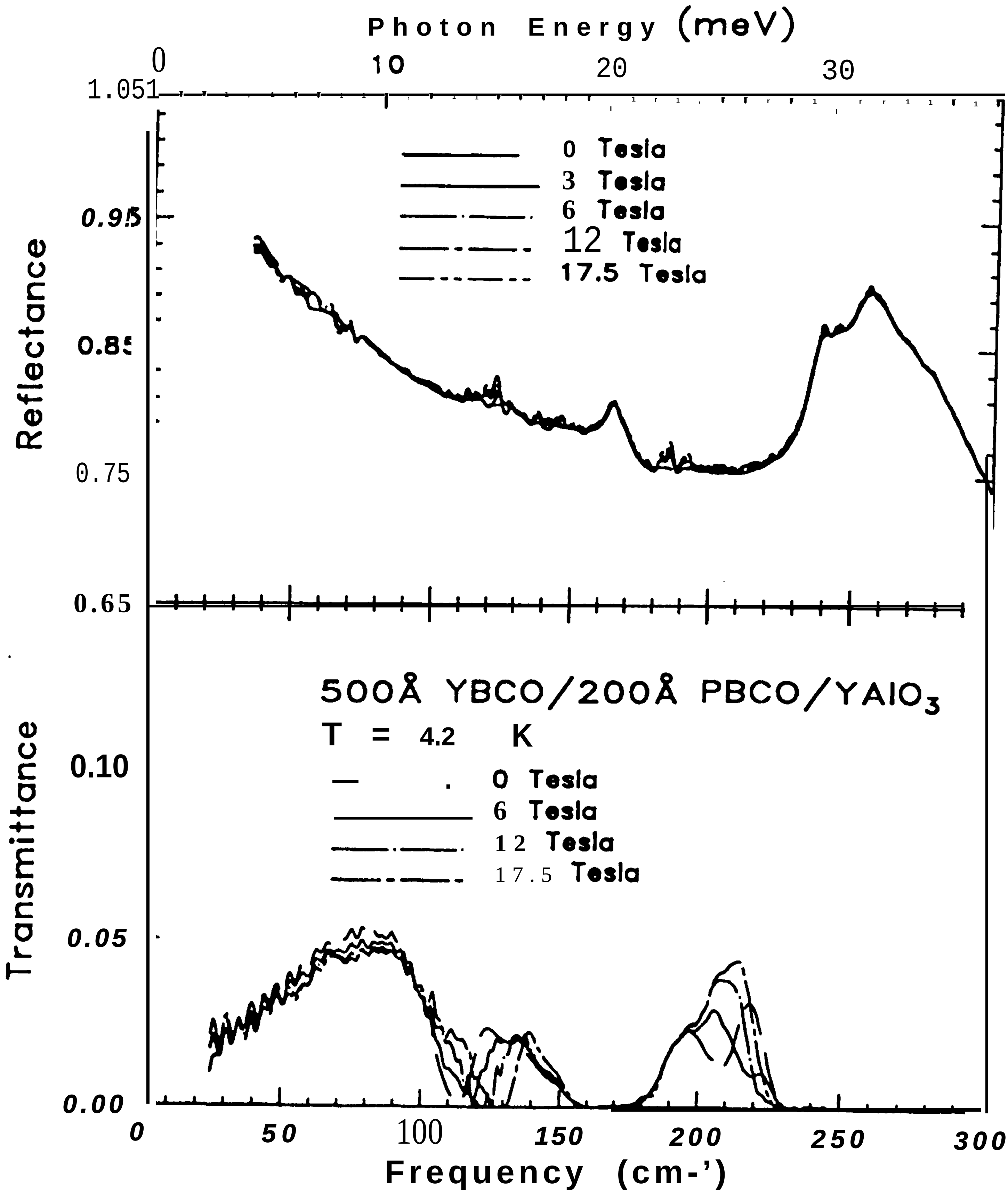
Figure captions

Fig. 1. The 4.2-K reflectance (upper) and transmittance (lower) *of a* 500Å YBCO/200Å PBCO/$YAlO_3$ film at several magnetic fields.

Fig. 2. The magnetic field and frequency-dependent conductivity of a 500Å YBCO/200Å PBCO/$YAlO_3$ film at 4.2 K.

Fig. 3. The measured zero-field reflectance (upper) of • SO (IOA YBCO/$SrTiO_3$ film at 10, 70, and 100 K. (Lower) displays the reflectance spectra at 4.2 K as a function of magnetic field.

