

## Determination of material parameters from THz measurements

BATOP GmbH, February 2018

*This paper helps to deduce material parameters like refractive index  $N(f)$  or the dielectric function  $\varepsilon(f)$  from Terahertz measurements in transmission or reflection. The experimental base is the absolute spectral amplitude of the Fourier transformed THz pulse. The dielectric function is determined by curve fitting using an appropriate material description and - if needed - also a calculation of thin film interference.*

### Table of contents:

1. The dielectric function $\varepsilon(f)$ of materials .....	1
1.2 Basic relations .....	1
1.3 Metals.....	2
1.4 Dielectric solids.....	2
1.5 Gases .....	2
2. THz transmission and reflection measurements .....	3
2.1 Transmission trough a gas .....	3
2.2 Transmission trough a solid.....	7
2.3 Reflection on a thick solid plate .....	12
2.4 Reflection on a thin solid plate.....	15

## 1. The dielectric function $\varepsilon(f)$ of materials

### 1.2 Basic relations

Refractive index  $N = n + i \cdot k$  n- real part, k – imaginary part (1)

Dielectric function  $\varepsilon(f) = N^2 = \varepsilon_r + i \cdot \varepsilon_i$   $\varepsilon_r$  - real part,  $\varepsilon_i$  – imaginary part (2)

Here  $\varepsilon$  means the relative permittivity.

Dispersion model 
$$\varepsilon(f) = \varepsilon_\infty + \frac{f_p^2}{f_0^2 - f^2 - i \cdot f \cdot \gamma}$$
 (3)

with  $f$  - frequency

$\varepsilon_\infty$  - high wavelength limit of the dielectric function  $\varepsilon(f)$ . It can be 1.

$f_0$  - resonance frequency

$f_p$  - plasma frequency

$\gamma$  - damping constant

The resonance frequency  $f_0$  is characterized by the bonding energy of electrons in solids or the vibration energy in molecules  $E_0$  and connected with this energy by  $E_0 = h \cdot f_0$ .

The plasma frequency  $f_p$  is determined by the density  $n_D$  of charged particles with charge  $q$ , for instance the electrons in a metal, but also in a dielectric or semiconductor material after excitation of bound electrons into the conduction band. With the particle charge  $q$ , the particle mass  $m$  and the vacuum permittivity  $\varepsilon_0$  the plasma frequency  $f_p$  can be calculated as

$$f_p = \sqrt{\frac{n_D \cdot q^2}{\varepsilon_0 \cdot m}} \quad (4)$$

The damping constant  $\gamma = 1/(2 \cdot \pi \cdot \tau)$  is related to the relaxation time  $\tau$  of the excited particles and therefore with the energy exchange with the surroundings (photon or phonon emission). We use here the frequency  $f$  instead of the angular frequency  $\omega = 2 \cdot \pi \cdot f$  to simplify the equations.

### 1.3 Metals

In case of free electrons in a metal with electrical conductivity  $\sigma$  the electrons can undergo free translation, but their movement is damped by resistive joule heating,.

Because these electrons are free there does not exist a resonance frequency. Therefore is  $f_0 = 0$ . The resulting dielectric function for metals is

$$\varepsilon(f) = \varepsilon_\infty - \frac{f_p^2}{f^2 + i \cdot f \cdot \gamma} = \varepsilon_\infty + \frac{i \cdot \sigma(f)}{2 \cdot \pi \cdot f \cdot \varepsilon_0} \quad (5)$$

Here the optical conductivity  $\sigma(f)$  is introduced with

$$\sigma(f) = \frac{\sigma_0}{1 - i \cdot 2 \cdot \pi \cdot f \cdot \tau} = \frac{n_e \cdot e^2 \cdot \tau / m_e}{1 - i \cdot 2 \cdot \pi \cdot f \cdot \tau} \quad (6)$$

$\tau$  is the time between two collisions of the free electrons and is related to the damping constant  $\gamma$  by

$$\gamma = \frac{1}{2 \cdot \pi \cdot \tau} \quad (7)$$

In a metal is  $\tau \approx 10^{-14}$  s and therefore the damping constant  $\gamma \approx 2 \cdot 10^{13}$  Hz. Therefore in the low frequency range  $f \ll \gamma$  around 1 THz the real part of the dielectric function can be neglected so that it can be approximated by

$$\varepsilon(f) \approx \frac{i \cdot f_p^2}{f \cdot \gamma} \quad (8)$$

Because of the low damping of incident electromagnetic wave in the THz range a metal is a nearly perfect reflector and can be used as reference mirror for calibration of reflection measurements.

### 1.4 Dielectric solids

In a low frequency region the response of a dielectric material can be described using equation (3). In a wider spectral range more than one energy transition can be involved, so the dielectric function must be extended as

$$\varepsilon(f) = \varepsilon_\infty + \sum_j \frac{f_{p,j}^2}{f_{0,j}^2 - f^2 - i \cdot f \cdot \gamma_j} \quad (9)$$

to include different electronic transitions with resonance frequencies  $f_{0,j}$ . The dispersion formula (9) ensures, that the real and imaginary parts of the dielectric function are related according to the Kramers-Kronig relation.

### 1.5 Gases

A gas with vibrational transitions in the THz region can be described with equation (9) and  $\varepsilon_\infty = 1$ .

$$\varepsilon(f) = 1 + \sum_j \frac{f_{p,j}^2}{f_{0,j}^2 - f^2 - i \cdot f \cdot \gamma_j} \quad (10)$$

## 2. THz transmission and reflection measurements

In a THz time-domain spectrometer at first the time dependent transmitted or reflected pulse is measured. In the next step the measured time dependent signal is Fourier transformed into the frequency domain to get the frequency dependent phase  $\varphi(f)$  and the absolute value of the transmittance  $|t(f)|$  or reflectance  $|r(f)|$ . Because of the ambiguity of the phase modulo  $2 \cdot \pi$  the use of the phase function  $\varphi(f)$  for the determination of material parameters is difficult. Therefore we use for this determination only the functions  $|t(f)|$  or  $|r(f)|$ .

