Determining thickness independently from optical constants by use of ultrafast light

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We show that the application of ultrafast techniques, especially terahertz time-domain spectroscopy, allows simultaneous measurements of material thickness and optical constants from transmission measurements, by analyzing not only the phase difference between the main terahertz pulse through the medium but also the subsequent multireflection pulse (an echo) from the medium. Such a method provides a fast and precise characterization of the optical properties and can extract thickness information and hence other optical constants in a broad bandwidth. It may have applications in science and engineering such as *in situ* film thickness and quality monitoring, optical constants measurement, medical imaging, noninvasive detection, and remote sensing. © 2004 Optical Society of America

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Ultrafast laser techniques have opened a tremendous research opportunity in the study of the interaction of short pulses of light with matter. With the discovery of picosecond photoconducting Hertzian dipoles[1] and high-brightness terahertz (THz) beams characterized with an ultrafast detector,[2] we have seen more and more applications of ultrafast light in the study of material optical properties[3] in the THz range. The index of refraction of materials in the THz range has been investigated by Fourier transform infrared (FTIR) spectroscopy,[4] asymmetric FTIR,[5] and THz time-domain spectroscopy[6–8] (THz-TDS). Standard methods are used to measure the material's thickness and index of refraction[9] separately. The proposed method differs from other imaging techniques, such as contrast difference in optical coherent tomography[10–12] or the peak-to-peak intensity ratio in THz imaging, in determining and estimating the index of refraction.[13] We show that the application of ultrafast techniques allows simultaneous measurements of material thickness and optical constants from transmission measurements.

We first use THz-TDS to demonstrate the method. Although the THz-TDS as a special ultrafast technique provides information on both the amplitude and the phase of the electromagnetic radiation directly, the amplitude and phase are traditionally used to determine the real and imaginary index of refraction with the material thickness determined by other methods.[14,15]

Recent developments in THz time-domain analysis have demonstrated that it is possible to extract material thickness L independent of complex index of refraction.[16,17] However, existing methods were limited by iterative algorithms. In this Letter a method is described that permits the separation of the medium thickness L from the complex index of refraction. We illustrate the technique with the following derivation.

Consider the experimental geometry of Fig. 1. The reference signal is

$$E_{\mathrm{ref}}(\omega) = E_{\mathrm{in}}(\omega)\exp(-j\,\vec{n}_{\mathrm{air}}\omega L/c)\,, \tag{1}$$

where $\vec{n}_{\mathrm{air}}(w)$ (arrow implies that the value is a complex constant), the complex index of refraction of air, is nearly a constant number,[17] except for the sharp features from water absorption.[6] The primary THz pulse through the sample can be expressed as

$$E_{\mathrm{primary}}(\omega) = \vec{t}_{12}\vec{t}_{23}E_{\mathrm{in}}(\omega)\exp(-j\,\vec{n}_{\mathrm{sample}}\omega L/c)\,, \tag{2}$$

and the echo (first multiple reflections) through the sample is

$$E_{\mathrm{echo1}}(\omega) = \vec{r}_{12}\vec{r}_{23}E_{\mathrm{primary}}(\omega)\exp[-j\,\vec{n}_{\mathrm{sample}}\omega(2L)/c]\,, \tag{3}$$

where $\vec{r}$ and $\vec{t}$ are the reflection and transmission coefficients on the interface, determined by the Fresnel equations (normal incidence).

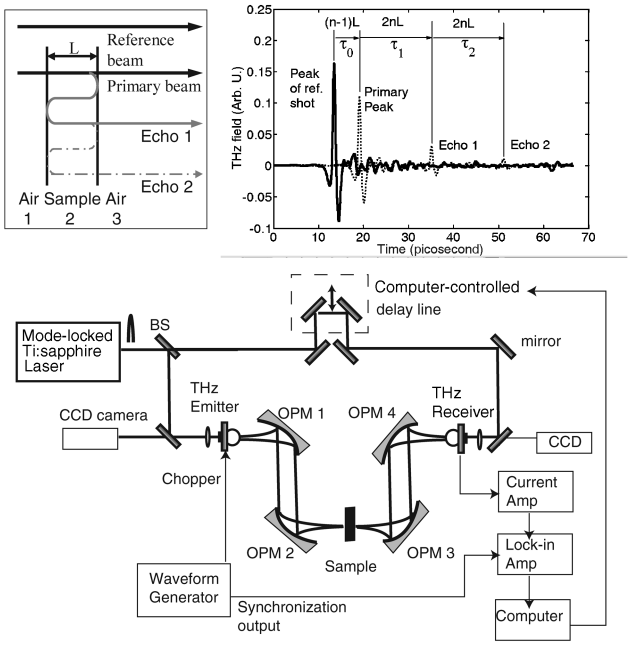

Fig. 1. THz-TDS system and echo analysis method. The echo analysis is shown in the upper left with the THz-TDS signal shown on the right. The lower figure shows the THz-TDS system: OPM1–OPM4, optical parabolic mirrors; BS, beam splitter.

