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Citation: Appl. Phys. Lett. **109**, 021105 (2016); doi: 10.1063/1.4955407
View online: http://dx.doi.org/10.1063/1.4955407
View Table of Contents: http://aip.scitation.org/toc/apl/109/2
Published by the American Institute of Physics

APPLIED PHYSICS LETTERS **109**, 021105 (2016)

# Highly accurate thickness measurement of multi-layered automotive paints using terahertz technology

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(Received 8 May 2016; accepted 22 June 2016; published online 12 July 2016)

In this contribution, we present a highly accurate approach for thickness measurements of multi-layered automotive paints using terahertz time domain spectroscopy in reflection geometry. The proposed method combines the benefits of a model-based material parameters extraction method to calibrate the paint coatings, a generalized Rouard's method to simulate the terahertz radiation behavior within arbitrary thin films, and the robustness of a powerful evolutionary optimization algorithm to increase the sensitivity of the minimum thickness measurement limit. Within the framework of this work, a self-calibration model is introduced, which takes into consideration the real industrial challenges such as the effect of wet-on-wet spray in the painting process. *Published by AIP Publishing*. [http://dx.doi.org/10.1063/1.4955407]

Due to their unique properties, terahertz waves attract more and more interest in real time quality control, especially for non-destructive inline thickness sensing during the production process. Besides others, terahertz radiation penetrates a wide variety of non-conductive materials, such as paint coatings in automotive and aircraft industry, ceramics, papers, glass-fiber reinforced plastics, and dielectric substrates. One of the promising industrial applications of terahertz technology is the vehicle paint quality control in the automotive industry. Conventional approaches for automotive paint thickness measurements such as the eddy current method, magnetic gauges, and the ultrasonic approach are limited due to the complexity of the multi-step painting process, where five layers of thin coatings, including zinc phosphate, e-coat, primer or filler, basecoat, and clearcoat, are deposited on the vehicle surface. New possibilities to overcome these restrictions have been shown by terahertz radiation. One of the first remarkable works in this field was published in 2007 by Yasuda *et al.*[1] They proposed a multiple regression analysis approach using the least-square fitting algorithm in order to decrease the minimum measurable paint film thickness for THz spectrometers. The introduced model has shown the great potential of THz technology by decreasing the minimum measurable film thickness from 108 $\mu$m down to 20 $\mu$m. Recently, Su *et al.*[2] extended the numerical parameter fitting method proposed by Yasuda *et al.* by integrating the etalon effect, the absorption and dispersion of materials in the simulation algorithm. However, they continued to use the deterministic least-square fitting to determine the thicknesses. Using the proposed approach, the minimum thickness was further decreased down to 18 $\mu$m for both single and multilayer automobile paints.[2] A second very interesting approach to investigate multilayer coatings using TDS-reflection and transmission geometries was presented recently by van Mechelen *et al.*[3] The main benefit of this model is the use of stratified dispersive model, which reduces the number of sought variables significantly, enabling a simultaneous extraction of the individual optical material parameters in multilayer structures.

In principle, the individual thicknesses of multi-layered coatings can be directly determined using time of flight measurements of ultrashort THz pulses. As shown in Figure 1, the absolute thickness of a layer $z$ can be determined from the phase information between two pulse echoes, which are reflected from its front and back surface. For optically thick samples, the reflected adjacent echoes are separated in time domain so that the individual thicknesses can be calculated directly from the time delay $\Delta t_z$ as follows:

$$d_z = \frac{c\Delta t_z}{2n_z \cos\theta_z}, \quad (1)$$

where $\theta_z$ is the angle of incidence and $n_z$ is the refractive index. The minimal thickness resolution $d_{min}$ with this model is limited by the terahertz pulse length $\Delta T$ and can be derived from Equation (1) as follows:

