TEMPERATURE-DEPENDENT FAR-INFRARED ABSORPTANCE OF THIN $YBa_2Cu_3O_{7-\delta}$ FILMS IN THE NORMAL STATE

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This work presents the absorptance of high-temperature superconducting $YBa_2Cu_3O_{7-\delta}$ (YBCO) films, deposited on Si substrates, in the far infrared from 15 to 95 cm^{-1} (wavelength from 667 to 105 μm) at temperatures of 100, 200, and 300 K (i.e., in the normal state). Our experiments show a significant difference in the absorptance for radiation incident on the film side as compared to radiation incident on the substrate side. Interference fringes associated with the Si substrate are observed from the measurement and used to analyze the interaction of radiation with the film–substrate composite at the interface. The film thickness is found to have a strong effect on the absorptance of the film–substrate composite, especially for radiation incident on the substrate side.

High-temperature superconducting (HTSC) thin films have been used to construct highly sensitive infrared radiation detectors [1–5]. These devices operate above liquid-nitrogen temperature using a HTSC film as the sensing element. Knowledge of the amount of radiation absorbed within the film is crucial for the detector design. A larger absorptance yields a higher responsivity and detectivity as well as a smaller noise equivalent power [2, 6]. The absorptance of the HTSC film depends not only on the wavenumber of the incident radiation and temperature of the film–substrate composite but also on the thickness of the film and properties of the substrate. Analyses of radiative properties of YBCO films are presented mostly for thick films [7, 8], which have an absorptance that is small compared to that of thin films. Considering that high absorptance in the YBCO film is a crucial requirement in the construction of radiation detectors, there is a need to determine the absorptance of thin films. Phelan et al. [9] investigated the thickness-

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NOMENCLATURE

c_0	speed of light in vacuum ($= 2.9979 \times 10^8$ m/s)
d	thickness, m
i	$(-1)^{1/2}$
n	real part of the refractive index
N	refractive index
r	reflection coefficient
R	reflectance
t	transmission coefficient
T	transmittance
T_c	critical temperature, K
α	absorptance
γ	scattering rate, cm^{-1}
δ_f	complex phase change in the film
ε	dielectric function
ε_h	high-frequency dielectric constant
κ	imaginary part of the refractive index
ν	frequency, cm^{-1}
ν_p	plasma frequency, cm^{-1}
$\Delta\nu$	free spectral range, cm^{-1}
ϕ_s	phase change in the substrate

Subscripts

f	film, film side
s	substrate, substrate side
0	vacuum

dependent absorptance of YBCO films deposited on MgO substrates. Their measurements are in the wavelength region from 10 to 50 μm (1000 to 200 cm^{-1}) at room temperature.

Higher absorptance of the film–substrate composite occur when radiation is incident on the substrate side (backside illumination) [2]. Backside illumination requires that the substrate be transparent in the measured spectral region. Silicon is an ideal substrate for the fabrication of HTSC bolometers because of its high thermal conductivity and infrared transparency [3, 4]. Kumar et al. [10] presented transmittance and reflectance of YBCO films on Si substrates and obtained complex dielectric functions of the YBCO film at different temperatures.

In the present study, the absorptance of a YBCO film (35 nm thick) deposited on a Si substrate ($\approx$ 200 μm thick) is obtained from the transmittance and reflectance spectra measured at temperatures of 100, 200, and 300 K in the spectral region from 15 to 95 cm^{-1}. Because of the interference effects in the Si substrate, the absorptance depends strongly on the wavenumber. The dielectric function determined by Kumar et al. [10] is used to model the absorptance, which is then compared with the measured results. Furthermore, the effect of film thickness on the absorptance of the film–substrate composite is investigated.