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



Photon Energy (meV)
0
10
20
30
1.051
0.95
0.85
0.75
0.65
Reflectance
0 Tesla
3 Tesla
6 Tesla
12 Tesla
17.5 Tesla
500Å YBCO/200Å PBCO/YAlO3
T = 4.2 K
0.10
0.05
0.00
Transmittance
0 Tesla
6 Tesla
12 Tesla
17.5 Tesla
0
50
100
150
200
250
300
Frequency (cm-')

Fig

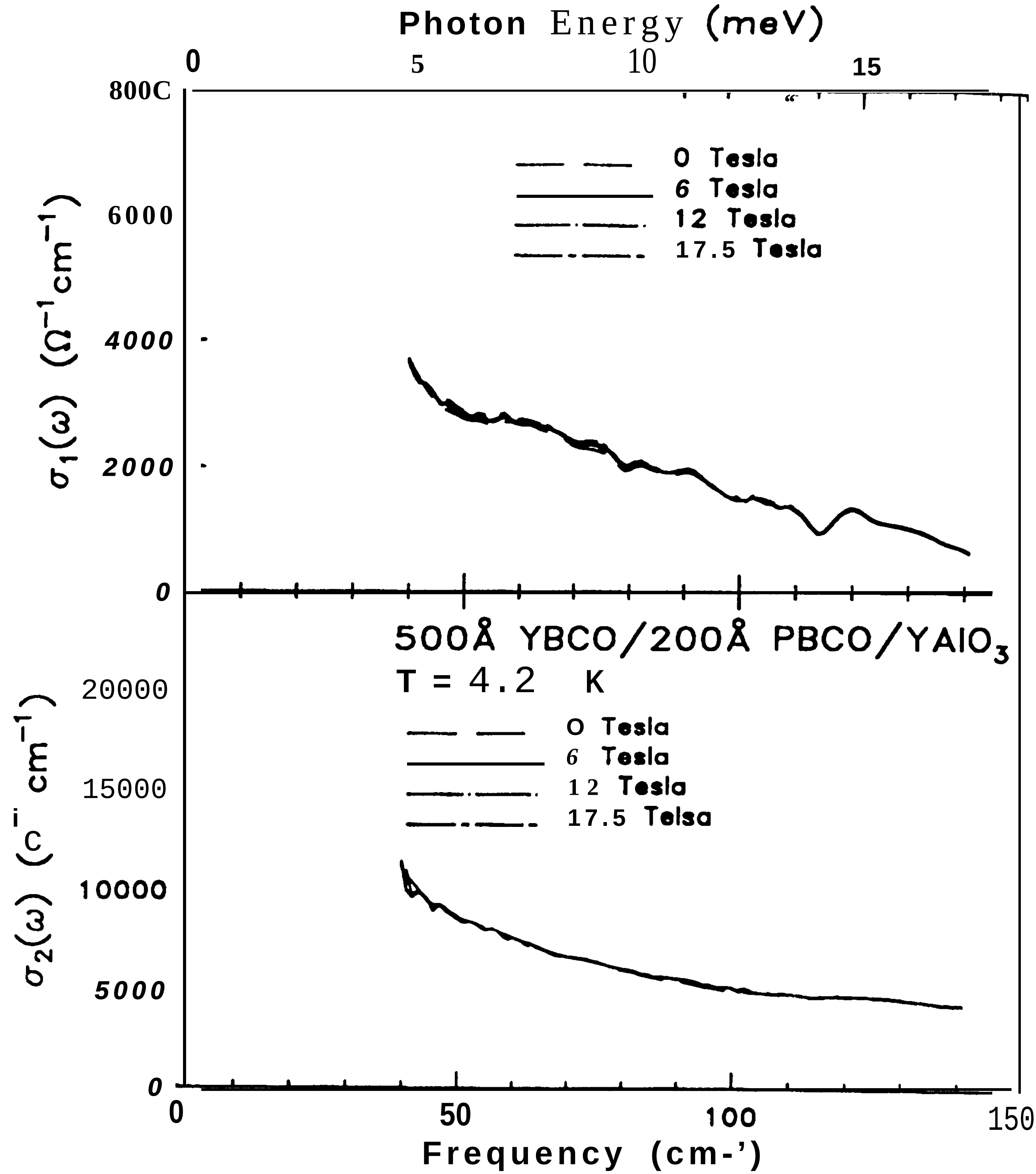
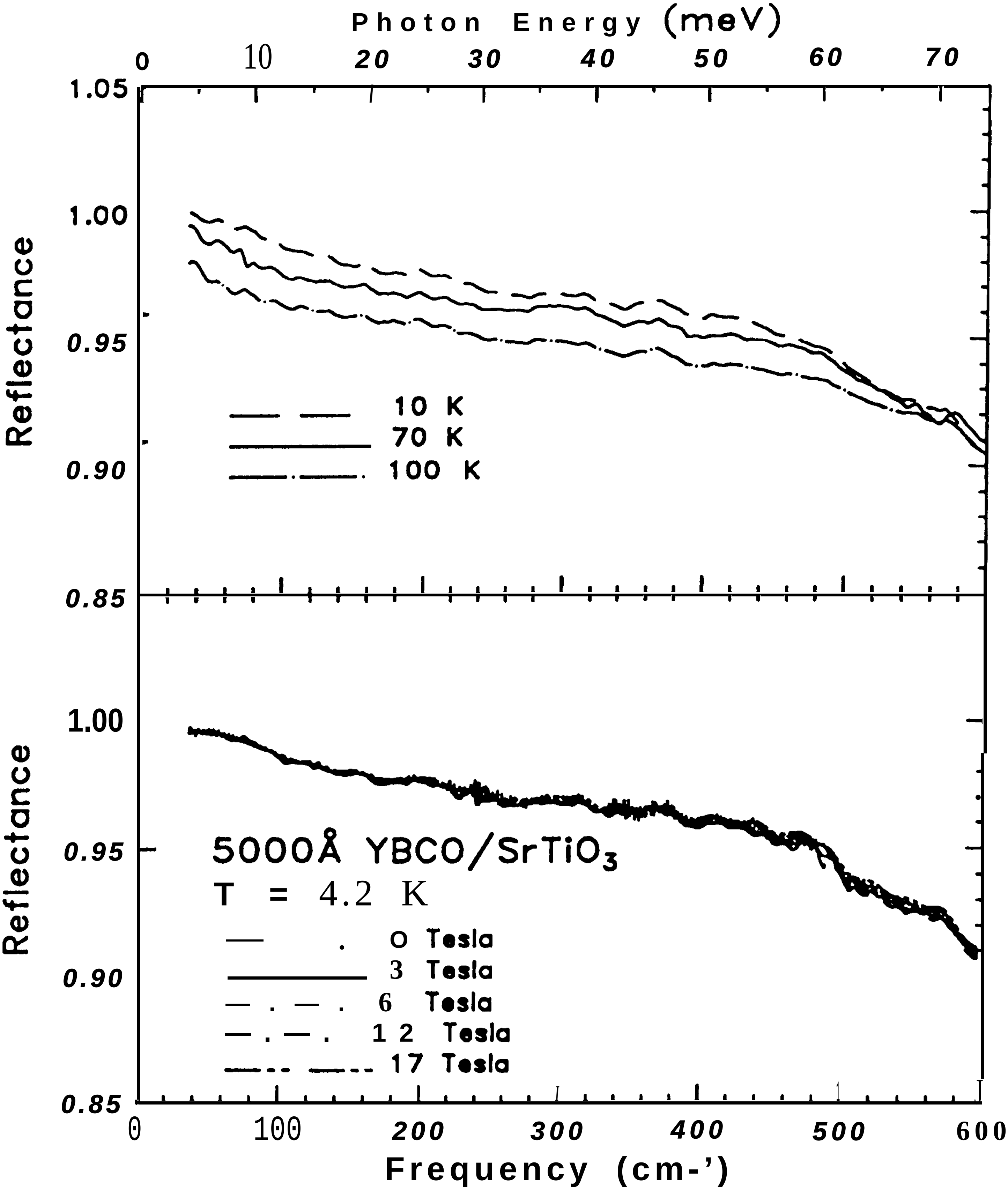

