

A Reliable Method for Extraction of Material Parameters in Terahertz Time-Domain Spectroscopy

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Abstract—This paper introduces a novel method that allows fast and reliable extraction of material parameters in terahertz time-domain spectroscopy. This method could be applied for most of materials and requires neither simplifying assumptions nor samples of different thickness for the extraction. The presented extraction procedure operates either on truncated terahertz signals when temporal windowing is possible, or on full ones otherwise. Some experimental examples covering all practical cases are given. In particular, the extraction procedure treats the tedious case of samples for which internal reflections of the terahertz pulse slightly overlap.

I. INTRODUCTION

TERAHERTZ spectroscopy is a powerful technique for material studies over a wide range of frequency which extends from a few tens of gigahertz to a few terahertz. Using ultrashort electromagnetic pulses, terahertz spectroscopy gives access to the complex refractive index of a wide variety of materials, such as dielectrics [1], semiconductors [2], liquids [3], and superconductors [4]. For parameters extraction, the temporal profiles of the terahertz pulses are recorded twice, the first time without the sample to be characterized, and the second time with the sample. The recorded profiles correspond to the temporal dependence of the electric field associated with the terahertz pulses. Then, the ratio of their Fourier transforms gives the complex transmission coefficient of the sample as function of frequency. The final step of the study is concerned with the extraction of the complex index of refraction from the measured transmission coefficient, that is to solve an inverse problem. The resolution in the general case is a little tricky, especially because of mathematical convergence problem. For example, classical methods like the Newton–Raphson one [5], although efficient in many cases, become unstable when multiple reflections of the terahertz pulse in the material are not temporally separated.

We present, in this paper, a very reliable method of extraction which converges in all practical cases and which is comparable to the Newton–Raphson method in terms of computation speed, this latter method being the fastest when it converges [5]. In Section II, we describe the problem and we indicate the assumptions we have made. The next section presents the extraction method we have developed in the case of optically thick materials. The general case is treated

in Section IV. Some measures and parameters extractions, illustrating the different cases, are given in Section V. We close by discussing the obtained results and the validity of the assumptions that have been made.

II. DESCRIPTION OF THE PROBLEM AND ASSUMPTIONS

We consider a sample of thickness L , referred as 2, placed between two media 1 and 3 (see Fig. 1). We make the following assumptions.

- The sample is an homogeneous slide with two flat and parallel sides.
- The sample and the neighboring materials are magnetically isotropic without surface charges.
- The electromagnetic response of the three media is linear.

The terahertz beam goes through the sample from medium 1 to medium 3. We suppose that it is a plane wave impinging on the sample at normal incidence. We also assume that its polarization is linear and parallel to the optical axis of the device (in the case of an optical anisotropy of the sample). With the preceding assumptions, the spectral component at angular frequency ω of the electric field of the terahertz wave transmitted through the sample is written as

$$S_{\text{SAMPLE}}(\omega) = \eta(\omega) \cdot T_{12}(\omega) \cdot P_2(\omega, L) \cdot T_{23}(\omega) \cdot \underbrace{\sum_{k=0}^{+\infty} \{R_{23}(\omega) \cdot P_2^2(\omega, L) \cdot R_{21}(\omega)\}^k}_{\text{FP}(\omega)} \cdot E(\omega) \quad (1)$$

where $E(\omega)$ is the electric field of the terahertz wave emitted by the transmitting antenna,

$$R_{ab}(\omega) = \frac{\tilde{n}_a - \tilde{n}_b}{\tilde{n}_a + \tilde{n}_b} \quad (\text{reflection coefficient at the } a\text{--}b \text{ interface})$$

$$T_{ab}(\omega) = \frac{2\tilde{n}_a}{\tilde{n}_a + \tilde{n}_b} \quad (\text{transmission coefficient from medium } a \text{ to medium } b)$$

$$P_a(\omega, d) = \exp\left[-i\frac{\tilde{n}_a\omega d}{c}\right] \quad (\text{propagation coefficient in medium } a \text{ over a distance } d)$$

with $\tilde{n}_a = n_a - i\kappa_a$ being the complex refractive index of medium a which depends on the angular frequency ω . Media 1 and 3 can be stratified. All the reflection, transmission and propagation coefficients in these media are included in the term $\eta(\omega)$: we simply suppose that the echoes of the terahertz wave created in media 1 and 3 are negligible or occur on a time scale different from that corresponding to the signal of

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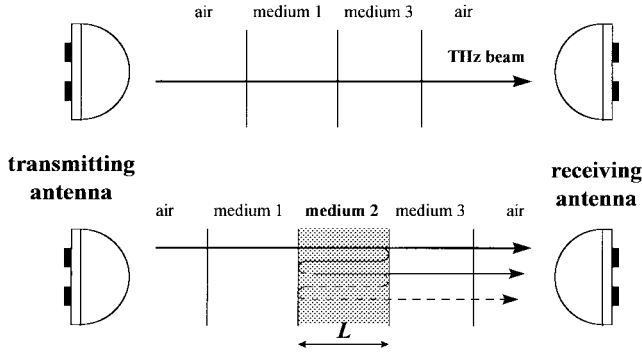


Fig. 1. Optical path of the terahertz beam between the transmitting and receiving antennas before (upper diagram) and after (lower diagram) sample insertion between the media 1 and 3.

interest. The backward and forward reflections in the sample (Fabry-Pérot effect) are represented by the summation $FP(\omega)$.

