THz spectroscopy and source characterization by optoelectronic interferometry

Stephen E. Ralph and D. Grischkowsky
IBM Watson Research Center, P.O. Box 218, Yorktown Heights, New York 10598

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We demonstrate a new type of THz optoelectronic interferometer, by fully characterizing a recently developed THz source to beyond 6 THz, and by measuring the absorption coefficient of high-resistivity GaAs from 1 to 5 THz. The two source THz interferometer is driven with two 4 mW beams of 60 fs dye-laser pulses and produces interferograms with exceptional signal-to-noise ratios.

Recently, many new optoelectronic sources of freely propagating THz radiation have been demonstrated.[1–14] Common to all these sources has been the problem to properly characterize the emitted THz radiation with receivers of limited bandwidths. The broadest bandwidth receivers have used ion-implanted, silicon-on-sapphire (SOS), photoconductive switches. In general, the limiting response of photoconductive receivers will be determined by the rise-time of the photocurrent itself, if they are not constrained to lower frequencies by their THz optical properties or electrical circuitry. For an ultrafast ion-implanted SOS receiver, this limiting photoconductive rise time has been experimentally determined to be 190 fs,[15] giving an ultimate response time of 150 fs.

An alternative method of source characterization, which bypasses the problems of receiver bandwidth, is based on interferometric techniques using a power detector. This approach was first demonstrated for THz radiation sources by Greene *et al.*[16] They measured autocorrelation signals with a full width at half maximum (FWHM) of 230 fs for the THz radiation pulse from laser-created carriers accelerated by the surface field of a photoconductive semiconductor.[11] In their approach, they used a single-THz radiation source, illuminated by 10 Hz repetition rate, amplified, 100 fs pulses from a colliding-pulse, mode-locked (CPM) dye laser, together with a Martin–Puplett interferometer and a liquid-helium-cooled bolometer.

Here, we report an alternative interferometric approach, which uses unamplified, 100 MHz repetition rate, CPM dye-laser pulse excitation of a two-source interferometer. Via this approach, together with fast, scanning-delay-line averaging, we obtain orders-of-magnitude improvements in the signal-to-noise ratio of the measured interferograms. Consequently, we were able to demonstrate the applicability of the technique to spectroscopy by measuring the absorption of GaAs to 5 THz. We used the newly developed THz radiation source,[12] based on the photogeneration of carriers within trap-enhanced electric fields,[17] to produce radiation to beyond 6 THz. With this THz source, we measured a 230 fs FWHM autocorrelation signal and an average power of 30 nW for 4 mW of laser power. These results extend earlier characterizations of this source by broad-band photoconductive receivers.[12,15]

The optoelectronic THz configuration [Fig. 1(a)] is identical for both sources, and has been described earlier;[12] the coplanar transmission-line structure consists of two 10-μm-wide metal lines separated by 80 μm, fabricated on high-resistivity GaAs. Typically, 100 V are applied across the lines. Illuminating the edge of the positively biased line with focused (5 μm diam) ultrafast laser pulses produces synchronous bursts of THz electromagnetic radiation. A CPM dye laser provides the 623 nm, 60 fs excitation pulses in two beams with average powers of 4 mW at the excitation spots.

The optoelectronic interferometer is illustrated in Fig. 1(b), where the two identical THz radiation sources are depicted. It is important to notice that the interferometer is

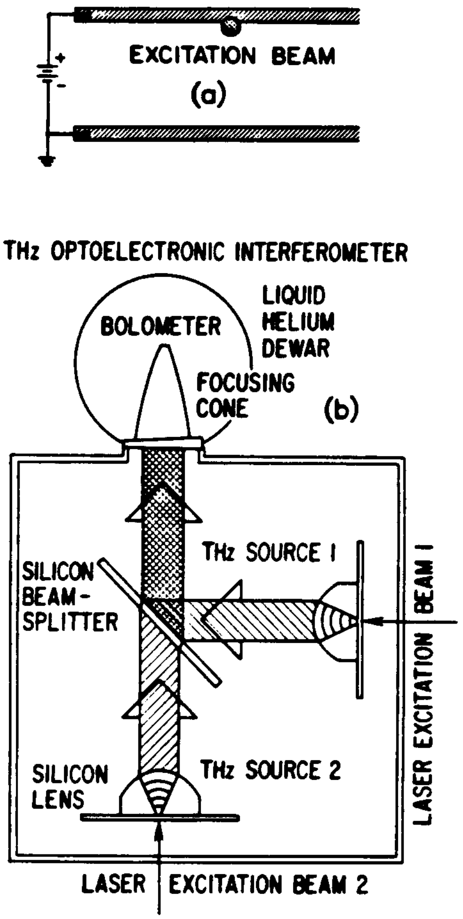

FIG. 1. (a) Configuration used to generate the freely propagating pulses of THz radiation. (b) Two-source THz optoelectronic interferometer shown in the horizontal plane.

