

Back-side reflectance of high T_c superconducting thin films in the far infrared

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(Received 6 July 1998; accepted for publication 27 July 1998)

We have measured the far-infrared reflectance of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ high T_c superconducting (HTS) films, for radiation incident on the substrate side (back-side illumination), in the spectral region from 15 to 650 cm^{-1} and at temperatures between 10 and 300 K. The HTS films were deposited on Si substrates (with YSZ/CeO₂ buffer layers) by pulsed laser ablation. The extremely large temperature dependence of the reflectance experimentally demonstrates the feasibility of using HTS films to construct far-infrared intensity modulators. In this letter, we present the measured results, as well as an analysis based on thin-film optics and a simple two-fluid model. © 1998 American Institute of Physics. [S0003-6951(98)04039-X]

Optical modulators have important applications in remote sensing, spectroscopy, and other terahertz technologies.¹ A new device concept was recently proposed that uses high-temperature superconducting (HTS) films as intensity modulators for far-infrared radiation.² The HTS intensity modulator utilizes the abrupt change of the reflectance of HTS thin films as they switch from the superconducting to the normal state. The change between the two states can be made extremely fast by using pulsed current or optical excitations.³ Calculations show that the change in the reflectance of the film/substrate composite is much greater for radiation incident on the substrate side (back side) than for radiation incident on the film side.¹ Although the optical properties of HTS materials have been investigated by many groups, most studies are for crystals and opaque films.⁴⁻⁶ Some researchers have also measured the transmittance and film-side reflectance of HTS films on various substrates.⁷⁻¹³ Genzel *et al.*¹⁴ studied the reflectance of a thick HTS film by placing a Si wafer on the top of the film and observed the optical resonance effect caused by multiple reflections inside the Si wafer. At present, however, there exist no data on the back-side reflectance of thin HTS films.

In the present work, a Fourier transform spectrometer was employed to measure the back-side reflectance of a $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO) film deposited on a Si substrate with two buffer layers: Y₂O₃-stabilized ZrO₂ (YSZ) and CeO₂. The dielectric buffer layers have negligible effect on the optical properties of the film/substrate composite because their thicknesses are much less than the wavelength.¹⁵ The measured spectral range extended from 15 to 650 cm^{-1} with a spectral resolution of 1 cm^{-1} . A 4.2 K liquid-He-cooled bolometer measured the spectrometer output signal. The specimen was mounted in a liquid He cryostat sealed with polyethylene windows. The specimen was maintained at fixed

temperatures by using a heater and a temperature controller. The angle of incidence was fixed at 7.5° , close to normal incidence. The reflectance was measured using a Au mirror, which has a reflectance greater than 0.995 in the far infrared, as the reference. The expanded uncertainty of the reflectance is estimated to be 0.1 (95% confidence), possibly due to the large oscillations in the spectra caused by interference effects.

The YBCO film and buffer layers were deposited by pulsed laser ablation in an optimized temperature and gas environment.¹⁶ The $12\text{ mm} \times 12\text{ mm}$ substrate was cut from a large Si wafer. The thickness of the substrate was approximately $204\text{ }\mu\text{m}$, determined from the refractive index of Si and the interference fringes measured with a spectrometer. The thickness of the film was 35 nm, determined from the deposition time and previously calibrated growth rate. The as-grown film had a critical temperature T_c of 88 K. However, after a back-side cleaning process removed the Ag paste that was used to hold the substrate in the deposition chamber, the T_c of the YBCO film dropped to $\approx 82\text{ K}$. Detailed discussions of the experiments are given by Kumar *et al.*¹⁵

Figure 1 presents the back-side reflectance of the film/substrate composite at 300, 200, 100, 50, and 10 K. The reflectance oscillates with a free spectral range of $\approx 7\text{ cm}^{-1}$, caused by the interference effects in the Si substrate. Here, the free spectral range is equal to $\Delta\nu = 1/(2n_s d_s)$, where $n_s \approx 3.4$ is the refractive index of Si and d_s (cm) is the thickness of the Si substrate. In the normal state, the average reflectance over a free spectral range, is nearly constant for all temperatures. However, the fringe contrast, defined as the difference divided by the sum of the maximum and minimum reflectances, decreases from 300 to 200 K and then increases from 200 to 100 K. Furthermore, the peak locations for the interference maxima and minima interchange between 100 and 300 K. These effects are caused by the

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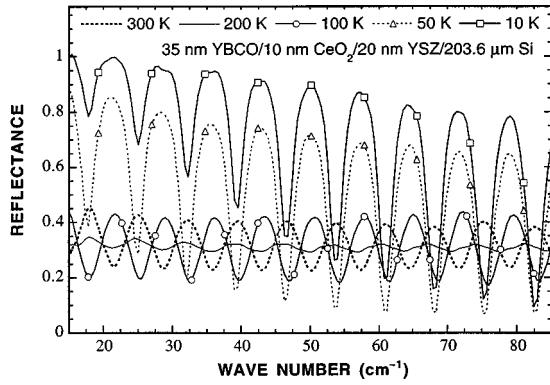


FIG. 1. Back-side reflectance of the film/substrate composite at different temperatures.

temperature-dependent changes of the reflection coefficient of the film.