The spectral component of the electric field of the terahertz wave transmitted without the sample is given by

$$S_{REF}(\omega) = \eta(\omega) \cdot T_{13}(\omega) \cdot P_{air}(\omega, L) \cdot E(\omega). \quad (2)$$

The complex transmission coefficient $T(\omega)$ of the sample is obtained by dividing the signal $S_{SAMPLE}(\omega)$ recorded with the sample by $S_{REF}(\omega)$ recorded without the sample

$$T(\omega) = \frac{S_{SAMPLE}(\omega)}{S_{REF}(\omega)} = \frac{2\tilde{n}_2(\tilde{n}_1 + \tilde{n}_3)}{(\tilde{n}_2 + \tilde{n}_1)(\tilde{n}_2 + \tilde{n}_3)} \cdot \exp \left[-i(\tilde{n}_2 - \tilde{n}_{air}) \frac{\omega L}{c} \right] \cdot FP(\omega) \quad (3a)$$

with

$$FP(\omega) = \frac{1}{1 - \left(\frac{\tilde{n}_2 - \tilde{n}_1}{\tilde{n}_2 + \tilde{n}_1} \right) \left(\frac{\tilde{n}_2 - \tilde{n}_3}{\tilde{n}_2 + \tilde{n}_3} \right) \cdot \exp \left[-2i\tilde{n}_2 \frac{\omega L}{c} \right]}. \quad (3b)$$

In the case of the characterization of thin films made by a modification of the substrate surface (ionic implantation, chemical attack, etc.) one can show easily that the refractive index of the substrate \tilde{n}_{subst} has to be used instead of \tilde{n}_{air} in (3a). In fact, the sample takes the place of a part of the substrate. It is also the case for the characterization of hygroscopic materials placed in a cell filled with an inert substance which refractive index is \tilde{n}_{subst} . Here again, we suppose that the temporal echoes from the substrate do not interfere with the ones from the thin film. From now on, we will consider this latter case because it is more general ($\tilde{n}_{air} = 1$ represents actually a particular value of \tilde{n}_{subst}).

III. SOLUTION FOR OPTICALLY THICK SAMPLES

This is the most common case in practice and also the easiest to solve. We define an “optically thick sample” as a sample for which the echoes of the terahertz pulse, caused by multiple reflections into the sample, are temporally well separated. In other words, the terahertz signal drops to zero between two echoes. So it becomes possible to keep only the first directly transmitted terahertz pulse thanks to a temporal windowing. Thus, we do not have to take into account the

multiple reflections into the sample any more and consequently the term $FP(\omega)$ is removed from (3a). Nevertheless, there is no analytical solution to (3a) with $FP(\omega) = 1$, except if absorption can be considered as negligible. So, we have to solve (3a) with $FP(\omega) = 1$. In terms of computation speed, one of the most efficient methods to solve such a root finding problem is to use the Newton-Raphson method. Unfortunately, this method could diverge easily if we are looking for the roots of an oscillating function. That is precisely the case of the real and imaginary parts of $T(\omega)$, as function of n_2 and κ_2 . Nevertheless, the Newton-Raphson method is applicable but only if one starts the calculation with values of n_2 and κ_2 very close to the final solution.

Our method has no requirement on the starting values. Instead of searching for the zeros of $T(\omega) - T_{meas}(\omega)$, where $T(\omega)$ and $T_{meas}(\omega)$ are respectively the calculated and measured complex transmission coefficients, we use functions that are smooth and monotonous. The logarithm of $|T(\omega)|$ and the argument of $T(\omega)$ are functions that look like planes when plotted versus n_2 and κ_2 . Then we define the following error function

$$\delta(n_2, \kappa_2) = \delta\rho^2 + \delta\varphi^2 \quad (4a)$$

where

$$\delta\rho = \ln(|T(\omega)|) - \ln(|T_{meas}(\omega)|)$$

and

$$\delta\varphi = \arg(T(\omega)) - \arg(T_{meas}(\omega)). \quad (4b)$$

This error function is similar to a paraboloid (see Fig. 2), so its expression can be approximated by the general equation of a paraboloid [6]¹

$$\delta(n_2, \kappa_2) \equiv \delta(\vec{r}) \cong f(\vec{r}) = \frac{1}{2} \vec{r} \cdot \vec{A} \cdot \vec{r} - \vec{b} \cdot \vec{r} + c \quad (5)$$

