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Pulsed THz Imaging for Thickness

Characterization of Plastic Sheets

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Highlight:

- Terahertz reflection and transmission has been applied to measure the thickness of polycarbonate and poly-methyl methacrylate sheets.
- The negative dispersion is found for both PC and PMMA, which broadens the width of THz reflected signal from the back interface of sample, degrading the depth resolution.
- Cross-correlation accounting for dispersion is an effective approach of extrapolating the structure of thicker samples whose second echo is hidden in noise floor, compared with frequency-wavelet domain deconvolution.
- A factor-of-two improvement after accounting for dispersion, enables commercial pulsed THz systems to measure thicknesses of plastics in the few -*cm* range.

Abstract

The thickness of 2, 4, 6, 8, 10, and 12 mm thick polycarbonate (PC) and poly-methyl methacrylate (PMMA) sheets were characterized nondestructively by terahertz time-of-flight tomography in reflection. Due to terahertz attenuation and dispersion in the material, first characterized in transmission, features in the reflected signal associated with structure deep in the sample (i.e., the back interface) may be difficult to identify, and therefore signal processing is employed, namely frequency-wavelet domain deconvolution and a cross-correlation approach in which we account for dispersion. The refractive indices are found to be $n(\nu) = 1.67 - 0.0096\nu$ for PC and $n(\nu) = 1.61 - 0.0085\nu$ for PMMA, while the absorption coefficients, which follow the form of the universal dielectric response, are $\alpha(\nu) = 0.628 + 4.64 \nu^{1.675}$ for PC and $\alpha(\nu) = 0.388 + 13.15 \nu^{1.777}$ for PMMA, where ν is measured in THz and the absorption coefficient is in units of cm^{-1} . We show that straightforward analysis techniques that neglect dispersion are limited to measuring layer thicknesses in these materials of less than ~ 17 mm, while accounting for dispersion results in a factor-of-two improvement, allowing us to measure samples as thick as ~ 36 mm.

Index Terms— Nondestructive testing, terahertz imaging, optical constants, polycarbonate, PMMA

1. INTRODUCTION

Plastic materials are so widely used that any attempt to list their applications would be fruitless. Nonetheless, for a range of problems in manufacturing and inspection, as well as for electromagnetic applications, specifically from tens of GHz to a few THz, characterization of the optical constants in the terahertz (THz) range of these materials is essential. The aim of this study is to present a methodology whereby relatively thick plastic sheeting can be characterized insofar as its thickness d in a nondestructive and noncontact fashion utilizing pulsed THz tomography. This methodology requires, as an intermediate step, the characterization of the frequency-dependent refractive index $n(\nu)$ and absorption constant $\alpha(\nu)$ in this frequency range.

THz time-of-flight (TOF) tomography is an attractive nondestructive approach for thickness characterization of electrically nonconducting objects [1] due to several advantages over other techniques, such as cross-sectional imaging (destructive), x-ray imaging (ionizing radiation, low contrast for some materials, wavelength much shorter than the relevant length scales) [2], [3], transmission electron microscopy (problematic in electrically insulating samples, limited to thin samples, and may require vacuum) [4], and ultrasonic imaging (may require a coupling fluid) [5]. THz electromagnetic waves penetrate many electrically nonconductive materials, such as textiles [6], paper [7], polymers [8], leather [9], wood [10], and ceramics [11], canvas paintings [12], and frescos [13]. In some cases, THz radiation can penetrate deeper than mid-, near-infrared, visible, or ultraviolet light. In THz TOF tomography one subjects a sample to short-duration THz pulses and measures the time-dependent reflected signals, or *echoes*, due to reflections off spatial discontinuities in the dielectric function.

In the simplest case when attenuation and dispersion can be neglected, depth information in samples that are not too thin is obtained from the optical delay between the echoes coupled

with a knowledge of the refractive indices of the constituents. Considerable effort has been expended on characterizing optical thin samples—in practice in the THz regime $\ll 100 \mu\text{m}$ [14], [15]. Here, the main issue is that successive echoes of the incident THz pulse from adjacent material interfaces overlap in time, and thus necessitate advance signal-processing techniques to reconstruct the impulse-response function $h(t)$ of the sample in reflection, in some cases below the axial resolution limit based on the experimental THz bandwidth.

For thick samples the problems are quite different. In such cases, attenuation and dispersion may make identification of echoes arising from interfaces distant from the front surface weak and distorted and thus difficult to identify. Attenuation causes echoes associated with weak features to be lost in noise particularly in thicker samples through which the THz pulses must propagate over relatively long distances; dispersion results in time-delay dependence of the echo's temporal profile, in terms of amplitude reduction and spreading of the echo.

In this work, we start by analyzing the transmitted THz signal through polycarbonate (PC) and poly-methyl methacrylate (PMMA) sheets of thicknesses 2, 4, 6, 8, 10, and 12 mm to characterize the frequency ν dependent attenuation constant $\alpha(\nu)$ and refractive index $n(\nu)$. While these plastics find applications in a broad range of fields, their optical constants in the THz regime are of interest in strategic technologically focused applications. PC comprises a class of thermoplastics, one application of which is to serve as an insulator in electronics, including in capacitors. PMMA, for example, serves as a resist in microelectronic processing. The values found are $n(\nu) = 1.67 - 0.0096\nu$ for PC and $n(\nu) = 1.61 - 0.0085\nu$ for PMMA and $\alpha(\nu) = 0.628 + 4.64\nu^{1.675}$ for PC and $\alpha(\nu) = 0.388 + 13.15\nu^{1.777}$ with ν measured in THz. Once the optical constants are characterized, we use this information to carry out THz TOF tomography in reflection to extract the thickness d of the samples. The analysis to obtain d is carried out using two different techniques: frequency-wavelet domain

deconvolution (FWDD) [16], [17] and cross correlation (CC) between the reflected and incident THz signals [18], [19]. Our CC approach is motivated by the matched-filter technique widely employed in radar. We show that naïve analysis techniques that neglect the effects of dispersion are limited to measuring layer thicknesses in these materials of less than ~17 mm, while accounting for dispersion results in a factor-of-two improvement, allowing us to measure samples as thick as ~36 mm. Indeed, accounting for dispersion is conceptually similar to filter-bank matched-filter approaches developed for detection of Doppler shifted radar signals [20]. Note that some of these results were reported in summary form in Ref. [21].