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mechanically fixed; the interferogram is obtained by scanning the relative time delay between the two laser-excitation beams. The interferometer is situated in an airtight enclosure filled with dry-nitrogen vapor in order to mitigate the strong absorption due to ambient water vapor.[18] After synchronized laser excitation of each THz source chip, a large fraction of the laser-generated burst of THz radiation is emitted into the GaAs substrate in a cone normal to the interface and is then collected and collimated by a crystalline-silicon lens attached to the back side of the chip.[10,19] The generated THz radiation is vertically polarized. The two emergent collimated THz beams from the silicon lenses propagate approximately 15 cm to a 3-mm-thick, 50-mm-diam, crystalline-silicon disk used as a beam splitter oriented at 45° with respect to both beams. The beams overlap at the beam splitter and from there on travel together 15 cm to an Infrared Laboratories liquid-helium-cooled bolometer.

The remarkable transparency and lack of dispersion of high-resistivity (10 kΩ cm), zone-refined, crystalline silicon is key to the high performance of our interferometer.[19,20] A power absorption coefficient α of less than 0.05 cm^{-1} has been measured up to 2 THz.[19] Recent measurements in our laboratory have placed an upper limit at 6 THz of less than 0.3 cm^{-1} on α. Correspondingly, the index of refraction shows an exceptionally small frequency dependence, changing from 3.418 at 0.25 THz to 3.420 at 6 THz.[19,20] Consequently, the absorption and dispersion in our matched pair of 6.5-mm-thick silicon lenses and 3-mm-thick beam splitter are negligible. Thereby, the Fresnel reflection from the beam splitter is essentially frequency independent.

The THz interferogram was obtained by measuring the amplified-bolometer output with a digital, averaging oscilloscope, while the relative time delay between the two laser-excitation beams was scanned at a 0.5–2 Hz repetition rate using the newly developed Edelstein rapid scanner.[21] Usually, 512 scans were averaged together, although the signal could be easily observed on a single scan. In fact, the signal-to-noise ratio of a single scan was often greater than 20:1, thereby permitting iterative adjustments in real time.

We show a measured interferogram in Fig. 2(a) composed of 8192 individual data points averaged over 1024 scans. The bolometer signal is proportional to the sum of the cw average powers of the two beams plus twice the convolution of the corresponding freely propagating THz pulses.[16] Making use of the calibrated sensitivity of the bolometer, we show the average power incident on the bolometer versus the relative time delay between the two sources. Clearly, a strong interference is observed when the THz pulses overlap in time and space. The negative going interferometric signal shows that the THz pulses have opposite polarity; this is caused by the polarity change of the THz pulse which reflects from the silicon beam splitter. The overall symmetry of the signal indicates a convolution between two similar pulses, while the minor asymmetry is considered to be due to some remaining misalignment of the two interfering beams and perhaps slightly different pulse shapes generated by the two sources. The minimum

