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Towards the elimination of water in THz-TDS data

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Abstract—One of the most problematic points in THz-TDS is the absorption induced by water lines. This absorption complicates and biases the fitting algorithms. We propose here an approach to eliminate water from TDS data. The 1-parameter model can eventually be used to quantify water present in different samples.

Keywords—THz, TDS, gas-phase, data treatment

I. INTRODUCTION

THz Time Domain Spectroscopy (TDS) has witnessed great enhancements since its start around three decades ago. The non-stop advances in optics and electronics permit to achieve higher performance of the readily available compact TDS systems. The broadband capabilities of the THz-TDS systems along with their ~ 100 dB dynamic range, make it a versatile tool for a broad field of applications for characterizing different types of materials and molecules including biomolecules, liquids, gases and compounds of environmental interest. Despite its diverse targeted systems, the TDS is still not considered a standard analytical technique mainly due to the tedious error analysis required to estimate the uncertainties. Multiple efforts in the last few years have been employed in this context to derive a robust approach for error estimation in TDS experiments [1, 2, 3, 4].

Another effect that may limit the TDS performance is the high absorption of water within the operational frequency ranges (0.2-6 THz). Water lines dominate the THz-TDS spectra and many approaches have been followed to decrease the water amounts in the effective THz path the most common being the use of N_2 purge boxes and vacuum gas cells [5]. Even though these methods nearly eliminate the presence of water, trace amounts of water are always present in the published spectra, originating from different sources including imperfect purging, water inside the sample itself; desorption phenomena, etc. In some cases, like fragile biological samples or real-life atmospheric samples, it is impossible to remove water, where the integrity of the sample is indispensable. Water, even in trace amounts can complicate the extraction of the complex index of refraction, especially through likelihood parameter minimization approaches.

In addition, the presence of water can diminish the detection limits of the TDS especially when interfering with numerical data analysis protocols that are implemented for trace detection. Moreover, the main effect of water absorption

in the THz range in general can be seen in the choice of telecom windows. All of this motivated us to propose a robust and systematic approach to isolate water contribution in TDS time traces and spectra. Our group has introduced custom-made Python codes capable of maximising the parameters and information extracted from time domain data through numerical treatment. The first one, called Correct@TDS is aimed to reduce the error in time domain data through extracting uncertainties on delay line position, laser amplitude fluctuations, dilatation in the fiber optics and periodic sampling. The extracted parameters are used to realign the individual spectra before averaging. The second is called Fit@TDS and its functionalities to extract material parameters and refractive index as well as their uncertainties, have been described previously [6].

In this conference, we will propose an approach for the systematic removal of water lines from THz-TDS spectra based on experimental measurements at different relative humidity conditions and data extracted from databases.

II. EXPERIMENTS

The experiments were conducted using a stainless steel 1 m gas cell of diameter 50 mm, placed in the center of a collimated THz beam. The source of this beam is a MenloSystems TeraSmart TDS system. The THz beam is collimated through the use of two 50 mm 90° parabolic mirrors. The traverse of the beam through the cell is ensured through two 2mm TPX windows fixed on the two extremities of the gas cell. Water gas was introduced into the cell through evaporation from a liquid sample and fed into the vacuum-pumped cell

Relative humidity, which is defined as the ratio of the vapour pressure of air to its saturation vapour pressure, is usually the measurable property that is reported along with TDS measurements. Following the same spirit, Time traces were recorded at different %RH. The quantity of water necessary to achieve a certain RH value was calculated based on the saturation vapor pressure at 23°C (~ 27 mbar). The gas cell was flushed/pumped for at least 30 minutes after each experiment to ensure complete desorption of water from the walls.

III. RESULTS

Figure 1 below shows the absorption spectra of water at different %RH.

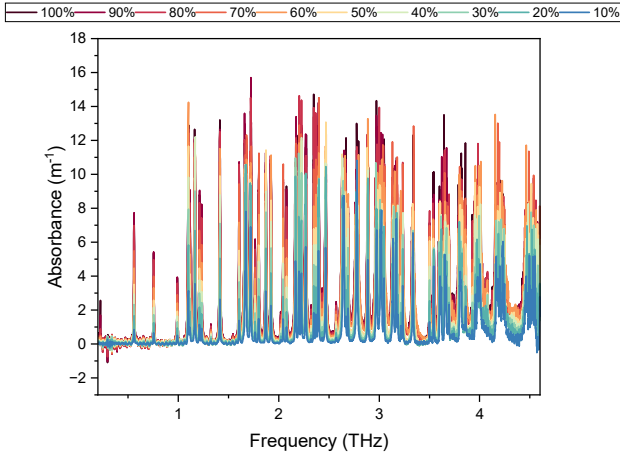


Figure 1 Absorbance (m^{-1}) of water at different %RH (0-100%).

The raw time traces will be fitted using the publicly available Fit@TDS software. The fit output for all the data will be used as an input matrix for the water quantification/removal approach. This permits to have a 1 parameter fit for all water lines and will be explained after.

IV. METHODOLOGY

The core of this approach is to be able to quantify or remove water based on a one-parameter fit of all the water lines in the covered spectral range. To achieve this, a multi-oscillator time domain fit is required for each recorded time trace at a defined %RH. The initial guesses are extracted from W2020 water lines database[7] and/or HITRAN database[8]. The fit is done through a minimisation of an objective function containing the parameters describing Lorentz oscillators. The permittivity of the system can be described, using a Lorentz model, by the following equation:

$$\tilde{\epsilon} = \tilde{\eta}^2(\omega) = \tilde{\epsilon}_\infty + \sum_{k=1}^{k_{max}} \frac{\Delta\epsilon_k \omega_{0k}^2}{\omega_{0k}^2 - \omega^2 + i\omega\gamma_k}$$

Where $\tilde{\epsilon}_\infty$ is the dielectric permittivity at high frequency compared to the range of interest, k_{max} is the number of considered oscillators, $\Delta\epsilon_k$, ω_{0k} and γ_k are the strength (units of permittivity), resonant frequency and damping rate of the k th oscillator, respectively

The output of the fits, i.e. a list of experimentally extracted Lorentz line strength, frequency and linewidth parameters, for each oscillator, will be stored as a matrix of reference.

The water removal or quantification algorithm will be implemented in the Correct@TDS. This means it will be processed along the different proposed corrections as a one

parameter option. The idea is to perform a minimization fit of the sum of all Lorentz oscillators in the time trace, starting from the experimental reference matrix. The obtained fitted time trace will be subtracted from the experimental time trace to ensure the full removal of water lines. Another option will be matching the fitted parameters to the reference matrix and deducing the quantity of water present in the time trace.

V. CONCLUSION

The current work serves as a step in the path of efficient and systematic TDS data analysis. In this conference, we will present in more detail our approach to eliminate water from time domain data, even in trace amounts. This approach acts as a proof of concept for the quantification of water in TDS spectra towards its elimination.

We expect these efforts to widen the field of applications of this versatile technique and to push one step further to becoming an analytical method of reference.

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