where \vec{r} defines a position in the plane (n_2, κ_2) and \vec{A} , \vec{b} , and c are constant parameters. Let \vec{r}_0 be the vector corresponding to the minimum of the paraboloid ($\vec{\nabla} f(\vec{r}_0) = \vec{0}$). \vec{r}_0 is a good estimation of the solution of the equation $\delta(n_2, \kappa_2) = 0$ we are looking for. We derive from (5) and from $\vec{\nabla} f(\vec{r}_0) = \vec{0}$

$$\vec{A} \cdot \vec{r}_0 = \vec{b}. \quad (6)$$

Using (6), the gradient of the function $f(\vec{r})$ can be written as

$$\vec{\nabla} f(\vec{r}) = \vec{A} \cdot (\vec{r} - \vec{r}_0). \quad (7)$$

As a result, if we know the \vec{A} tensor and if we can calculate the gradient of $f(\vec{r})$, (7) gives us directly \vec{r}_0 . The solution is

$$\vec{r}_0 = \vec{r} - \vec{A}^{-1} \cdot \vec{\nabla} f(\vec{r}) \quad (8)$$

¹To find the value of the variables for which the function is minimum, it is quite common to approximate the function by its second order Taylor's series. In two-dimension, the approximated function is a paraboloid. For more details, see [6].

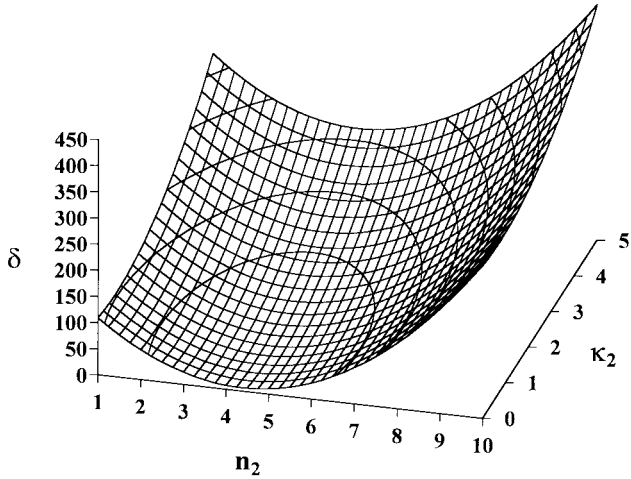


Fig. 2. Error function δ versus real and imaginary parts of the complex refractive index \bar{n}_2 for $\omega L/c = 3$, $|T_{\text{meas}}(\omega)| = 0.05$ and $\arg(T_{\text{meas}}(\omega)) = 10$ rad.

where \bar{A} is the tensor of second derivatives of the function $f(\vec{r})$ with respect to n_2 and κ_2 . But $\delta(\vec{r})$ is not perfectly a paraboloid, so \vec{r}_0 is only an approximate value of the minimum of the function $\delta(\vec{r})$. However, with successive iterations, using the above method at each step p , we will quickly converge to the solution. The following relation sums up this iterative procedure

$$\vec{r}_{p+1} = \vec{r}_p - \bar{A}^{-1} \cdot \vec{\nabla} \delta(\vec{r}_p) \quad (9)$$

with \vec{r}_p being the solution at the level of the p th iteration and where \bar{A}^{-1} is calculated at position \vec{r}_p .

Because the analytical expression of the error function $\delta(\vec{r})$ is quite simple and of low dimension, the calculation of the inverse Hessian matrix \bar{A}^{-1} can be made directly, either analytically or numerically, without requiring complex procedures like the ones involved in variable metric methods [6]. We consider that the extraction procedure is finished when $\delta\rho$ and $\delta\varphi$ are respectively at least ten times lower than the corresponding experimental uncertainties. These two latter uncertainties are determined by calculating the total differentials of $\delta\rho$ and $\delta\varphi$

$$\Delta\delta\rho = \frac{\Delta(|T_{\text{meas}}(\omega)|)}{|T_{\text{meas}}(\omega)|} + |\kappa_2 - \kappa_{\text{subst}}| \frac{\omega}{c} \cdot \Delta L \quad (10a)$$

$$\Delta\delta\varphi = \Delta(\arg(T_{\text{meas}}(\omega))) + |n_2 - n_{\text{subst}}| \frac{\omega}{c} \cdot \Delta L \quad (10b)$$

where $\Delta(|T_{\text{meas}}(\omega)|)$, $\Delta(\arg(T_{\text{meas}}(\omega)))$ and ΔL are, respectively, the uncertainties over the modulus and the argument of the measured transmission coefficient and over the thickness of the sample.

In order to validate this extraction procedure, we have tested it on a large number of simulated cases, with real and imaginary parts of the complex refractive index, of both the sample and the media 1 and 3, varying, respectively, from 1 to 10 and from 0 to 10. These values cover all practical cases. The extraction procedure always succeeds with an average number of iterations varying between 2 and 3.

