Removal of Spectral Distortion Due to Echo for Ultrashort THz Pulses Propagating Through Multilayer Structures with Thick Substrate

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Abstract

Given the wide range of applications of time-domain spectroscopy, and particularly THz time-domain spectroscopy, the modelling of a probe pulse propagating through a multilayered structure is often required. Due to the fact that the multilayers are usually grown on a substrate much thicker than the other layers, the transmission of a probe pulse includes a series of echo pulses caused by the multiple reflections at the substrate interfaces over a long time. However, experiments often measure only a small time window and construct the transmitted spectrum only from the first transmitted pulse. Due to the fact that typical substrates lead to times of crossing comparable to the spectral bandwidth, the first transmitted pulse’s spectrum and the full transmitted spectrum can importantly differ. It is therefore important to theoretically model the transmission without the echo, to be able to make a direct comparison with experimental results. Here, we propose a method to elegantly and easily theoretically remove the echo from the transmission spectrum. The spectrum of the transmitted pulse without echo will be produced analytically without additional numerical steps.

Keywords Multilayer · Terahertz time-domain spectroscopy (THz-TDS) · Transfer matrix · Light propagation · Transmission and absorption

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1 Introduction

In the field of material sciences, researchers are mostly aiming at developing new kinds of materials with specific or customized characteristics using newly designed complex structures. These complex structures are usually multilayered system [1, 2]. These specially tailored multilayers are key components to many different devices that have been widely used in modern technology [3]. They are also very important components of photodetectors, transistors, sensors, photo-voltaic cells [4] as well as THz emitters [5–7]. To study the properties of these specially designed multilayers, an electromagnetic probe in different range of frequencies (for example, the optical range, RF range, THz range) is usually used. Different probes give access to, for instance, thermal properties of the materials, such as the electron-phonon and phonon-phonon interactions [8, 9] as well as the transport [10–13].

However, the thickness of the materials used to construct these kinds of multilayers are usually down to nanometer scale. Hence, to hold and stabilize these materials, they are generally grown on another layer, called substrate, with a thickness much larger than the active layers. When performing time-domain THz spectroscopy (THz-TDS) [14–16] on a multilayer, a THz probe pulse is sent through it. The transmitted pulse is not measured on a spectrometer, but the time profile of the field is directly probed. When the multilayer is held on a substrate, as the pulse propagates through the whole system, echoes are produced by the air-substrate and substrate-active multilayer interface reflections. Experimentally only the first transmitted pulse is usually relevant, and very often the following echo pulses are not analysed and not even measured. These samples, when the main transmitted pulse and the echoes are clearly separable, are usually called optically thick samples and the way of ignore measuring the echo information is generally referred as temporal windowing [17].

However standard theoretical approaches to the propagation of electromagnetic waves through multilayers do not distinguish those pulses and produce the full spectrum of the transmitted radiation [18–20]. This is usually not a problem, if the time it takes the pulse to traverse the substrate is comparable with the pulse time-width: in this case the first transmitted pulse and echoes overlap temporally and are measured together. On the other hand if the substrate traverse time is considerably longer than the pulse width, the echoes are not recorded and the full theoretical spectrum is not directly comparable with experimental results that measure only the first transmitted pulse. Therefore any theoretical treatment cannot overlook this issue and must provide the spectrum of the first pulse only and not that of the full train of pulses.

A straightforward way to remove the echoes is by doing additional Fourier transforms and time filtering steps. However, the time filtering step may cause unwanted losses in spectral features and a distortion of the spectrum in certain situations. Theoretically, such process also can result only in numerical data, even when analytical expressions could be available and desirable. ‘Adjacent averaging’ is another technique commonly used to address these issues [21]. However, this simple way may lead to the loss of spectrum details. A further commonly used approach to remove the echo in THz-TDS is the de-convolution algorithm [21, 22]. This is commonly used to remove echo pulses from experimental results; however it can achieve at most a reduction of the effect of the echoes rather than truly removing them. Similarly to
the other used algorithms, the de-convolution algorithm cannot be used to construct usable analytic expressions. Another approach is to simply assume the substrate as semi-infinite. Even if it is possible to construct analytical expressions in this case, the important change in the geometry leads to unpredictable errors in the spectrum [18].