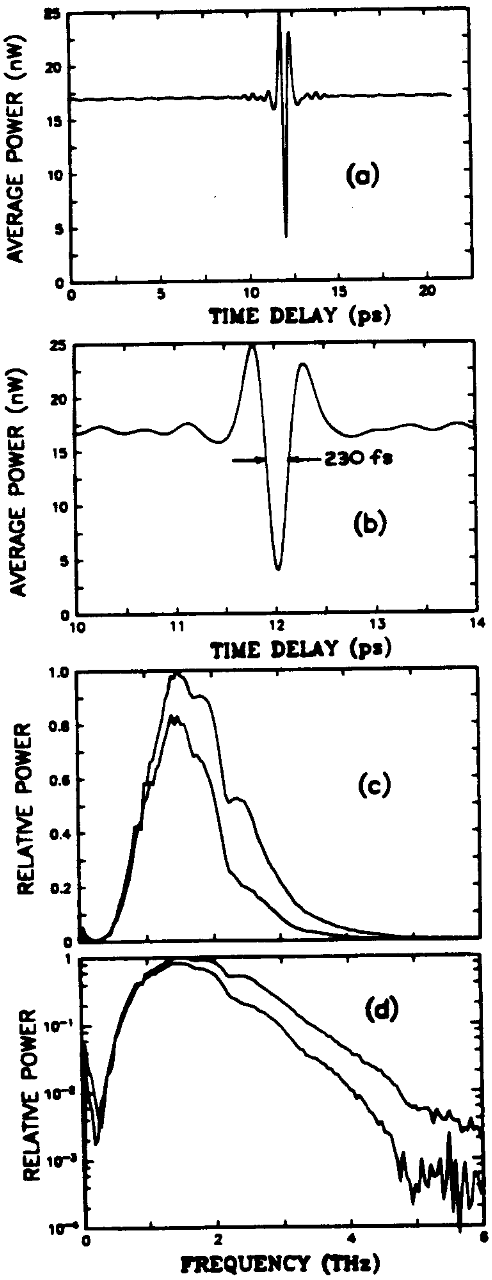

FIG. 2. (a) Measured THz interferogram. (b) Expanded view of the measured THz interferogram. (c) The upper curve is the numerical Fourier transform of (a), which is equal to the relative power spectrum of the freely propagating THz pulses. The lower curve is the same but with a 2-mm-thick, high-resistivity GaAs sample in front of the bolometer. The amplitude of the lower curve has been divided by the Fresnel transmission coefficient 0.46 (assuming a frequency independent index of refraction of 3.60) to compensate for the reflective losses from the sample. (d) Logarithmic presentation of (c).

of the interference signal approaches the theoretical limit of zero power, thereby indicating reasonably good matching of the spatial overlap and curvature of the two beams and parallel polarizations. The best symmetry and the deepest minimum were obtained as follows. Each individual THz source was adjusted for maximum power, and the two sources were aligned to follow a common path to the bolometer. Finally, the interference signal was optimized by slight adjustments of the laser illumination spots and

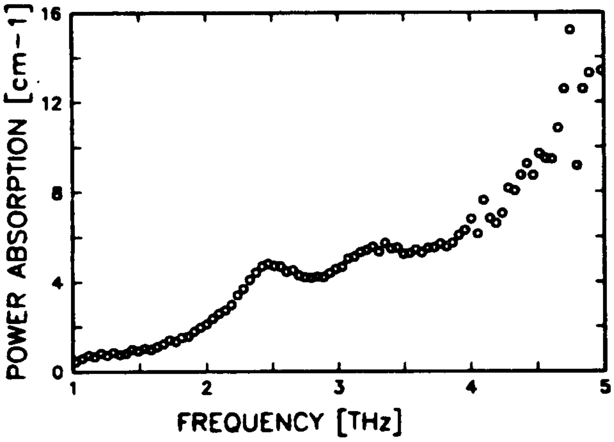

FIG. 3. Measured power absorption coefficient in cm^{-1} for 10 MΩ cm GaAs.

small changes in the relative bias voltages. Even though the interferometer is in a nitrogen-filled enclosure, the symmetric structure about the main interferometric feature is due to coherent radiation from some residual water vapor excited by the passage of the THz pulses.[18] In Fig. 2(b) [an expanded view of Fig. 2(a)], the FWHM of the THz interferogram is seen to be 230 fs, equal to that of the earlier measurement[16] made with a different type THz source.

The upper curve of Fig. 2(c) displays the numerical Fourier transform of Fig. 2(a) with the base line set equal to zero to eliminate the dc component. This transform gives the power spectrum of the freely propagating and interfering THz pulses. However, the spectral feature at 2.25 THz is due to absorption by the entrance window to the bolometer, while the artificial rise at frequencies below 0.3 THz is due to some remaining misalignment of both the interferometer and the scanning-delay line. A logarithmic presentation of this same spectrum is shown as the upper curve of Fig. 2(d), where is is seen that useful power extends to beyond 6 THz = 200 cm^{-1} = 24.8 meV.