1 EXPERIMENTS

The YBCO films were deposited by pulsed laser ablation on a double-side-polished Si wafer approximately 200 μm thick. A 20-nm-thick yttria-stabilized zirconia (YSZ) film and a 10-nm-thick CeO_2 film were deposited on the substrate prior to the deposition of the YBCO film, in order to grow high-quality superconducting film. The thickness of the YBCO film is 35 nm, determined by previous calibrations of the deposition rate. The YBCO films formed this way are a–b plane oriented with a critical temperature (T_c) of 88 K. The critical temperature dropped

by a few K after a backside cleaning process. Detailed descriptions were given in [4] and [10].

The transmittance spectra of the specimen were measured using a slow-scan Michelson interferometer with a mercury-arc source. A liquid-helium-cooled Si bolometer and lock-in electronics were used to measure the output signal. The spectral resolution was approximately 0.5 cm^{-1}. A liquid-helium-cooled cryostat was used to cool the sample. Polyethylene windows, which are transparent in the far-infrared region, were used to seal the cryostat. Silicon diode sensors were used to measure the temperatures of the cold finger and the specimen. The temperature is displayed and controlled by a temperature controller. The reflectance was measured using a fast-scan Fourier transform spectrometer with a resolution of 1 cm^{-1}. A gold mirror was used as the reference for the reflectance measurements. The reflectivity of this mirror, which is greater than 0.995 in the far-infrared region, was taken to be unity. Calibrations with a bare Si wafer indicated an expanded uncertainty (95% confidence) of 0.1 for both transmittance and reflectance measurements. Hence, the combined expanded uncertainty in the absorptance is 0.14. The unusually large uncertainties are possibly due to interference effects in the sample [10].

Figure 1 shows the measured absorptance at various temperatures for radiation incident on both the film side and the substrate side. The measured absorptance data are much noisier near the cutoff wavenumbers. The interference pattern appears to be independent of the wavenumber except for the data at 300 K, where the absorptance decreases with the wavenumber below 40 cm^{-1}. This could have been caused by the uncertainty in the transmittance measurement. The interference fringes are caused by the Si substrate, because the free spectral range $\Delta\nu$ (i.e., the frequency interval between adjacent interference maxima) of slightly greater than 7 cm^{-1} is consistent with that calculated from $(2n_s d_s)^{-1}$, where n_s is the refractive index and d_s is the thickness of silicon.

For radiation incident on the film side, there is very little change in the absorptance from 300 K to 100 K. The fringe-averaged absorptance, defined as

$$\bar{\alpha}(\nu) = \frac{1}{\Delta\nu}\int_{\nu-\Delta\nu/2}^{\nu+\Delta\nu/2} \alpha(\nu')\, d\nu' \tag{1}$$

is between 0.21 and 0.29, where ν is the wavenumber. The peak locations shift slightly toward higher wavenumbers as the temperature decreases, because of the change in the refractive index of silicon with temperature.

The absorptance is greater for radiation incident on the substrate side than for radiation incident on the film side. For the backside illumination, the absorptance increases as temperature decreases, as shown in Figure 1*b*. The average absorptance is 0.4 at 300 K, 0.48 at 200 K, and 0.55 at 100 K. There is a phase shift of π rad in the absorptance spectra between 300 K and 100 K; in other words, the interference maxima at one temperature correspond to the absorptance minima at the other. Furthermore, the spectrum at 200 K has no distinct interference fringes. The experimental data are compared with theoretical calculations given in the next section.

