



## **[Invited] Terahertz time domain spectroscopy data processing: analysing uncertainties to push boundaries**

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# Terahertz time domain spectroscopy data processing: analysing uncertainties to push boundaries

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## ABSTRACT

Terahertz spectroscopy provides information on the motion of the charges in a sample at a picosecond scale. To recover this information from Terahertz time-domain spectroscopy (THz-TDS), one usually extracts the experimental refractive index then fits these curves. This approach suffers from several limitations, among them the difficulty to compare models of motions, provide the error bar associated with the extracted magnitude and a resolution limitation coming from the Fourier criteria of the fast Fourier transform. By adopting a Bayesian framework taking into account the experimental uncertainties and directly fitting the time-domain trace, we overcame these limitations. When correlated and epistemic uncertainties/noise are present, the algorithm considers its distribution as part of the data to fit and can mistake it for real physical features. Hence, it offers poor discrimination between good models and bad ones. After a thorough analysis of the experimental noise, we developed a preprocessing software removing epistemic noise on the time traces and providing an estimate of the noise correlation matrix (generalization of the standard deviation). It allows the proper weighting of the error function of the fit using these uncertainties and therefore the derivation of the Akaike information criteria, a metric enabling to calculate the most probable model from a set of models one wants to compare. In addition, by being in the time domain we avoid the Fourier criteria for the resolution and thus could get information on experimental lines down to 30 MHz with a commercial THz-TDS system.

**Keywords:** Terahertz, Time Domain Spectroscopy, signal processing, super-resolution

## 1. INTRODUCTION

The method of time-domain spectroscopy (TDS) began to develop rapidly about 30 years ago with the birth of the femtosecond laser as sub-picosecond sampling [1]. The idea was quickly used for spectroscopy [2] and a few years later, the integration of photoconductive antennas [3, 4] brought it to a practical broadband THz spectroscopic system. They enabled myriads of applications from semiconductor spectroscopy [5, 6], 2D materials [7], liquids [8], biological samples [9, 10] and carbohydrates [11, 12]. More recently, the measurements of band-pass filters [13] and metasurfaces embedded in microfluidic circuitry [14] were presented along with more fundamental studies of, for instance, ultra-strong coupling [15, 16].

The basic specificity of this spectroscopic technique reads in its name: measurements are not done in the frequency domain using a dispersive element, but in the time domain. In addition, in contrast to Fourier transform infrared spectrometers, in which the measured function is the self-correlation of the time data through interferometry, TDS is based on a direct measurement of the electric field versus time in the THz frequency range. Today, TDS is the main tool for broadband terahertz (THz) spectroscopy, these systems are now commercial and several companies offer products with more than one decade of bandwidth with a resolution up to  $\sim 1$  GHz and a sensitivity defined by its dynamic range of  $\sim 100$  dB. These figures are very impressive though, they do not fully reflect the actual capacities of the systems. First, the dynamic range compares the signal without any samples in the beam to one without any beam in the sample. It says how the systems are able to handle very lossy samples but absolutely nothing about the signal to noise ratio of the measurements. Second, parameter extraction for spectroscopic measurements has been a focus of several groups since the beginning, but there is still no method able to retrieve the parameter fully taking into account the uncertainties on the signal. Third, the resolution given is linked to the Fourier transform used in the system and one can overcome this limitation.

## 2. FROM THE DYNAMIC RANGE TO THE SENSITIVITY

Impact of the noise on THz-TDS measurements is a long haul effort [17] and we propose here to use a very common but still not standardized methods based on statistics and repetition of the measurements. As written above, the datasheet of the THz-TDS systems includes dynamic range data that compare the signal at the peak frequency to the noise at frequency where the system does not operate. This clearly helps make the difference from a first generation system with the new ones. However, it is not a clear figure on how reliable your measurements will be. Since most of the TDS measurements are done thanks to an accumulation of tenths or hundreds of time traces, one step further is to pay attention not only to the mean of the measurements with as well to the standard deviation as in fig 1 both in the time and frequency domain.

