

Far-infrared transmission spectra of granular free-standing thin films of $\text{YBa}_2\text{Cu}_3\text{O}_x$ near the percolation threshold

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We report far-infrared transmittance measurements on granular free-standing thin films of $\text{YBa}_2\text{Cu}_3\text{O}_x$ in the frequency range $10\text{--}650\text{ cm}^{-1}$. The transmittance was measured both on metallic films, in the normal (110-K) and superconducting (10-K) states, and on insulating films. The metallic samples behave as a mixture of conducting and insulating small grains in the vicinity of the percolation threshold. We obtained a fit to the measured transmittance with the two-dimensional effective-medium approximation. The frequency-dependent effective conductivity was calculated and the absorption coefficient was observed to change as $\omega^{0.87}$.

I. INTRODUCTION

The electromagnetic properties of inhomogeneous composites have been a subject of numerous studies.¹ The reason partially lies in the indications that composites have a behavior very different from that of their constituents.^{2,3} In systems where the typical spatial dimensions (particle size) are small compared with the wavelength of the incident electromagnetic field, the inhomogeneous medium can be viewed as being uniform. The propagation of radiation is then described by an effective dielectric function, ϵ_{eff} . The electromagnetic response of the composite depends on whether a connected path of one of the constituents is present through the material. In a metal-insulator composite the conducting path is formed at a critical metal concentration $f = f_c$. The effective response of the system depends on whether f is near to, or much different from, the percolation threshold.⁴

In this paper we study the far-infrared (FIR) transmission properties of free-standing thin films of $\text{YBa}_2\text{Cu}_3\text{O}_x$, consisting of a random mixture of small, roughly spherical, metallic and insulating particles. The size of an individual particle (about $0.5\ \mu\text{m}$) was large compared to atomic dimensions but small compared to the wavelength of the FIR radiation ($15\text{--}1000\ \mu\text{m}$). The films were prepared by a technique described previously.⁵ Since no substrate was present, we were able to cover a frequency range $10\text{--}650\text{ cm}^{-1}$, wider than in previous transmission measurements on these materials.^{6,7}

Previous FIR measurements on oriented $\text{YBa}_2\text{Cu}_3\text{O}_x$ films were done using the single-bounce reflectance method,⁸⁻¹⁴ with a few transmittance,^{6,7,15} and direct absorptivity¹⁶ measurements. The average grain size of samples previously studied was around $10\ \mu\text{m}$, about 20 times the particle size in our experiments. Commonly used lattice-matched substrates, such as SrTiO_3 , KTaO_3 , LaAlO_3 , ZrO_2 , and MgO ,^{17,18} allow formation of predominantly $a-b$ plane oriented films. However, all of the known suitable substrates are either opaque in the FIR

region or have strongly temperature-dependent transmission. Thus it has previously been difficult to observe small changes in transmission due to the films. Williams *et al.*⁶ did perform the transmission measurements on $\text{YBa}_2\text{Cu}_3\text{O}_x$ films. However, the presence of an absorbing substrate in these measurements required the use of the Brookhaven Synchrotron Light Source, which provided radiation 100–1000 times stronger than a conventional mercury lamp.

The FIR properties of metal-insulator mixtures have been previously studied, with samples prepared as metal smokes,^{3,19} metal particles embedded in an insulating host,^{2,20,21} or granular films.^{20,22-25} The smokes consist of unsupported metal particles, at typical volume fractions less than 0.1, with voids between the grains playing the role of the insulator. The preparation of samples consisting of metal particles embedded in an insulator allows good control over the metallic volume fraction. At low metal concentrations, the particles are well separated from each other. The granular metal films are typically formed by simultaneous deposition of insulating and conducting particles onto a substrate. The degree of granularity can be affected by the substrate temperature, the deposition rate, and the pressure during the deposition.

The FIR properties of an inhomogeneous mixture change when the metallic component undergoes the superconducting transition. The properties of such a mixture can be described by a model appropriate for a system in the normal state, with the adequate inclusion of the dielectric function for the superconducting part. As long as it can be assumed that the superconducting grains have bulk characteristics, the effective response can be calculated using the dielectric function of a bulk superconductor.