At the extrema of the frequency range, the signal is noisy because of the dramatic reduction of its amplitude. However, as $\arg(T(\omega))$ is mainly proportional to the frequency, the relative noise on $\arg(T(\omega))$ is much smaller for the highest frequencies than for the lowest ones. Therefore, the extraction of n_2 and κ_2 over the full frequency range is performed from the highest frequency down to the lowest one. As n_2 and κ_2 are continuous functions of the frequency for any material, each pair of values is calculated using the previous one as starting point. Thus, the full extraction is very fast: for example, the determination of n_2 and κ_2 from 50 GHz up to 1.5 THz with a resolution of 5 GHz takes about 50 ms using a computer equipped with a Pentium 90 processor.

The direct calculation of $\arg(T_{\text{meas}}(\omega))$ from the temporal profiles of the recorded terahertz pulses gives a value varying between $-\pi$ and $+\pi$; $\arg(T_{\text{meas}}(\omega))$ shows then some artificial phase jumps. As the complex transmission coefficient $T(\omega)$ is a continuous function of the frequency for any material, the calculated values of $\arg(T_{\text{meas}}(\omega))$ are $2p\pi$ -shifted (p being an integer) each time the absolute difference of the values of $\arg(T_{\text{meas}}(\omega))$ for two consecutive frequencies exceeds π . Despite having made $\arg(T_{\text{meas}}(\omega))$ continuous, the excess of noise at low frequencies can produce a $2p\pi$ -shift of $\arg(T_{\text{meas}}(\omega))$ over the entire frequency range. This possible phase shift is eliminated using a linear approximation of $\arg(T_{\text{meas}}(\omega))$ derived in the middle frequency-range for which the level of noise is very low: $\arg(T_{\text{meas}}(\omega))$ is $2p\pi$ -shifted such as this linear approximation tends, as necessary, toward zero for $\omega = 0$.

IV. SOLUTION FOR THIN SAMPLES

When the samples are optically thin, terahertz pulse echoes get superimposed. If the signal is cut off at a nonzero value, it gives unexploitable results because the number of echoes to be taken into account is not an integer. The only solution consists in recording the signal over the entire time domain (i.e., until the signal goes down to zero), which at first glance seems to be restricting but is not in practice. In fact, only the characterization of slightly absorbing materials with refractive index very different from the ones of the neighboring materials raises problems. Indeed, in this case, the successive echoes of the terahertz signal exhibit an amplitude decreasing very slowly with time: therefore, one needs to record the whole signal on a very large temporal scale.

As the extraction procedure in the case of optically thick materials is reliable and fast, we have chosen to treat thin samples in a similar way and therefore we consider the Fabry-Pérot effect as a perturbation. We start with approximate values of n_2 and κ_2 , as described below, to estimate the value of function $\text{FP}(\omega)$. Dividing the measured complex transmission coefficient $T_{\text{meas}}(\omega)$ by this value gives a complex transmission coefficient with strongly reduced echoes. Then, the extraction procedure presented in Section III applies again to extract n_2 and κ_2 . Starting with a good estimation of n_2 and κ_2 and repeating the whole procedure described in this paragraph, one will converge to the solution after a few iterations. The principle of this new extraction procedure that takes into

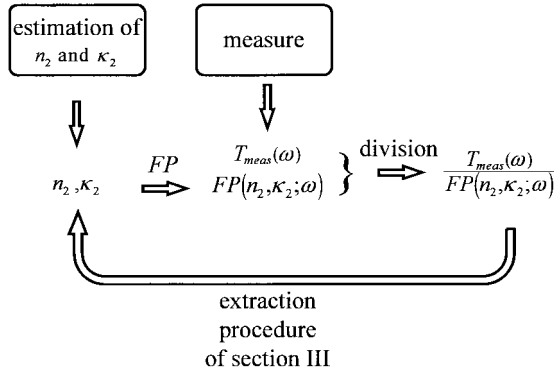


Fig. 3. Description of the extraction procedure in the case of thin samples.

account the Fabry–Pérot effect is depicted on Fig. 3. We have taken the same criterion of convergence than in Section III (i.e., when $\delta\rho$ and $\delta\varphi$ are respectively at least ten times lower than the corresponding experimental uncertainties).²

Let us now focus on the determination of the starting values of n_2 and κ_2 . In order to obtain a good estimation of these two values, we are looking for approximate analytical expressions of n_2 and κ_2 as function of the measured transmission coefficient $T_{\text{meas}}(\omega)$. The propagation in the sample introduces a dephasing which is mostly due to the difference ($n_2 - n_{\text{subst}}$) of refractive indices between the sample and the medium that has been substituted by the sample, while it is only slightly modified by both the Fabry–Pérot effect and the absorption. Therefore, we obtain a first analytical estimation of n_2

$$n_2 \cong n_{\text{subst}} + \frac{\arg(T_{\text{meas}}(\omega))}{\omega L/c}. \quad (11)$$