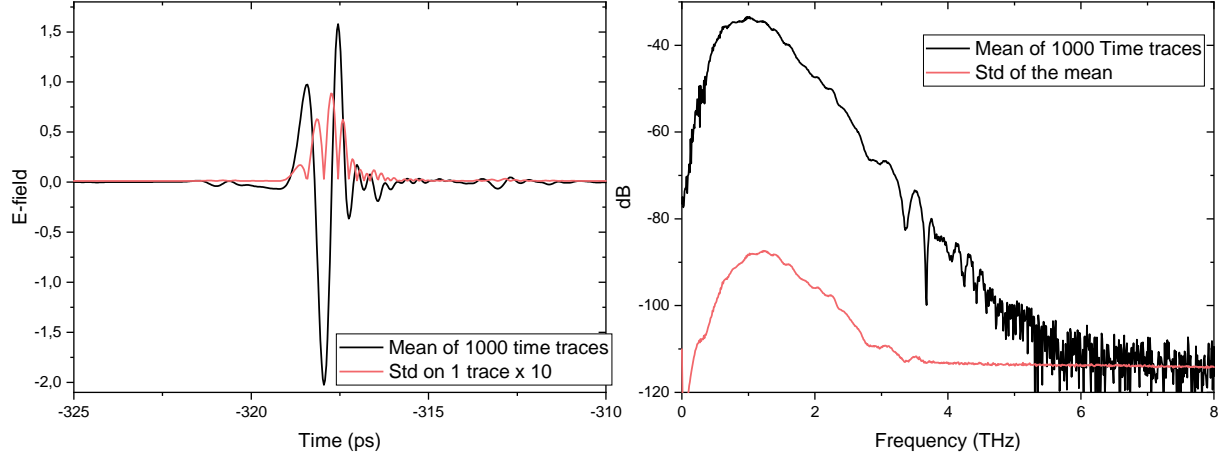


Figure 1: On the left the mean of time traces for a TDS experiment without samples and its standard deviation (the time traces were 100 ps long but we zoomed on the main pulse). On the right FFT of this mean and its standard deviation – 30dB to render the standard deviation on the mean.

There one can see that the dynamic range vision tremendously underestimates the noise in the range where the signal is high and a further look in the time domain shows that the standard deviation appears to be proportional to the derivative of the time signal [18]. In fact, the system we are using is fully based on optical fibers with a total length of almost ten meters. The length and refractive index of these fibers slightly depend on the temperature with an immediate consequence of slightly changing the delay between the laser pulse coming on the emitter and the one coming on the receiver. This will shift the time trace by a small amount (less than a time step), enough to introduce an additional standard deviation. Gratefully, since we expect the pulse to be identical, we can shift back all the time traces and resynchronize them as shown in fig 2. To achieve this resynchronization, we computed the classical average, compared all the traces (mean square) to this average and chose the more similar as the reference. Then we shift all the traces from a small delay ( $\sim 10$  fs) to synchronize it to the reference again by mean square similarity.

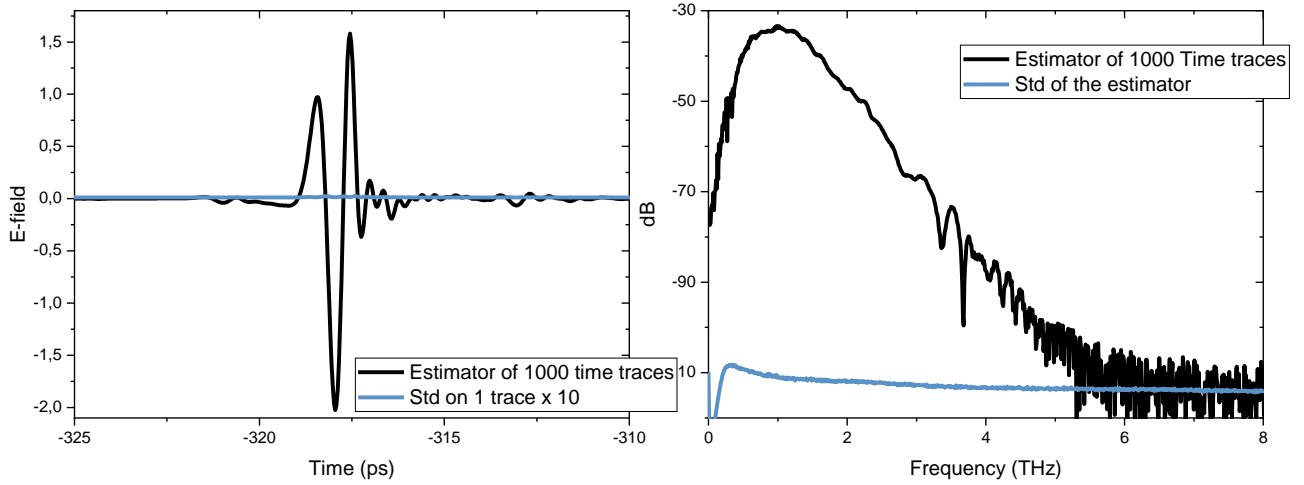


Figure 2 : On the left the estimator of time traces for a TDS experiment without samples and its standard deviation (the time traces were 100 ps long but we zoomed on the main pulse). The estimator is based on a resynchronization of the time trace before taking the mean. On the right FFT of this estimator and its standard deviation – 30dB to render the standard deviation on the mean.