### 2.1 Transmission trough a gas

As an example the time-domain transmission measurement of humid air is considered. As reference the transmission trough dry nitrogen gas is used. The figure 1 below shows the result of the time-domain measurement.

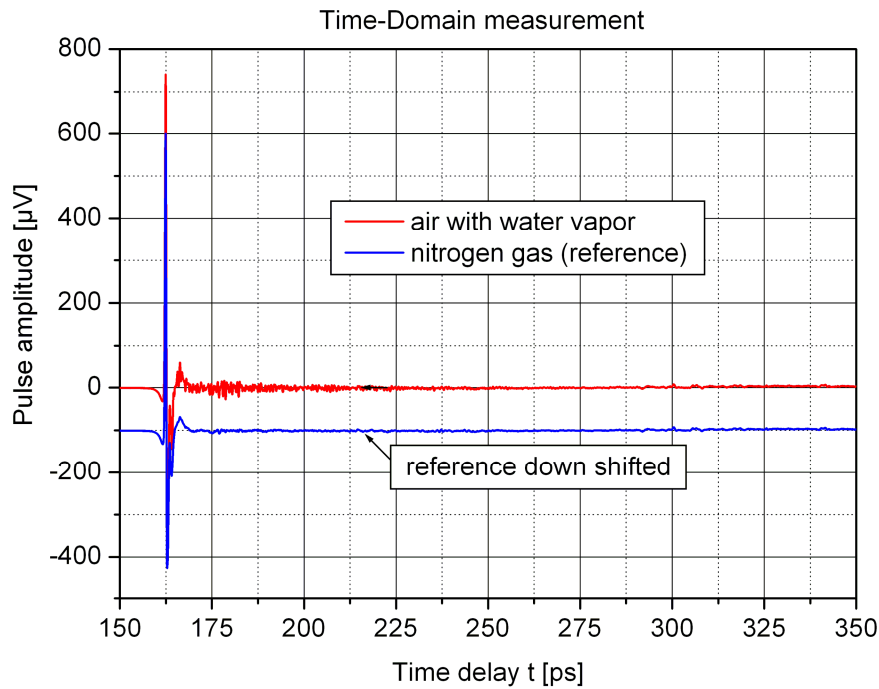


Fig. 1: Time-domain transmission of humid air and dry nitrogen gas with 20 cm measurement path

The spectral transmission  $t(f)$  can be calculated from Fourier transformed time-domain data of humid air  $t_{air}(f)$  and reference  $t_{nitrogen}(f)$  by

$$|t(f)| = \frac{|t_{air}(f)|}{|t_{nitrogen}(f)|} \quad (11)$$

The result of this division is shown in figure 2a.

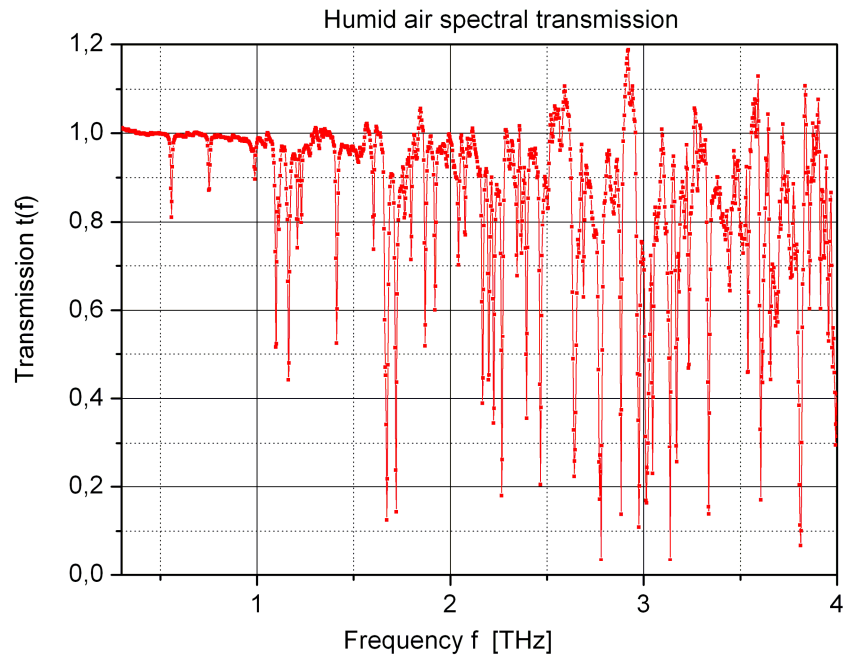


Fig. 2a: Spectral transmission  $|t(f)|$  of humid air with 20 cm measurement path length,, calculated via Fourier transformation of time-domain data in fig. 1. The calculated points by Fourier transformation are connected with lines.

There are many water vapor absorption lines in the measured spectral region. Transmission values  $> 1$  can be only explained by uncertainties of the time-domain measurements which increase with frequency.

For determination of the dielectric function  $\varepsilon(f)$  we restrict ourselves on a smaller spectral region from 300 GHz to 1.8 Hz.