Besides depending on n_2, κ_2 is mainly dependent on $|T_{\text{meas}}(\omega)|$. Starting from the (3), it can be shown, after some quite long and tedious calculations, that κ_2 obeys the following relation

$$AX^3 + X \cong D \quad \text{with} \quad X = \exp\left[-\kappa_2 \frac{\omega L}{c}\right] \quad (12a)$$

$$A = \frac{(n_2 - n_1)(n_2 - n_3)}{(n_2 + n_1)(n_2 + n_3)} \cdot \cos\left(2n_2 \frac{\omega L}{c}\right) \quad (12b)$$

$$D = \frac{(n_2 + n_1)(n_2 + n_3)}{2n_2(n_1 + n_3)} \cdot \exp\left[-\kappa_{\text{subst}} \frac{\omega L}{c}\right] \cdot |T_{\text{meas}}(\omega)|. \quad (12c)$$

To obtain this result, we suppose that the media on both sides of the sample exhibit low absorption, therefore, we have neglected κ_1 and κ_3 in (3). We have also considered κ_2 is small enough to allow us to detect a signal in transmission. Thus, higher order terms in κ_2 in (3) can be neglected. For materials of very high absorption, that do not meet this requirement, terahertz spectroscopy in reflection is much more suitable [7].

²As in Section III, the experimental uncertainties are determined by calculating the total differentials of $\delta\rho$ and $\delta\varphi$. However, even if the calculation is simple, the expressions are much more complex than in relations 10 because of the term $\text{FP}(\omega)$ in (3a), and therefore, they are not reported here.

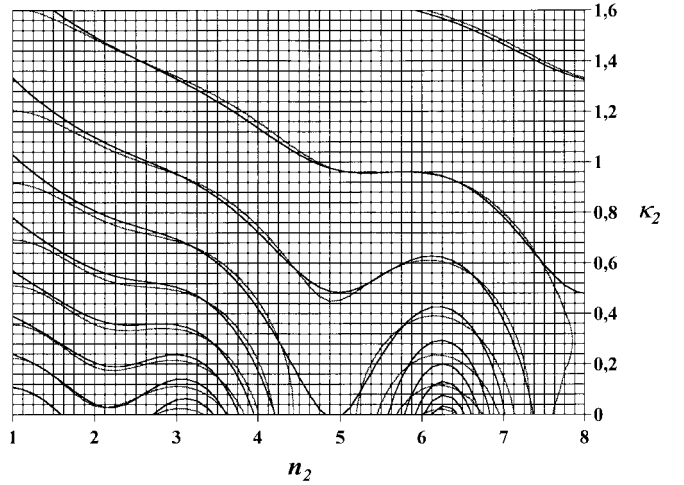


Fig. 4. Contour plots versus n_2 and κ_2 of the calculated moduli of both the rigorous (in black) and the approximate (in gray) complex transmission coefficients $T(\omega)$, for $\omega L/c = 1$ and $\bar{n}_1 = \bar{n}_3 = 1$. The represented contours correspond to values of $|T(\omega)|$ going from 0.1 to 0.9, by step of 0.1.

Using the Cardan method [8], (11a) can be solved easily. We show on Fig. 4 the contour plot versus n_2 and κ_2 of the calculated moduli of both the rigorous [see (3)] and the approximate [from (12)] transmission coefficients. As one can see, there is a very good agreement between the rigorous and approximate values of $|T(\omega)|$ over the full range of n_2 and κ_2 plotted here, this agreement becoming excellent when the difference between the refractive indices of the sample and the neighboring media is small. Let us notice that this range covers almost all the parameters of materials one can measure in practice. A similar agreement is also observed in the case of $\arg(T(\omega))$.

We have tested with success this method on a large number of simulated cases, with n_2 varying from 1 to 5 and κ_2 from 0 to 10 (i.e., the power absorption $\alpha_2 = 2\omega\kappa_2/c$ varying from 0 to more than 10^2 cm^{-1} for all the studied frequency band). This range of values corresponds to almost all the materials. Thanks to a precise estimation for the starting values of n_2 and κ_2 , the presented extraction procedure is not only highly reliable but also very rapid (taking the Fabry–Pérot effect into account only increases the execution time by a factor lower than 4). Finally, we would like to point out that this method, based on a perturbation theory, works even in cases for which the Fabry–Pérot effect modifies quite strongly the sample transmission coefficient.

V. MEASURES

A. The Setup

As our experimental setup is a common one, we refer the reader to [9] and [10] for a more complete description of the experiment. Terahertz pulses are produced by optical excitation of a LT-GaAs [11], [12] photoconductive switch with a self-mode-locked Ti:Sapphire laser. The collimation of the terahertz wave is achieved with a 20-mm-diameter hyperhemispherical lens fabricated in high-resistivity-silicon. The laser beam used to excite the transmitting antenna is

chopped at 1 kHz and a lock-in amplifier is used to improve the signal-to-noise ratio (SNR) of the detected current. The delay-line control, data acquisition and data processing are driven by a computer. For each sample, we make several measurements; taking their average enables us both to improve the accuracy and to estimate the uncertainties of the measurements.