$$d_{min} = \frac{c\Delta T}{2n}. \quad (2)$$

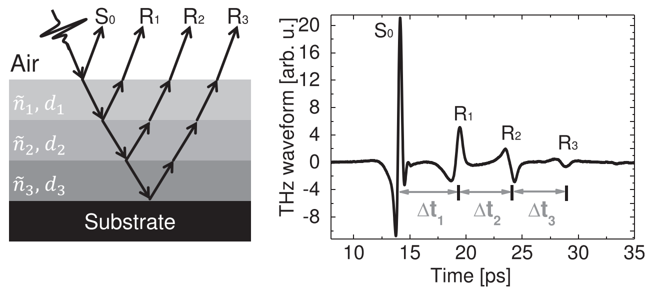


FIG. 1. Schematic representation of THz radiation interaction with a multi-layered structure (left) and the corresponding reflected THz waveform (right).

For commercially available pulsed terahertz systems, the minimal thickness resolution $d_{min}$ is estimated to be about 80 $\mu$m. However, the typical paint thickness for the automobile industry lies between 5 $\mu$m and 60 $\mu$m. For these thin films, the pulse echoes, which describe the boundaries of the single layers within the multilayer system, overlap in the time domain so that their time separation cannot be determined precisely. In order to resolve the thicknesses of the individual layers, a numerical regression approach has to be applied. In this work, an advanced regression approach is presented, which provides a high robustness even for very thin layers with complex search spaces, taking into consideration the real industrial challenges in the painting process.

The numerical regression approach consists of calibration, simulation, and optimization.The calibration deals with the extraction of the complex refractive index of all layers in the investigated sample. In a widely used classical calibration, the spectral optical material parameters of each individual coating are extracted from single layers in a dry state. However, the paint layers in real applications are not always deposited on dry coatings. The underlying coating can be dry, wet, or in an intermediate state, depending on the coating process. Consequently, the adjacent layers merge partly causing a change of the material parameters compared to the dry single layers. To circumvent this source of uncertainty, a self-calibration approach is developed to extract the material parameters of all layers from the sample at its final state, simultaneously. Hence, we take into consideration all effects that may occur during the painting process such as wet-on-wet spray. A stratified dispersive Debye model provides an accurate approximation for most automotive paints. The spectral permittivity function of each layer is given as

$$\epsilon(\omega)^z = \epsilon_\infty^z + \frac{\epsilon_s^z - \epsilon_\infty^z}{1 + i\omega\tau^z}, \tag{3}$$

where $\epsilon_s$, $\epsilon_\infty$, and $\tau$ are the static relative permittivity, the permittivity at infinite frequency, and the relaxation time, respectively. The variable $z$ is an integer index, which labels each discrete layer. In this case, the optimization parameters are $\epsilon_s$, $\epsilon_\infty$, and $\tau$. Figure 2 shows the obtained best graphical results after the thickness optimization for the same four-layer sample based on the classical calibration on single layers and the developed self-calibration approach. The graphical agreement demonstrates the effect of the wet-on-wet painting. Due to the change of the material parameters in the multi-layered sample compared to the single layers, the calibration on single layers is not able to simulate the effect of multiple reflections while the self-calibration model provides a very good graphical agreement.

The reflected waveform of arbitrary multilayer thin films $E_r(t)$ can be obtained using the inverse Fourier transformation $\mathscr{F}^{-1}$ according to the following formula:

$$E_r(t) = \mathscr{F}^{-1}[H(\omega)\mathscr{F}[E_0(t)]], \tag{4}$$

where $\mathscr{F}[E_0(t)]$ is the Fourier transform of the incident terahertz pulse, also known as reference pulse, and $H(\omega)$ is the transfer function. A suitable method to model multilayer structures was published in 1937 by Rouard,[4] which outperforms the classical transfer matrix method with a faster computation time and less memory allocation.[5,6] For the explanation of this model, let us take into account only the first coating that is deposited onto the substrate. The modeling process starts from this layer, which is first considered as a single layer with a simple transfer function $H_l(\omega)$