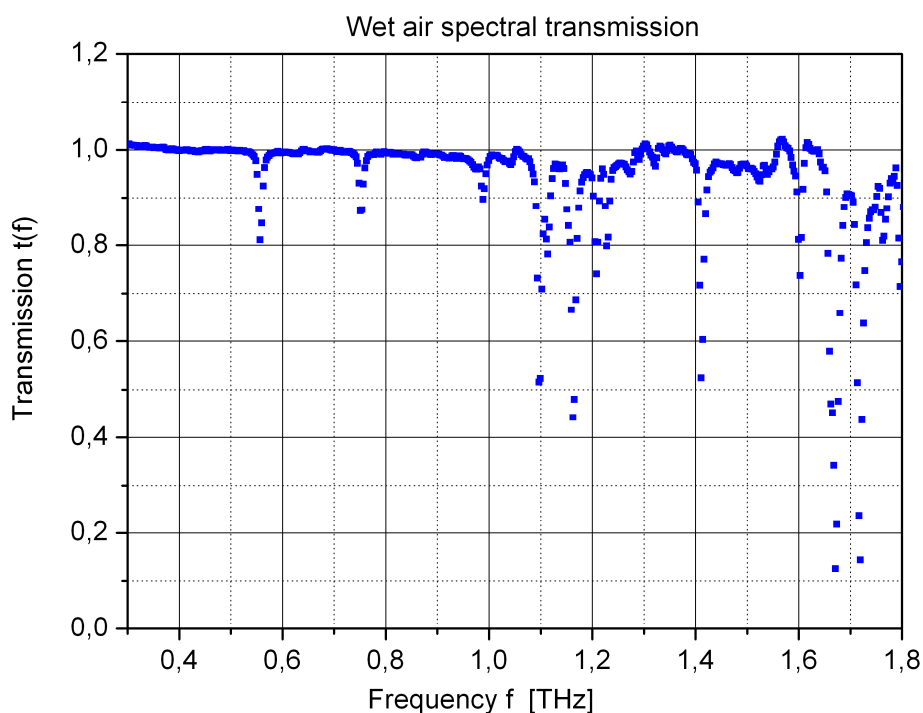


Fig. 2b: Spectral transmission  $|t(f)|$  of humid air with 20 cm measurement path length with restricted spectral region.

The transmission of an electromagnetic wave through a material can be described by the transmission coefficient

$$t(f) = e^{i\Phi(f)} = e^{i \cdot 2\pi \cdot N \cdot d \cdot f / c} \quad (12)$$

With  $\Phi(f)$  - frequency dependent phase  
 $d$  - thickness of the (gas) sample  
 $c$  - speed of light in vacuum.

By using relations (1) and (2) we get for the absolute value of transmission

$$|t(f)| = \left| e^{-2\pi \cdot k(f) \cdot d \cdot f / c} \right| = \left| e^{-2\pi \cdot \text{Im}(N) \cdot d \cdot f / c} \right| = \left| e^{-2\pi \cdot \text{Im}(\sqrt{\varepsilon}) \cdot d \cdot f / c} \right| \quad (13)$$

For determination of the dielectric function (10) of the measured gas two steps are needed:

- The start parameters  $f_{0,j}$ ,  $f_{P,j}$  and  $\gamma_j$  are estimated from measured transmittance  $|t(f)|$ .
- A curve fitting of  $|t(f)|$  with the dielectric function (10) in equation (11) must be done.

The start parameters can be estimated from measured transmittance  $|t(f)|$  as follows:

- $f_{0,j}$  = frequencies at the transmission minima  $t_{\min} = |t(f_{0,j})|$
- $\gamma_j$  = FWHM - full width of half maximum of the spectral absorption minima  $t_{\min}$
- $f_{P,j} = \sqrt{\frac{c \cdot (1 - t_{\min}) \cdot \gamma_j}{2 \cdot \pi \cdot d}}$  (14)

Fig. 2c shows the result of a curve fitting to approximate the measured spectral transmission in fig. 2b.

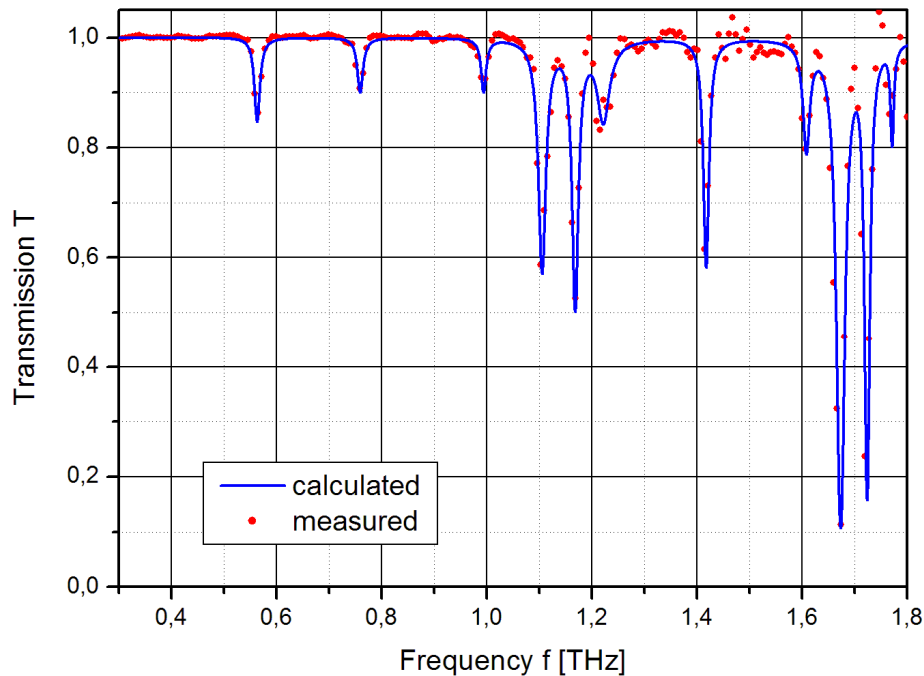


Fig. 2c: Calculated (blue line) and measured (red points) transmittance  $|t(f)|$  of water vapor using fitted parameters  $f_{0,j}$ ,  $f_{P,j}$  and  $\gamma_j$  for the dielectric function  $\varepsilon(f)$ .