The obtained signal to noise ratio of the spectra is typically 50 dB at the maximum of the spectrum, i.e., for a frequency between 300 and 400 GHz. Without considering the effect of the divergence of the terahertz beam (see Section VI), neither the uncertainty of the sample thickness, the measurement accuracies on the refractive index and the absorption (given by the measurement standard deviations), are respectively given by $\Delta\alpha$ [cm⁻¹] · L [mm] \approx 0.1–1 and $\Delta n \cdot L$ [mm] \approx 0.002–0.005. These relations are verified in the range 150–750 GHz, for a sample thickness varying from 10 μ m up to 1 cm. The accuracy on the absorption is limited by the measurement dynamic (50 dB), which is itself mainly limited by the noise of the transmitting antenna in our set up. The uncertainty of $\arg(T_{\text{meas}}(\omega))$ is also mainly due to the noise on the spectra of the terahertz pulses, this first uncertainty being responsible of the accuracy on the refractive index.

Although having characterized a large number of materials, we have chosen to present three typical cases in this paper.

B. Thick Samples

As first example, we are interested in the case of an optically thick material. The sample is a 1.07-mm-thick BK 7 glass slide. The temporal profiles of the terahertz pulse measured with and without the sample are shown in Fig. 5. The terahertz signal transmitted by the sample falls down to zero for times larger to 7 ps after the first pulse. Thanks to the sample thickness, the first echo is rejected far (\sim 12 ps) from the first pulse. Thus, the extraction could be rigorously done with the data values recorded only up to 10 ps after the first pulse. The refractive index and the power absorption of BK 7 are plotted in Fig. 6. BK 7 presents no dispersion on the entire studied frequency band; its refractive index is almost constant. However, its absorption increases rapidly with the frequency and reaches very high values. This makes usual glasses like BK 7 unsuitable to be used as transmission windows for terahertz spectroscopy.

C. Thin Films

We present here, as an example of thin film characterization, some results for porous silicon. As the index of this material depends strongly on its porosity, it can vary over a wide range of values and therefore, in the present example, it is completely unknown. This fully justifies to use the method developed in this paper. The sample has been made from a p-doped silicon wafer with a 380 μ m thickness and a 30 Ω ·cm resistivity. The porous silicon layer obtained by anodic attack in an HF acid solution is 43 μ m thick and its porosity (percentage of the volume of the void space to the total volume) is 55%. The dashed and solid curves of Fig. 7 have been respectively measured before and after the porous layer fabrication. First, the silicon substrate before processing has been characterized

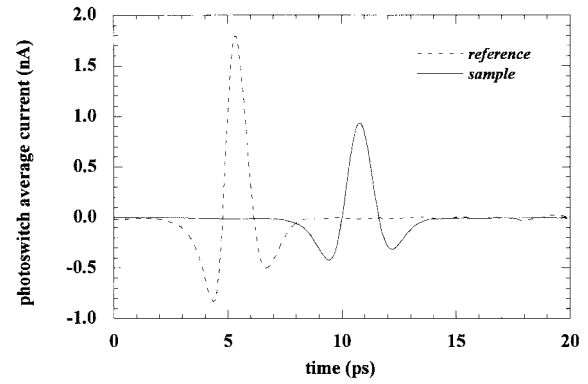


Fig. 5. Measured terahertz pulse for both reference (dashed curve) and 1.07-mm-thick BK 7 sample (solid curve).

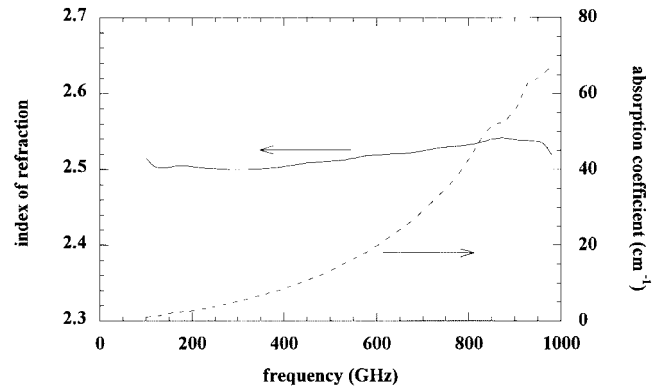


Fig. 6. Refractive index (solid curve) and power absorption in cm⁻¹ (dashed curve) of BK 7 extracted from the temporal curves plotted in Fig. 5.

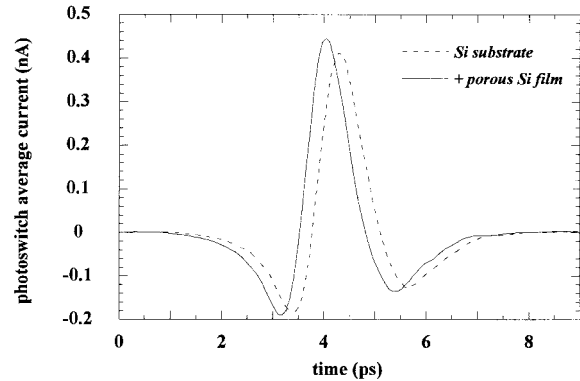


Fig. 7. Measured terahertz pulse for both reference (dashed curve) and thin film of porous silicon (solid curve) with a thickness of 43 μ m made from a 380 μ m thick p-type doped silicon wafer (which material parameters are plotted in Fig. 8).

in order to get its material parameters. They are absolutely needed to extract the parameters of the porous layer. As shown on Fig. 8, both the refractive index (3.4) and the absorption (4.7 cm⁻¹) of the silicon substrate vary not much.