$$H_l(\omega) = r_{l-1,l} + \frac{t_{l-1,l}t_{l,l-1}r_{l,l+1}e^{\frac{2i\tilde{n}_l\omega d_l}{c}}}{1 - r_{l,l-1}r_{l,l+1}e^{\frac{2i\tilde{n}_l\omega d_l}{c}}}, \tag{5}$$

where $l$ denotes the layer index, $d_l$ is the coating thickness, $n_l$ is the complex spectral refractive index, $c$ is the speed of light in vacuum, and $t_{l,l+1}$ and $r_{l,l+1}$ are the Fresnel transmission coefficient and the Fresnel reflection coefficient, respectively. Now, we interpret the overlying layer $l-1$ again as a single layer with the simple transfer function like the previous layer $l$

$$H_{l-1}(\omega) = r_{l-2,l-1} + \frac{t_{l-2,l-1}t_{l-1,l-2}\boldsymbol{r}_{1-1,l}e^{i\beta}}{1 - r_{l-1,l-2}\boldsymbol{r}_{1-1,l}e^{i\beta}}, \tag{6}$$

where $\beta = \frac{2\tilde{n}_{l-1}\omega d_{l-1}}{c}$. So far, the two layers are interpreted as simple single layers. The idea of Rouard is to replace the Fresnel reflection coefficient in 6 with the complete transfer function of the adjacent layer $l$. So, we integrate the entire effects together, which provide a full description of the multilayer structure. The transfer function of a two-layer sample using the Rouard's method is then given by

$$H^2_{Total}(\omega) = r_{l-2,l-1} + \frac{t_{l-2,l-1}t_{l-1,l-2}\boldsymbol{H}_l(\omega)e^{i\beta}}{1 - r_{l-1,l-2}\boldsymbol{H}_l(\omega)e^{i\beta}}. \tag{7}$$

For an arbitrary number of layers, it is necessary to "follow the path back" from the substrate to the incident medium.

The fitting algorithm, also known as optimization algorithm, is a core step to determine the efficiency of the thickness

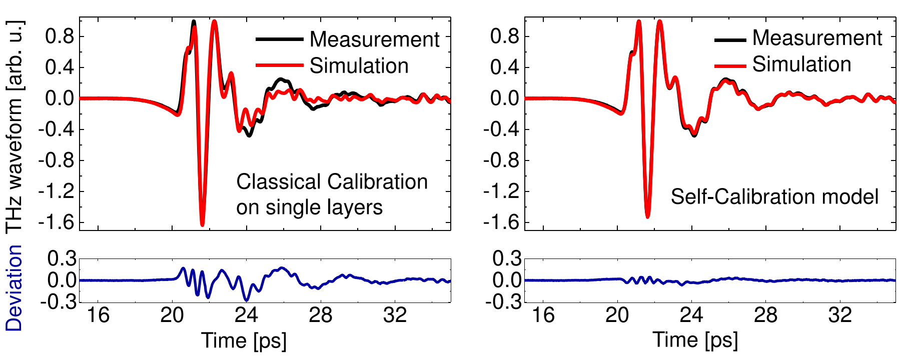


FIG. 2. A graphical comparison between obtained results after the optimization process: (left) the classical calibration on single layers and (right) the direct self-calibration model on the multilayer system.