2. SAMPLES

Six solid PC and PMMA plastic sheets, nominally of thicknesses $d = 2, 4, 6, 8, 10$, and 12 mm for each plastic, were studied. The relevant physical, optical and mechanical properties for measured PC and PMMA plastic sheets are shown in Table. 1. The top and bottom surfaces are diamond polished and shaped so as to provide visually smooth surfaces. THz scattering at the surfaces is expected to drop significantly after polishing.

THz reflection and transmission spectra were measured with a Teraview TPS Spectra 3000, a broadband THz time-domain system, shown schematically in Fig. 1, which generates roughly single-cycle THz pulse with bandwidth extending from 60 GHz to 3 THz (2 cm^{-1} to 100 cm^{-1}). The transverse spot size at the focus of the THz beam is frequency-dependent, and is about 300 μm at 1 THz. The ultrafast laser used is an Er-doped fiber laser that emits 780-nm pulses of sub-100 fs duration at a repetition rate of ~100 MHz with an average output THz power > 65 mW. On the detection side, by varying the time delay of the variable delay line, a time-delayed optical pulse from the fiber laser temporally overlaps with the THz pulse to be detected which coincide on another photoconductor; the time-dependent THz field thus modifies the optical properties of the delayed ultrafast optical pulse. By these means, both the

time-dependent amplitude and phase of the THz pulse are recorded. The numerical aperture of the THz optics is 1/8, giving a depth of focus in these materials of ~ 10 mm (though depending on frequency). We conclude that there might be some complications arising in our thickest samples; however, the trends discussed below are consistent with well-grounded models and the analysis is self-consistent. We therefore neglect any possible effects due to the THz beam shape.

3. EXPERIMENTAL APPROACH

Our aim is to measure the thickness d of relatively thick samples using THz TOF tomography. To do so, we implement a two-step procedure. In the first step, we characterize the optical constants $\alpha(\nu)$ and $n(\nu)$. This is conveniently studied based on the THz propagation in a transmission geometry. Once $\alpha(\nu)$ and $n(\nu)$ are known, we carry out THz TOF tomography in reflection to measure sample thicknesses d .

3.1 Transmission and Optical Constants

The dielectric function of a material is determined in transmission by measuring a reference pulse $E_r(t)$ propagating through an empty spectrometer and a sample pulse $E_s(t)$ propagating through the spectrometer with the sample thickness of thickness d placed in the beam path at normal incidence [22]. We are most interested in the THz pulse propagation within the plastic sheet itself; however, we also need to describe what happens when the propagating pulse encounters either the top air/plastic or the bottom plastic/air interface. The relative amplitude of the signal reflected or transmitted at an interface compared with the amplitude of the reference signal is determined by the reflection and transmission coefficients $r(\nu)$ and $t(\nu)$,

$$r_{inc/trans}(\nu) = \frac{1 - n_{rel}(\nu)}{1 + n_{rel}(\nu)}$$

$$t_{inc/trans}(\nu) = \frac{2}{1 + n_{rel}(\nu)}$$

where $n_{rel}(\nu) = n_{inc}(\nu)/n_{trans}(\nu)$ is the frequency-dependent ratio of the refractive index on the side of incidence to that on the side of transmission.

We define $A_r(\nu)$ and $A_s(\nu)$ as the amplitude of Fourier transforms of $E_r(t)$ and $E_s(t)$, and $\phi_r(\nu)$ and $\phi_s(\nu)$ as the phase of Fourier transforms of $E_r(t)$ and $E_s(t)$, respectively. The relative phase change $\phi_r(\nu) - \phi_s(\nu)$ of the ν frequency component of the signal transmitted through the sample, is used to calculate the refractive index $n(\nu)$ of a plastic of thickness d ,

$$n(\nu) = 1 + \frac{c}{2\pi\nu d} [\phi_r(\nu) - \phi_s(\nu)] \quad [1]$$

(We neglect any dispersion in the Fresnel coefficients.) The ratio $A_s(\nu)/A_r(\nu)$ of the amplitudes can be used to calculate the absorption coefficient through the Beer-Lambert law

$$A_s(\nu)^2 = A_r(\nu)^2 \exp [-\alpha(\nu) d] \quad [2]$$

giving

$$\alpha(\nu) = -\frac{2}{d} \ln \left\{ \frac{A(\nu)}{A_0(\nu)} \frac{[n(\nu) + 1]^2}{4n(\nu)} \right\} \quad [3]$$

where the factor

$$t_{air/plastic}(\nu)t_{plastic/air}(\nu) = \frac{[n(\nu) + 1]^2}{4n(\nu)}$$

accounts for transmission coefficients t at the front air/plastic and back plastic/air interfaces ($n_{air} = 1$) [23]. While there may also be a scattering contribution to the apparent reflection and transmission coefficients at the interfaces, we have verified that any such effects are sample (and position) independent. From the point of view of the analysis, that is all that is important, since the analysis ultimately eliminates the effects of surface reflections/scattering.

Due to the roughly single-cycle nature of the THz pulse, the pulse bandwidth extends from ~60 GHz to 3 THz. In the high-frequency range, the spectrum is characterized by a gradual-roll-off, until the signal level approaches the noise floor of the THz system. The noise floor is

relatively frequency-independent and corresponds to the spectrum recorded by blocking the THz beam path completely. The origin of the noise is predominantly electronic.

Apodization functions, which smoothly bring a sampled signal down to zero at the edges of the sampled region, are employed to suppress leakage sidelobes produced as an artefact of the discrete Fourier transform. However, this suppression is at the expense of broadening the waveform, resulting in decreased spectral resolution of THz system [24]. Many apodization functions have been reported and discussed in the literature [25],[26],[27],[28]. Of those explored, boxcar apodization functions appear to be the best to resolve peaks, but produce more ripple artifacts in the spectrum; Happ-Genzel and triangle functions perform poorly with regard to resolving peaks and peak height, although ripples are significantly reduced. Balancing the noise level against spectral resolution, we choose apodization functions of the Blackman-Harris 3-term form, a generalization of the Hamming family; more details can be found in Ref. [29].

