Dielectric Constant Measurement of Thin Films Using Goniometric Terahertz Time-Domain Spectroscopy

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Abstract—Goniometric time-domain spectroscopy (GTDS), employing an ultrashort electromagnetic (EM) pulse technique, has been developed for measuring the dielectric constant of thin films in a broad band of gigahertz to terahertz. An ultrafast optoelectronic system, including an emitter and a detector unit, is constructed with a θ–2θ goniometer. A silicon wafer was analyzed as the reference substrate material. A sharp π phase-shift of the reflected EM wave was observed at the Brewster angle of 73.5° for a bare silicon wafer. The phase shift for a film on the Si substrate is relatively smooth due to its two surfaces’ providing a complex reflectance. The dielectric constant of the film on Si, related with angular dependency of the phase shift, can be extracted by means of fitting the curve or measuring slope of the curve near the Brewster angle. The measured dielectric constants of FLARE, TiOx, and PZT film are reported.

Index Terms—Brewster angle, dielectric constant, phase shift, terahertz beam, thin film.

I. INTRODUCTION

ANY NEW dielectric materials are being developed for potential use in high-speed ultralarge-scale integration (ULSI) microelectronics [1]. The proper propagation of a clock signal through the device interconnects requires that the surrounding dielectric materials do not have any significant dispersion in the band of ten times of the base frequency. Metrology tools for the measurement of the frequency-dependent dielectric constant of materials in the gigahertz to terahertz range are urgently needed in this development effort. Conventional methods of film characterization have frequency limits. For example, a capacitance measurement via an impedance analyzer is used to determine the dielectric constant at low frequency (<1 MHz), and it is possible to use a network analyzer to measure dielectric response into the low gigahertz region [2]. Sending an optical beam to the film near the Brewster angle, the dielectric information (refractive index) can be extracted by measuring the elliptical polarization properties of the reflected beam. The technical core of the ellipsometry is to obtain the ratio of s- and p-polarized complex reflectance. Ellipsometry is used for the measurement at visible light and near infrared region (20 to 800 THz) due to the detection limitation [3].

Terahertz time-domain spectroscopy (TDS) is a well-established method employing electromagnetic (EM) pulse emission and detection with applications in the gigahertz to terahertz range [4], [5]. The temporal shapes of the input and propagated/ reflected terahertz pulses are measured by the cross-correlation between an electromagnetic pulse and a probe optical pulse. The powerful advantages can be summarized as follows.

1) The detection of an EM pulse is extremely sensitive. A nanowatt signal can be easily detected. The reported dynamic range of an EM pulse waveform exceeds 10⁶ with microwatt average power of the optical probe beam by using an electrooptic (EO) sampling technique [6].
2) TDS provides information in a very broad band. The 37 THz bandwidth of ZnTe EO sensor has been previously demonstrated [7]. It provides a tool for studying materials at frequencies up to tens of terahertz.
3) The thermal background is absent because the femtosecond optical gated probe pulse is only modulated by the detected EM pulse in a short time period.
4) Coherent measurement provides both amplitude and phase information.

TDS has been demonstrated to measure the dielectric constant of bulk materials and freestanding thick films using the transmission geometry [8], [9]. The fundamental idea of goniometric time-domain spectroscopy (GTDS) is to apply the advanced detection technique to ellipsometry. Unlike the conventional ellipsometry, the complex reflectance is measured not only at a certain angle but also at multiple points within a range around the Brewster angle. In addition, the terahertz beam is tuned to be p-polarized but the probe optical beam is s-polarized. The angular dependence of the terahertz reflectance determines the dielectric constant of thin film under measurement. GTDS is applicable for measuring thin films on a substrate.

The GTDS concept was addressed in [10]. In this paper, we report a detailed discussion of the technology, the computation methods, and measured results on three samples of different nature.

II. PRINCIPLES

Fig. 1 illustrates the experimental arrangement of the GTDS apparatus. A femtosecond laser beam is split in two, with the majority of the power exciting the terahertz pulse emitter and the rest going to an EO field sensor for field detection [10]. The time delay for waveform scanning is controlled by a pair of mirrors mounted on a translation stage with a resolution of 25 nm. The p-polarized terahertz beam is collimated by a paraboloidal...
mirror. The terahertz beam and the reference beam are combined by a pellicle and sent to the sample under test. A \( \theta \rightarrow 2 \theta \) goniometer with 0.01° angular resolution is integrated into the terahertz system. The film sample is mounted on the \( \theta \) rotation stage of the goniometer. A detection unit including an EO crystal, a quarter-wave plate, a Wollaston polarizer, and a balance detector pair is placed on the 2\( \theta \) stage. After the pellicle, the terahertz beam is guided by the optical probe beam so that the rotation can be performed accurately even though it has a relatively large divergent angle. The input terahertz waveform can be measured at position 1 with the sample and its mount removed. When the sample is placed in the system, the reflected terahertz waveform can be scanned by moving the delay stage. The detection unit at position 2 is rotated by 2\( \theta \) while the sample is rotated by \( \theta \). Although the reference beam is set to be \( s \)-polarized to avoid its phase shift, its intensity changes with the angle of incidence. The terahertz signal has to be normalized by the intensity of the reference optical beam [11].