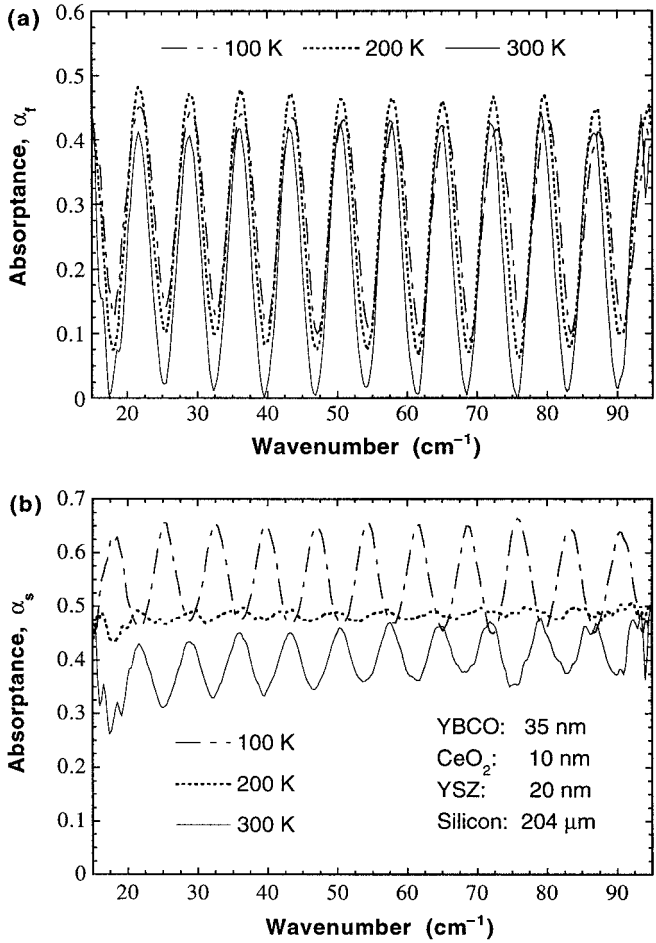

Figure 1. Measured absorptance of YBCO film for radiation incident on: (*a*) the film side; (*b*) the substrate side.

2 ANALYSIS

The dielectric function $\varepsilon(\nu)$ of YBCO can be modeled as the sum of the Drude term, $\varepsilon_{\text{Drude}}$, which accounts for the temperature-dependant free carrier absorption and a real constant ε_h that accounts for the mid-infrared and high-frequency contributions. The Drude term is

$$\varepsilon_{\text{Drude}} = -\frac{\nu_p^2}{\nu^2 + i\nu\gamma} \tag{2}$$

where ν_p is the temperature-independent plasma frequency and γ is the temperature-dependent scattering rate. Considering that phonons and mid-infrared band have little contribution to the far-infrared region below 100 cm^{-1}, the value of ε_h

is set to be 100. Kumar et al. [10] obtained ν_p and γ values by fitting the calculated transmittance to the measured spectra. The fitted value of ν_p is 5700 cm^{-1}, while γ is 600, 400, and 230 cm^{-1} at 300, 200, and 100 K, respectively. The complex refractive index of the YBCO film is related to the dielectric function by $N_f = n_f + i\kappa_f = \sqrt{\varepsilon(\omega)}$, where n_f and κ_f are the real and imaginary parts of the refractive index.

The absorptance within the Si substrate is negligible because it is of high purity (resistivity of 10 Ω m). The Si thickness is determined from the interference patterns in the measured transmittance and reflectance by assuming that $n_s = 3.42$ at wavenumbers from 10 to 100 cm^{-1}, because the thickness of Si can be assumed constant with negligible thermal expansion [11]. The calculated transmittance and reflectance are then compared with the measured spectra to determine the n_s at lower temperatures. The results are $n_s \approx 3.405$ at 200 K and 3.395 at 100 K. The far-infrared dielectric constants of YSZ and CeO_2 are approximately 25 and 17, respectively, with negligible absorption [12]. Initially, we have used the matrix method to compute the radiative properties of the sample film consisting of four layers: a 35-nm YBCO film, a 20-nm YSZ layer, a 10-nm CeO_2 layer, and a 204-μm Si substrate [13, 14]. Because the buffer layers are very thin and dielectric in nature, it was found that they have no effect on the radiative properties of the sample film. Our calculations also show that the angle of incidence of the infrared beam, less than 10°, has negligible effects on the transmittance and reflectance. In order to explain the observed features in the absorption spectra, simple equations for the radiative properties are derived here, considering a thin absorbing film on a thin dielectric substrate.