Thanks to this new estimator, we reduced the standard deviation by about a factor 100 in the region of the data. It follows an increase of the precision by the same factor. In addition, the noise that was clearly pink became much more white even if some pink residues remain.

### 3. FITTING TDS DATA WITH A

The goal of every spectroscopy is to extract information from the recorded data. This information lies in models fitting a transfer function from a measured reference to a measured sample [19, 20, 21]. A figure of merit or a criterion is needed for both using an optimization algorithm to get the good parameters for the model and comparing two models fitting the same set of data [22]. This quantitative figure is for instance very important if one wants to compare two shapes for an absorption peak. The general principle here is that the more noisy a data point the less you can trust it. It leads to weight the data with the inverse of the error bar of each data point the so called the precision. This comparison is straightforward in frequency-domain spectroscopy because the models are also in the frequency model, making the transfer function a simple multiplication. Therefore, because the noise is often an uncorrelated white noise one can use its envelope as a weighing function for the fit.

However, as well explained in [23, 24], one of the specificities of THz-TDS is that the data are in the time domain. It means that when one performs a FFT to get the spectrum, the FFT will transform the noise as well to the frequency domain. By construction, the noise at a given frequency will be correlated to the one at another, making the weighting quite difficult in the frequency domain.

To go a step further into modeling, all the models for refractive indices or dielectric constant that we know are expressed in the frequency domain. It follows that the transfer function from the reference to the sample signal that was a multiplication in the frequency domain, becomes convolutions by a transfer function in the time domain. Therefore, the non-correlated hypothesis for noise is never true even in the time domain. E.g. for a slab sample, the model transforms the reference pulse into several Fabry-Perrot echoes, each of them getting the same, consequently correlated, noise. Thus, the weighting function does not apply any more, but one needs to use the noise correlation matrix (precision matrix) as weighting in the fit as explained in [23, 24]. It means that the knowledge of the noise both the magnitude and its correlation is of importance and is needed for a proper weighting of a fit.

We implemented this precision matrix into the fit@tds software we already developed [25, 21]. The approach is very similar to the one already described but instead of having a constant weighting function rendering for an hypothetical constant white noise, we multiply the time domain difference between the sample time trace and the model time trace by the precision matrix [23, 24]. As a result, the criterion or function of merit is now normalized to the noise and one can quantitatively compare the models. We used this approach on the usual golden sample lactose as shown on fig 3.

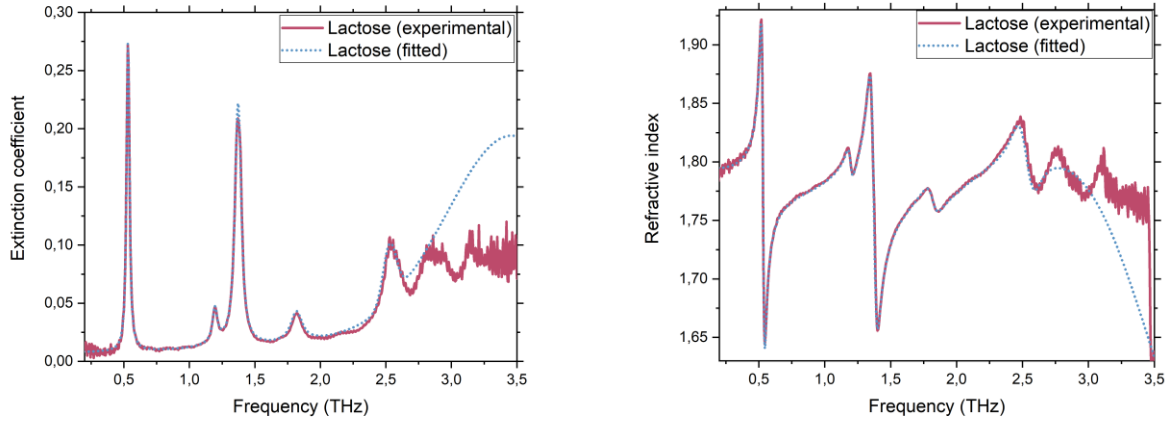


Figure 3 : Lactose experiments fit. On the left extracted extinction coefficient and its fit, on the right real part of the refractive index and its fit.