The dielectric function  $\varepsilon(f)$  with fitted parameters  $f_{0,j}$ ,  $f_{P,j}$  and  $\gamma_j$  is shown in figures 4 and 5.

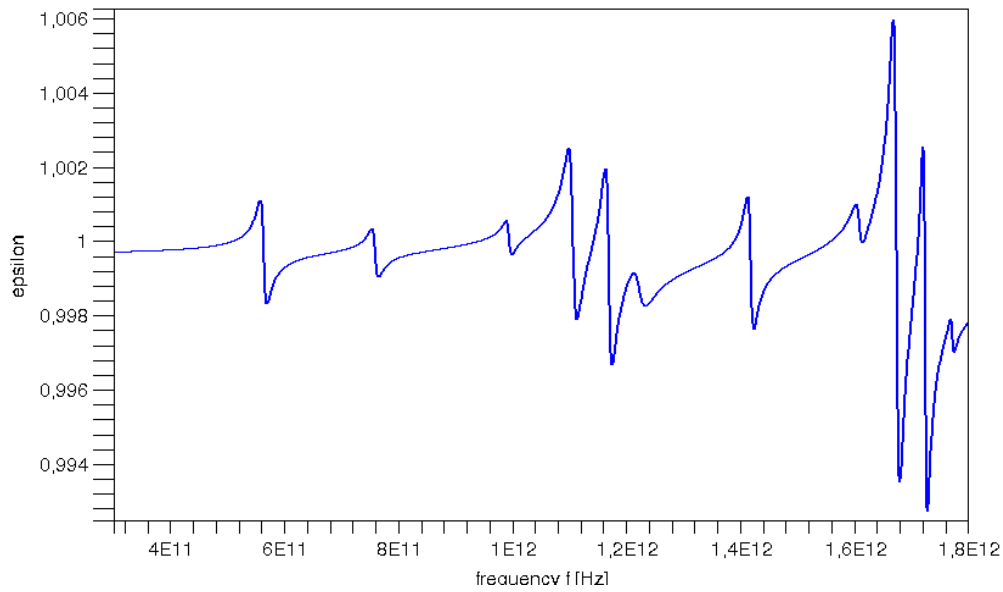


Fig. 3: Real part of dielectric function  $\varepsilon_r(f)$  with fitted parameters  $f_{0,i}$ ,  $f_{P,i}$  and  $\gamma_i$

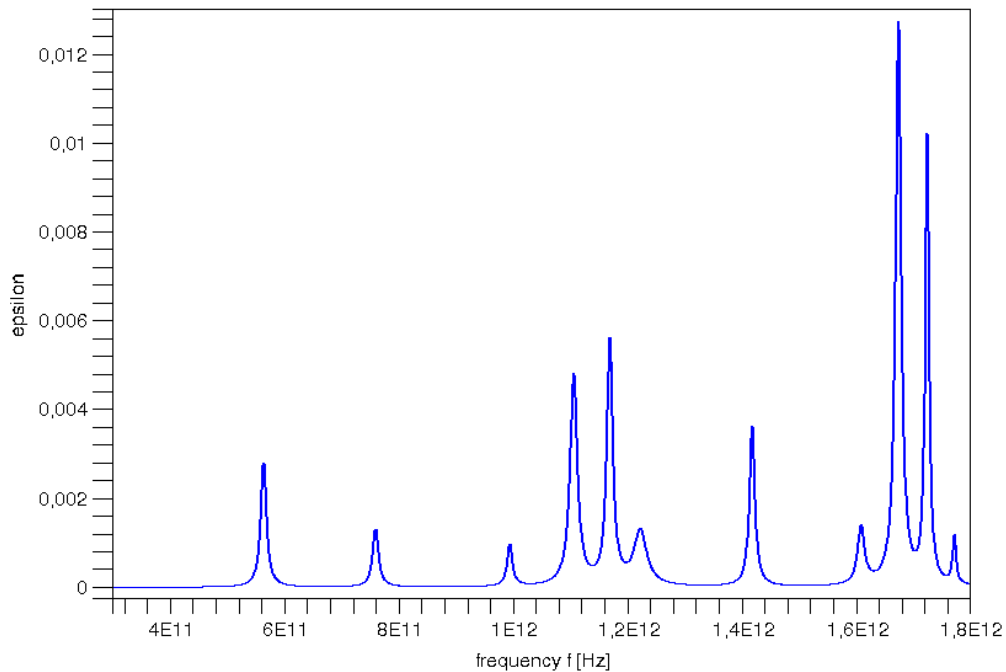


Fig. 4: Imaginary part of dielectric function  $\varepsilon_i(f)$  with fitted parameters  $f_{0,j}$ ,  $f_{P,j}$  and  $\gamma_j$

### Interpretation of the measured absorption lines in water vapor

The fitted  $f_{0,j}$ ,  $f_{P,j}$  and  $\gamma_j$  parameters for the absorption lines are listed in table 1. If we consider the measured resonances as vibrations of the H atoms in the H<sub>2</sub>O molecule, then we can use relation (4) to determine the atomic polarization from the measured plasma frequencies  $f_{P,j}$ . The hydrogen and oxygen atoms are in the first stage electric neutral, so that the charge would be zero. But the charge transfer in the atomic bonds results in dipoles, which can vibrate and rotate.