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Dividing Eq. (2) by Eq. (1) and Eq. (3) by Eq. (2), one obtains

$$T_1 = \frac{E_{\text{primary}}}{E_{\text{ref}}} = \vec{t}_{12}\,\vec{t}_{23}\exp[-j(\vec{n}_{\text{sample}} - \vec{n}_{\text{air}})\omega L/c], \tag{4}$$

$$T_2 = \frac{E_{\text{echo}}}{E_{\text{primary}}} = \vec{r}_{12}\,\vec{r}_{23}\exp[-j(\vec{n}_{\text{sample}}\omega(2L)/c]. \tag{5}$$

Phase shift from sample interfaces can be accounted for as an almost constant value compared with the contributions from the thickness-dependent term in T_1 and T_2.

One can summarize from the above equations that the phase of T_1 ($\angle T_1$) is determined by optical path-length difference $(n_{\text{sample}} - n_{\text{air}})L$, and $\angle T_2$, contrary to one's intuition, actually has different information as $2n_{\text{sample}}L$. When phase information from the primary pulse and echo are combined, one extracts the material thickness directly. The sample thickness is just

$$L = [(\angle T_2 - 2\angle T_1)/2n_{\text{air}}](c/\omega). \tag{6}$$

After the material thickness is determined, the material's index of refraction can be deducted from the same phase measurement as in Eqs. (4) and (5).

To illustrate the method, we analyze the transmission through highly resistive silicon, a typical nonpolar material. The time-domain data are shown in Fig. 1. The resulting optical parameters are shown in Fig. 2. Notice that the thickness can be extracted from one frequency (such as at 0.5 THz), instead of being frequency dependent. Unlike the low-loss, low-dispersion silicon, many of the interesting materials (doped semiconductors, electro-optic materials, etc.) are dispersive and lossy in the THz range.[9] The proper separation of the primary pulse and the echoes may require a thin sample as in Fig. 3.

With broader-bandwidth THz pulses (shorter-pulse lasers and broadband generation and detection techniques) the extent of the amplitude fluctuations related to E_{primary} can extend well beyond the beginning of echo. If this happens, one can use the interferometer method of Johnson *et al.*,[13] which utilizes a THz pulse version of optical coherence tomography; with zero-delay destructive interference one can distinguish two or more pulses that overlap in time. This technique improves the sensitivity and ability to distinguish contributions from thin layers by interferometrically removing the overlapping pulses.

The proposed metrology can be useful for thickness measurements of materials in a spectral range that are not strongly absorbing and dispersive.[9] It is therefore useful to assess the applicability of such a technique in other shorter-wavelength ranges. Compared with other laser-based ultrafast techniques,[18] THz-TDS is unique since it provides both amplitude and phase information. However, although the method described here for THz-TDS analysis relies only on the phase information directly, it is equally applicable to other ultrafast laser systems with the advancement of laser waveform diagnostic techniques.[19]

Using the interferometry method, we examine the transmission of near-infrared laser pulses through a piece of glass as in Fig. 4. The interference fringe in the spectral domain (time domain in the THz case) is due to the interference of three delayed light pulses: the reference pulse (air transmission), the primary transmission pulse, and the primary pulse with the echoes.

The ultrafast echo analysis provides both the index of refraction and the sample thickness that one cannot achieve from a single measurement. Although the method is derived from THz-TDS, it can apply to other pulse-based spectroscopy, such as ultrafast laser spectroscopy and asymmetry FTIR.[20,21] The ability to