As an example, thanks to this quantitative criterion, we fit the time traces with 5 peaks + two phenomenological peaks to take into account for the high frequency absorption. We performed this fit with either a Lorentz model or a Voigt model (Voigt being the convolution of a Lorentzian shape by a Gaussian distribution). The conclusion was that the Voigt profiles was more suited for our date for the all the peak with an increasing importance when going to high frequency.

#### 4. SUPER RESOLUTION IN TDS SYSTEMS

The application of THz-TDS to study gas phase samples is attractive as the broad coverage up to 6 THz should enable fast multi-species analysis. In fact, this has been tried as early as in 1989 [26]. Nevertheless, gas spectroscopy studies employing THz-TDS are uncommon as the length of the delay line limits the resolution. Indeed, the time domain signal is recorded over a short time window corresponding the mechanical translation of the delay line. The spectrum is obtained using a Fast Fourier transform. FFT projects the signal on a basis of functions that is a frequency comb. The spacing between the teeth of the comb is equal to the inverse of the time windows length is the resolution limitation  $Fc$ . For gas spectroscopy and sensing applications, the needs for specificity and selectivity require to reduce the collisional broadening up to the Doppler linewidth limit which is reached at low pressure ( $<10$  mbar). In this case, the apparatus face the challenge of narrow and sparse, fully or partially, resolved rotational lines requiring high-resolution techniques [27, 28]. In THz-TDS, one idea could be to use very long delay lines. However, this would terribly degrade the signal to noise ratio, the stability and the compactness of the systems. Other experimental approaches based on the use of optical frequency combs enabled new broadband spectroscopic setups of particular relevance for THz molecular spectroscopy [29]. However, these systems are still, extremely complex to operate, costly and the experiments last hours if not days. Signal processing attempts were made [30] to unravel THz-TDS potential for high-resolution measurements. In fact, in other fields, as quantum chemistry [31], nuclear magnetic resonance [32], or photonic simulations [33] methods, such as wavelet transform [34], filter diagonalization [35] or harmonic inversion [31] allowed resolution performance improvements far beyond  $Fc$  because they do not use basis functions without pre-fixed central frequency.

To overcome this issue we implemented a Constrained Reconstruction Super Resolution (CRSR) algorithm [36, 37, 38]. It is based on our previous fitting algorithm and software. We are fitting the data with spectral Lorentz lines. However, instead of applying the FFTs in our loops on the recorded time window, we zero padded the reference (and only the reference) up to the repetition time of the laser, and performed the FFT on this signal. Then we compare the transformed reference and the sample only where we recorded the signal for the sample as shown on figure 4.