3.2 THz Time-of-Flight Tomography

After carrying out transmission studies to determine $\alpha(\nu)$ and $n(\nu)$, we conducted THz TOF tomography in reflection at almost normal incidence. The aim is to identify the echo originating in the reflection from the back plastic/air interface. The time delay between the echoes from the top air/plastic interface and from the bottom plastic/air interface together with a knowledge of $n(\nu)$ of the plastic provides a measure of the sample thickness d . In reflection, the time-dependent electric field of the THz reference signal $E_r(t)$ produced by the apparatus was recorded by reflecting the incident signal from a steel plate (excellent THz reflector), as shown in Fig. 2. Note that the sampling period in the measurement is set to $T_s = 0.0116$ ps, and each recorded temporal THz signal $E_s(t)$ reflected from the sample contains 4096 sampled data points, and these signals are averaged over 10 shots per pixel at a given

time delay. Each plastic sample was raster-scanned by a set of motorized stages moving in the x and y directions in 0.2 mm steps over 10 mm x 10 mm regions, which corresponds to 50×50 pixels to verify the uniformity of our measurements and analysis. The effect of THz absorption due to atmospheric water vapor was suppressed by carrying out the experiments in dry N_2 . More detailed descriptions can be found in Ref. [30]. Of course, this is complicated by attenuation—the thicker the sample, the weaker the back reflection—and by dispersion, which causes the back reflection both to spread and to reduce in amplitude. In order to identify the second echo, we apply two techniques: frequency-wavelet domain deconvolution (FWDD) and cross correlation (CC) between the reference pulse and the reflected signal modified to account for dispersion.

4. RESULTS AND DISCUSSION

4.1 Optical constants

First, the optical constants of the plastic samples are determined in transmission as described above. The absorption spectra for PC and PMMA of various d are shown in Fig. 3. The absorption mechanism is related to relaxation and the amorphous state of the plastics and is manifested within the spectral range lower than 3 THz [31]. Note that the curves for different d agree at lower frequencies (for $\nu < 1.2$ THz), and the absorption coefficients at the frequency of 1 THz of PC and PMMA is 9.2 cm^{-1} and 13.02 cm^{-1} , in excellent agreement with published values 11 cm^{-1} for PC and 12.5 cm^{-1} for PMMA [31], [32]. As d increases, the strength of the transmitted signal decreases due to the attenuation and thus falls below the noise floor at a frequency that decreases with increasing d . The fact that $\alpha(\nu)$ agrees for various d in each plastic where the signal-to-noise ratio exceeds unity confirms the consistency of the approach.

The absorption coefficient and conductivity in broad range of materials show a power-law

dependence on ν as described by the universal dielectric response [33], [34], [35]. We find that the frequency dependence of $\alpha(\nu)$ can be well fit by $\alpha(\nu) = \alpha_0 + \beta \nu^m$ where $\alpha_0 = 0.5642$, $b = 9.075$, and $m = 1.669$ for PC and $\alpha_0 = 0.04057$, $b = 13.66$, and $m = 1.776$ for PMMA, respectively, where ν is measured in THz and α is in units of cm^{-1} .

Frequency-dependent absorption coefficients in plastics showing behavior reminiscent of universal dielectric response have been widely observed [8]. In particular, results in Ref. [8] for PC are quantitatively quite close to what we observe. Some, but not all, plastics studied in [36] were observed to exhibit similar behavior. Again, this tendency is confirmed by the results of Refs. [37] and (including PMMA) [38].

The refractive index of PMMA and PC sheets of various thicknesses are shown in Fig. 4. Comparing results for sheets of different thickness d (but the same material), we find (as it must be) that $n(\nu)$ is independent of sample thicknesses, as presented in Fig. 4(a) and (c). Figure 4(c) shows some apparent nonlinear behavior with respect to ν of $n(\nu)$, mainly at low and high frequency, likely where spectral density is low. Moreover, $n(\nu)$ for PC in the frequency range $80 \text{ GHz} < \nu < 3 \text{ THz}$ exhibits approximate linear behavior with a somewhat more restricted band for PMMA. Fitting the experimental results for d , the thickness for which signal to noise is highest, gives $n(\nu) = -0.0096 \nu + 1.673$ for PC and $n(\nu) = -0.0085 \nu + 1.613$, with ν measured in THz, as shown in inset figure in Fig. 4(b) and (d), the noticeable negative dispersion in low frequency are also confirmed by published data from FTIR measurement [31],[32]. The values 1.66 and 1.61 are the $\nu = 1 \text{ THz}$ refractive indices for PC and PMMA, which are in good agreement with values of refractive index 1.651 for PC and 1.596 PMMA reported in the literature [36],[39]. First, we comment on the overall value of the refractive index, not accounting for dispersion. Ref. [8] finds n in the relevant frequency range to be 1.65 for PC in good agreement with our results. Ref. [36] finds $n \sim 1.6$ for PMMA and ~ 1.63 for PC. Ref. [38] finds for PMMA, $n \sim 1.6$.

We now turn our attention to THz TOF tomography to measure the thickness d of the PMMA and PC sheets. Typical reflected THz signals for PC and PMMA sheets with various d are shown in Figs. 5 and 6. We observe that the reflected signal contains two echoes, the first from the top air/plastic interface, the second from the bottom plastic/air interface. The second echo, compared with the first, has opposite sign due to the reflection from a high- to a low-index medium. (Multiple reflections are very weak and occur at larger time delays.) Apart from any scattering due to interface roughness, the amplitude of the first echo with respect to $E_r(t)$ is determined by the relevant Fresnel coefficient, whereas the amplitude of second peak is additionally affected by dispersion as well as the attenuation coefficient and the Fresnel coefficient of the plastic/air interface. The plastic-sheet thickness d is calculated as the product of the refractive index and the difference of optical delay between two echoes. Of course, dispersion complicates the precise determination of the time delay for the second echo. We now discuss the mechanism of dispersion during the propagation.