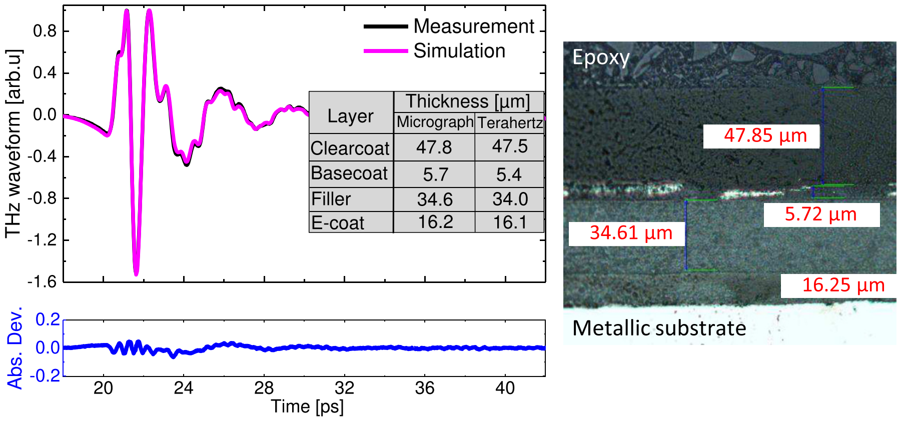


FIG. 3. The obtained graphical and numerical results after the optimization (left) as well as a comparison with the values obtained from the micrograph (right).

measurement. In contrast to the deterministic fitting algorithms, the stochastic optimization methods are increasingly applied for practical problems because they provide many benefits compared to deterministic methods. They are based on randomness, statistics, and probability, which increase the probability to find the global minimum, even for objective functions that are non-differentiable, non-continuous, nonlinear, noisy, and multi-dimensional, having a complex search space with several local minima. For this work, the stochastic differential evolution (DE) approach, which was introduced by Storn and Price in 1997,[7] has been selected. The stochastic optimization approach improves with its random population initialization, crossover, and mutation operations the minimum measurable thickness limit.

In this contribution, a variety of automotive specimens with different basecoat-colors, including up to four layers, has been investigated. A micrograph at the same position as THz measurement has been made for most samples to check the accuracy of the obtained results. The presented results are generated using a compact fiber-coupled spectrometer, which provides high stability for the rough industrial environment comprising strongly varying temperature, high humidity, and acoustic vibrations. The measurement time amounts to 1 s with simultaneous evaluation of data from the previous measurement. The evaluation uses general-purpose computing on graphics processing units and thanks to the developed highly parallelized algorithm that lasts less than 300 ms. Hence, the every-second-cycle for thickness measurement desired from the industry is fulfilled.

The first sample consists of four layers, including an electrocoat, a filler, a metallic silver basecoat containing aluminium flakes, and a clearcoat. Figure 3 shows the result of the regression procedure, and the measured thicknesses, which are compared to rated values obtained by a micrograph. The results obtained after the optimization process show a very good graphical agreement between the measured THz signal and the simulated one. Moreover, the measured values agree nicely with the thicknesses obtained with the micrograph. Furthermore, it can be seen that our approach was applied to resolve very thin individual layer thicknesses within multilayered paint samples down to 5 $\mu$m.

Besides samples on metallic substrates, coated carbon-fiber-reinforced polymers (CFRP) samples have also been investigated using the developed approach. An example of the achieved results for a two-layer sample is sketched in Fig. 4. The excellent graphical fit results and the close match of the thickness values demonstrate the robustness of the developed approach for the coated CFRP.

Figure 5 shows the obtained results for a two-layer sample on a dielectric substrate. Unlike conductive materials such as metal or CFRP, dielectric substrates provide a front reflection from the front side and back reflections from the back side of the substrate. Both reflections include information about the thickness values of the applied paint coatings. The very interesting result in the investigation of such samples is the ability to measure thick substrates with a thickness of about 3 mm and a very thin coating having a thickness of about 10 $\mu$m, simultaneously.

Besides the in-line thickness monitoring where a point measurement is required, the developed approach can also be applied for online and offline applications where a complete surface control is desired. Figure 6 shows an example