GTDS employs the reflection information of an EM wave to determine the dielectric properties of a thin film. The temporal waveforms of an input pulse \( E_{\text{in}}(t) \) and a reflected pulse at an incident angle of \( \theta \), \( E_{\text{ref}}(t, \theta) \), are measured in a GTDS system. The incident and reflected waveforms are gathered, and the electric fields in the time domain are expressed as

\[
E_{\text{in}}(t) = \int G(\nu) S(\nu) \exp(-i 2 \pi \nu t) \, d\nu
\]

\[
E_{\text{ref}}(t, \theta) = \int r(\nu, \theta) \exp[i \varphi(\nu, \theta)] \cdot G(\nu) S(\nu) \cdot \exp(-i 2 \pi \nu t) \, d\nu
\]

respectively, where
- \( \nu \) frequency;
- \( \theta \) incident angle;
- \( S(\nu) \) frequency component of the incident signal;

\( G(\nu) \) response function of terahertz system, which is related with the sensor and the geometry of the system and independent on the incident angle.

The complex reflectance of a film on a substrate can then be obtained

\[
r(\nu, \theta) \exp(i \varphi(\nu, \theta)) = \frac{\int E_{\text{ref}}(t, \theta) \exp(i 2 \pi \nu t) \, dt}{\int E_{\text{in}}(t) \exp(i 2 \pi \nu t) \, dt}.
\]

Two curves of \( r(\nu, \theta) \) versus \( \theta \) and \( \varphi(\nu, \theta) \) versus \( \theta \) can be plotted. The dielectric constant can be extracted by the analysis of these curves because the reflectance is related to the dielectric properties of the material.

A film on a substrate has two surfaces, shown in Fig. 2. When a polarized electromagnetic wave is reflected from such a material, the complex reflectance is expressed by the Drude equation [12]

\[
r e^{i \delta} = \frac{r_1 + r_2 e^{i \delta}}{1 + r_1 r_2 e^{i \delta}}
\]

where \( r_1 \) and \( r_2 \) are the reflectance of the two interfaces (air-to-film and film-to-substrate, respectively) of the film and \( \delta \) is the phase difference. The reflectance is mathematically described for \( p \)-polarized waves by the Fresnel formulas

\[
r_1 = \frac{n_2 \cos \theta_1 - n_1 \cos \theta_2}{n_2 \cos \theta_1 + n_1 \cos \theta_2}
\]

\[
r_2 = \frac{n_3 \cos \theta_2 - n_2 \cos \theta_3}{n_3 \cos \theta_2 + n_2 \cos \theta_3}
\]

where \( n_1 \), \( n_2 \), and \( n_3 \) are the refractive indexes of air, the film, and the substrate, respectively, and the \( \theta_1 \)s are the incident and refraction angles, obeying Snell’s law

\[
n_1 \sin \theta_1 = n_2 \sin \theta_2 = n_3 \sin \theta_3.
\]

In (1)

\[
\delta = \frac{4 \pi}{\lambda} n_2 d \cos \theta_2
\]

where \( \lambda \) is the EM wavelength and \( d \) is the film thickness. Placing (5)–(8) into (4), one can express the complex re-
reflectance as a function of the incident angle \( \theta_i \). Utilizing this theory, we can calculate the amplitude and phase separately for films with different dielectric constants on silicon substrates. The calculated results are compared with the experimental results. The experimental results show that the amplitude of the reflectance drops to a minimum value, at which we define the Brewster angles, and then increases as the incident angle increases. The phases change by \( \pm \pi \) as the incident angle increases, where the + and – signs are for the films with dielectric constants smaller and larger than that of silicon, respectively. The phase shifts for the films are relatively smooth compared with that of the bulk silicon, and the changing “speed” depends on the dielectric constant of the film and its thickness.