Though dispersion in the frequency range of interest has been found to be quite weak by a number of investigators, the influence of dispersion on THz propagation becomes important when the samples are sufficiently thick as in our study. The tendency for the dispersive term to be overall negative for these plastics is not universally observed. Ref. [8] appears to show weak positive dispersion for PC and a range of other plastics. Negative dispersion at somewhat higher frequencies is seen in Ref. [38] for PMMA and PC. A range of plastics (not including PMMA or PC) show weak negative dispersion in Ref. [40] though Ref. [41] finds positive and negative dispersions for various common plastics (again not including PMMA or PC). PMMA and various other plastics are found to have negative dispersion in Ref. [36]. Ref. [42] shows considerable scatter in their frequency-dependent refractive index data and we therefore deem it not reliable to ascertain the sign of the dispersion. The lack of detailed agreement between the results of various authors may also have to do with the rather generic

nature of the terms PC and PMMA. The materials investigated in the studies may have been fabricated by different methods, with different chemical composition, and with different surface treatments, any or all of which may lead to different optical constants.

In light of the various results reported in the literature, we comment on the approach above and then present a test of the results we have obtained. The frequency-dependent refractive index $n(\nu)$ was found above directly from the transfer function. The approach is both conceptually and experimentally straightforward, and involves relatively little in the line of sophisticated interpretation. Still, we confirm the dispersive effects we found in the frequency domain by analyzing the temporal pulse shape upon propagation through the plastic in reflection. As we see, this provides us with a confirmation of the values for $|n_1|$ given above, but does not provide the sign of the dispersive term.

We now describe a simple time-domain model accounting for the effects of dispersion to lowest order. Instead of the actual temporal shape of $E_r(t)$, for the purposes of our analysis, we assume a broadband temporally Gaussian reference THz pulse,

$$E_r(t, 0) = e^{-\pi\nu_0^2 t^2} \quad [4]$$

where ν_0 is a measure of its bandwidth. Note that this model pulse's bandwidth is centered at $\nu = 0$, whereas the pulse spectrum in Fig. 2 lacks spectral content below ~ 100 GHz. We expect that pulse spreading will not depend too critically on the pulse shape so long as it covers the relevant spectral bandwidth. After Fourier transformation, the electric field of THz pulse in the frequency-domain after propagating over distance d , can be expressed as

$$A_s(\nu, z) = A_s(\nu, 0)e^{-\alpha(\nu)z/2}e^{-in(\nu)k(\nu)z} \quad [5]$$

The absorption coefficient originates in the real part of the exponent that only controls the amplitude of reflected signal. We include only the lowest-order dispersion term (second order) in $n(\nu)$, i.e., $n(\nu) = n_0 + n_1\nu$. In addition, for the application of this pulse-propagation

model, we will neglect the dispersion in the absorption coefficient within the pulse bandwidth.

After taking the inverse Fourier transform, the THz pulse in the time domain is

$$A_s(t, z) = \int_{-\infty}^{+\infty} d\nu A_s(\nu; z) e^{2\pi i \nu t} = e^{-\frac{\alpha z}{2}} \int_{-\infty}^{+\infty} d\nu A_s(\nu, 0) e^{-2\pi i n_1 \nu^2 z/c} e^{2\pi i \nu \tilde{t}} \quad [6]$$

where $\tilde{t} = t - n_0 z/c$ includes the propagation delay. Thus,

$$\begin{aligned} A_s(t, z) &= v_0^{-1} e^{-\frac{\alpha z}{2}} \int_{-\infty}^{+\infty} d\nu e^{-\frac{\pi \nu^2}{v_0^2}} e^{-\frac{2\pi i n_1 \nu^2 z}{c}} e^{2\pi i \nu \tilde{t}} \\ &= v_0^{-1} e^{-\alpha z/2} \sqrt{\frac{\pi}{p}} e^{-p} \end{aligned} \quad [7]$$

with $p = \pi(v_0^{-2} + 2in_1 z/c)$.

From Eq. (7), the temporal pulse width is determined by

$$v_0^{-1} [1 - (\frac{2n_1 z v_0^2}{c})^2]^{-1/2} \approx v_0^{-1} [1 + (\frac{2n_1 z v_0^2}{c})^2] \quad [8]$$

with respect to the incident pulse amplitude, the amplitude of the pulse is attenuated as

$$[1 - (\frac{n_1 z v_0^2}{c})^2] \exp(-\alpha z) \approx \exp(-\alpha z) \quad [9]$$

Here, α is a representative value of the absorption coefficient within the pulse bandwidth.

This gives us a rough way to find the dispersion $|n_1|$ based on the temporal shape of the THz pulse following propagation through the plastic sheet. For PC we find $|n_1| = 0.0093$, and for PMMA $|n_1| = 0.0086$, respectively. Note that the modification to the pulse shape for the Gaussian depends only on n_1^2 ; therefore, we can only determine $|n_1|$ by these means. Nevertheless, the model result is in surprisingly good agreement with the rigorous value obtained in the frequency domain. As shown in Fig. 7. Due to the THz pulse shape, the spectral density at low frequency is greater than at high frequency, and the power falls off to the noise floor. This enables us to estimate the noise floor from the measure signal the THz beam path is blocked.

The second echo is reduced in amplitude with increased d as shown in Fig. 8 both due to attenuation and to pulse spreading caused by dispersion [43]. These effects can be distinguished because dispersion also causes the pulse to spread as well as to become reduced in peak height. Based on the Cauchy formula [44], the width of the reflected signal displays an excellent fit to the model $A + Bd^2$. We find for PC $A = 0.706$, $B = 0.0053$, and for PMMA $A = 0.721$, $B = 0.0078$, enabling us to estimate n_1 as 0.009 for PC and 0.008 for PMMA. These values for n_1 agree well with the slope of frequency-dependent refractive index obtained from the frequency-domain analysis of the transmission data. Note again, however, that this mode does not determine the sign of n_1 .