Let d_f and d_s be the thicknesses of the film and substrate, respectively, and $N_j = n_j + i\kappa_j$ be the complex refractive index of vacuum ($j = 0$), film ($j = f$), and substrate ($j = s$). Note that $N_0 = 1$ and $N_s = n_s$. At normal incidence, the complex Fresnel reflection and transmission coefficients are $r_{jk} = (N_j - N_k)/(N_j + N_k)$ and $t_{jk} = 1 + r_{jk}$, where $j, k = 0, f$, or s. The transmission and reflection coefficients of the film for infrared beam coming from the vacuum (subscript a) and from the substrate (subscript b) are [13]

$$t_a = \left(t_{0f} t_{fs} e^{i\delta_f}\right) / \left(1 - r_{f0} r_{fs} e^{i2\delta_f}\right) \tag{3}$$

$$t_b = \left(t_{sf} t_{f0} e^{i\delta_f}\right) / \left(1 - r_{f0} r_{fs} e^{i2\delta_f}\right) \tag{4}$$

$$r_a = \left(r_{0f} + r_{fs} e^{i2\delta_f}\right) / \left(1 - r_{f0} r_{fs} e^{i2\delta_f}\right) \tag{5}$$

and

$$r_b = \left(r_{sf} + r_{f0} e^{i2\delta_f}\right) / \left(1 - r_{f0} r_{fs} e^{i2\delta_f}\right) \tag{6}$$

where $\delta_f = 2\pi\nu N_f d_f$ is the complex phase change in the film. The transmittance and reflectance are

$$T = \left| \frac{t_a t_{s0}\, e^{i\phi_s}}{1 - r_b r_{s0}\, e^{i2\phi_s}} \right|^2 \tag{7}$$

$$R_f = \left| \frac{r_a + (t_a t_b - r_a r_b) r_{s0}\, e^{i2\phi_s}}{1 - r_b r_{s0}\, e^{i2\phi_s}} \right|^2 \tag{8}$$

$$R_s = \left| \frac{r_{0s} + r_b\, e^{i2\phi_s}}{1 - r_b r_{s0}\, e^{i2\phi_s}} \right|^2 \tag{9}$$

where $\phi_s = 2\pi\nu n_s d_s$ is the phase change in the substrate, which is responsible for the interference fringes. It can be shown that $t_a t_b - r_a r_b = (e^{i2\delta_f} - r_{f0} r_{fs})/(1 - r_{f0} r_{fs} e^{i2\delta_f})$. The absorptance for incidence on the film side is $\alpha_f = 1 - R_f - T$, and the absorptance for incidence on the substrate side is $\alpha_s = 1 - R_s - T$.

In the far-infrared limit, where the wavenumbers are much smaller than the scattering rate, $\varepsilon_{\text{Drude}}$ given in Eq. (2) becomes purely imaginary, with an absolute value much greater than ε_h. Furthermore, in the normal state, d_f is much smaller than the radiation penetration depth in the film. It can be shown that r_b is real and changes from negative, when the sheet resistance (i.e., the dc resistivity divided by the thickness of the film) is greater than $\sim 157\ \Omega$, to positive, when the sheet resistance is smaller than $\sim 157\ \Omega$ [10, 15]. From Eq. (9), if $r_b = 0$, $R_s = r_{0s}^2 \approx 0.3$ is the reflectance at the vacuum–substrate interface and is independent of the wavenumber. This explains why α_s at 200 K has no distinct fringes. The dc resistivity of the YBCO film decreases with temperature. Thus, r_b is positive at 300 K but negative at 100 K. This sign change results in a phase shift of π rad in the absorptance spectra.

The absorptance calculated from the above equations is compared with our experimental results in Figure 2. Because the reflectance for radiation incident on the film side is greater than that for backside illumination and the transmittance is the same in both cases, the absorptance is greater for the backside illumination. The predicted and measured absorptance values match closely, considering the experimental uncertainty, except for the interference pattern in α_s at 200 K. The actual phase at 200 K is very complicated because the electron scattering rate of the YBCO film may be complex and dependent on the wavenumber [16].