To our knowledge, there have been no attempt at addressing the problem of the removal of the echo analytically in real geometries. Notice that for continuous wave (CW) excitations, echoes are part of the signal and should not be removed. As we will show later, removing the echo leads to the disappearance of periodic peaks and valleys in the spectrum of the transmission. These features are often absent even in CW experiments, yet for very different reasons. The propagation through the substrate might be incoherent, and accounting for this effect allows for a spectrum that more closely reproduces CW experiments [19, 23–25]. Different authors attempt some suitable averaging procedure to obtain CW spectra including incoherence [24, 26]. We however warn that, even if removing the echo and describing the incoherence of transmission through the substrate modify the spectrum in ways that might look qualitatively similar at a first glance, the two physical effects are different and, therefore, the resulting spectra are not the same.

In the simulation of THz-TDS for different purposes such as material characterization and material properties extraction, the traditional Transfer Matrix Method (TMM) is often used as the theoretical basis [17, 27–32]. We provide here a modified TMM with an direct analytical expression to remove the echo which can be applied to the simulations and provide more reliable calculations when dealing with optically thick samples in THz-TDS experiments. As shown in Fig. 1, rather than solving the transmission through the whole multilayer, we first calculate the transmission through the substrate and analytically extract the first pulse that crosses the substrate/active layers interface, while discarding the further propagation that will give rise to echo pulses. We then propagate the mentioned pulse through the active multilayer (now including all multiple reflections) to obtain the transmitted wave. Our method provides an elegant analytical expression of the final transmitted pulse,

![Fig. 1](image-url) The comparison of the traditional removal of the echo method and our method
without the need of avoidable numerical steps, as well as being numerically stable over a huge range of layers thicknesses.

2 Recapitulation of TMM

2.1 Single Layer Treatment

Before considering the echo removal for the whole multilayer system, let us recapitulate the standard transfer matrix approach. We begin with a single layer treatment with orthogonal incidence. As the field within one layer can be represented as the superposition of a left-propagating and right-propagating wave \( k = \omega \sqrt{\mu \varepsilon} \). So that if we consider the Fourier components of the time Fourier transform (FT), then the general solution can be written as,

\[
\begin{bmatrix}
E(\omega, z) \\
H(\omega, z)
\end{bmatrix}
= \begin{bmatrix}
 e^{i\omega \sqrt{\varepsilon \mu} z} & e^{-i\omega \sqrt{\varepsilon \mu} z} \\
\sqrt{\frac{\mu}{\varepsilon}} e^{i\omega \sqrt{\varepsilon \mu} z} & -\sqrt{\frac{\varepsilon}{\mu}} e^{-i\omega \sqrt{\varepsilon \mu} z}
\end{bmatrix}
\begin{bmatrix}
f^> [\omega] \\
f^< [\omega]
\end{bmatrix}
\]  \( (1) \)

and \( f^> \) and \( f^< \) represent the amplitude of the right and left moving waves respectively. For brevity we will write the Fourier transformed equation (1) as,

\[
\tilde{F}(\omega, z) = \bar{a}[\omega, z] \tilde{f}[\omega] .
\]  \( (2) \)

so that for a single layer,

\[
\tilde{F}[d] = M[d] \tilde{F}[0]
\]  \( (3) \)

with

\[
M[d] = \bar{a}[d] (\bar{a}[0])^{-1} = \begin{bmatrix}
\cos(\omega \sqrt{\varepsilon \mu} d) & i \sqrt{\frac{\mu}{\varepsilon}} \sin(\omega \sqrt{\varepsilon \mu} d) \\
i \sqrt{\frac{\varepsilon}{\mu}} \sin(\omega \sqrt{\varepsilon \mu} d) & \cos(\omega \sqrt{\varepsilon \mu} d)
\end{bmatrix} .
\]  \( (4) \)