Our films differ from granular samples studied previously in several ways. The previous FIR measurements on granular metals and low- T_c superconductors^{2,3,20} were done on systems where the metal volume fraction f was well below the critical volume for percolation, f_c . However, our samples were very close to the theoretical per-

colative threshold concentration. It is expected that the behavior of the system close to f_c will be much different from the behavior at small f .⁴ Some of the previous metal-insulator-mixture measurements were also done³ on small metal particles, $\sim 100 \text{ \AA}$ in radius. The particles in the systems described here, however, were more than an order of magnitude larger. While the properties of the larger particles should be well described by the bulk dielectric function,²⁶ this need not be the case for smaller particles, where quantum size effects could play an important role.

The frequency dependence of the transmission, as well as the position of the absorption peaks of our granular free-standing films are explained in terms of a two-dimensional effective-medium approximation (EMA). The results of the EMA were then used to calculate the frequency-dependent conductivity and the absorption coefficient of the films. We found that the behavior of the conductivity was consistent with previous calculations²⁷ for a composite near the percolation transition. However, the absorption coefficient did not follow an ω^2 dependence, previously reported²⁰ for very dilute systems. The Mattis-Bardeen model²⁸ predicts zero absorption in the superconducting state for the radiation frequency ω less than the energy gap, 2Δ . In this model the absorption becomes nonzero at 2Δ , increasing at higher frequencies, but always staying smaller than in the normal state. Carr, Garland, and Tanner²⁰ observed an anomalous infrared absorption in granular low- T_c superconductors with metal volume fraction $f \ll f_c$. They found that for $\omega \sim 2\Delta$ the granular superconductors become more absorbing in the superconducting state than in the normal state.

The typical area of our free-standing thin films was around 100 mm^2 with thicknesses $0.1\text{--}0.5 \text{ }\mu\text{m}$. We studied the transmission properties of several metallic and insulating films in the range from 10 K to room temperature. The samples were characterized using a JEOL JXA-840 electron-probe microanalyzer and a Siemens D500 x-ray diffractometer. The energy-dispersive x-ray analysis indicated that both the metallic and the insulating samples had concentrations of Y, Ba, and Cu close to the stoichiometric values. X-ray results showed that all expected polycrystalline lines were present in the spectra. The metallic films showed the narrow orthorhombic phase splitting, while insulating films were tetragonal. The superconducting transition of the metallic films was measured using both an ac inductive technique and a superconducting quantum interference device magnetometer. The first method gave a slightly higher onset temperature, around 85 K, with a transition width of about 15 K. The T_c for our free-standing films was taken to be 78 K, i.e., at the midpoint of the transition. The fragility of the films did not allow resistivity measurements.

Transmission spectra were obtained using a step-and-integrate fast-Fourier-transform spectrometer. For frequencies between 10 and 200 cm^{-1} the spectrometer was configured as a polarizing interferometer (Specac) with a $12.5\text{-}\mu\text{m}$ wire grid-spacing beamsplitter. From 100 to 650 cm^{-1} the spectrometer was arranged as a conven-

tional Michelson interferometer, and the frequency range was covered using a 12-G Mylar beamsplitter. In our measurements two 1.5-l ^4He Dewars from Infrared Laboratories, Inc. were used. The dual Dewar arrangement enabled the samples to be heated up to room temperature without affecting the temperature of the detector Dewar. The samples with the associated heater controls were placed in the first Dewar, while cold filters mounted on a slide and a composite-doped Si bolometer operating at 1.6 K were placed in the second ^4He Dewar. The filter slide was located in front of the bolometer. The filters could be selected externally, which enabled us to choose the frequency range to be studied without warming up the detector Dewar.