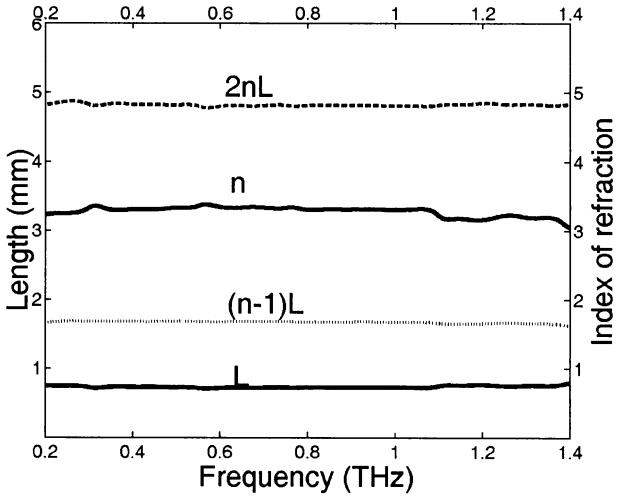

Fig. 2. Sample thickness can be deducted through a difference of two phase measurements. The fluctuation of the thickness around 1.2 THz was a result of a poor signal-to-noise ratio in this frequency range. One can also estimate n and L of the silicon wafer from the peak-to-peak positions of the primary and echo pulses. The measured n and L are 3.4 and 0.71 mm, respectively, which are in agreement with the values determined in Ref. 17.

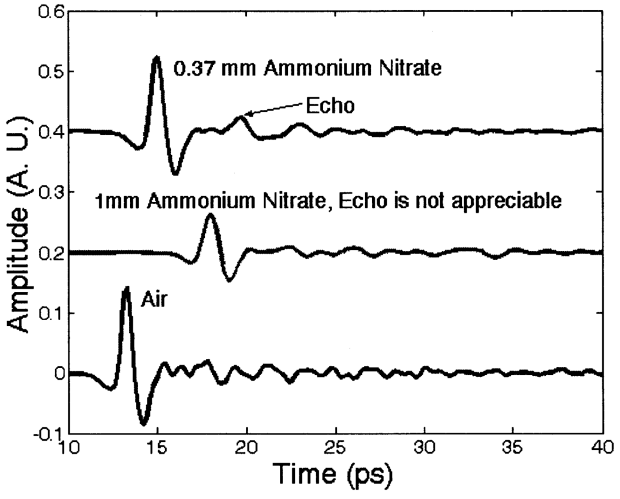

Fig. 3. Echo analysis of ammonium nitrate. The echo does not appear in the 1-mm sample, but it appears in a 0.37-mm sample, a one-third reduction in thickness.

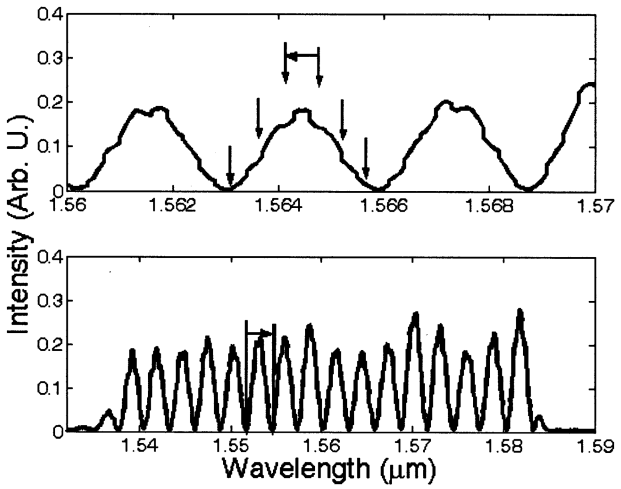

Fig. 4. Larger oscillation corresponds to a delay of 2.9 ± 0.2 ps, the delay between the air peak and the glass peak. The small oscillation in the top figure is estimated to be 15.5 ± 0.2 ps, corresponding to a delay between the air peak and the echo. The resulting n is ~1.82 with a thickness of ~1.04 ± 0.06 mm, and it agrees well with the measured n and L values of the flint glass. The precision in this determination is limited by the simple algorithm used.

extract the sample thickness through the transmission spectra suggests that the method may be applied to noninvasive detection and remote sensing. Another potentially promising application of the described method might be in the biomedical imaging of human tissue (such as skin, drier tissues such as bones or teeth, or the *in vivo* image of the human eye). Using an ultrafast light source, one can characterize a material's optical properties and physical thickness to a very high dynamic range and precision, e.g., from 3 mm to 25 nm for a 150-fs laser in the near infrared.[18]

In summary, we have described a new method that uses short pulses of light in a transmission mode to independently characterize the optical properties of a material and the medium's physical thickness. Such a method provides a fast and precise characterization of the optical properties and can extract thickness information and optical constants in a broad bandwidth. It may have applications in science and engineering, such as *in situ* film thickness and quality monitoring, medical imaging, noninvasive detection, and remote sensing. Increased use of ultrafast light in metrology and spectroscopy applications is expected with the advancement of ultrafast techniques with shorter pulses at higher frequencies[22,23] and frequency precision.[24]

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