For a simple interpretation of the measured plasma frequencies  $f_{P,j}$  equation (4) with the hydrogen atom mass  $m_H$  and  $q=e_0$  can be used.

The density  $n_W$  of the water vapor atoms in the THz measurement path and can be calculated by using the ideal gas equation for the water molecules

$$n_W = \frac{p_W}{k_B \cdot T} \quad (15)$$

Here  $p_W$  is the partial pressure of water vapor at temperature  $T$  and  $k_B$  the Boltzmann constant. At a temperature of 22 °C ( $T = 295$  K) and 50 % relative humidity the water vapor pressure is  $p_W = 1.3$  kPa.

With these values we can calculate  $n_W = 3.19 \cdot 10^{23}/m^3$  in the measurement path.

If we consider the plasma frequency  $f_{P,j}$  as oscillator strength with transition probabilities  $p_j$ , then we can these probabilities deduce from the measured plasma frequencies as follows:

$$p_j = \frac{f_{P,j}^2 \cdot m_H \cdot \varepsilon_0}{e_0^2 \cdot n_W} = \frac{f_{P,j}^2 \cdot m_H \cdot \varepsilon_0 \cdot k_B \cdot T}{e_0^2 \cdot p_W} \quad (16)$$

According to equation (7) we can calculate also the relaxation time  $\tau_j$  for the  $j$ -th transition. The results are included in table 1.

Measured water vapor resonances			Calculated resonance parameters		
Resonance frequencies $f_{0,j}$ [THz]	Plasma frequencies $f_{P,j}$ [GHz]	Damping constant $\gamma_j$ [GHz]	Transition energies $E_{0,j} = f_{0,j} \cdot h$ [meV]	Transition probabilities $p_j$ * $10^6$	Relaxation time constants $\tau_j$ [ps]
0.5635	2.1713	11.906	2.335	8.54	13.37
0.7593	1.7625	12.413	3.146	5.63	12.82
0.9942	1.6974	10.990	4.119	5.22	14.48
1.1061	4.4438	14.952	4.583	35.77	10.64
1.1692	4.4286	12.216	4.844	35.53	13.03
1.2225	3.2014	26.86	5.065	18.57	5.92
1.4181	3.7876	11.130	5.876	25.99	14.30
1.6088	2.7593	14.507	6.666	13.79	10.97
1.6739	7.7914	11.437	6.936	109.97	13.92
1.7243	5.8436	7.8733	7.145	61.86	20.22
1.7718	1.8708	7.3127	7.341	6.34	21.77

*Table1: Fitted parameters  $f_{0,j}$ ,  $f_{P,j}$  and  $\gamma_j$  for the measured water vapor sample according to equations (2), (10) and (12) and the calculated parameters transition energy  $E_{0,j}$ , transition probability  $p_j$ , and relaxation time  $\tau_j$  after equations (14), (15) and (7) .*

The calculated relaxation time constants  $\tau_j$  are in the region of several hundred picoseconds. This shows, that for an accurate determination of such molecule resonances a long time scan of about 1 ns for the THz measurement is needed.

## 2.2 Transmission trough a solid

The transmission of an electromagnetic wave trough a solid material with thickness  $d$  can be considered in a first approximation as a three step process. Using the symbols in figure 5 below we can write for the total transmission  $t$  considering possible multiple reflections within the resonator like solid

$$t = \frac{t_f \cdot t_d \cdot t_b}{1 - r_f \cdot r_b \cdot t_d^2} \quad (17)$$

The three transmission coefficients and the two reflection coefficients can be calculated using the Fresnel equations with zero degree angle of incidence as follows:

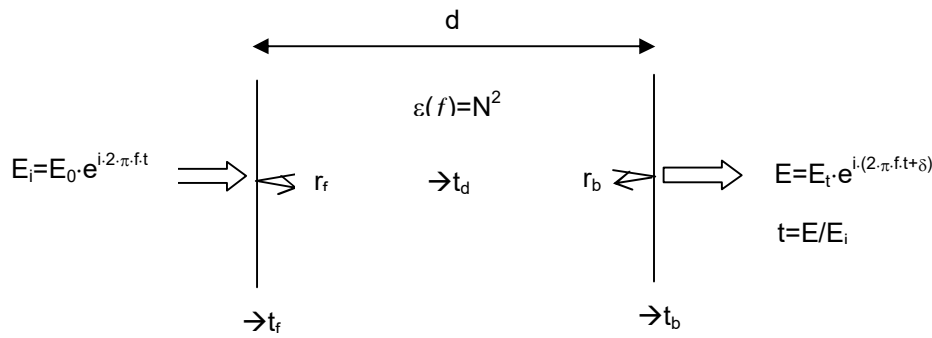


Fig. 5 Schematic for transmission of an electromagnetic field  $E$  through a solid with dielectric function  $\varepsilon(f)$  and thickness  $d$ .

$$t_r = \frac{2}{N+1}, \quad t_d = e^{i\Phi}, \quad \Phi = \frac{2 \cdot \pi \cdot f \cdot N \cdot d}{c}, \quad t_b = \frac{2 \cdot N}{N+1}, \quad r_f = r_b = \frac{N-1}{N+1} \quad (18)$$

The interference term in the denominator in eq. (17) is only significant for small thickness, low material absorption at low frequencies because the exponential damping of the transmitted field amplitude is proportional to  $d \cdot k \cdot f$ . Using equations (17) and (18) the absolute value of the transmittance through a solid body can be written as

$$|t(f)| = \left| \frac{4 \cdot N \cdot e^{i \cdot 2 \cdot \pi \cdot f \cdot N \cdot d / c}}{(N+1)^2 - (N-1)^2 \cdot e^{i \cdot 4 \cdot \pi \cdot f \cdot N \cdot d / c}} \right| \quad (19)$$