In the normal state, the dielectric function of the YBCO film can be modeled by a simple Drude model in the far-infrared region. In the far-infrared region where the wave number is much smaller than the electron scattering rate, the real part (n_f) and the imaginary part (κ_f) of the refractive index are approximately the same, that is,

$$n_f \approx \kappa_f \approx \left(\frac{2.998 \times 10^7}{\nu \rho_{dc}} \right)^{1/2}, \quad (1)$$

where ν (cm^{-1}) is the wave number and ρ_{dc} ($\mu\Omega \text{ cm}$) is the dc resistivity.¹⁷ In this case, the reflection coefficient of the film for incidence from the substrate side is purely real: $r_b = (n_s - 1 - y)/(n_s + 1 + y)$, where $y = 37.67 d_f / \rho_{dc}$ [d_f (nm) is the film thickness] is the dimensionless admittance of the film in the Hagen–Rubens limit.^{15,18} For normal incidence, the reflectance of the film/substrate composite for radiation incident on the substrate side can be obtained by tracing the multiply reflected waves in the Si substrate. The expression is simplified when r_b is real, as is the case here. Then,

$$R = 1 - \frac{(1 - r_s^2)(1 - r_b^2)}{1 + r_s^2 r_b^2 + 2 r_s r_b \cos(4 \pi \nu n_s d_s)}, \quad (2)$$

where $r_s = (1 - n_s)/(1 + n_s)$ is the Fresnel reflection coefficient at the vacuum/substrate interface.¹⁹ If $y = n_s - 1$, then $r_b = 0$; the film serves as an antireflection coating that eliminates the interference fringes in the back-side reflectance spectrum.¹⁸ This is the case with the 200 K data, where ρ_{dc} is close to $550 \mu\Omega \text{ cm}$. The reflectance is $R \approx 0.3$, equal to that at the vacuum/substrate interface. Because $r_s < 0$, the interference minima will be located at $\nu = m \Delta \nu$ and maxima at $\nu = (m + \frac{1}{2}) \Delta \nu$ if $r_b < 0$, where m is an integer; this is the case at 300 K. As the temperature is decreased, the electrical resistivity decreases and the admittance increases. Below 200 K, $r_b > 0$, and the interference maxima are at $\nu = m \Delta \nu$ and minima at $\nu = (m + \frac{1}{2}) \Delta \nu$. The thickness of Si is almost independent of temperature, and its refractive index is taken to be 3.42 at 300 K, 3.405 at 200 K, 3.395 at 100 K, and 3.39 at 50 and 10 K.^{15,20} The actual spectra are a little more complicated because of the finite electron scattering rate. The antireflection effect may be further explored to enhance the absorptance of the YBCO film for the design of infrared detectors.^{21,22}

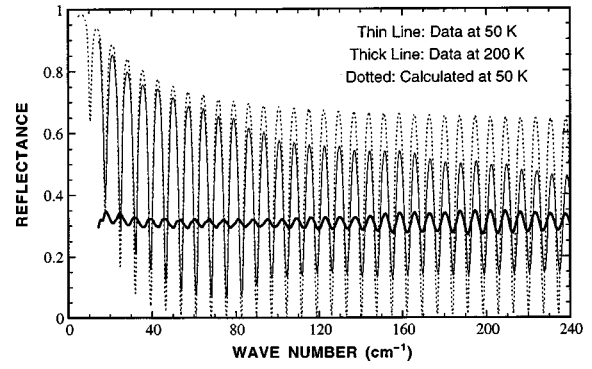


FIG. 2. Reflectance at 200 and 50 K in the region from 15 to 240 cm^{-1} .

The reflectance increases significantly in the superconducting state, as seen from Fig. 1. The fringe contrast is much greater below T_c due to the optical resonance effect.¹⁴ At 10 and 50 K, the average reflectance increases as the wave number decreases. The difference in the back-side reflectance between the normal state and the superconducting state is extremely large at certain wave numbers. For example, the reflectance at $\approx 22 \text{ cm}^{-1}$ increases by a factor of 2 from 100 to 50 K. This experimentally demonstrates the feasibility of using HTS thin films for the construction of far-infrared modulators.

