

TIME-RESOLVED TERAHERTZ TRANSMISSION
SPECTROSCOPY OF DIELECTRICS

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Using the method of time-domain terahertz-transmission spectroscopy we measured the far-infrared complex dielectric function at room temperature in the range of 3 to 80 cm^{-1} for a variety of samples: $\text{Li}_2\text{Ge}_7\text{O}_{15}$ (LGO), $(\text{CH}_3\text{NHCH}_2\text{COOH})_3 \cdot \text{CaCl}_2$ (TSCC), high-permittivity microwave ceramics, etc. The precision of the measurements and subsequent evaluation of the dielectric function is discussed.

Keywords Terahertz pulses; far infrared; time-resolved spectroscopy

INTRODUCTION

The time-domain terahertz-transmission spectroscopy (TDTTS) is a method less than a decade old; it could have been developed due to the availability of highly coherent long-wavelength radiation in form of extremely short pulses. Starting from the mid - 80s, a variety of materials or structures was reported to emit sub-picosecond THz pulses (i.e. ultra-broadband electromagnetic transients with a central frequency near 1 THz): crystals with large second order nonlinearity^[1,2], photoconductive switches integrated in dipole antenna structures^[3,4], electric field biased and unbiased semiconductor surfaces in bulk semiconductor wafers^[5,6,7], etc. The detection techniques which are based on either photoconductive^[8,9] or electro-optic^[10,11] time-resolved sampling allow a phase sensitive determination of the electric-field waveform of the THz pulses. Large

recent progress in the development of the THz radiation emitters and detection systems made possible to successfully use the described technique as a time domain spectroscopic method for investigation of polar excitations in millimeter and sub-millimeter spectral range^[12]. This spectral range in fact constitutes a bridge between the classical IR spectroscopy domain and the frequencies accessible by microwave techniques; up to now it was covered practically only by a costly technique in a more restricted spectral range using the backward-wave oscillators (BWO)^[13].

TDTTS experiments has already been applied to different kinds of materials: to study vibration-rotational spectra of gases and liquids^[14], to investigate polar excitations in optical materials like sapphire and quartz^[15], in semiconductors (Ge, GaAs, Si)^[15,16], to study the complex conductivity spectra in superconductors^[17], etc.

In this contribution we describe our experimental setup, we present the room temperature measurements for some dielectric materials, we compare them to the previously published results obtained by other techniques and briefly discuss the observed differences.

EXPERIMENTAL SETUP

Our experimental setup (see Fig. 1) uses a Lexel 480 Ti:sapphire laser oscillator (80 fs, 800 nm) as the light source. Horizontally polarized THz pulses were generated in a biased large-aperture photoconducting antennas made of LT GaAs or semi-insulating GaAs:Cr. The electro-optic sampling technique^[18] with a 1 mm thick $\langle 110 \rangle$ ZnTe sensor crystal was employed for the time-resolved detection of the pulses. The dynamic range of the measurements was better than 10^3 . The spatial transformations of the THz beam were obtained using two aluminum ellipsoidal mirrors with one common focus in the sample position. The spectral resolution of the system depends on the length of the time runs: typically, a resolution of 0.5 cm^{-1} can be achieved.

TIME-RESOLVED TERAHERTZ SPECTROSCOPY

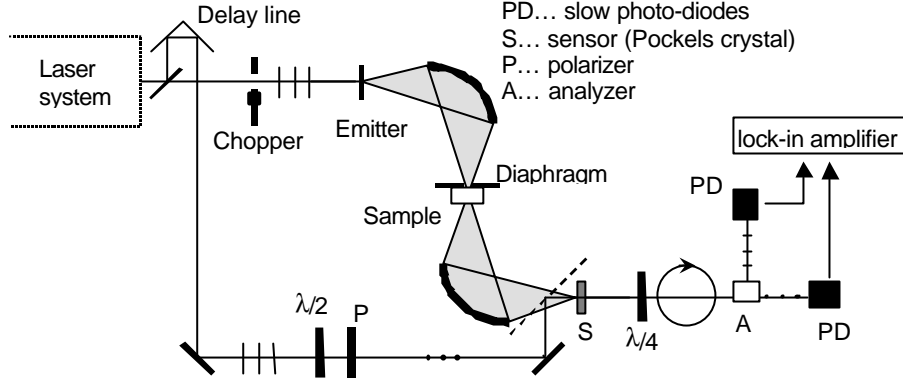


FIGURE 1 Scheme of the TDTTS experiment.

