THz time-domain spectroscopy of high T_c substrates

D. Grischkowsky and Søren Keiding
IBM Watson Research Center, P.O. Box 218, Yorktown Heights, New York 10598

(Received 7 May 1990; accepted for publication 6 July 1990)

Using the method of time-domain spectroscopy, we have measured the absorption and dispersion from 0.2 to 2 THz of the high T_c substrates, magnesium oxide, yttria-stabilized zirconia (YSZ), and lanthanum aluminate. Our measurements on YSZ and $LaAlO_3$ at both room temperature and 85 K show unacceptably large absorptions for high-speed transmission line applications. At 85 K, MgO is shown to be an excellent material in terms of its low loss at THz frequencies.

One of the most promising applications of the new high T_c superconducting materials is for high bandwidth coplanar transmission lines.[1-9] This application is important because the resistive losses of ordinary metal lines significantly degrade the bandwidth of a transmission line when submicron linewidths are used.[2] Ordinary superconductors have band gaps of 0.7 THz or less, which precludes the transmission of electrical pulses shorter than 1 ps because of the absorption of frequencies above the band gap.[2] However, for the high T_c materials the band gap should be higher than 15 THz. Therefore, subpicosecond pulses could be transmitted down high T_c lines, if the radiation losses were sufficiently reduced,[7,9-11] and if the dielectric substrates did not absorb at these frequencies. At present the required THz absorption data for the high T_c substrates is not available in the literature. However, a new optoelectronic THz beam system[12] has become available for time-domain spectroscopy (TDS) of these substrates in the relevant frequency range from 0.2 to 2.0 THz.[13] This system is based on the optoelectronic generation and reception of a beam of subpicosecond THz pulses. By inserting a sample in the beam and comparing the shape of the original subpicosecond THz pulses with the shapes of pulses that have propagated through the sample, one is able to deduce the frequency-dependent absorption and dispersion.

In this letter we present such TDS measurements of three dielectric substrates used with high T_c coplanar transmission lines, magnesium oxide,[8] yttria-stabilized zirconia (YSZ),[5] and lanthanum aluminate.[6,7] These materials were measured at both low temperature (85 K) and room temperature. From our measurements, YSZ is seen to be unsuitable due to its unusually high absorption at THz frequencies. MgO is dielectrically a good material with a relatively low value of the static dielectric constant and at 85 K sufficiently low loss to allow for the extended propagation of subpicosecond electromagnetic pulses. However, epitaxial films of the quality required for high bandwidth, transmission lines have not yet been grown on MgO. The new substrate lanthanum aluminate[14] allows good film growth for transmission lines, but has a relatively high value of the static dielectric constant, and significant loss at THz frequencies. Consequently, the optimal substrate for high T_c high bandwidth transmission lines remains to be discovered.

The setup used to generate and detect the short pulses of THz radiation is depicted in Fig. 1(a) and has been described earlier.[12,13] The transmitter and receiver each consist of a micron-sized dipole antenna embedded in a coplanar transmission line and optoelectronically driven by 70 fs pulses from a colliding-pulse mode-locked dye laser. The generated pulses of THz radiation are collimated by a silicon lens attached to the transmitting chip and directed onto a paraboloidal mirror that recollimates the radiation into a beam directed towards the receiver, where it is focused onto the receiving antenna. The time-dependent voltage induced across the receiving antenna is determined by measuring the collected charge (current) versus the time delay between the laser excitation pulses and the laser detection (gating) pulses. Such a measured transmitted THz pulse (0.6 ps minimum to maximum) is shown in Fig. 1(b). The amplitude spectrum presented in Fig. 1(c) is obtained from a numerical Fourier transform of the measured pulse of Fig. 1(b) and illustrates the 2 THz bandwidth available for spectroscopy.

For the low-temperature measurements, a dewar with 10-mm-thick, 50-mm-diam high-resistivity (10 kΩ cm) crystalline silicon windows was centered between the transmitter and receiver. In the THz frequency range, high-resistivity silicon is almost completely transparent with negligible dispersion.[13] The dewar was a homemade liquid-nitrogen container with a cylindrical opening, in the middle of which the sample was mounted using a compressed indium ring for thermal contact. The average equilibrium temperature of 85 K for our samples was measured for a typical cooling cycle by a calibrated diode thermometer mounted at the center of the sample.