One can determine the dielectric constant of the film by using the complex reflectance curve data. There are multiple methods to extract the dielectric information from the curves. First, a curve fit of the phase can be used. For this curve fit, the phases of reflectance at some discrete angles for the frequency \( \nu \), \( \varphi_{\text{ex}}(\theta_i) \), are measured from the experiment. They are placed with theoretical phase data \( \varphi(\theta_i, n_2) \) calculated from (1) in the fitting function

\[
f(n_2) = \sum_i (\varphi_{\text{ex}}(\theta_i) - \varphi(\theta_i, n_2))^2.
\]  

Changing the curve fit parameter \( n_2 \) to achieve the minimum value of \( f(n_2) \), we obtain the refractive index of the film. Its dielectric constant simply is \( \varepsilon(\nu) = \varepsilon_0 n_2^2(\nu) \). The phase shift is completed in a region near the Brewster angle. The dielectric constant can be extracted from the slope of the angle-phase curve in this sensitive region. A phase relaxation angle width (PRAW) is defined by two phases near the Brewster angle, for example, \( -0.5 \) and \( -2.5 \) for a high-\( K \) material. From the Drude equation, the relation between the dielectric constant and PRAW can be obtained. Alternatively, we can use the phase difference between two angles near the Brewster angle to define the slope of the angle-phase curve. Consequently, the dielectric constant is determined. This technique also utilizes the Drude equation. In this process, the measurement can be carried out only at two angles, and once the phase difference is known the dielectric constant can be calculated.

The analysis above is appropriate for a nonabsorptive film, for which only one parameter, \( n_2 \), needs to be determined. Another quantity, the extinction coefficient, has to be tested for an absorptive material. In (5) and (6), \( n_2 \) has to be replaced by \( n_2 + k_2 i \). Since the amplitude and phase curves are high-order functions with the rank of their polynomial Taylor expansions greater than two, \( n_2 \) and \( k_2 \) can be determined by the analysis of the angular dependence of the complex index. Curve fitting is the best method used to characterize an absorptive film.

**III. EXPERIMENTAL RESULTS**

We must know the dielectric constant of the substrate in order to measure a film deposited on its surface. Our substrate was silicon with a resistivity of \( >20 \ \Omega \text{cm} \) and a dopant concentration of about \( 5 \times 10^{14} \ \text{cm}^{-3} \). Fig. 3 illustrates the waveforms of the incident and the reflected EM pulses at different incident angles. As expected, the signal decreases as the incident angle increases, and at 73.5°, the reflected signal vanishes. At angles smaller than 73.5°, the reflected waves have no phase shift with respect to the incident pulse, implying that their relative phases remain zero. At incident angles larger than 73.5°, the pulses are flipped from their interaction with the substrate. Using fast Fourier transform, these pulses are converted into frequency-domain signals. They provide information in a wide frequency band from 600 GHz to 1.7 THz. The dielectric constant is determined at each frequency component. For example, the amplitude and phase of reflectance at 1 THz are depicted in Fig. 4, showing a zero-amplitude minimum value and the \( \pi \)-phase jump at 73.5°. The refractive index of silicon is then determined at this frequency from \( n = \tan \theta_B = \tan(73.5°) = 3.38 \), and the dielectric constant is then calculated as \( \varepsilon = n^2 = 11.4 \). It was observed that the Brewster angles for the frequency range from 0.6 to 1.7 THz are all about 73.5°, implying that silicon does not
have a significant dispersion in this frequency range. This result agrees with published data [13].

Following the same process as used for silicon wafer, the complex reflectance of a fluorinated poly(arylether) FLARE with 3.3 μm thickness on a silicon substrate is obtained and depicted in Fig. 5. In contrast to the case of a bare silicon wafer, the phase shift is relatively smooth. We fit the phase and amplitude of the measure waveforms by changing the dielectric constant in the Fresnel equations. From the best fit, we obtained the dielectric constant of 2.8 ± 0.1 for FLARE at 1 THz. The measured result at other frequencies in the 700 GHz to 1.4 THz range does not show the dispersion of this material.

We have measured a 980-Å titanium oxide (TiOx) film on a silicon substrate and used the PRAW and the phase difference near the Brewster angle to calculate the dielectric properties in the THz region. For this film, the phase shift from zero to −π shown in Fig. 6 is observed. This indicates that its dielectric constant is higher than that of the silicon substrate. We selected ϕ1 = −0.5 and ϕ2 = −2.5 for the PRAW method. We inserted points linearly between these two phases to get a curve with 1000 points, from which we obtain PRAW = 1.13. Substituting the known values of frequency and refractive index of Si, 3.38, into (4)–(8), one numerically obtains the relation between the PRAW and the dielectric constant of the film. Fig. 7 depicts this dependency at 1.05 THz for films with different thicknesses. From the “+” point, we determined the dielectric constant at a frequency of 1.05 THz of the TiOx film on silicon to be 47.