The free-standing films of $\text{YBa}_2\text{Cu}_3\text{O}_x$ were mounted on a four-position copper carousel, each with a clear aperture of about 3 mm diameter. Our transmission reference was a blank aperture. Both the sample and the detector Dewar were cooled to liquid-helium temperature. A heating element, mounted near the sample wheel, allowed the sample temperature to be raised continuously from liquid-helium temperatures to room temperature. The sample temperature was monitored using a calibrated silicon-diode sensor from Lake Shore Cryogenic, Inc. We studied the transmission spectra at the temperatures from 10 to 300 K and concentrated at the frequency region $10\text{--}650 \text{ cm}^{-1}$. In all, three metallic free-standing films of different thicknesses were studied in their superconducting and normal states. The results presented here were taken from one film as a representative of the behavior. In addition, we have also measured several insulating $\text{YBa}_2\text{Cu}_3\text{O}_6$ films. Some of the measurements were done on films that were insulating as deposited. However, the results given here were from a metallic film that had been reannealed in argon to remove the oxygen. This allowed us to compare the spectra of the insulating and metallic states of the same film. The spectra of both the as-deposited and the reannealed insulating films were similar.

II. EXPERIMENTAL RESULTS

In Fig. 1 the solid lines show the transmittance of a metallic $4800 \pm 500\text{-}\text{\AA}$ -thick free-standing $\text{YBa}_2\text{Cu}_3\text{O}_x$ film in the frequency range $10\text{--}650 \text{ cm}^{-1}$ and at two temperatures, 10 and 110 K. The frequency dependence of the transmittance at 110 K can be divided into two regions. Below 100 cm^{-1} , there is a strong frequency dependence of the transmittance as it increases by a factor of 4 as the frequency decreases. Above 100 cm^{-1} , the transmittance varies by less than 2%, with a slight increase in transmittance as the frequency increases. At 10 K, however, the transmittance increases initially for frequencies decreasing below 100 cm^{-1} but flattens off at about 30 cm^{-1} and then drops sharply [Fig. 1(b)].

Note that for frequencies between 30 and 300 cm^{-1} the transmittance at 10 K is higher than at 110 K. An interesting feature in the superconducting state (10 K) is the sharpening of the phonon line at about 156 cm^{-1} . Shown

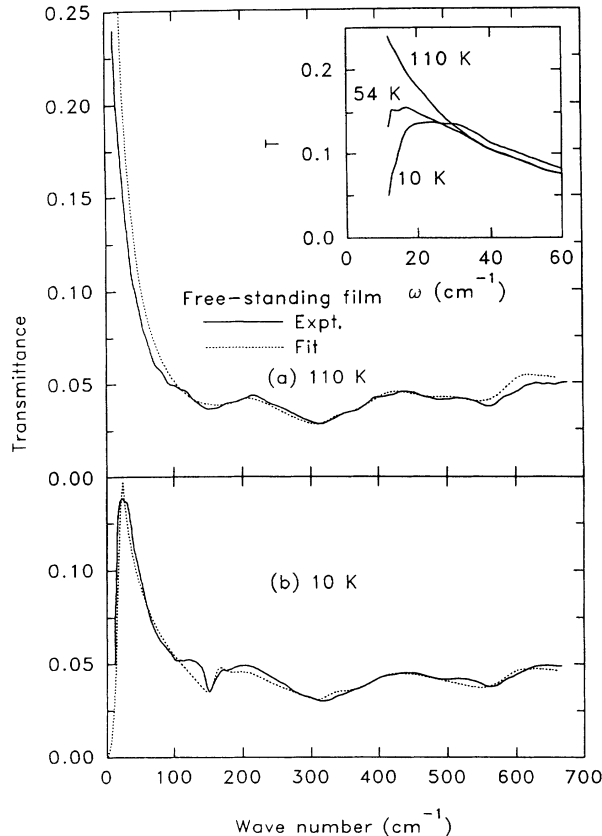


FIG. 1. Measured and calculated transmittance of a granular free-standing metallic film: (a) 110 K; (b) 10 K. The inset shows the measured transmittance below 60 cm^{-1} at three temperatures.