Another measure of the temperature of the sample was the change upon cooling of the apparent index of refraction n_a. We introduce the term apparent index, because we do not know the length change upon cooling. Therefore as a simple diagnostic approach, we consider the sample length to be constant and mathematically attribute all the change in the relative transit time of the pulse through the sample to the change in n_a. Consequently, at 85 K the measured n_a includes an offset error due to the unknown length changes of the samples, but the frequency dispersion is accurate. Restated more precisely, the actual index of refraction n at

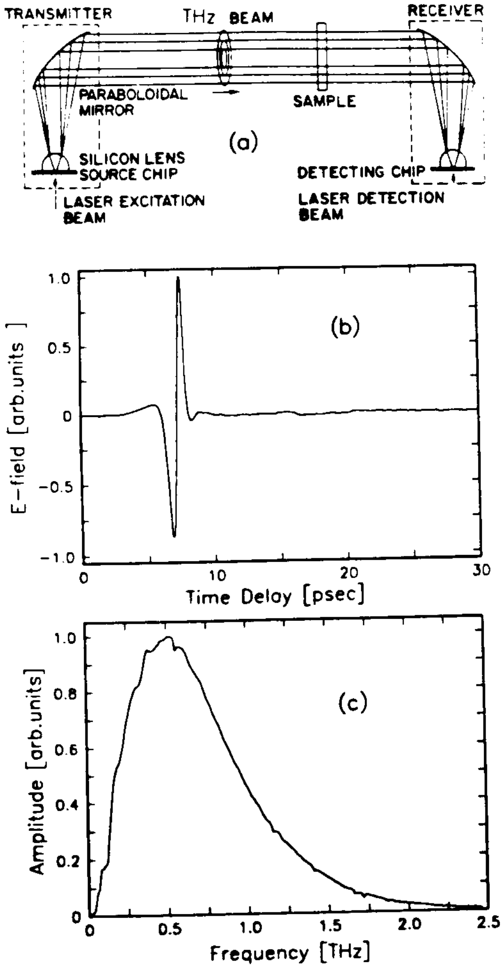

FIG. 1. (a) Schematic of the experiment; (b) measured transmitted THz pulse; (c) amplitude spectrum of (b).

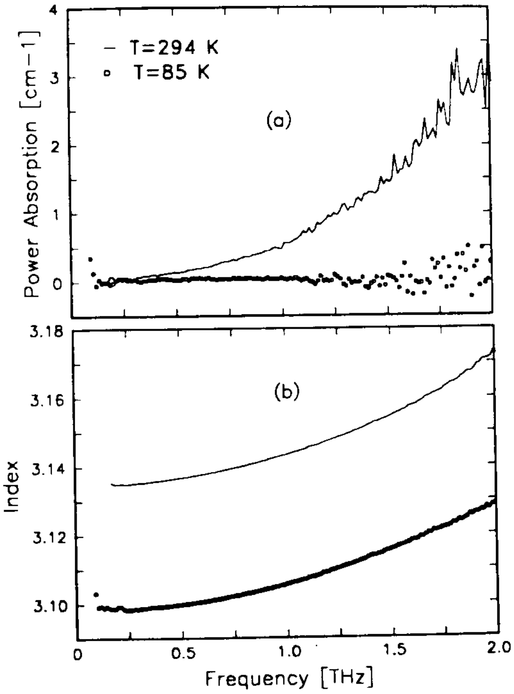

FIG. 2. TDS measurements of MgO. (a) Power absorption coefficient at room temperature(solid line) and at 85 K (circles); (b) Index of refraction at room temperature (solid line) and apparent index of refraction at 85 K (circles).

low temperatures is given by $n = n_a + (n_a - 1)\delta/L$, where δ is the change in the sample length and L is the original length at room temperature. By monitoring the reduction in the transit time through the sample upon cooling, we could determine when the temperature had equilibrated.

In Fig. 2 we present TDS measurements on crystalline magnesium oxide. The sample used was a 38-mm-diam, 4.7-mm-thick polished, single-crystal disk obtained from the Valpey–Fisher Company. The fact that this material is optically isotropic eliminates concern about crystal orientation and polarization of the THz beam. The measured absorption coefficient at room temperature is given by the solid line in Fig. 2(a). The absorption is significantly less than sapphire, but not as low as silicon or quartz.[13,15] From the relative phase of the spectral components, the index of refraction versus frequency is obtained in Fig. 2(b). Here, the index shows a quadratic dependence with frequency and the comparatively large dispersion of MgO is evident.[13,15] Upon cooling the sample to 85 K, the absorption is dramatically reduced as shown by the circles in Fig. 2(a), while the frequency dependence of the index of refraction remains approximately the same as for room temperature. The measured absorption at 85 K is sufficiently low to allow for the extended (many centimeter) propagation of subpicosecond electromagnetic pulses in MgO.