Although multiple reflections in the porous layer exist, they are absolutely not detectable on the temporal curve partly because of the small thickness of the porous layer. Nevertheless, if we make two extractions, taking and not taking the Fabry-Pérot effect into account, the results are

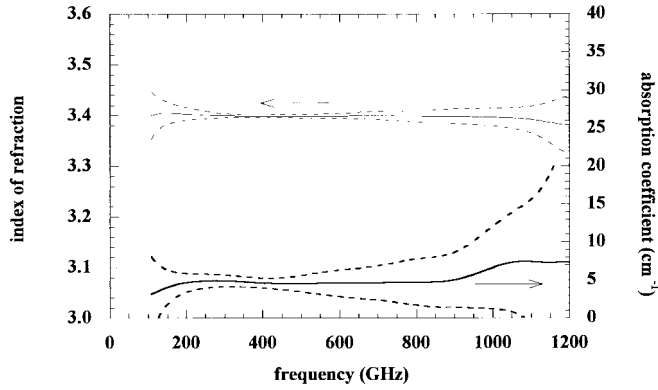


Fig. 8. Refractive index (thin curves) and power absorption in cm^{-1} (thick curves) for 30 $\Omega\cdot\text{cm}$ p-type doped silicon. The solid curves represent the mean value obtained by a 10-measure averaging. The mean values of n_2 and κ_2 plus or minus the measure standard deviation are shown as dashed curves.

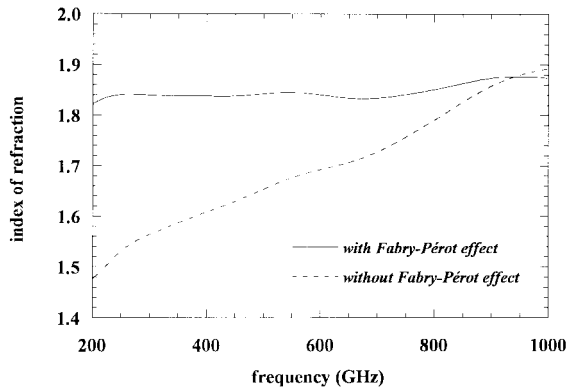


Fig. 9. Refractive index for porous silicon with a porosity of 55%. The right values of the refractive index are plotted as solid curve and have been obtained by taking the Fabry-Pérot effect into account. The dashed curve represents the refractive index extracted without taking the echoes of the terahertz pulse into account.

completely different. Among other things, the refractive index, obtained without echo contribution, increases with the frequency while the true refractive index is nearly constant (see Fig. 9). We made also some measurements with much thinner porous silicon layers, down to a thickness of 10.2 μm . For this latter thickness, the refractive index of the layer is determined with a precision of 0.2–0.3 over the 200–600-GHz frequency band, as derived from the measured standard deviation. We also observed a great difference between the two extractions concerning the absorption (see Fig. 10). Indeed, the absorption calculated with the wrong extraction procedure becomes negative (i.e., non physical) for the high frequencies. But, because of the small thickness and absorption of the layer, even the true absorption value is obtained with a poor accuracy (uncertainty $\sim 100\%$): only its order of magnitude has to be considered here. To get a better precision on both refractive index and absorption, one has to use thicker layers following the relations given in Section V-A.

Even undetectable effects on the temporal curves can radically modify the results. Although it is obvious, the presence of echoes must never be determined by an analysis on temporal curves but only from elementary considerations on the physical

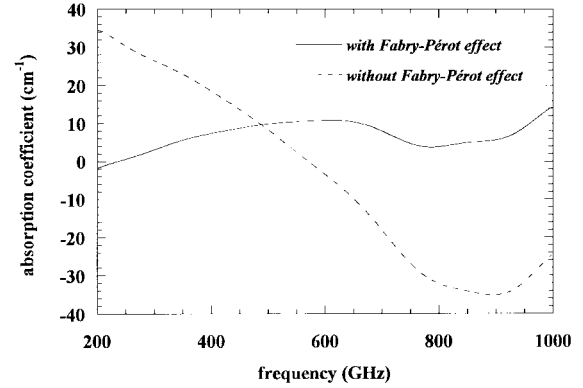


Fig. 10. Power absorption in cm^{-1} of a thin film of porous silicon (which refractive index is represented in Fig. 9). The right values are plotted as solid curve and have been obtained by taking the Fabry-Pérot effect into account. The dashed curve represents the absorption extracted without taking the echoes of the terahertz pulse into account.