An example of time-domain spectroscopy is presented in Fig. 3 for the measured power absorption of high-resistivity GaAs. These higher precision results are compatible with the earlier measurements by Stolen[22] and Perkowitz,[23] and extend the frequency range of the previous time-domain spectroscopy measurements on GaAs.[19] The 50-mm-diameter, 2-mm-thick, LEC grown, 10 MΩ cm GaAs sample was obtained from Sumitomo Electric. For the measurement of Fig. 3, the reference interferogram is that described by Fig. 2. The lower curves in Figs. 2(c) and 2(d) give the corresponding spectrum (adjusted for reflectivity losses) for an interferogram taken with the GaAs sample placed (in both beams) in front of the bolometer. The absorption spectrum of Fig. 3 is obtained from a numerical analysis of these actual spectra. For our semi-insulating sample, two well-defined features at 2.5 and 3.3 THz are observed, compared to the single-absorption peak observed by Stolen[22] for a Cr-doped sample. These two features also appeared in the measurement of Perkowitz,[23] but not in the detail shown here. An additional remark concerns the THz sources, for which some of the generated THz radiation is absorbed upon traversing the 0.46-mm-thick, high-resistivity GaAs chips. As seen from Fig. 3, the absorption by the GaAs chip will reduce the source power at 5 THz by two times (more at higher frequencies), thereby indicating the need for characterizing electronic materials in the THz frequency range.

In conclusion, we have demonstrated a high signal-to-noise optoelectronic THz interferometer system capable of characterizing THz sources to well beyond our attained 6 THz value, and with a demonstrated dynamic range of more than 1000 for the power spectrum. The applicability of this system to THz time-domain spectroscopy was demonstrated by measuring the absorption coefficient of high-resistivity GaAs from 1 to 5 THz.

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[1] G. Mourou, C. V. Stancampiano, A. Antonetti, and A. Orszag, Appl. Phys. Lett. **39**, 295 (1981).
[2] D. H. Auston, K. P. Cheung, and P. R. Smith, Appl. Phys. Lett. **45**, 284 (1984).
[3] A. P. DeFonzo, M. Jarwala, and C. R. Lutz, Appl. Phys. Lett. **50**, 1155 (1987); **51**, 212 (1987).
[4] P. R. Smith, D. H. Auston, and M. C. Nuss, IEEE J. Quantum Electron. **24**, 255 (1988).
[5] Ch. Fattinger and D. Grischkowsky, Appl. Phys. Lett. **53**, 1480 (1988); **54**, 490 (1989).
[6] Y. Pastol, G. Arjavalingam, J-M. Halbout, and G. V. Kopcsay, Electron. Lett. **24**, 1318 (1988).
[7] M. van Exter, Ch. Fattinger, and D. Grischkowsky, Appl. Phys. Lett. **55**, 337 (1989).
[8] Ch. Fattinger and D. Grischkowsky, IEEE J. Quantum Electron. **QE-25**, 2608 (1989).
[9] B. B. Hu, X.-C. Zhang, and D. H. Auston, Appl. Phys. Lett. **56**, 506 (1990).
[10] M. van Exter and D. Grischkowsky, IEEE Trans. Microwave Theory Tech. **38**, 1684 (1990).
[11] X.-C. Zhang, B. B. Hu, J. T. Darrow, and D. H. Auston, Appl. Phys. Lett. **56**, 1011 (1990).
[12] N. Katzenellenbogen and D. Grischkowsky, Appl. Phys. Lett. **58**, 222 (1991).
[13] B. B. Hu, N. Froberg, M. Mack, X.-C. Zhang, and D. H. Auston, Appl. Phys. Lett. **58**, 1369 (1991).
[14] D. R. Dykaar, B. I. Greene, J. F. Federici, A. F. J. Levi, L. N. Pfeiffer, and R. F. Kopf, Appl. Phys. Lett. **59**, 262 (1991).
[15] D. Grischkowsky and N. Katzenellenbogen, *OSA Proceedings on Psec. Elect. and Optoelect.*, edited by T. C. L. Gerhard Sollner and J. Shah (Optical Society of America, Washington, DC, 1991), Vol. 9.
[16] B. I. Greene, J. F. Federici, D. R. Dykaar, R. R. Jones, and P. H. Bucksbaum, Appl. Phys. Lett. **59**, 893 (1991).
[17] S. E. Ralph and D. Grischkowsky, Appl. Phys. Lett. **59**, 1972 (1991).
[18] M. van Exter, Ch. Fattinger, and D. Grischkowsky, Opt. Lett. **14**, 1128 (1989).
[19] D. Grischkowsky, S. Keiding, M. van Exter, and Ch. Fattinger, J. Opt. Soc. Am. B **7**, 2006 (1990).
[20] E. V. Loewenstein, D. R. Smith, and R. L. Morgan, Appl. Opt. **12**, 398 (1973).
[21] D. C. Edelstein, R. B. Romney, and M. Scheuermann, Rev. Sci. Instrum. **62**, 579 (1991).
[22] R. H. Stolen, Appl. Phys. Lett. **15**, 74 (1969).
[23] S. Perkowitz, J. Phys. Chem. Solids **32**, 2267 (1971).