An example for a measured transmission  $|t(f)|$  through a 1 mm thick glass plate is shown in figure 6 below

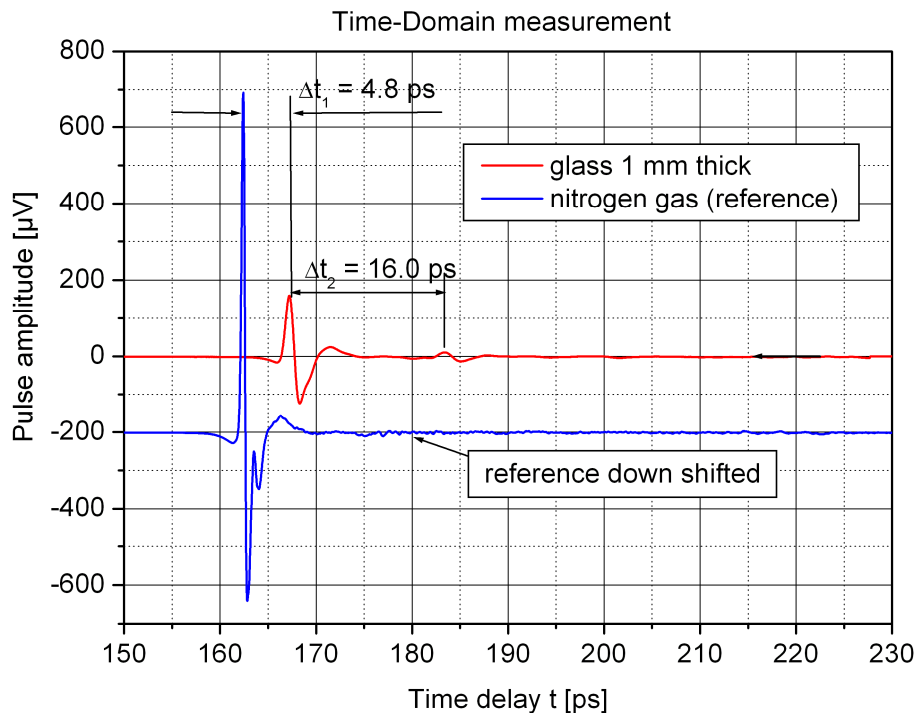


Fig. 6a: THz pulse transmission through a 1 mm thick absorbing glass plate



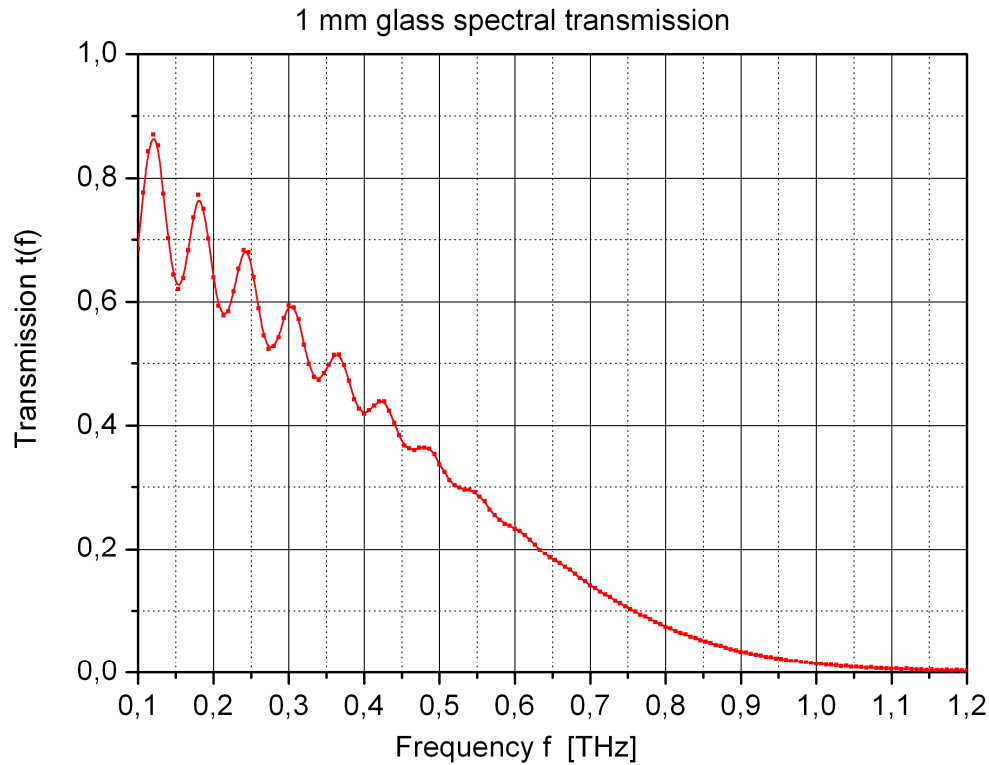


Fig. 6b: Spectral transmission curve  $|t(f)|$  of 1 mm thick glass plate.

In the measured spectral region up to 1.2 THz only one resonance absorption dip can be seen. Therefore the dielectric function can be formed with only one set of parameters  $\epsilon_\infty$ ,  $f_0$ ,  $f_P$  and  $\gamma$  in equation (9).

The needed start parameter set for a subsequent curve fitting can be found in two consecutive steps.