on Fig. 1 are three temperature-independent transmission minima at about 380 , 490 , and 576 cm^{-1} . The lines at 156 , 190 , 270 , 320 , and 576 cm^{-1} are close to the phonon lines observed by reflectance measurements on polycrystalline samples.⁸ The line at 490 cm^{-1} was observed by only a few groups,^{29,30} and may be due to the impurity phases, while the 380 cm^{-1} line was observed in the insulating phase of $\text{YBa}_2\text{Cu}_3\text{O}_x$,³¹ and may be due to the oxygen-deficient grains. The discussion on the lattice vibrations in the $\text{YBa}_2\text{Cu}_3\text{O}_x$ system had been summarized previously.³²

The inset of Fig. 1 shows the temperature dependence of the transmittance below 60 cm^{-1} . One notes that at 10 K the transmittance tends towards zero as the frequency decreases, indicating that there was supercurrent screening. This behavior is consistent with a conventional superconductor, which gives transmission in the superconducting state $T_s \rightarrow 0$ as $\omega \rightarrow 0$. At an elevated temperature (54 K) the transmittance showed reduced screening.

A frequency dependence of the transmission, similar to that observed here, was reported by Carr, Garland, and Tanner²⁰ on granular Pb films with the grain size on the order of $0.1 \mu\text{m}$. They found that the normal-state transmission at low frequencies decreased with in-

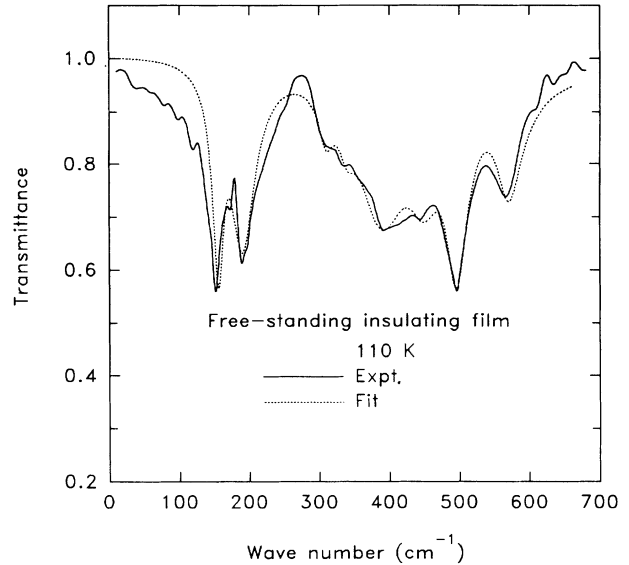


FIG. 2. Measured and calculated transmittance of an insulating granular thin free-standing film. This film is the same film shown in Fig. 1 except it was reannealed to a nominal oxygen concentration of 6.5.

creasing frequency. Transmission measurements in the spectral region 40 to 290 cm^{-1} on partially oriented $\text{YBa}_2\text{Cu}_3\text{O}_x$ films deposited on MgO substrates were reported by Williams *et al.*⁶ Also, Forro *et al.*¹⁵ studied the FIR transmission of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ single crystals. The frequency and temperature dependence of the transmission of these systems differs from that obtained for our systems. We account for the differences by accounting for the percolation effects that govern the behavior of our films.

Next, we measured the FIR response of several oxygen-deficient films. The energy-dispersive x-ray analysis showed that all of the insulating films were close to the stoichiometric ratio. X-ray results showed that they were of the tetragonal phase. With their thicknesses taken into account, all of the insulating films show similar FIR response. Some of the films were insulating as deposited. The results presented here were from the metallic film described earlier, after it was annealed at 550°C in an argon atmosphere. The measurements were done at 10 , 54 , 110 , and 298 K . Within the experimental error the results were temperature independent, and we present here the transmittance at 110 K . As shown in Fig. 2, the transmittance of the insulating films is markedly higher than of the metallic ones, with strongly pronounced absorption peaks. The removal of the oxygen in these films creates a material with no free carriers. The screening of the phonons is therefore reduced and the absorption lines become sharper.