4.2 Thickness estimation

Characterization of $n(\nu)$, beyond its intrinsic interest, is of importance in using THz TOF tomography to measure the thickness of thick samples where the reflection from the back interface may be very weak. But before we see how this information can be applied to this problem, we employ FWDD for thickness determination to provide a benchmark. FWDD can assist in suppressing the effects of high- and low-frequency noise and to better distinguish weak features (i.e., echoes) in the reflected signal. In this study, Hanning filters are selected serving as the frequency-domain filters, with transfer function $F(\nu)$ given by

$$F(\nu) = \begin{cases} e^{i2\pi\nu t_0} \cos^2\left(\frac{\pi\nu}{2\nu_c}\right) & |\nu| \leq \nu_c \\ 0 & |\nu| > \nu_c \end{cases} \quad [10]$$

where t_0 corresponds to the arrival time of the main peak in the time-domain and ν_c is the cutoff frequency. The transfer function for the filter is manipulated by optimizing the cutoff frequency ν_c . The spectrum of the Hanning window function applied in these works is shown in Fig. 9. In the wavelet denoising procedure, *symlet* (sym4) wavelets are selected with level 8 for the wavelet decomposition. More details can be found in Ref [16]. The reconstructed

impulse response function for the PC and PMMA samples are shown in Fig. 10 and 11. Due to the elimination of anomalous spikes in low- and high-frequency regions, the frequency components within the band [0 THz, 3.5 THz] are retained. Consequently, the signal-to-noise ratio of the reconstructed signal is significantly improved, and two pulses corresponding to the air/plastic and plastic/air interfaces can be clearly identified for both plastic sheets. Focusing attention on the second pulse of reflected THz signals for various PC and PMMA sheets, broadening of this second reflection from the back interface is evident due to the collective effect of narrowing the bandwidth with frequency-domain filter as well as dispersion effect, together with the low signal-to-noise of the second echo resulting from obvious fluctuations corresponding to the low frequency noise as residual noises, making it increasingly difficult to precisely identify the time delay to the second echo as sample thickness increases.

FWDD, which decomposes of the detected signal $E_s(t)$ into wavelets that best match the features of the received signal, is not easily adapted to account for dispersion because the complexity of employing the dispersion model into wavelet. Instead, we consider the CC between the detected reflected signal $E_s(t)$ and the reference pulse $E_r(t)$ suitably modified to account for dispersion; in essence, CC quantifies the degree of linear similarity between the reference and the reflected signal at various time delays [45]. Pulses in $h(t)$ recovered from FWDD are much wider than the ideal impulses because of the dispersion as well as the narrow of bandwidth by frequency-domain filtering, and consequently limit the depth resolution of FWDD. CC, however, provides an intuitively simple way to account for dispersion. It only determines the time-delay between the reference and reflected signals. After calculating the CC between two signals, the maximum (or the minimum if the signals are negatively correlated) of the cross-correlation function indicates the point in time where the signals are best aligned; furthermore, these techniques are also powerful for conditions

with low signal-to-noise where the reflected signal is weak. The CC between the reflected signal $E_s(t)$ and reference signal $E_r(t)$ can be expressed as

$$\omega(t) = E_s(t) \otimes E_r(t) \triangleq \int_{-\infty}^{\infty} E_s^*(\tau) E_r(\tau + t) d\tau \quad [11]$$

In practice, we should consider the discrete form of Eq. [11], which is

$$\omega[n] = \sum_{m=1}^N E_s[m] E_r[m + n] \quad [12]$$

where N the number of data points [19],[46].

In order to enhance the depth resolution, balancing the efficiency and complexity, a linear time-invariant (LTI) system [47] was designed to simulate the effect of pulse-spreading [48]. The temporal spreading was assumed caused by the propagating THz pulse traveling a distance Δz in an attenuative/dispersive medium. Hence, the reference THz pulse $h_{\Delta z}(t)$ can be expressed as

$$h_{\Delta z}(t) = \rho_{\Delta z}(t) \otimes h_0(t) \quad [13]$$

where $h_0(t)$ is the THz signal at $\Delta z = 0$, corresponding to the reference THz signal, and $\rho_{\Delta z}(t)$ is the impulse response function of plastic samples.

A quasi-Dirac function was used to simulate the impulse-response function is $\rho_{\Delta z}(t)$, which is of the form

$$\rho_{\Delta z}[n] = \begin{cases} a & n = 0 \\ 1 - 2a & n = 1 \\ a & n = 2 \\ 0 & \text{otherwise} \end{cases} \quad [14]$$

where the parameter a determines the degree of pulse-spreading, and its range should be $0 < a < 1/2$ [48]. Note that when $a = 0$, $\rho_{\Delta z}[n] = \delta[n - 1]$, indicating that it only displays a simple time delay of one sampling period without pulse-spreading. The criterion for the fitting parameter a is based on the mean square error (MSE) between width of the convolved signal

$\rho_{\Delta z}(t) \otimes h_0(t)$ and the width of raw measured signal at 2, 4, 6, 8, 10, and 12 mm. The comparison between simulated and measured width of second pulse for PC and PMMA samples are shown in Fig. 12. The optimal value for parameter a in this model for PC is determined to be 0.23 and for PMMA is 0.28. For high thicker samples, $d > 10$ mm, the error between the simulated width and measured width is increases, which attributes to the low signal-to-noise in that region. The CC results for PC and PMMA with and without accounting for dispersion are shown in Fig. 13 and 14. The comparison of amplitude of second pulse of reflected THz signal between FWDD result and CC result with and without accounting for dispersion is shown in Fig. 15. For both PC and PMMA, the amplitude of the CC peak of the reflected pulse from the back surface is enhanced significantly when dispersion is included as opposed to when dispersion is neglected, indicating that pulse spreading is a significant factor that degrades the ability to identify weak reflected pulses in thicker samples. Moreover, pulse spreading will also limit the accuracy of thickness determinations in thicker samples employing analysis techniques that do not account for dispersion.

While FWDD was successful in identifying the echo from the back plastic/air interface of all sheets studied, it is clear that this approach is near its limit for the 12 mm sheets shown in Figs. 10 (f) and 11(f); CC will thus enable sheet thickness determination for somewhat larger values of d than were available for study here. We can quantify this by extrapolating our results. The extracted FWDD and CC peak values, with and without dispersion, in Fig. 15 are fit to exponentials (since the falloff is dominated by absorption). The noise floor is determined by the mean and standard deviation of system noise and is indicated in the panels in Fig. 15. When the amplitude of second echo (obtained by FWDD or CC with or without dispersion) descends to the noise floor, the signal reflected from sample can no longer not be distinguished from system noise. The extrapolated value of d where the peak amplitude is

equal to the noise floor defines the thickest sample for which a given analysis technique can be used to detect the echo from the bottom plastic/air interface.