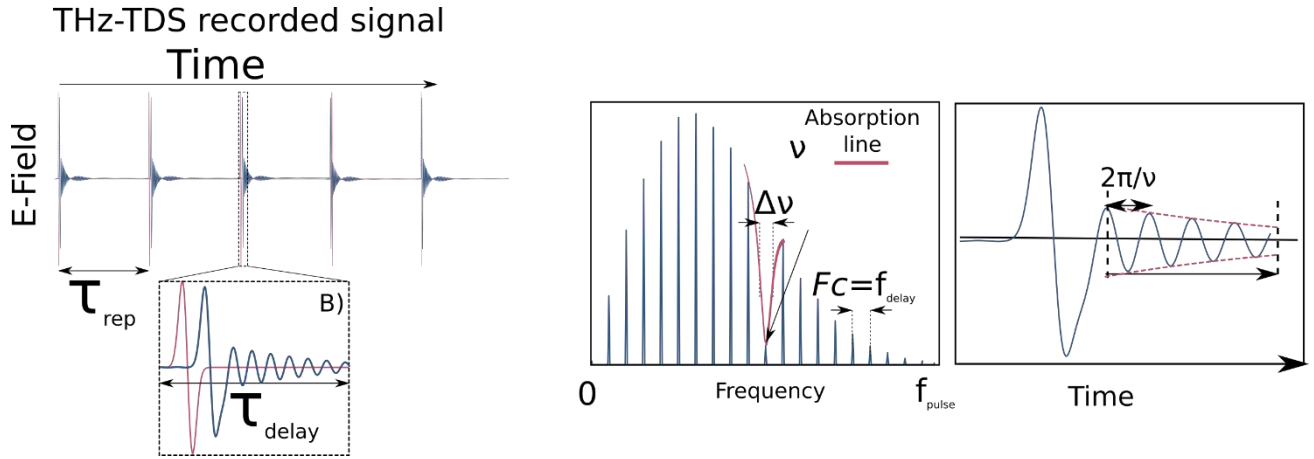


Figure 4. Principle of the methods. In THz-TDS the signal recorded in the time domain (left) is limited by the time windowing coming from the travel range of the delay line (see zoomed part). Usually, a discrete Fourier transform is applied to this signal (center). The CRSR (right)) uses the same data and perform a full analysis in the time domain to retrieve the parameters of each spectral lines, which correspond in the time domain to the damped sinu and thus achieve super resolution

It leads to super-resolution spectroscopy on a commercial broadband ( $\sim 0.2 - 6$  THz) THz Time Domain Spectrometer with a Fourier transform limited spectral resolution of  $F_c = 1.2$  GHz. We validated the methods on rotational spectra of gas phase ammonia ( $\text{NH}_3$ ) with the measurements of Doppler limited and self-broadened rotational lines in the 1 – 700 mbar pressure range as shown on fig 5.

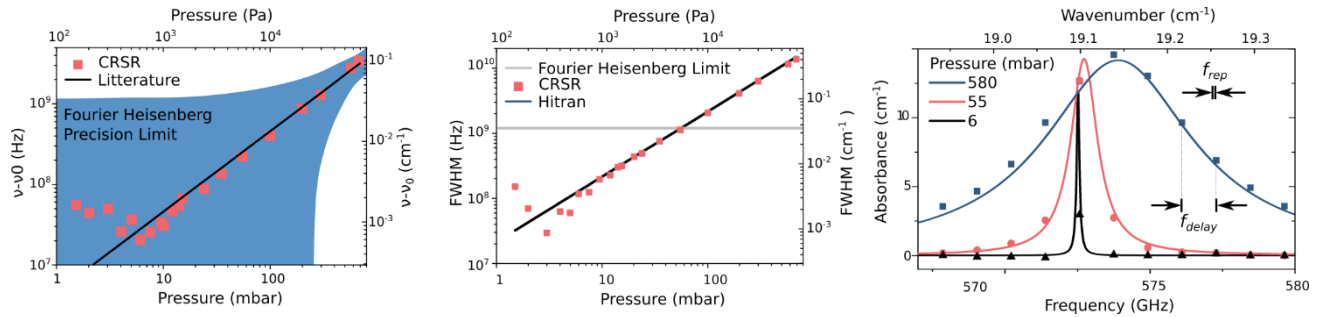


Figure 5 : Super resolution of the line around 570 GHz of gaseous  $\text{NH}_3$ . Right central frequency difference to the literature one at  $P=0$  bar. Center, width, right example of the reconstructed line for 3 pressures. All data are compared with Hitran database based on literature results.

Here one sees that the precision of the CRSR surpasses the usual Fourier criteria of 1.2 GHz and reaches a few tens of GHz. It is also important to note that the precision of the retrieval remains better than the Fourier criteria even for the highest pressure. These results were done on the most prominent line of the  $\text{NH}_3$  THz spectrum, yet we manage to get similar results up to 3.5 THz and even enable the separation of a doublet around 1.2 THz [38]. Finally, we insist on the fact that these experimental results were achieved on a 7 cm long gas cell because of the path length limitation due to the TDS. Further experiments are ongoing on a 1 M long cell and we expect much better performances on several different gasses.

## CONCLUSION

Time domain Terahertz spectroscopy became a commercial apparatus a decade ago and is now spreading in many fields of science. The system technology is nowadays evolving with the objective to remove the delay line as in Asynchronous Optical Sampling System (ASOPS) [39], Optical sampling by cavity tuning (OSCAT) [40] or electronically controlled optical sampling principle (ECOPS) [41]. At the same time, several groups are making efforts into signal processing to

enable more quantitative approaches by introducing quantitative criteria [40] and more realistic modeling [42]. We are following this trend and manage; -to reduce noise by a factor of about 100; to implement the Akaike criteria to quantitatively compare models; and finally to implement a super resolution method able to take the full benefit of the time domain data.

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