2.2 Multilayer Treatment

The transmission across a multilayer is computed requiring that the fields at the interfaces have to be continuous. We assume the cases when the multilayer is sandwiched by air or vacuum, as shown in Fig. 2, since it is the most common case (the formulas are easily generalised). We will use subscripts to denote matrix and vector properties belonging to a given layer. In particular, the semi-infinite left air layer will be denoted with 0, the layers of the multilayer will be denoted with increasing numbers from left to right and the semi-infinite air layer on the right will be denoted with \( \infty \). A local axis for the \( z \) coordinate in each layer, with origin on the left surface (implying that the coordinate of the right surface will be the thickness of the layer) will be used. The only exception is the semi-infinite left air layer where the origin is set at the right surface (since the other surface is at \( -\infty \)). Figure 2 shows a general case of fields within a multilayer system.
By enforcing field continuity at the interfaces of the multilayer, we can obtain all the fields and the amplitudes of the propagating waves. In particular we obtain

\[
\tilde{f}_\infty = (a_{\infty}[0])^{-1} \left( \prod_{j=N}^{1} M_j[d_j] \right) a_0[0] \tilde{f}_0 = T_{[0,\infty]} \tilde{f}_0
\]

where we draw the attention to the inverse order in the multiplication. And the \( T_{[0,\infty]} \) matrix here is the transfer matrix of the whole multilayer system.

### 2.3 Transmission and Reflection

A transmission experiment is easily modelled by requiring that there is no left-propagating wave in the rightmost air layer. This is achieved by solving the system

\[
\begin{bmatrix}
    f_\infty^> \\
    f_0^<
\end{bmatrix} = T_{[0,\infty]} \begin{bmatrix}
    f_0^>
    f_0^<
\end{bmatrix},
\]

where \( f_0^> \) represents the incoming, \( f_0^< \) the reflected, and \( f_\infty^> \) the transmitted wave respectively. The transmission coefficient \( t \) is defined as the ratio between the amplitude of the electric field due to the right going wave in the right air layer and the amplitude of the electric field due to the right going wave in the left air layer. The reflection \( r \) coefficient is analogously defined as the ratio between the amplitude of the electric field due to the left going wave in the left air layer and the amplitude of the electric field due to the right going wave in the left air layer[33]. In the considered case when the multilayer is surrounded by air on both sides the two coefficients simplify to the ratio of the corresponding amplitudes of the waves as

\[
t_{[0,\infty]} = \frac{f_\infty^>}{f_0^>} = \frac{T_{[0,\infty],11} T_{[0,\infty],22} - T_{[0,\infty],12} T_{[0,\infty],21}}{T_{[0,\infty],22}}
\]
where the further subscripts represent matrix elements. Thus, we are able to write out the transmitted and reflected wave as,

\[ f_{\text{\(\text{\(\rightarrow\)\)}}}^{\infty} = t_{[0,\infty]} \cdot f_{\text{\(\text{\(\leftarrow\)\)}}}^{0}, \]

and

\[ f_{0}^{\text{\(\text{\(\leftarrow\)\)}}} = r_{[0,\infty]} \cdot f_{\text{\(\text{\(\rightarrow\)\)}}}^{\infty}. \]

### 3 Removal of the Echo Within the Substrate in the Transmission

For the removal of the echo, we consider a sample grown on a layer, the substrate, much thicker than the other ones. We will use the subscript \(S\) for the substrate layer, and let the layer indices start at 1 for the first layer after the substrate.