PRINCIPLE OF THE METHOD

The above described detection system allows for measuring of the time profile of the electric field of the THz pulses. The spectroscopic method consists in the measurement of a reference waveform $E_r(t)$ with an empty diaphragm and a signal waveform $E_s(t)$ with the sample attached to the diaphragm and filling the whole aperture. The Fourier components of the two signals are obtained through the fast Fourier transform and they define the complex transmission function of the sample:

$$t(\omega) = E_s(\omega) / E_r(\omega). \quad (1)$$

In the case of a homogeneous sample the complex refractive index $N = n + ik$ is related to the complex transmission through

$$t(\omega) = \frac{4N \exp[i\omega(N-1)d/c]}{(N+1)^2} \sum_{k=0}^m \left[\left(\frac{N-1}{N+1} \right) \exp(i\omega Nd/c) \right]^{2k}, \quad (2)$$

where d is the sample thickness, c is the light velocity and m is the number of reflections in the sample. These reflections are experimentally resolved (at least for thick samples) and form separate

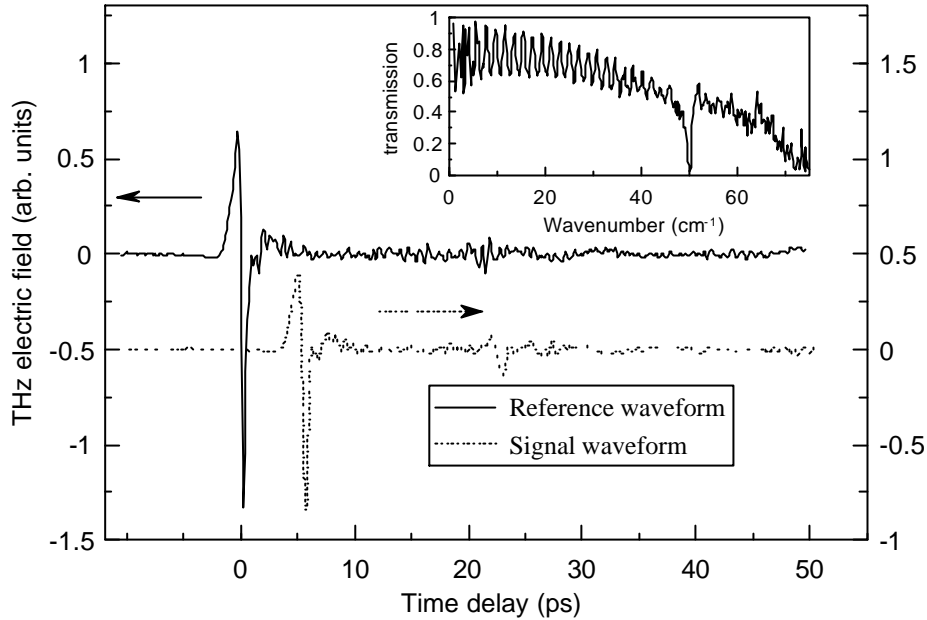


FIGURE 2 Waveforms obtained for the 0.97 mm thick LGO sample.
Inset: amplitude of the calculated transmission function.

pulses in the measured signal so that the value of the coefficient m can be easily determined; for thin samples the geometrical series should be summed up to the infinity.

Eq. (2) is then numerically solved: it constitutes two real equations for two real parameters n and κ . The TDTS usually allows to avoid the problem of the periodicity of the phase: the order of the phase for the spectral components near the central frequency is easily determined from the measured time delay between the signal and the reference pulses. The only problem can arise near a very sharp dielectric resonance: it is not always possible to determine from a single measurement how many orders were swept during the sharp increase or decrease of the dielectric function, i.e. what is the magnitude of the dielectric anomaly. It is then necessary to compare the results for two samples with different thicknesses (this is discussed below in the case of the lithium germanate). As a result one obtains the complex dielectric function of the sample in the whole spectral range transmitted by the THz pulse without any ambiguity.