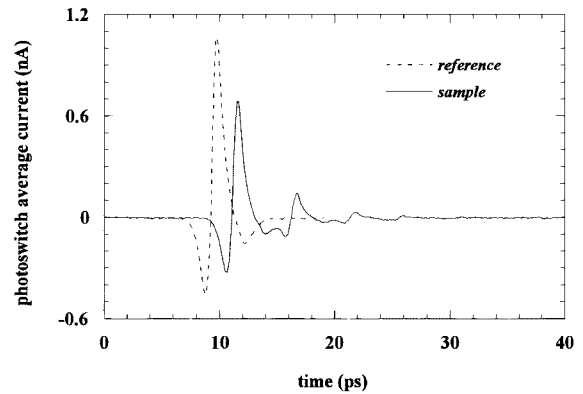


Fig. 11. Measured terahertz pulse for both reference (dashed curve) and 8.4 $\Omega\cdot\text{cm}$, 225- μm -thick p-type doped silicon (solid curve).

characteristics of the sample (mainly its optical thickness). We can notice that the amplitude of terahertz pulse obtained with the sample in presence of porous layer is lightly larger than the one obtained prior to the fabrication of the porous layer. With a refractive index in between the air and substrate ones, the porous layer serves as an antireflection coating.

D. Samples of Thickness Halfway Between

In contrast, the following example corresponds to the typical case for which no straightforward solution exists. Temporal windowing can not be used because of echoes overlap. Moreover, the sample thickness is too large and consequently a simplification of (3) is not possible. Thus, the whole temporal signal must be recorded. Fig. 11 shows a measure obtained with on a n-doped silicon wafer of a 225- μm thickness and a 8.4 $\Omega\cdot\text{cm}$ resistivity. On the terahertz signal measured with the sample (solid curve), one distinguishes three echoes of the terahertz pulse, the fourth one being under the noise floor. This rapid decrease of the echoes amplitude is mainly due to weak Fresnel reflection. The refractive index of the sample is plotted in Fig. 12 and is nearly constant over the complete frequency band, which extends from 50 GHz to 1.5 THz. Its value is 3.4 and its standard deviation, calculated from six measures,

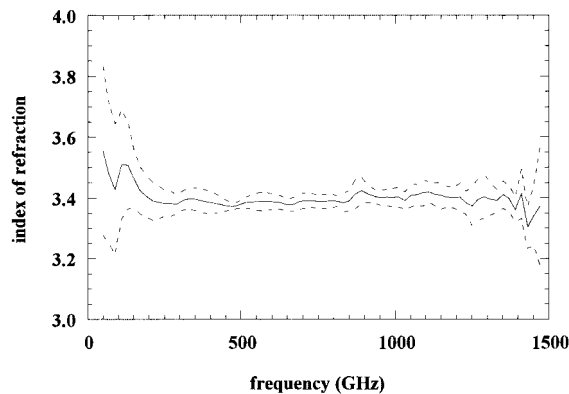


Fig. 12. Refractive index of 8.4 Ω -cm p-type doped silicon extracted from the temporal curves plotted in Fig. 11. The solid curve represent the mean value obtained by a four-measure averaging. The mean values of the refractive index plus or minus the measure standard deviation are shown as dashed curves.

is less than 0.05 between 250 and 1400 GHz. One can notice that the standard deviation is periodically slightly reduced for frequencies corresponding to Fabry–Pérot transmission peaks. The absorption (a few cm^{-1}), not represented here, has the same rough estimate as the setup sensitivity.

VI. DISCUSSION

Initially, we have made a few assumptions. To our knowledge and until now, the first three assumptions (homogeneous plate slide with a linear electromagnetic response and which is magnetically isotropic), concerning the sample and the neighboring media, have been satisfied for all the materials that have been characterized by terahertz time-domain spectroscopy. In contrast, the plane-wave hypothesis is not entirely satisfied. Especially for the lowest frequencies, the divergence of the terahertz beam is too large and the beam can not be considered as a plane wave when it goes through the sample. One of the ways used to partially solve this problem is to focus the terahertz beam with off-axis parabolic mirrors and to place the sample at the waist location [1], but it makes optical adjustments tricky. We have chosen to use collimation lenses of large size in order to limit the divergence of the terahertz beam for lower frequencies. In the case of our experimental setup, Jepsen *et al.* [13] have shown that the terahertz beam can be represented by a Gaussian beam emitted from a circular aperture equal to the diameter of the collimation lens of the transmitting antenna. Using Gaussian optics we have calculated that the divergence of our terahertz beam is lower than 50 mrad for frequencies higher than 200 GHz. But, for lower frequencies, a great care is necessary to interpret the results.

VII. CONCLUSION

We have presented a novel method for the extraction of material parameters in terahertz time-domain spectroscopy. This general method, which applies to all practical cases, is not only highly reliable but also very fast, allowing a real-time extraction. We have demonstrated its validity on three concrete examples. In the case of thin films, we stress

that the Fabry–Pérot effect has to be taken into account, even if no temporal signatures of this effect are seen in the recorded signal. The next step to go toward a full extraction procedure will be concerned with a rigorous determination of the uncertainties on both the real and imaginary parts of the refractive index. Mainly, the influence of the divergence of the terahertz beam should be treated.

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