III. CALCULATIONS

The transmittance results can be understood by examining the characteristics of the free-standing films. As

an electron micrograph picture of the film showed, and due to the nature of the preparation technique, the free-standing films consisted of particulates about half a micron in diameter. The transmission of the metallic films could be described assuming a random mixture of conducting and insulating grains. The particle size ($\simeq 0.5 \mu\text{m}$) was small compared to the wavelength of the incident electromagnetic field. The optical properties of such a medium can be described by an effective dielectric function, which depends on the dielectric functions of a metal and an insulator, the volume fraction of the metal, and the particle shape. Since the dimensions of the particles were comparable to the film thickness ($\simeq 0.5 \mu\text{m}$), we modeled the data with the two-dimensional EMA.³³ The particles were roughly spherical in shape and had a narrow size distribution centered around a half micrometer. The effective dielectric function of the medium, $\epsilon_{\text{eff}}(\omega)$, was then obtained from the equation

$$(1-f) \frac{\epsilon_i(\omega) - \epsilon_{\text{eff}}(\omega)}{\epsilon_i(\omega) + \epsilon_{\text{eff}}(\omega)} + f \frac{\epsilon_m(\omega) - \epsilon_{\text{eff}}(\omega)}{\epsilon_m(\omega) + \epsilon_{\text{eff}}(\omega)} = 0, \quad (1)$$

where ϵ_m and ϵ_i are the dielectric functions of the metal and the insulator respectively, and f was the fraction of the metallic component. The physical solution of Eq. (1) is determined by the requirements that ϵ_{eff} be continuous and that $\text{Im}\epsilon_{\text{eff}} > 0$.

The presence of transmission minima in our spectrum suggests that the complete description of ϵ_m should include the bound Lorentz oscillators in addition to the Drude function

$$\begin{aligned} \epsilon_m(\omega) &= \epsilon_0 + \sum_j \frac{\omega_j^2}{\omega_{0j}^2 - \omega^2 - i\gamma_j\omega} - \frac{(\omega_p\tau)^2}{(\omega\tau)(\omega\tau + i)} \\ &\equiv \epsilon_0 + \epsilon_L + \epsilon_D, \end{aligned} \quad (2)$$

where the oscillator parameters are given in Table I. The lines at 490 and 380 cm^{-1} were ascribed to the impurity phases and the oxygen deficiency. They were included in the dielectric function, ϵ_i , of the insulating grains but not in the Lorentz term for the metallic grains.

The free-carrier parameters were as follows: plasma frequency, $\omega_p = 1 \text{ eV}$ ($= 8000 \text{ cm}^{-1}$) and $\tau^{-1} = 200 \text{ cm}^{-1}$, while ϵ_0 was 13. The value of f was 0.48, close to the percolation threshold for the two-dimensional system,

TABLE I. Fitting parameters for metallic films.

| ω_{0j} (cm^{-1}) | 10 K | | 110 K | |
|------------------------------------|---------------------------------|------------------|---------------------------------|------------------|
| | γ_j (cm^{-1}) | Ω_j (meV) | γ_j (cm^{-1}) | Ω_j (meV) |
| 156 | 20 | 21 | 100 | 38 |
| 190 | 60 | 25 | 75 | 32 |
| 270 | 80 | 6 | 100 | 6 |
| 320 | 45 | 31 | 65 | 49 |
| 380 ^a | 120 | 66 | 120 | 73 |
| 490 ^a | 90 | 7 | 90 | 37 |
| 576 | 85 | 58 | 90 | 66 |

^aNot included in the Lorentz term for the metallic grains.

$f_c = 0.50$. The plasma frequency required for the fit was close to the reported values determined via reflectance measurements.^{11,13} The value of τ^{-1} was higher than previously reported.^{15,11} However, this increased scattering was attributed to percolation effects.

In writing a dielectric function for the insulating component of the metallic films it was assumed that the response came only from the phonons and that no free carriers were present. Some of the phonon lines in Table I were observed also in the insulating films, and, besides from the c axis, they most likely came from oxygen-deficient grains. The transmittance through a thin film of thickness $d \ll \lambda$ was derived by Tinkham.³⁴

$$\mathcal{T} = \frac{4n}{(n+1+y_1)^2 + y_2^2}, \quad (3)$$

where $y_1 + iy_2 = (4\pi/c)d(\sigma_1 + i\sigma_2)$ is the film admittance. This thin-film approximation is good when the penetration depth $\delta \gg d$, as applies here. Since no substrate was present, the index of refraction n for the surrounding area was set equal to 1. In Fig. 1(a) we show the fit to the normal-state transmittance obtained by first determining the effective conductivity using Eqs. (1) and (2) and then finding \mathcal{T} from Eq. (3).