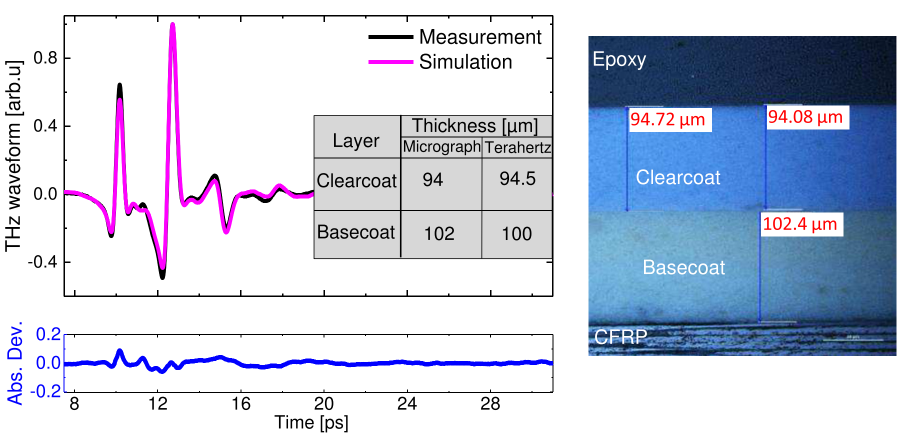


FIG. 4. The obtained graphical and numerical results after the optimization (left) as well as a comparison with the values obtained from the micrograph (right).

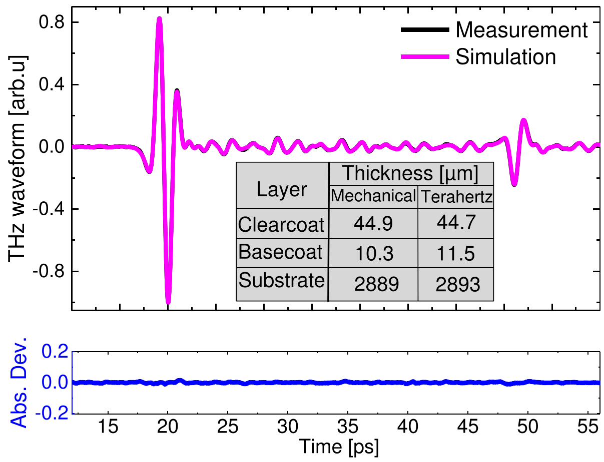


FIG. 5. The obtained graphical and numerical results after the optimization.

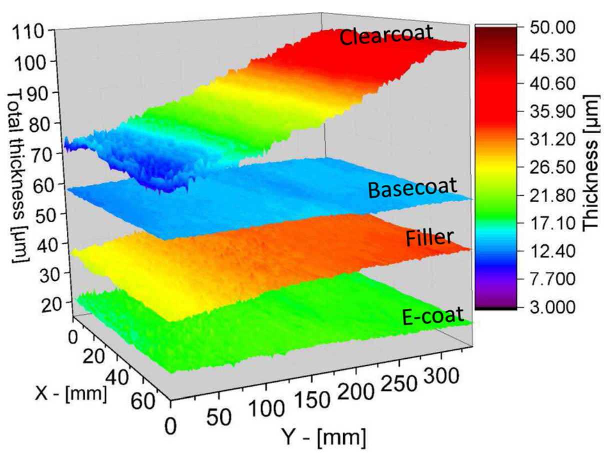


FIG. 6. A 3D thickness image of a four-layer specimen with two wedges in clearcoat and filler.

of a surface measurement on a steel sheet having the dimension 40 cm $\times$ 8 cm. The sample includes four paint coatings with wedges in the clearcoat and the filler. The total number of measured pixels with a resolution of 2 mm amounts 11 229, which results in an overall measurement time of 3.1 h, including raw data acquisition and data evaluation.

In summary, we have introduced an advanced regression procedure with a self-calibration model to measure individual automotive paint coatings in complex multi-layered structures, taking into consideration the real industrial challenges such as the effect of wet-on-wet spray in the painting process. Due to the high robustness of the proposed self-calibration method and the genetic optimization algorithms, the approach has been applied to resolve individual layer thicknesses within multi-layered paint samples down to 4 $\mu$m. Furthermore, we have shown that the developed approach is suitable to measure individual paint coatings on metallic substrates, on carbon-fiber-reinforced polymers, and on dielectric substrates with an accuracy, which is usually less than 1 $\mu$m. For imaging applications, the approach provides a complete surface control.

We would like to acknowledge financial support from the Innovation Center of Applied System Modeling in Kaiserslautern.

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