Step 1:

At first a dispersion free complex refractive index  $N = n + i \cdot k$  can be estimated in the following way:

- a. The real part of the refractive index  $n$  can be deduced from the time delay  $\Delta t_1$  or  $\Delta t_2$  in fig. 6a or from the frequency period  $\Delta f$  in the low frequency part of the spectrum in fig. 6b. With the difference frequency  $\Delta f$  between two consecutive transmission maxima or minima and the dedicated phase difference  $\Delta\Phi = 2 \cdot \pi$  the real part of the refractive index  $n$  can be estimated by neglecting the imaginary part to

$$n \approx \frac{c}{2 \cdot d \cdot \Delta f} \approx \frac{c \cdot \Delta t_2}{2 \cdot d} \approx 1 + \frac{c \cdot \Delta t_1}{d} \quad (20)$$

From transmission measurement of  $d = 1$  mm thick glass in figure 6b frequency period  $\Delta f = 61$  GHz can be deduced. With this value and formula (20) we get  $n = 2.46$ . With  $\Delta t_2 = 16$  ps in fig. 6a we get  $n = 2.4$  and with  $\Delta t_2 = 4.8$  ps the result is  $n = 2.44$ .

- b. The imaginary part of the refractive index  $k$  can be deduced from the transmitted amplitude. In the region of interferences the maxima can be used to determine  $k$  values. The transmission maxima can be calculated approximately by using eq. (18), where the imaginary part  $k$  of the complex refractive index is only considered in the exponential terms and the real part of the phase  $\Phi$  is a multiple  $m$  of  $2 \cdot \pi$ . This results in the following approximation for the maximum transmission:

$$t_{max} = \frac{4 \cdot n \cdot e^{-2 \cdot \pi \cdot f \cdot k \cdot d / c}}{(n+1)^2 - (n-1)^2 \cdot e^{-4 \cdot \pi \cdot f \cdot k \cdot d / c}} \quad (21)$$

If  $n$  is known, then  $k$  can be calculated from  $t_{max}$  using relation (21) by

$$k = \frac{c}{2 \cdot \pi \cdot f \cdot d} \cdot \ln \left( \frac{t_{max} \cdot (n-1)^2}{\sqrt{4 \cdot n^2 + t_{max}^2 \cdot (n^2 - 1)^2} - 2 \cdot n} \right) \quad (22)$$

With  $t_{max} = 0.363$  at  $f = 486$  GHz we get from equation (22)  $k = 0.0925$ .

A test calculation of  $|t(f)|$  using equation (19) with the values  $n=2.5$  and  $k=0.0925$  shows figure 7. A comparison between the measured transmission in figure 6b and the calculated transmission in fig. 7 shows, that dispersion must be included to describe the experimental results. The lower measured transmission with increasing frequency can be only explained by an increasing imaginary part  $k$  of the refractive index with frequency.

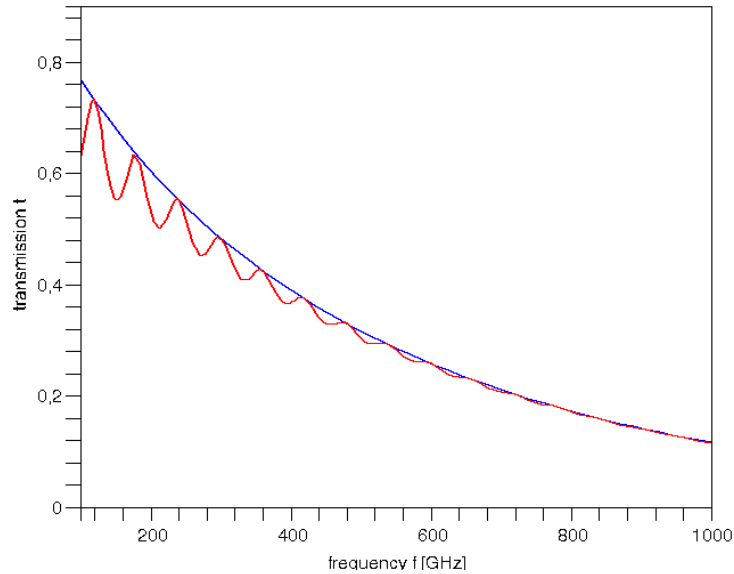


Fig. 7: Calculated transmission  $|t(f)|$  (red curve) of a 1 mm thick glass plate with  $N=2.5 + i \cdot 0.057$  using equation (19). The blue curve is for the maximum transmission  $t_{max}$  according to equation (21).

Step 2:

Using the estimated  $n$  and  $k$  values from step 1 the parameters  $\epsilon_\infty$ ,  $f_0$ ,  $f_P$  and  $\gamma$  in equation (9) must be estimated to start the curve fit with equation (19). The start parameters can be found as follows:

- The value of  $\epsilon_\infty$  must be between 1 and  $n^2$ . A possible start value is  $\epsilon_\infty = n$ . In this case some dispersion is included. In the example above it means  $\epsilon_\infty = 2.5$ .
- Because the minimum transmission in fig. 6b is at frequencies  $> 1.2$  THz we can set  $f_0 = 1.4$  THz as start value.
- The damping constant  $\gamma$  can be estimated from the spectral absorption bandwidth FWHM:

$$\gamma = \frac{FWHM}{2} \quad (23)$$

The full width at half maximum (FWHM) absorption bandwidth in fig. 6b may be about 1.6 THz.

The start value of the damping constant can be assumed as  $\gamma = 0.8$  THz.

- The plasma frequency  $f_P$  can be estimated in dependency on the measured transmission  $t_0$  on the resonance frequency  $f_0$  and the damping constant  $\gamma$  as

$$f_P \approx \sqrt{\frac{f_0 \cdot \gamma}{1 - t_0}} \tag{24}$$

For the 1 mm glass sample with the transmission  $t_0 \approx 0$  at the resonance frequency  $f_0 \approx 1.4$  THz we get from (24)  $f_P \approx 1.1$  THz.