For the superconducting state we used the same EMA equation, Eq. (1), assuming that the Drude contribution remains due to some fraction of normal carriers. Thus, we have rewritten the dielectric function ϵ_m as

$$\epsilon_m(\omega) = \epsilon_0 + \epsilon_L + (1 - f_{\text{sc}})\epsilon_D + f_{\text{sc}}\epsilon_{\text{sc}}, \quad (4)$$

where the expressions for ϵ_D , ϵ_L , and ϵ_0 are the same as in Eq. (2). The value f_{sc} denotes the fraction of the superconducting carriers within the metallic grains. Also, as in Ref. 7, we obtain

$$\epsilon_{\text{sc}} = -\frac{\omega_p^2}{\omega^2} + i\frac{\pi\omega_p^2}{2\omega}\delta(\omega), \quad (5)$$

where $\delta(\omega)$ is a Dirac δ function. The real part of Eq. (5) was obtained from the normal-state Drude formula under the assumption that for the superconductor $1/\tau \rightarrow 0$. The imaginary part was then calculated from the Kramers-Kronig relation.

We obtained a good fit for the transmission at 10 K by leaving ω_p temperature independent (8000 cm^{-1}), and $\epsilon_0 = 13$. We fixed τ^{-1} for the normal component at 180 cm^{-1} . The parameter allowed to vary was f_{sc} , and we obtained a fit for $f_{\text{sc}} \simeq 0.20$. The position of the absorption lines were unchanged from the normal state. The oscillator parameters are given in Table I. In Fig. 1(b) we show the EMA fit for the transmittance.

The data for the insulating films were modeled assuming that there were no free-carriers present, so that the contribution to the dielectric function came only from phonons; thus only the first two terms of Eq. (2) were considered. The parameters for the phonon lines are given in Table II, and the value of ϵ_0 was 15. In Fig. 2 we show the fit to the transmittance obtained by first determining the effective conductivity and then calculating \mathcal{T}

TABLE II. Fitting parameters for insulating films.

| ω_{0j} (cm ⁻¹) | γ_j (cm ⁻¹) | Ω_j (meV) |
|-----------------------------------|--------------------------------|------------------|
| 156 | 15 | 21 |
| 190 | 35 | 30 |
| 310 | 25 | 12 |
| 340 | 27 | 11 |
| 390 | 75 | 38 |
| 450 | 55 | 26 |
| 495 | 30 | 28 |
| 570 | 40 | 23 |

from Eq. (3).

In the superconducting state the absorption lines up to about 320 cm⁻¹ are more narrow than the corresponding lines in the normal state, with especially noticeable sharpening at 156 cm⁻¹ (Table I). However, at higher frequencies the width of the peaks stays about the same both above and below T_c . In the insulating films the phonon lines become sharper. The lines at 156 and 190 cm⁻¹ do not change from the metallic state. They represent an in-phase motion of O(1)-Cu(1) and an Y vibration parallel to the c direction, respectively. The line corresponding to 450 cm⁻¹ was not observed in the metallic state.

The metallic samples had a metal-volume fraction $f = 0.48$, very close to the percolation threshold concentration for two dimensions. When the value f_c in a metal-insulator composite is reached from below, a closed conducting path is formed and the mixture as a whole becomes conducting at zero frequency. Although for the two-dimensional case the theoretical critical volume fraction is 0.50, the actual composite might show the transition at lower metal concentrations. Also, any variation in the size of the metallic particles could account for the formation of the connected path at f lower than 0.50. A value of f_c lower than the theoretically predicted value was also previously observed in three-dimensional systems.³⁵