In Fig. 3 we present TDS measurements on crystalline YSZ. The sample used was a 50-mm-diam 1-mm-thick disk of YSZ obtained from Ceres Corporation. This material is optically isotropic. The unusually high absorption of this material at room temperature is shown by the solid line in Fig. 3(a). The index of refraction versus frequency is presented in Fig. 3(b). Upon cooling to 85 K, the absorption is reduced to that shown by the circles in Fig. 3(a). Unfortunately, the low-temperature absorption is still much too high for this material to function as a substrate at THz frequencies. At 85 K the frequency dependence of the index has significantly changed from that at room temperature.

In Fig. 4 we present TDS measurements on crystalline lanthanum aluminate. The sample used was a 50-mm-diam, 1-mm-thick disk obtained from AT&T Nassau Metals. This material is classified as a perovskite-like compound because at room temperature it has a slight rhombohedral distortion. The faces of the disk were the (100) crystallographic faces. Although with this orientation we detected some optical birefringence, we could not detect any birefringence at THz frequencies. At room temperature the measured absorption coefficient is shown as the solid line in Fig. 4(a). The measured index of refraction versus frequency is presented in Fig. 4(b) and is significantly higher than that corresponding to the dielectric constant of 15.3 measured at 1 kHz.[14] Upon cooling to 85 K the absorption is significantly reduced to that shown by the circles in Fig. 4(a), while the frequency dispersion of the index of refraction remains approximately the same as

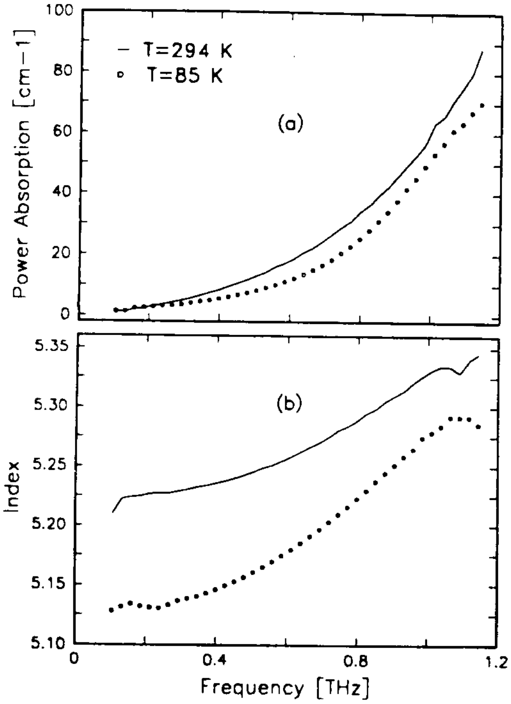

FIG. 3. TDS measurements of yttria-stabilized zirconia. (a) Power absorption coefficient at room temperature (solid line) and at 85 K (circles); (b) Index of refraction at room temperature (solid line) and apparent index of refraction at 85 K (circles).

for room temperature. At 85 K the absorption at 0.5 THz is consistent with the previously estimated[7] upper limit of 0.25 cm^{-1}. However, with increasing frequency the absorption increases to values that would not allow for the extended propagation of subpicosecond electromagnetic pulses in this material.

We have performed similar measurements on lanthanum gallate, recently introduced as a high T_c substrate.[16] Lanthanum gallate has the same crystal structure with somewhat more distortion than LaAlO_3. Consequently, for LaGaO_3 the birefringence at THz frequencies is an observable and complicating factor for clean measurements. In addition, LaGaO_3 is strongly dichroic at THz frequencies. Roughly speaking, the observed features were similar to LaAlO_3 with a comparable index of refraction, dispersion, and absorption. The absorption is definitely too high to allow for applications as a substrate for high bandwidth transmission lines.

We acknowledge some early TDS measurements on the MgO and YSZ samples by Martin van Exter. Hoi Chan was responsible for the excellent masks and wafer fabrication.