Figure 2 shows the reflectance at 200 and 50 K at wave numbers up to 240 cm^{-1} (the measured spectral region extends to 650 cm^{-1}). The predicted reflectance using a two-fluid model (discussed below) at 50 K is also shown as the dotted line. As the wave number increases, the antireflection effect becomes weaker and the fringe contrast for the 200 K data increases, because the wave number is closer to the electron scattering rate ($\gamma \approx 400 \text{ cm}^{-1}$ at 200 K). The calculated reflectance agrees well with the measured reflectance at 50 K below 80 cm^{-1} , considering the experimental uncertainty. The measured fringe contrast is smaller than predicted, suggesting that the spectral resolution used in the experiments may be insufficient near the sharp interference minima. The calculated interference pattern is unchanged at wave numbers from 100 to 240 cm^{-1} , but the measured fringe contrast decreases as the wave number increases. This decrease can be explained by the partial coherence of the reflected waves inside the Si substrate.²³ The effects of the beam divergence and thickness variation of the substrate become stronger at higher wave numbers.²⁴

The dielectric function $\epsilon(\nu)$ of the YBCO film can be modeled as:^{4,5}

$$\epsilon(\nu) = (n_f + i \kappa_f)^2 = \epsilon_h - f_s \frac{\nu_p^2}{\nu^2} - (1 - f_s) \frac{\nu_p^2}{\nu(\nu + i \gamma)}, \quad (3)$$

where f_s is the fraction of superconducting electrons ($f_s = 0$ in the normal state), ϵ_h (≈ 100) accounts for high-frequency and mid-infrared contributions, ν_p is a temperature-independent plasma frequency, and γ is the scattering rate that depends only on temperature. These parameters were obtained at different temperatures by comparing the calculated transmittance with the measured data.¹⁵ The results are $\nu_p = 6700 \text{ cm}^{-1}$; $f_s = 0.35$ at 10 K and 0.25 at 50 K; $\gamma = 600, 400, 230, 150$, and 190 cm^{-1} at 300, 200, 100, 50, and 10 K, respectively. There exists a large fraction

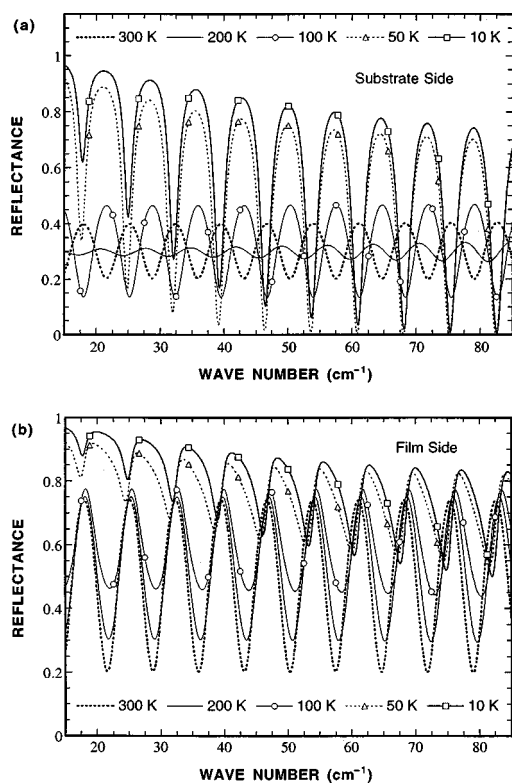


FIG. 3. Calculated reflectance at different temperatures: (a) back side; (b) film side.

of normal electrons at very low temperatures, which may be explained by the existence of thin layers near the substrate or the surface that are not superconducting as well as defects in the film. Similar results have been reported for thin HTS films.^{8–10}

The reflectance for radiation incident on the substrate side and on the film side are computed using a transfer matrix method. The results are shown in Fig. 3. The predicted back-side reflectance is consistent with the measured values at all temperatures. At 10 K, however, the predicted reflectance has much lower minima than the measured, which may be attributed to the insufficient resolution and the partial coherence effect.

For incidence on the film side, the locations of the reflectance maxima and minima depend little on temperature in the normal state, but shift significantly from the normal state to the superconducting state. The fringe contrast decreases as the temperature is reduced. This is explained by the reduction of the radiation penetration depth in the YBCO film with decreasing temperature. Note that the wave that is reflected by the back surface of the substrate must pass twice through the film before it could interfere with the wave reflected directly from the film. At 200 K, the wave reflected by the film and that reflected by the back surface of the Si substrate can still interfere. Measurements of film-side reflectance agree well with the calculated values in the normal state. Additional measurements are being performed for the film-side reflectance in the superconducting state.

For the first time, the reflectance of HTS film has been measured for radiation incident on the substrate side. The large change in the back-side reflectance from the superconducting state to the normal state at certain wave numbers experimentally demonstrates the feasibility of constructing

HTS far-infrared intensity modulators. Notice that the change from the superconducting state to the normal state can also be achieved using a large electric current or optical irradiation. The intensity of the reflected radiation can be rapidly modulated by changing the film material between the superconducting state and the normal state. The reflection coefficient of the film for incidence from the substrate has a strong effect on the fringe pattern and contrast. An antireflection effect has been observed at 200 K for the measured specimen. The simple two-fluid model can be used in the very far infrared (below 100 cm^{-1}). However, at higher wave numbers, the interference fringes in the substrate have been suppressed due to partial coherence between multiply reflected waves. Future research is needed to study the effect of electric current and optical excitations on the back-side reflectance.

This research was partially supported by the University of Florida through an Interdisciplinary Research Initiative award and by the National Science Foundation through Grant Nos. DMR-9705108 and CTS-9812027.

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