TIME-RESOLVED TERAHERTZ SPECTROSCOPY

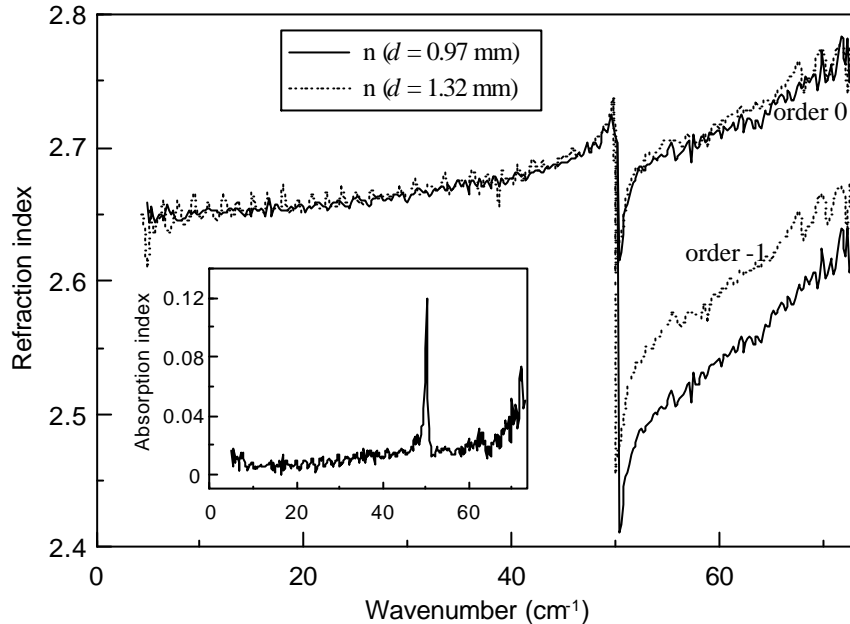


FIGURE 3 Calculated refractive index of LGO. Two solutions corresponding to different orders in the phase determination are shown. The zero order is the correct one. Inset: calculated absorption index.

The refractive index n is mainly dependent on the phase of the transmission function [cf. eq. (2)]. As the phase itself is measured with a high accuracy, the absolute value of n can be very precisely determined: the limiting factors are the plane-parallelism of the sample and the precision of its thickness determination.

EXPERIMENTAL RESULTS

Lithium germanate (LGO)

We studied the time-resolved transmission of this material along its polar c axis. The ferroelectric soft mode is detected only above the room temperature^[19], however the material presents another sharp low-frequency phonon (at about 50 cm^{-1}) which was the object of our study. An example of the obtained temporal waveform and the complex transmission function are shown in Fig. 2. The "noisy" signal