In Fig 15(a), (b) and (d), (e) we see that FWDD and CC without dispersion are limited to measuring PC and PMMA slabs with $d \sim 17$ mm. Accounting for dispersion, however, CC enables us to measure samples up to $d \sim 36$ mm. *This is a factor of ~ 2 improvement in layer thicknesses that can be measured by THz TOF tomography.*

CONCLUSIONS

In conclusion, we have measured the frequency-dependent refractive index and the attenuation constant of PC and PMMA. The refractive indices are $n(\nu) = 1.67 - 0.0096\nu$ for PC and $n(\nu) = 1.61 - 0.0085\nu$ for PMMA where ν is measured in THz; the absorption coefficient are $\alpha(\nu) = 0.628 + 4.64\nu^{1.675}$ in units of cm^{-1} for PC and $\alpha(\nu) = 0.388 + 13.15\nu^{1.777}$ for PMMA, where ν is measured in THz. These values are in reasonable agreement with those in the literature, though there is not universal agreement on the sign of the dispersion. We then turned our attention to using THz TOF tomography to measure the thickness of PC and PMMA plastic sheets within sample thickness d in the range 2 to 12 mm. Due to dispersion, which broadens the width of the THz reflection from the back interface of the sample, the depth resolution for FWDD is significantly degraded. We find that for thicker samples (here $d \gtrsim 8$ mm), the second echo arising from the back plastic/air interface is significantly broadened and reduced in amplitude, leading to this feature to become hidden in noise. We show by straightforward extrapolation that naïve analysis techniques that neglect the effects of dispersion (here FWDD and direct CC without dispersion) are limited to measuring layer thicknesses in these materials of less than ~ 17 mm. Accounting for dispersion, however, results in a factor-of-two improvement, allowing us to measure samples as thick as ~ 36 mm.

While our experimental study was limited to PMMA and PC sheets of maximum thickness 12 mm, our work shows that CC will enable THz TOF tomography to measure thicknesses of a variety of plastics to values of d in the few-cm range using commercial THz systems.

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Table 1:

Table 1. The physical, optical, and mechanical properties of the PC and PMMA plastic sheets studied.

Properties	Test Method	PC	PMMA
Density	ISO 1183	1.2 g/cm ³	1.19 g/cm ³
Water absorption (in water for 24hrs at 23°C)	ISO 62-1	0.35%	0.3%
Refractive index	ISO-489	1.586	1.49
Vicat softening point	ISO 306	106°C	148°C
Tensile strength	ISO 527-2	60MPa	65MPa
Elongation at break	ISO 527-2	6%	4%

Fig. 1

Fig. 1 Schematic diagram of THz time-domain system in both transmission and reflection mode. An ultrafast fiber laser produces near-infrared pulses that are incident on an electrically biased photoconductor (in the block marked THz Emission) thus converting them to approximately single-cycle THz pulses.

Fig. 2

Fig. 2 THz reference signal (pulse produced by the apparatus) with its power spectrum in the inset. The duration and dynamic range of THz pulse are ~ 2 ps and > 60 dB, while the corresponding spectrum extends from ~ 100 GHz to 3 THz before falling below the noise floor.

Fig. 3

Fig. 3 Frequency-dependent absorption coefficient $\alpha(\nu)$ for (a) PC and (b) PMMA. The different level of the reflected signal with respect to the noise floor for samples with different d is responsible for the frequency at which $\alpha(\nu)$ appears to level off (and the curves diverge). The absorption is described by the universal dielectric response (power-law behavior) as discussed in the text.

Fig. 4

Fig. 4 (a) Frequency-dependent refractive index $n(\nu)$ for PC; (b) Comparison between $n(\nu) = 1.61 - 0.0096\nu$ and measurement; (c) Frequency-dependent refractive index $n(\nu)$ for PMMA; (d) Comparison between data for the 6-mm thick PMMA sheet and $n(\nu) = 1.67 - 0.008476\nu$.

Fig. 5

Fig. 5 Typical measured THz signals for PC plastic sheets of various thicknesses. Insets show THz pulse reflected off back plastic/air interface on expanded scale.

Fig. 6

Fig. 6 Typical measured THz signals for PMMA plastic sheets of various thicknesses. Insets show THz pulse reflected off back plastic/air interface on expanded scale.

Fig. 7

Fig. 7. Plastic-sheet thickness-dependence of width of the first pulse (THz pulses reflected off the front air/plastic interface) and the width of the second pulse (THz pulses reflected off the back plastic/air interface): first and second pulse for (a) PC and (b) PMMA. Note the different vertical scales for the widths of the first and second echoes.

Fig. 8

Fig. 8 Amplitude of second echo of reflected THz signal for all (a) PC and (b) PMMA sheets, indicating that attenuation follows Beer's law as indicated by black curves.

Fig. 9

Fig. 9. Hanning window function with typical values, $t_0=9.57$ ps and $\nu_c=3.5$ THz with inverse Fourier transform in inset.

Fig. 10

Fig. 10. FWDD results for the reconstructed impulse response function $h(t)$ for all PC samples. Insets show THz pulse reflected off back plastic/air interface on expanded scale.

Fig. 11

Fig. 11. FWDD results for the reconstructed impulse response function $h(t)$ for all PMMA samples. Insets show THz pulse reflected off back plastic/air interface on expanded scale.

Fig. 12

Fig. 12. Comparison between simulated and measured width of pulse reflected off back plastic/air interface for (a) PC and (b) PMMA samples.

Fig. 13

Fig. 13. CC results for all PC samples accounting for (black) and neglecting (red) dispersion. Insets show THz pulse reflected off back plastic/air interface on expanded scale.

Fig. 14

Fig. 14. CC results for all PMMA samples accounting for (black) and neglecting (red) dispersion. Insets show THz pulse reflected off back plastic/air interface on expanded scale.