An interesting feature of the percolative phenomena is the frequency dependence of the real part of the conductivity, $\sigma_1(\omega)$. According to the Mattis-Bardeen theory²⁸ the conductivity in the superconducting state, $\sigma_{1s}(\omega)$, which also governs the absorption, is zero up to the value of the energy gap, starts to rise at the gap frequencies, and meets the normal-state conductivity at higher frequencies. Measurements⁸ on the oriented samples of YBa₂Cu₃O_x showed that the frequency dependent conductivity was reduced in the superconducting state. However, measurements on conventional granular superconductors,²⁰ with the metallic fraction $f \ll f_c$, suggest that $\sigma_{1s} > \sigma_{1n}$ for frequencies around the energy gap 2Δ . The EMA model for the samples studied here gives the real part of the effective conductivity, Fig. 3, in the normal and superconducting states which increases with the frequency till about 300 cm⁻¹, and then, if the phonon lines are ignored, stays constant up to the highest measured frequencies.

The behavior of the real part of the effective conductivity

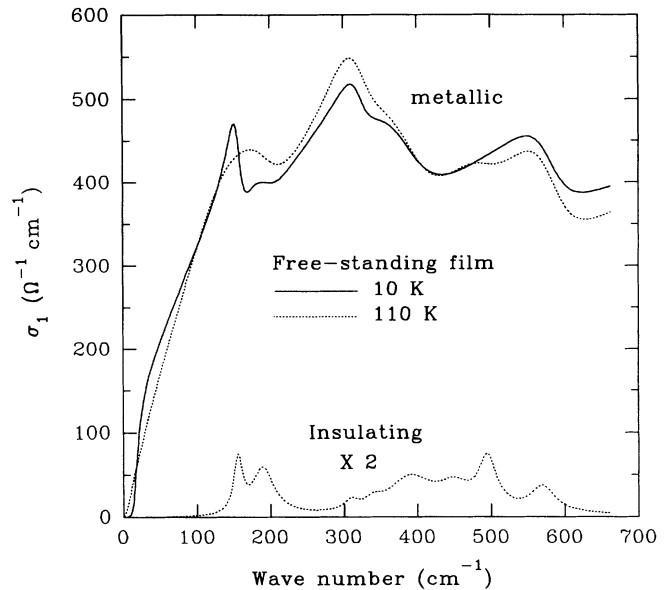


FIG. 3. Real part of the effective conductivity for the metal-insulator composite described in the text, calculated within the effective-medium approximation for the metallic film at 10 and 110 K. The difference in the two curves above 400 cm⁻¹ is the result of the fitting procedure.

ity had been studied previously by several authors.^{22,27} The EMA theory for metal-insulator systems predicts a broad resonant absorption peak centered at frequency, ω_r . The value of ω_r depends on the characteristics of the composite, such as the plasma frequency and the metal concentration. As f increases, the lower-frequency edge of this resonance moves toward lower frequencies, reaching zero at the percolation transition.²⁷ We observed this predicted behavior in our systems near the percolation threshold (Fig. 3).

For metallic films we found that the conductivity stayed almost unchanged with temperature. In the superconducting state the conductivity had a nonzero value down to the lowest measured frequencies (10 cm⁻¹). The reason might be the presence of the normal state component. An insulator is usually best characterized by the real part of the dielectric function, $\epsilon_1(\omega)$. We calculated this quantity from the fit for the insulating films, and found that it behaved as expected for insulators, approaching the value of 19 as $\omega \rightarrow 0$.

We obtained the absorption coefficient $\alpha(\omega)$ from the equation $\alpha(\omega) = 4\pi k\omega$, where k is the extinction coefficient calculated from EMA and ω is in cm⁻¹. The frequency dependence of α is shown in Fig. 4. For both the normal and the superconducting states of the metallic films the absorption increases with frequency and tends toward saturation at higher frequencies. In the previous study by Carr, Garland, and Tanner²⁰ on granular conventional superconductors, it was observed that for diluted systems $\alpha(\omega) \propto \omega^2$. However, at larger metal-volume fractions (~ 0.10 for a three-dimensional system) an $\alpha \propto \omega$ behav-