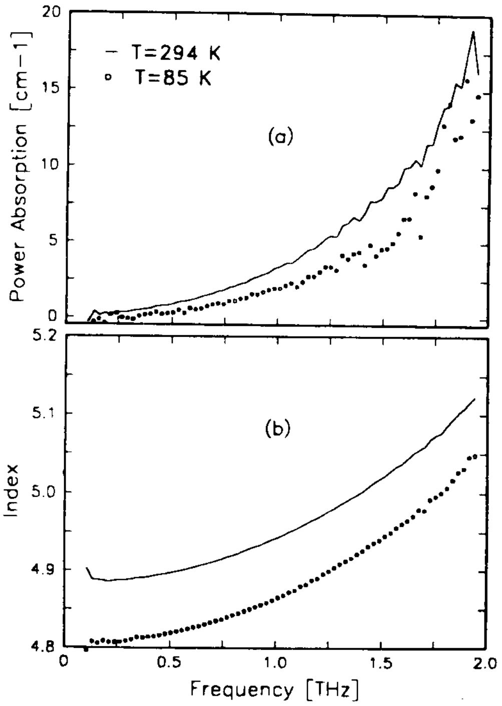

FIG. 4. TDS measurements of lanthanum aluminate. (a) Power absorption coefficient at room temperature (solid line) and at 85 K (circles); (b) Index of refraction at room temperature (solid line) and apparent index of refraction at 85 K (circles).

[1] M. B. Ketchen, D. Grischkowsky, T. C. Chen, C-C. Chi, I. N. Duling, III, N. J. Halas, J-M. Halbout, J. A. Kash, and G. P. Li, Appl. Phys. Lett. **48**, 751 (1986).

[2] W. J. Gallagher, C-C. Chi, I. N. Duling, III, D. Grischkowsky, N. J. Halas, M. B. Ketchen, and A. W. Kleinsasser, Appl. Phys. Lett. **50**, 350 (1987).

[3] D. Grischkowsky, M. B. Ketchen, C-C. Chi, I. N. Duling, III, N. J. Halas, J-M. Halbout, and P. G. May, IEEE J. Quantum Electron. **QE-24**, 221 (1988).

[4] D. Grischkowsky, R. Sprik, M. B. Ketchen, C-C. Chi, I. N. Duling, III, P. Chaudhari, R. H. Koch, and R. B. Laibowitz, J. Opt. Soc. Am. A **4**, (13) p130 (1987); *Conference on Lasers and ElectroOptics Technical Digest Series 1988* (Optical Society of America, Washington, DC, 1988), Vol. 7, pp. 386.

[5] D. R. Dykaar, R. Sobolewski, J. M. Chwalek, J. F. Whitaker, T. Y. Hsiang, G. A. Mourou, D. K. Lathrop, S. E. Russek, and R. A. Buhrman, Appl. Phys. Lett. **52**, 1444 (1988).

[6] M. C. Nuss, P. M. Mankiewich, R. E. Howard, B. L. Straughn, T. E. Harvey, C. D. Brandle, G. W. Berkstresser, K. W. Goossen, and P. R. Smith, Appl. Phys. Lett. **54**, 2265 (1989).

[7] M. C. Nuss and K. W. Goossen, IEEE J. Quantum Electron. **QE-25**, 2596 (1989).

[8] A. A. Valenzuela and P. Russer, Appl. Phys. Lett. **55**, 1029 (1989).

[9] D. S. Phatak and A. P. Defonzo, IEEE Trans. Microwave Theory and Techniques **38**, 654 (1990).

[10] D. B. Rutledge, D. P. Neikirk, and D. P. Kasilingham, in *Infrared and Millimeter Waves*, edited by K. J. Button (Academic, New York, 1983), Vol. 10, Pt. II.

[11] D. Grischkowsky, I. N. Duling, III, J. C. Chen, and C-C. Chi, Phys. Rev. Lett. **59**, 1663 (1987).

[12] Martin van Exter and D. Grischkowsky, IEEE Trans. Microwave Theory and Techniques (to be published).

[13] D. Grischkowsky, S. Keiding, M. van Exter, and Ch. Fattinger, to be published in JOSA-B. An extensive reference list covering other THz sources, techniques, and applications of time-domain spectroscopy is included in this paper.

[14] R. W. Simon, C. E. Platt, A. E. Lee, G. S. Lee, K. P. Daly, M. S. Wire, J. A. Luine, and M. Urbanik, Appl. Phys. Lett. **53**, 2677 (1988).

[15] D. E. Gray, ed, *American Institute of Physics Handbook*, third ed. (McGraw-Hill, New York, 1982).

[16] R. L. Sandstrom, E. A. Giess, W. J. Gallagher, A. Segmuller, E. I. Cooper, M. F. Chisholm, A. Gupta, S. Shinde, and R. B. Laibowitz, Appl. Phys. Lett. **53**, 1874 (1988).