3 THE EFFECT OF FILM THICKNESS

The film thickness has a significant effect on the absorptance, especially for the backside illumination [2, 9, 14]. The radiative properties of the film–substrate composite are calculated using the equations given in the previous section by varying d_f. The predicted absorptance at 100 K between 40 and 60 cm^{-1} is shown in Figure 3. For a bare Si substrate, the transmittance maximum ($T_{\max} = 1$) and the reflectance minimum ($R_{\min} = 0$) are located at $\nu = m\,\Delta\nu$, where m is an integer and $\Delta\nu = (2n_s d_s)^{-1} = 7.234$ cm^{-1} with $n_s = 3.395$ and $d_s = 203.6$ μm, whereas $T_{\min}$ and $R_{\max}$ are at $\nu = (m + 1/2)\,\Delta\nu$. The maximum of α_f is located near $m\,\Delta\nu$ (such as 43.4, 50.6, and 57.9 cm^{-1} in Figure 3*a*). The fringe-averaged absorptance and the fringe contrast increase with the film thickness up to $d_f \approx 20$ nm, and then decrease as d_f is further increased. The competing effects of a decreasing transmittance and an increasing reflectance as d_f increases result in an optimized d_f where the absorptance is maximum.

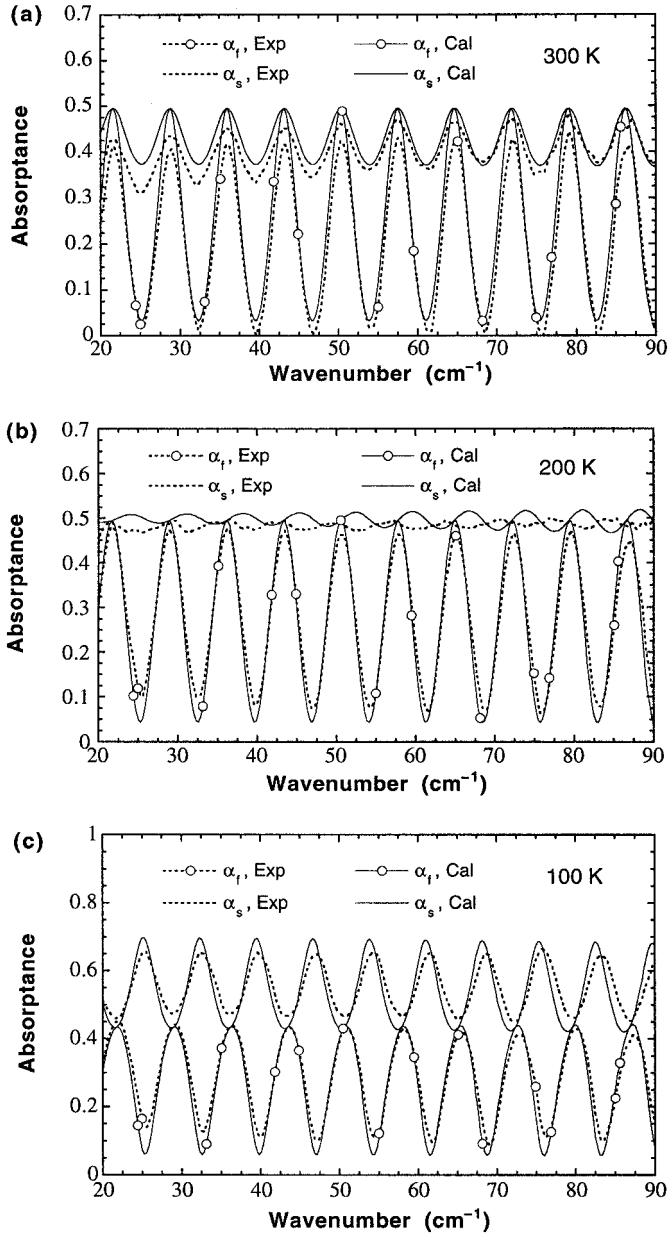

Figure 2. Comparison between the measured and calculated absorptance at: (*a*) 300 K; (*b*) 200 K; (*c*) 100 K.