To remove the echo, we first need to identify the first pulse travelling through the substrate, triggered by the incoming pulse on the left-hand air side and exclude the wave reflected at the substrate/layer 1 and all its multiple reflections. This can be simply constructed by imagining a sample formed only by the air/substrate interface. In that configuration the right-propagating wave in the substrate can be written as,

\[ f_{S}^{\text{\(\text{\(\rightarrow\)\)}}} = t_{[0,S]} \cdot f_{\text{\(\text{\(\leftarrow\)\)}}}^{0}. \]

where \(t_{[0,S]}\) is calculated from Eq. 7 with the transfer matrix \(T_{[0,S]}\) constructed as

\[ T_{[0,S]} = (a_{S}[0])^{-1} a_{0}[0]. \]

giving

\[ t_{[0,S]} = \frac{T_{[0,S],11} T_{[0,S],22} - T_{[0,S],12} T_{[0,S],21}}{T_{[0,S],22}}. \]

Notice that \(f_{S}^{\text{\(\text{\(\rightarrow\)\)}}}\) indeed is the first propagating pulse through the substrate (excluding the reflection pulse and all the subsequent ones due to multiple reflections within the substrate) even when the full sample is considered.

Such pulse will travel through the substrate and reach the interface with the rest of the sample. At that interface it will then undergo reflection, as well as propagation through the active part of the sample (inside which we need to consider multiple reflections). We can treat \(f_{S}^{\text{\(\text{\(\rightarrow\)\)}}}\) as an incoming wave for the rest of the multilayer and solve

\[
\begin{bmatrix}
 f_{\text{\(\text{\(\rightarrow\)\)}}}^{\infty, \text{\(\text{\(\text{\(\leftarrow\)\)}}\)} = \text{\(\text{\(\text{\(\text{\(\rightarrow\)\)}}\)}\)}}^{\text{\(\text{\(\text{\(\leftarrow\)\)}}\)}}^{\text{\(\text{\(\text{\(\rightarrow\)\)}}\)}} & f_{S}^{\text{\(\text{\(\rightarrow\)\)}}}\end{bmatrix} = T_{[S,\infty]} \begin{bmatrix}
 t_{[0,S]} \cdot f_{\text{\(\text{\(\leftarrow\)\)}}}^{0} & f_{S}^{\text{\(\text{\(\rightarrow\)\)}}} & \text{\(\text{\(\text{\(\text{\(\leftarrow\)\)}}\)}}\}
\end{bmatrix}
\]

where the transfer matrix \(T_{[S,\infty]}\) is

\[ T_{[S,\infty]} = (a_{\infty}[0])^{-1} \left( \prod_{j=N}^{1} M_{j}[d_{j}] \right) a_{S}[d_{S}]. \]
where we remind that $j = 1$ now refers to the first layer after the substrate. As we are interested only in the transmitted pulse, we obtain the overall transmission coefficient for the no-echo case as

$$t_{0,[0,\infty]}^{\text{no echo}} = t_{[0,S]}[t_{S,[S,\infty]},]$$

where

$$t_{S,[S,\infty]} = \frac{T_{S,[S,\infty]},12 T_{S,[S,\infty]},21 - T_{S,[S,\infty]},11 T_{S,[S,\infty]},22}}{T_{S,[S,\infty],22}}.$$  

### 4 Results and Discussion

We now show comparison of numerical results using the technique introduced above and experimental results for a THz wave propagating through a multilayer composed as Quartz (1 mm)/Pt (2 nm)/NiFe (3 nm). Let us first clarify that we do not measure the incoming pulse directly. Instead we perform a measurement without sample. We reconstruct the input wave (to be used for the TMM) by measuring the transmitted pulse without sample, and theoretically back-propagating it through air with same thickness as the sample. That is, in principle, not the real pulse that impinges on the sample: both the propagation through the optics after the sample and the detector responses should be removed as well (to be then added back when calculating the transmitted wave). We, however, assume these responses as both linear and scalar (meaning that they commute with the transfer matrix through the sample). Within this approximation (and thanks to the fact that the response of the sample is linear) it is not necessary to remove and then add optics’ and detector’s responses: the TMM can be applied directly to the partially reconstructed impinging pulse. We will show below how this approximation works extremely well in our case. Going beyond this approximation is possible, but it is not relevant to the main message of this work. The frequency-dependent material properties (refractive index $n$ and extinction coefficient $k$) used in the simulation for Quartz and NiFe were extracted from the experiment. The dielectric constants for Pt were calculated using the Drude model with plasma frequency 6.0 eV and damping frequency 0.21 eV [34].