Fig. 15

Fig. 15. Comparison between the peak amplitude associated with the echo from the bottom plastic/air interface for PC (a)-(c) and PMMA (d)-(e). Noise floor is indicated by horizontal dashed line. Blue curves show exponential fit to extracted values: (a) and (d) amplitudes of FWDD peak; (b) and (e) peak CC without dispersion; (c) and (f) peak CC with dispersion. Value of d where the exponential fit means the noise floor defines the thickest layer for which the second echo can be identified for a given analysis technique.

REFERENCE

- [1] L. Duvillaret, F. Garet, and J. L. Coutaz, "Highly precise determination of optical constants and sample thickness in terahertz time-domain spectroscopy," *Appl. Opt.*, vol. 38, no. 2, pp. 409-415, 1999.
- [2] M. Wormington, C. Panaccione, K. M. Matney, and D. K. Bowen, "Characterization of structures from X-ray scattering data using genetic algorithms," *Phil. Trans. R. Soc. Lond. A*, vol. 357, no. 1761, pp. 2827-2848, 1999.
- [3] K. Inaba, "X-ray thin film measurement techniques," *The Rigaku Journal*, vol. 24, no. 1, pp. 10-15, 2008.
- [4] S. Akhtar, S. Rubino, and K. Leifer, "A simple TEM method for fast thickness characterization of suspended graphene flakes," *Microsc. Microanal.*, vol. 22, no. 1, pp. 250-256, 2016.
- [5] A. Gallego, J. F. Gil, E. Castro, and R. Piotrkowski, "Identification of coating damage processes in corroded galvanized steel by acoustic emission wavelet analysis," *Surf. Coatings Technol.*, vol. 201, no. 8, pp. 4743-4756, 2007.
- [6] J. E. Bjarnason, T. L. J. Chan, A. W. M. E. Lee, M. A. Celis, and E. R. Brown, "Millimeter-wave, Terahertz, and mid-infrared transmission through common clothing," *Appl. Phys. Lett.* vol. 85, no. 4, pp. 519-521, 2004.
- [7] D. Banerjee, W. von Spiegel, M. D. Thomson, S. Schabel, and H. G. Roskos, "Diagnosing water content in paper by terahertz radiation," *Opt Expr.*, vol. 16, no. 12, pp. 9060-9066, 2008.
- [8] R. Piesiewicz, C. Jansen, S. Wietzke, D. Mittleman, M. Koch, and T. Kurner, "Properties of building and plastic materials in the THz range," *Int. J. Infrared Milli Waves*, vol. 28, no. 5, pp. 363-371, 2007.
- [9] A. I. Hernandez-Serrano, S. C. Corzo-Garcia, E. Garcia-Sanchez, M. Alfaro, and E. Castro-Camus, "Quality control of leather by terahertz time-domain spectroscopy," *Appl. Opt.*, vol. 53, no. 33, pp. 7872-7876, 2014.
- [10] Matthew Reid and R. Fedosejevs, "Terahertz birefringence and attenuation properties of wood and paper," *Appl. Opt.*, vol. 45, no. 12, pp. 2766-2772, 2006.
- [11] C. Lan, K. Bi, B. Li, Y. Zhao, and Z. Qu, "Flexible all-dielectric metamaterials in terahertz range based on ceramic microsphere/PDMS composite," *Opt Expr.*, vol. 25, no. 23, pp. 29156-29160, 2017.
- [12] J. Dong, A. Locquet, M. Melis, and D.S. Citrin, "Global mapping of stratigraphy of an old-master painting using sparsity-based terahertz reflectometry," *Sci. Rep*, vol. 7, pp. 1-12, 2017.
- [13] K. Fukunaga, T. Meldrum, W. Zia, M. Ohno, T. Fuchida, B. Blumich, "Nondestructive investigation of the internal structure of fresco painting," *Proc. Digit. Herit*, vol. 1, pp. 81-88, 2013.
- [14] E. P. J. Parrott, S. M. Y. Sy, T. Blu, V.P. Wallace, and E. Pickwell-MacPherson, "Terahertz pulsed imaging in vivo: measurements and processing methods," *J. Biomed. Opt.*, vol. 16, no. 10, pp. 106010-106018, 2011.
- [15] S. Krimi, J. Klier, J. Jonuscheit, G. V. Freymann, R. Urbansky, and R. Beigang, "Highly accurate thickness measurement of multi-layered automotive paints using terahertz technology," *Appl. Phys. Lett.* vol. 109, 021105, 2016
- [16] J. Dong, A. Locquet, and D.S. Citrin, "Terahertz quantitative nondestructive evaluation of failure modes in polymer-coated steel," *IEEE J. Sel. Top. Quantum Electron.*, vol. 23, no. 4, pp. 1-7, 2017.