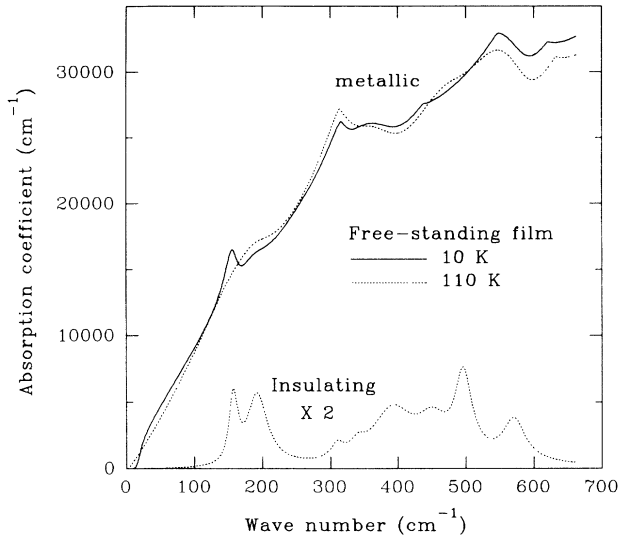


FIG. 4. Absorption coefficient for the metal-insulator composite described in the text, calculated within the effective-medium approximation for the metallic film at 10 and 110 K.

ior was found.²

Frequency-dependent percolation effects were also investigated theoretically by Stroud and Pan,³⁶ who calculated $\alpha(\omega)$ and $\text{Re}\sigma_{\text{eff}}(\omega)$ in granular metals. They found that for $f \simeq f_c$ and low frequencies ($\omega \ll \tau^{-1}$) these quantities depend on frequency as $\omega^{0.75}$ and $\omega^{0.5}$, respectively. Bergman and Imry⁴ suggested that at $f = f_c$ the conductivity of a system was proportional to ω^p , where $p \simeq 0.73$. For our system, we found that the calculated $\alpha(\omega)$ changes as $\omega^{0.67}$, in the measured frequency range, when the phonon contributions were subtracted. For frequencies $\omega \lesssim 300 \text{ cm}^{-1}$ we also obtained $\text{Re}\sigma_{\text{eff}}(\omega) \propto \omega^{0.5}$.

Carr, Garland, and Tanner²⁰ investigated a system of small Sn and Pb particles embedded in KCl host, with the metal concentration $f < 0.05$. In the superconducting state and frequencies much below 2Δ they found that the samples became less absorbing than in the normal state, while for $\omega \geq 2\Delta$ the absorption in the superconducting state was about 50% larger than that in the normal. Tan-

ner, Sievers, and Buhrman³ reported the FIR transmission measurements on granular free-standing smoke with the metal-volume fraction around 0.018. The smoke consisted of small Sn particles, $\simeq 140 \text{ \AA}$ diameter, surrounded by voids. No temperature dependence of the absorption coefficient was found, even for temperatures well below the bulk superconducting transition temperature. Our samples were free-standing films consisting of metallic and insulating particles in contact with each other. We did not observe any significant temperature dependence of the absorption coefficient.

Pham *et al.*¹⁶ measured the absorptivity of $\text{YBa}_2\text{Cu}_3\text{O}_x$ twinned crystals at 2 K in the frequency range $80\text{--}400 \text{ cm}^{-1}$. They found the ω^2 dependence and the values of the absorptivity less than 3%. Taking into account the reflection, and using the absorption coefficient calculated from EMA, we obtained the absorptivity for our system about an order of magnitude larger. Shown in Fig. 4 are the calculated absorption coefficients for the metallic films, in the normal and superconducting states, and the insulating films.

In conclusion, we presented FIR transmission measurements on granular thin free-standing films of $\text{YBa}_2\text{Cu}_3\text{O}_x$ in metallic and insulating states. The measurements were done without the presence of the substrate. This allowed us to study a wider frequency range than previously reported, and the interference from the substrate was not present. The data were described by the two-dimensional EMA. The metallic films appeared to be very close to the percolation threshold and showed behaviors similar to granular systems studied previously. The frequency dependence of the absorption coefficient and the real part of the effective conductivity agrees with some previous theoretical investigations of the percolative phenomena.

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