Figure 3a shows the experimental transmitted wave over a time window of over 40 ps. One can clearly observe the presence of an echo pulse with a time delay of $\sim 13$ ps. The same figure shows the theoretical transmitted pulse using the full TMM as well. Both the main transmitted pulse and the echo are accurately reproduced by the TMM. The spectra are, as expected, in very close agreement as well. The spectrum shows peaks and valley due to the effect of multiple reflections in the substrate (Fig. 3c).

However the situation reported in Fig. 3a is seldom reproduced in typical experiments, where much smaller time windows (not containing the echo pulses) are measured. To emulate that case, we select a 8-ps time window around the transmitted experimental pulse (represented by the grey area in Fig. 3b). For consistency we perform the same time windowing on the incoming pulse (not shown). When the 8-ps time window profile is used to calculate the spectrum (Fig. 3c), the latter is considerably altered compared to the original one (see Fig. 3b). It is clear that an accurate comparison of standard TMM and the experiment is no longer possible.
might be tempting to execute a convolution with a gaussian to smoothen the spectral oscillations, but that has no physical meaning and does not reproduce the correct spectrum.

We apply the method introduced in this work to remove the echo from the theoretical spectrum. We assume that the incoming pulse (actually the pulse in the absence of sample) is also measured over the same 8-ps time window. We then propagate this time windowed incoming signal and theoretically remove the echo. While the bare TMM and the time windowed experimental spectrum were extremely different, the theoretical transmission obtained removing the echo (red line) is instead in excellent agreement with the experimental data both in time (Fig. 3b) and frequency domain (Fig. 3d).

The above experiment and theoretical simulations are all done in the case of a thick substrate (1 mm), which is usually called ‘optically thick samples’. For these samples, temporal windowing is acceptable, and the removal of the echoes does not lead to information losses. However, for ‘optically thin’ samples, removing the echo experimentally by temporal windowing will cause loss of information in the spectrum due to the fact that echoes will interfere with the main pulse. We remind that a sample can be considered ‘optically thick’ if twice the time it takes a pulse to traverse it is
sufficiently longer than the time-width of the pulse itself. As an example of ‘optically thin’ samples, we compare experiments and theory for the case of a long THz pulse (∼10 ps) crossing a sample composed of only 0.5-mm Quartz (no metallic layers). Figure 4 shows a comparison of the transmission of a THz wave with the theoretical simulation of the transmission.

As shown in Fig. 4a and b, if the experiments can measure a long time window, the standard TMM does reproduce experiments very accurately. On the other hand, if a smaller time window is measured, there is considerable loss of information. First of all, any experimental time windowing becomes arbitrary as main pulse and echo pulses overlap in time. The time windowing does not remove the whole echo neither removes only the echo: it partially removes the echo as well as cutting part of the transmitted signal. As expected the method developed in the present article is not applicable (see Fig. 4b and d).

5 Conclusion

In conclusion, we have demonstrated a more effective way of removing the echo caused by the substrate on pulsed radiation propagating through a multilayer system grown on a thick substrate. This allows for a direct comparison with the experimental
transmission spectrum of a time dependent probe, where usually the limited time windows (which exclude echo pulses) are measured. This method allows for the direct use of an explicit analytic formula and therefore makes it unnecessary to perform additional numerical steps.

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Declarations

Conflict of interest The authors declare no competing interests.

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