- [17] Y. Chen, S. Huang, and E. P. MacPherson, "Frequency-wavelet domain deconvolution for terahertz reflection imaging and spectroscopy," *Opt. Express*, vol. 18, no. 2, pp. 1177-1190, 2010.
- [18] N. H. Matlis, G. R. Plateau, J. van. Tilborg, and W. P. Leemans, "Single-shot spatiotemporal measurement of ultrashort THz waveforms using temporal electric-field cross correlation," *J. Opt. Soc. Am. B*, vol. 28, no. 1, pp. 23-27, 2011.
- [19] S. Adrian-Martinez, M. Bou-Cabo, I. Felis, C.D. Llorens, J. A. Martinez-Mora, M.Saldana, and M. Ardid, "Acoustic signal detection through the cross-correlation method in experiments with different signal to noise ratio and reverberation conditions," International conference on Ad-Hoc Networks and wireless, 2015.
- [20] R. M. Lerner, "A matched filter detection system for complicated Doppler shifted signals," *IRE Trans. Information Theory*, vol. 6, no. 3, pp. 373-385, 1960.
- [21] M. Zhai, A. Locquet, C. Roquelet, and D. S. Citrin, THz Thickness Characterization of plastic sheets including dispersion, 44th International Conference on Infrared, Millimeter, and Terahertz Waves 2019.
- [22] W. H. Fan, A. Burnett P. C. Upadhy, J. Cunningham, E. H. Linfield, and A. G. Davies, "Far-infrared spectroscopic characterization of explosives for security applications using broadband terahertz time-domain spectroscopy," *Appl. Spectrosc*, vol. 61, no. 6, pp. 638-643, 2007.
- [23] P. U. Jepsen, and B. M. Fischer, "Dynamic range in terahertz time-domain transmission and reflection spectroscopy," *Opt. Lett*, vol. 30, no. 1, pp. 29-31, 2005.
- [24] R. J. Bell, *Introductory Fourier Transform Spectroscopy*, (Academic Press, New York, 1972).
- [25] A. S. Filler, "Apodization and Interpolation in Fourier-Transform Spectroscopy," *J. Opt. Soc. Am.* vol. 54, pp. 762-767, 1964.
- [26] R. H. Norton and R. Beer, "New Apodizing Functions for Fourier Spectrometry," *J. Opt. Soc. Am.* Vol. 66, pp. 259-264, 1976.
- [27] R. H. Norton and R. Beer, "Errata - New Apodizing Functions for Fourier Spectrometry," *J. Opt. Soc. Am.* vol. 67, pp. 419, 1977.
- [28] F. J. Harris, "On the Use of Windows for Harmonic Analysis with the Discrete Fourier Transform," *Proceedings of the IEEE*, vol. 66, pp. 51-83, 1978.
- [29] Blackman, R. B. and Tukey, J. W, *The Measurement of Power Spectra, From the Point of View of Communications Engineering*, 1959.
- [30] J. Dong, A. Locquet, D.S. Citrin, "Enhanced terahertz imaging of small forced delamination in woven glass fibre-reinforced composites with wavelet de-noising," *J Infrared Millim. Terahz Waves*, vol. 37: no. 3, pp. 289-301, 2016.
- [31] E. V. Fedulova, M. M. Nazarov, A. A. Angeluts, M. S. Kitai, V. I. Sokolov, and A. P. Shkurinov, "Studying of dielectric properties of polymers in the terahertz frequency range," Proc. SPIE 8337, Saratov Fall Meeting 2011: Optical Technologies in Biophysics and Medicine XIII, 833701 (28 February 2012).
- [32] Ryzhov V.A., "Low-energy libration excitations in glassy PMMA," *Solid State Physics*, vol. 44, no. 12, pp. 2229-2233, 2002.
- [33] A. K. Jonscher, "The 'universal' dielectric response," *nature.*, vol. 267, pp. 673-679, 1977.
- [34] D.K. Das-Gupta, and P.C.N. Scarpa, "Modeling of dielectric relaxation spectra of polymers in the condensed phase," *IEEE Electra Insul M*, vol. 15, no. 2, pp. 23-32, 1999
- [35] P. Lunkenheimer, and A. Loidl, "Response of Disordered Matter to Electromagnetic Fields" *Phys. Rev. Lett.*, vol. 91, no. 20, pp: 207601, 2003.
- [36] Y. Jin, G. Kim, and S. Jeon, "Terahertz Dielectric properties of polymers," *J. Korean, Phys. Soc*, vol. 49, no. 2, pp. 513-517, 2006.

- [37] W. He and Z. Guo-Zhong, "Terahertz spectroscopic inspection of several kinds of plastic," *Advances in Mathematics(China)*, vol. 39, no. 7, pp. 1185-1188, 2010.
- [38] P.D. Cunningham, N.N. Valdes, F.A. Vallejo, L. M. Hayden, B. Polishak, X. Zhou, J. Luo, A.K. Jen, J. C. Williams, and R. J. Twieg, "Broadband terahertz characterization of the refractive index and absorption of some important polymeric and organic electro-optic materials," *J. Appl. Phys.*, vol. 109, pp. 043505, 2011.
- [39] M. Naftaly, and R. E. Miles, "Terahertz time-domain spectroscopy for material characterization," *Proceedings of the IEEE*, vol. 95, no. 8, pp. 1658-1665, 2007.
- [40] S. F. Busch, M. Weidenbach, M. Fey, F. Schäfer, T. Probst, and M. Koch. "Optical properties of 3D printable plastics in the THz regime and their application for 3D printed THz optics," *J. Infrared Millim. Terahertz Waves* vol.35, pp. 993–997, 2014.
- [41] H. Wang, and G. Zhao, "Terahertz spectroscopic inspection of several kinds of plastic," *Acta Photonica Sinica*, vol. 39, no. 7, pp. 1185-1188, 2010.
- [42] W. R. Folks, S. K. Pandey, and G. Boreman, "Refractive Index at THz Frequencies of Various Plastics," in *Optical Terahertz Science and Technology*, OSA Technical Digest Series (CD) (Optical Society of America, 2007), paper MD10.
- [43] S. Zhou, D. G. Valchev, A. Dinovitser, J. M. Chappell, A. Iqbal, B. W. H. Ng, T. W. Kee, and D. Abbott, "Terahertz signal classification based on geometric algebra," *IEEE Trans. THz Sci. Technol.*, vol. 6, no. 6, pp. 793–802, Nov. 2016.
- [44] S. Wang, and G. Chen, "Cauchy problem of the generalized double dispersion equation," *Nonlinear Anal-Theor*, vol. 64, no. 1, pp. 159-173, 2006.
- [45] J. G. Proakis and D. G. Manolakis, *Digital Signal Processing*, 3rd prentice Hall, (1996).
- [46] L. Fillinger, A. Sutin, and A. Sedunov, "Acoustic ship signature measurements by cross-correlation method", *J. Acoust. Soc. Amer.*, vol. 129, no.2, pp.774-778, 2011.
- [47] T. Olofsson and T. Stepinski, "Minimum entropy deconvolution of pulse-echo signals acquired from attenuative layered media," *J. Acoust. Soc. Amer.*, vol. 109, no.6, pp. 2831-2839, 2001.
- [48] J. Dong, A. Locquet, and D.S. Citrin, "Terahertz superresolution stratigraphic characterization of multilayered structures using sparse deconvolution," *IEEE Trans. Terahertz Sci. Technol.*, vol. 7, no. 3, pp. 260-267, 2017.