We have found that the phase difference method is easier for the angle-phase slope determination. We selected θ1 as 73.3° and θ2 as 74°. The EM pulse waveform scanning was performed four times at each angle and averaged to obtain a higher signal-to-noise ratio. The phase difference was computed using the Fourier transform method. At 1.05 THz, the measured Δφ is −1.64. By solving the Drude equation numerically, the dielectric constant of film can be obtained. From this, we find that the dielectric constant with respect to the phase difference of −1.64 is 47. We determined the dielectric constant at other frequencies following the same procedure, and the data are shown in Table I.

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The refractive index and dielectric constant of an absorptive material are complex numbers. A 1.8-μm PbZr0.4Ti0.6O3 (PZT) film obtained from Ramtron International Corporation has been measured using GTDS. A 40-nm TiO2 barrier layer was deposited between the film and the silicon substrate to prevent interdiffusion of the PZT and silicon during processing. Calculations were performed treating the film as a two-layer and a one-layer film. However, no significant difference was seen in the results, and therefore it was determined that the barrier was so thin compared to the PZT film that the sample can be treated as a one-layer film. Fig. 8 shows the EM waveforms reflected from the PZT film and the waveform of the incident pulse for reference. The waveforms at the angles larger than 75° are distorted. The Brewster angle was found to be 82° at 670 GHz and
thin films in the gigahertz to terahertz frequency range. Curve fitting of the complex reflectance and the information on the phase shift near the Brewster angle are used to determine the dielectric constant. For demonstration, low-κ and high-κ films with thicknesses smaller than the terahertz wavelength have been measured.

The measurement accuracy is mainly dependent on the terahertz signal-to-noise ratio, the measurement accuracy of incident angle, and the thickness. The thickness measurement of dielectric film can be precisely obtained from a commercial metrology tool, for example, KLA-Tencor ASET F5, which has 2.0-Å accuracy. However, the measurement error bar $\delta \theta = \gamma \delta \theta$, where $\delta \theta$ is the inaccuracy of angle measurement. The slope $\eta$ is a function of the film thickness. From Fig. 7, the thicker film has less dielectric constant error. Limited by our condition, the current goniometer in the system can distinguish 0.01°. The signal-to-noise ratio of the terahertz signal at the Brewster angle is much smaller, due to the low reflectivity, than that at the small incident angle or reference signal. But it always can be improved by multi-time scan average. In additional, adding measurement at more angles also enhances the fitting accuracy.

In the analysis, the surfaces of films are assumed to be ideal planes. However, the roughness of the surface also affects the reflectance. When the roughness is considered, in (4), some scattering factors must multiply $r_1$ and $r_2$ for adjustment, $r_1 \rightarrow f_1 r_1$ and $r_2 \rightarrow f_2 r_2$, where $f_1$ and $f_2$ are related to the roughness of the two surfaces of the film, respectively, and they are normally smaller than one. In addition, our system also has the capability to measure a two-layer film. The theory of multilayer film is appropriate for such a material.

Fig. 8. The input THz beam and reflected waveforms reflected from a PZT film. From top to bottom, the incident angles are 45°, 50°, 55°, 60°, 65°, 70°, 75°, 78°, 80°, and 82.5°.

Fig. 9. The measured dielectric constant of the PZT film.

83.4° at 1.4 THz. The difference in the Brewster angles at different frequencies is caused by the dielectric dispersion. For this film, we used the curve-fitting method to determine the complex dielectric constant. (The slope of the phase shift cannot reflect both the real and imaginary parts of the dielectric constant, so we cannot use either of the phase methods.) Fig. 9 shows the dielectric constant of the PZT film. Clearly, it has dispersion in the frequency range from 670 GHz to 1.4 THz. The total noise from the curve fitting and the system performance gives 4~5% uncertainty. Its dielectric constant at low frequency from 10 KHz to 1 MHz characterized by the conventional electrical method is about 900. The refractive index at 632.8 nm is 2.58. Hence, as expected, the dielectric constant decays as the frequency increases.

IV. DISCUSSION AND CONCLUSION

Goniometric time-domain spectroscopy successfully extended the base theory of ellipsometry to very low frequency. It is a practical technique for measuring the dielectric constant of


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