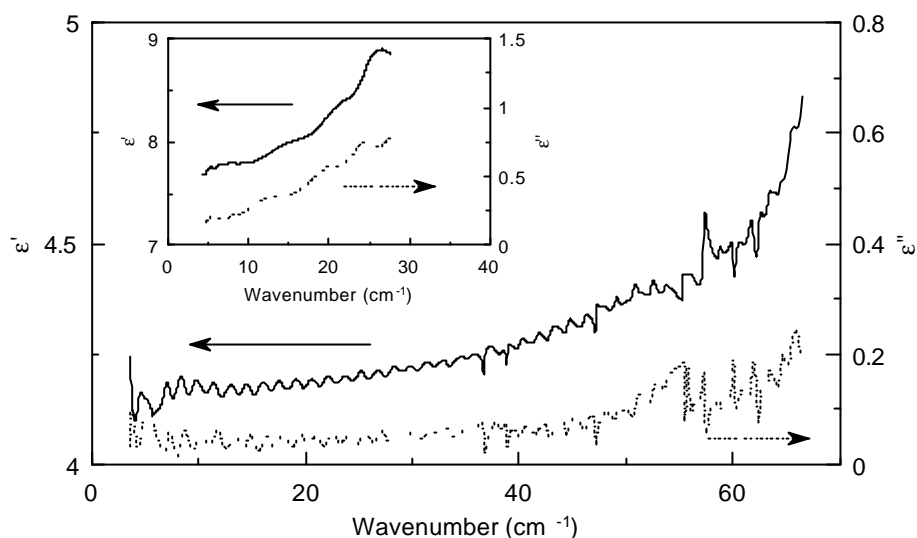


FIGURE 4 Dielectric function of TSCC versus wavenumber. Polarization parallel to the ferroelectric axis. Inset: polarization perpendicular to the ferroelectric axis.

after the main pulses corresponds mainly to the beats of the rotational absorption lines of the water vapor^[14]. The fringes in the transmission function are due to the internal reflections in the sample (echoes at 23 and 41 ps) and are exactly described by eq. (2) with $m = 2$. Fig. 3 plots the calculated refraction and absorption index. The values for the frequencies below the phonon resonance at 50 cm^{-1} are determined unambiguously from the temporal distance between the reference and signal pulses (Fig. 2). The correct value of the refraction index above 50 cm^{-1} can be determined from two independent measurements as shown in Fig. 3.

TSCC

TSCC is another weak ferroelectric where the soft mode was detected by BWO spectroscopy^[20,21]. In Fig. 4 we show our dielectric spectra parallel and normal to the ferroelectric axis. The measurements are in a good agreement with our recent Fourier transform IR measurements^[22] even if the overdamped soft mode at room temperature was not detected.

TIME-RESOLVED TERAHERTZ SPECTROSCOPY

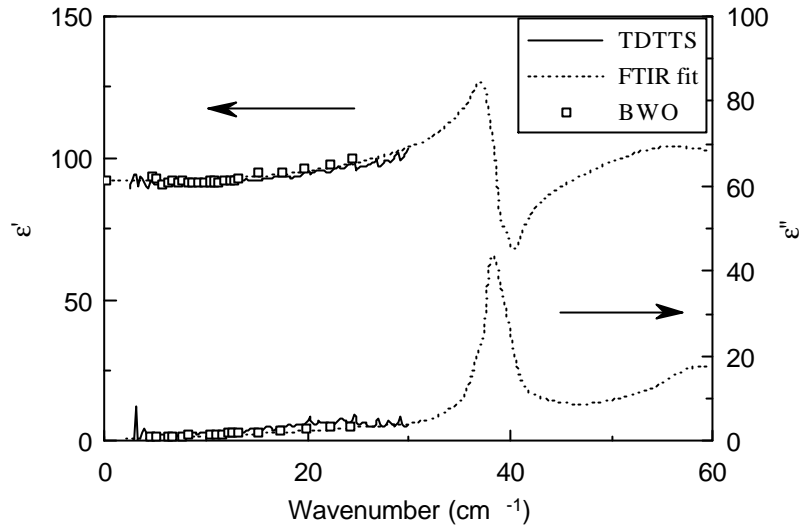


FIGURE 5 Dielectric function of $\text{Ba}_2\text{Nd}_5\text{Ti}_9\text{O}_{27}$ ceramics. TDTTS: our data; FTIR and BWO data: taken from Ref. 23.

Other materials

We studied a number of other dielectric and semiconductor materials at room temperature: high-permittivity ceramics, GaAs:Cr, Si, ZnTe (with different free carrier concentrations) etc. and we verified that the results obtained by our experimental setup are highly reproducible and correspond well to the previously published or expected results. We present as an example the data obtained for the ceramics $\text{Ba}_2\text{Nd}_5\text{Ti}_9\text{O}_{27}$ (Siemens)^[23] which was studied owing to its potential applications as a microwave high-quality factor ceramics (Fig.5)

CONCLUSION

We described the technique of the time-domain terahertz-transmission spectroscopy and our experimental setup. We discussed its possible applications on the study of the dielectric crystals in a number of examples.

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