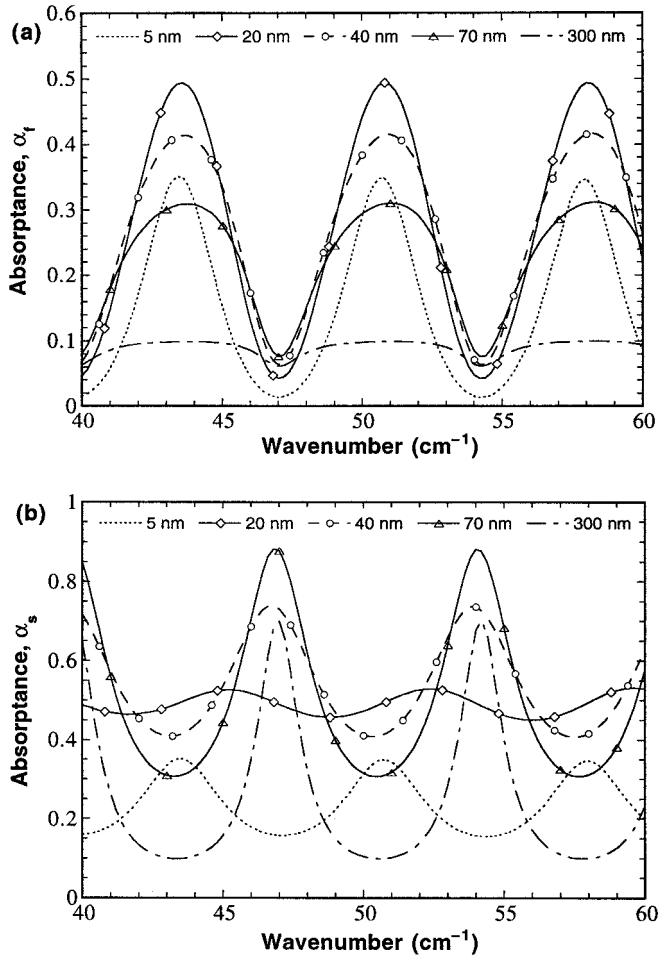

Figure 3. Calculated α_f (*a*) and α_s (*b*) at 100 K for film thickness $d_f = 5$, 20, 40, 70, and 300 nm.

As shown in Figure 3*b*, the value of the optimized d_f is different for the backside illumination as compared to that for incidence on the film side. The maximum of α_s is near $m\,\Delta\nu$ for $d_f <$ 20 nm and switches to near $(m + 1/2)\,\Delta\nu$ for $d_f >$ 20 nm. Note that $r_b \approx 0$ at $d_f =$ 20 nm, where an antireflection effect occurs at the substrate–film interface. The maximum absorptance and the fringe-averaged absorptance continue to increase beyond $d_f =$ 20 nm. The average absorptance reaches a maximum of ~ 0.6 when $d_f =$ 70 nm and the absorptance at $(m + 1/2)\,\Delta\nu$ is ~ 0.91 for 90 nm $< d_f <$ 120 nm.

The absorptance at $6.5\,\Delta\nu = 47.02$ cm^{-1} and $7\,\Delta\nu = 50.64$ cm^{-1} is plotted as functions of d_f in Figure 4. It is surprising to see that $\alpha_f = \alpha_s$ at 50.64 cm^{-1}. After a detailed examination of Eqs. (8) and (9), it can be shown that $R_f = R_s$ if $\phi_s = m\pi$, that is, $\exp(i2\phi_s) = 1$. This is true even for opaque films. The existence