Photon Energy (meV)
0
5
10
15
800C
6000
4000
2000
0
$\sigma_1(\omega)$ ($\Omega^{-1}cm^{-1}$)
0 Tesla
6 Tesla
12 Tesla
17.5 Tesla
500Å YBCO/200Å PBCO/YAlO$_3$
T = 4.2 K
20000
15000
10000
5000
0
$\sigma_2(\omega)$
0 Tesla
6 Tesla
12 Tesla
17.5 Telsa
0
50
100
150
Frequency (cm-')

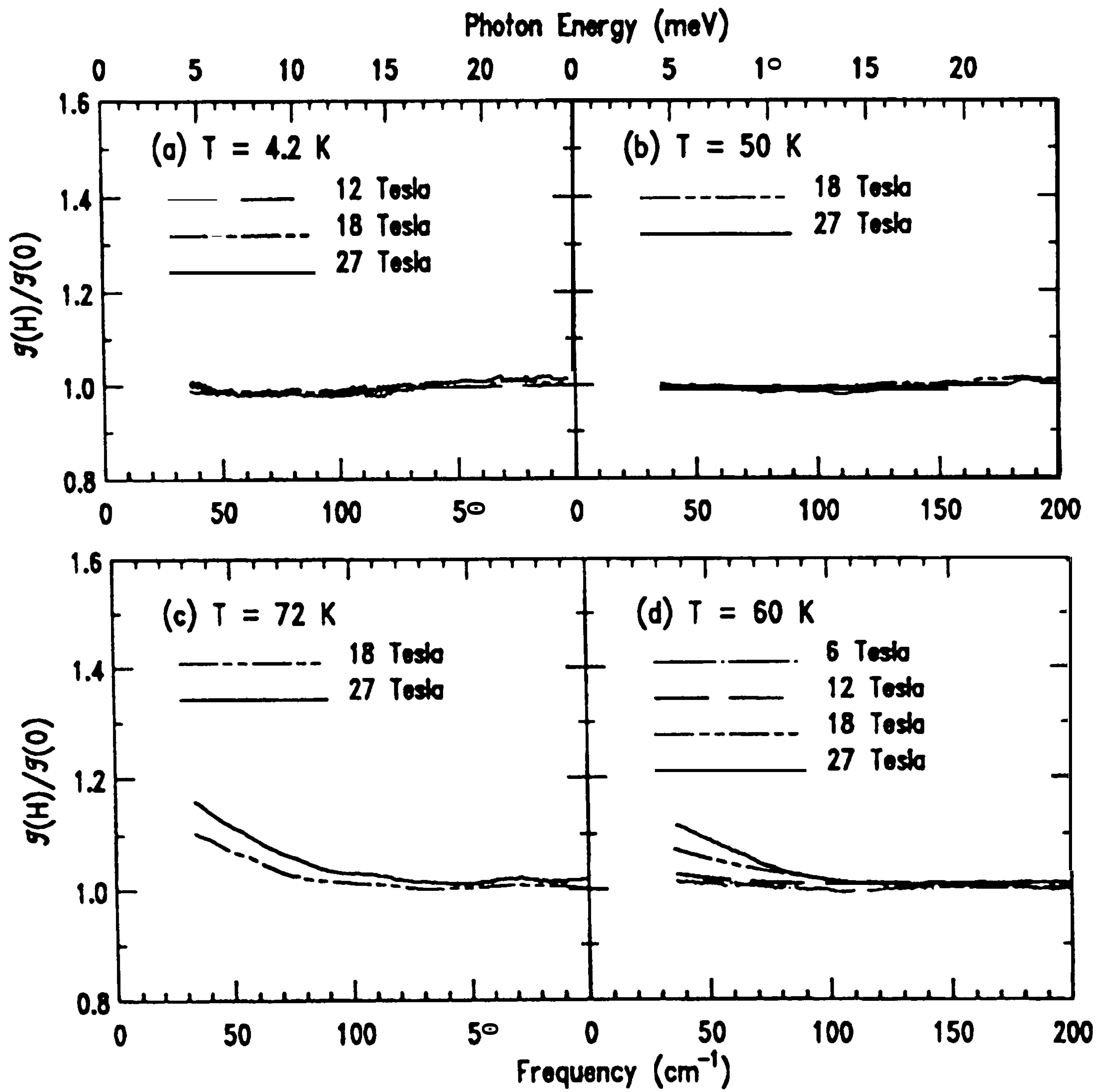
Fig. 2



Photon Energy (meV)
0
10
20
30
40
50
60
70
1.05
1.00
0.95
0.90
0.85
Reflectance
10 K
70 K
100 K
1.00
0.95
0.90
0.85
Reflectance
5000Å YBCO/SrTiO3
T = 4.2 K
O Tesla
3 Tesla
6 Tesla
1 2 Tesla
17 Tesla
0
100
200
300
400
500
600
Frequency (cm-')

Fig. 3

Fig 4



Photon Energy (meV)
(a) T = 4.2 K
12 Tesla
18 Tesla
27 Tesla
(b) T = 50 K
18 Tesla
27 Tesla
(c) T = 72 K
18 Tesla
27 Tesla
(d) T = 60 K
6 Tesla
12 Tesla
18 Tesla
27 Tesla
g(H)/g(0)
Frequency (cm^{-1})