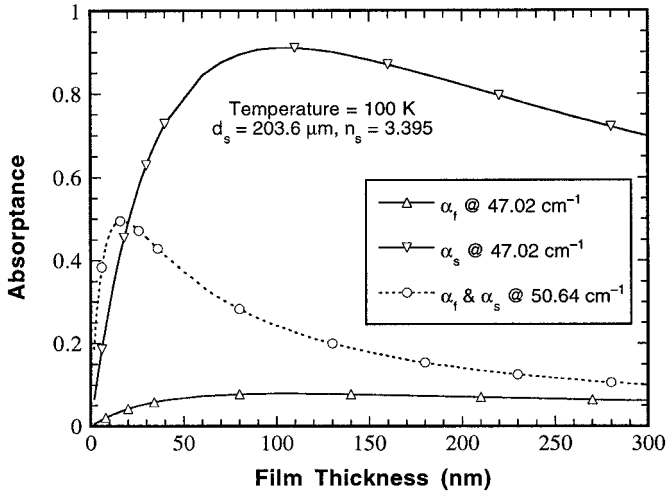

Figure 4. Effect of the film thickness on the absorptance at 47.02 cm^{-1} and 50.64 cm^{-1}.

of wavenumbers where $R_f = R_s$ can be used in practice to check the measurement uncertainty. It can be seen from Figure 2 that $\alpha_f = \alpha_s$ at wavenumbers equal to $m\,\Delta\nu$. The change in absorptance with d_f can be understood better by plotting the transmittance and reflectance at $6.5\,\Delta\nu$ and $7\,\Delta\nu$, as shown in Figure 5. Note that T decreases but R_f increases as d_f increases. When $r_b = 0$, which corresponds to $d_f \approx 20$ nm, R_s is approximately 0.3 and independent of the wavenumber. Because the wave reflected by the backside of the film interferes with the wave reflected by the surface of the substrate, R_s at 47.02 cm^{-1} continues to decrease as d_f is

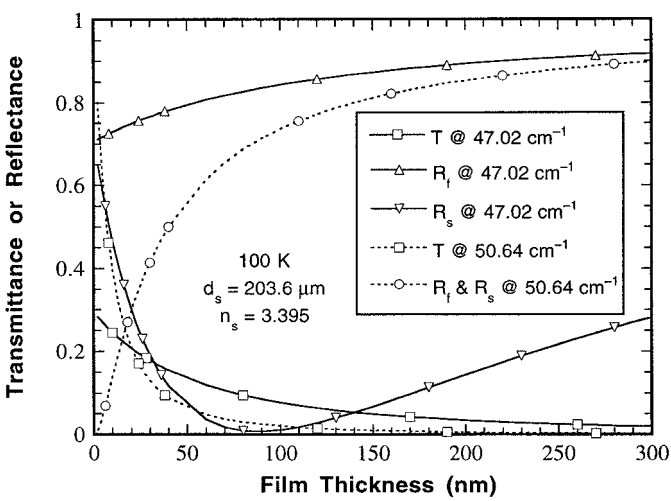

Figure 5. Transmittance and reflectance versus film thickness.

further increased above 20 nm. At wavenumbers equal to $(m + 1/2)\Delta\nu$, there exists a minimum R_s when $d_f \approx 90$ nm.

4 CONCLUSIONS

This work presents the absorptance of thin YBCO films deposited on transparent Si substrates in the far-infrared region at temperatures of 100, 200, and 300 K. The absorptance for backside illumination is found to be greater than that for radiation incident on the film side. Reflection at the interface of the film and substrate can greatly influence the fringe pattern of the observed absorptance spectra. Theoretical calculations using a simple Drude model are in good agreement with the measured absorptance. The effect of the film thickness on the absorptance is studied by calculating the radiative properties of the film–substrate composite as functions of d_f. There exists an optimal thickness for which the absorptance is maximum, although such a thickness for incidence on the film side is different from that for incidence on the substrate side. Furthermore, the optimized film thickness depends on the wavenumber due to interference in the substrate. At certain wavenumbers, the optimized absorptance for backside illumination can be greater than 0.9 at 100 K with a film thickness in the range from 90 to 120 nm. The absorptance data and analyses presented here for transparent substrates such as Si and for the case of backside illumination will facilitate the design of infrared detectors based on YBCO films. Experimental and theoretical investigations of the absorptance of YBCO films in the superconducting state will be presented in the future.

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