The frequency dependent transmission  $|t(f)|$  can be calculated with equation (19) using  $\epsilon_\infty, f_0, f_P, \gamma$  for the description of the frequency dependent refractive index  $N(f)$  according to equations (2) and (3). A curve fit to approximate the measured transmission in fig. 6b results in the final parameter set as follows:  $\epsilon_\infty = 2.54, f_0 = 1.59$  THz,  $f_P = 2.80$  THz,  $\gamma = 0.471$  THz. The calculated spectral transmission  $|t(f)|$  and the functions  $N(f)$  and  $\epsilon(f)$  with these parameters are shown in figures (8), (9), and (10).

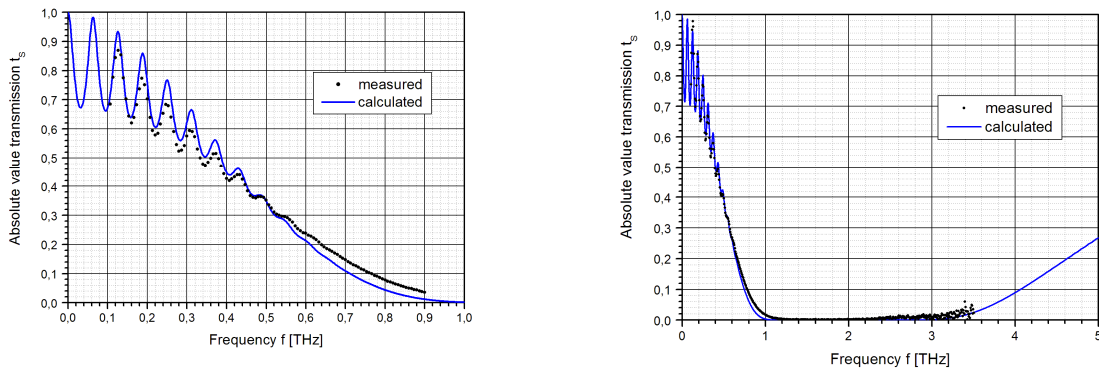


Fig. 8: Calculated transmission  $|t(f)|$  of a 1 mm thick glass plate with the dispersion parameters  $\epsilon_\infty = 2.54, f_0 = 1.59$  THz,  $f_P = 2.80$  THz,  $\gamma = 0.471$  THz after curve fitting using equations (2), (3) and (18).

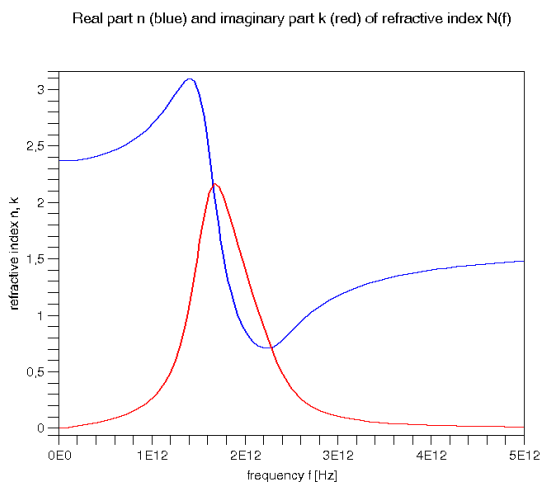


Fig. 9a Calculated function  $N(f)$

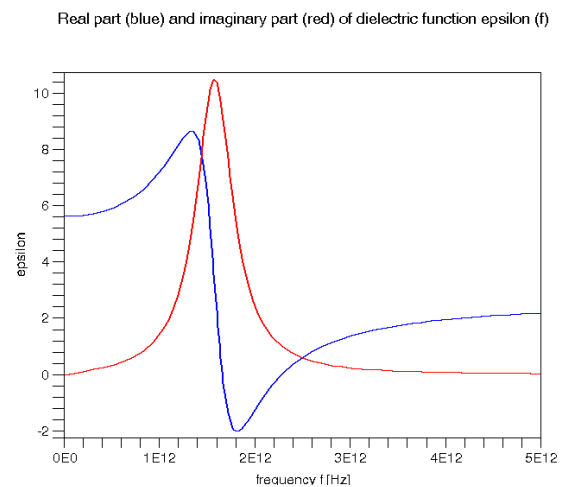


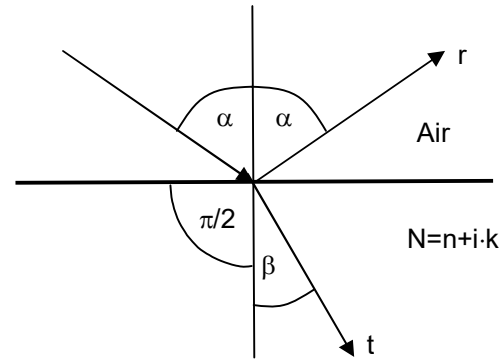
Fig. 9a Calculated function  $\epsilon(f)$

## 2.3 Reflection on a thick solid plate

In case of a reflection geometry the angle of incidence  $\alpha$  is typically non zero. Therefore we have to distinguish between the two reflection coefficients  $r_s$  and  $r_p$  corresponding to the polarization directions perpendicular and parallel to the plane of incidence. The reflected electric field amplitude  $r$  on an infinite thick sample can be described with the Fresnel coefficients as follows:

$$r_s = \frac{\cos(\alpha) - N \cdot \cos(\beta)}{\cos(\alpha) + N \cdot \cos(\beta)} \quad r_p = \frac{N \cdot \cos(\alpha) - \cos(\beta)}{N \cdot \cos(\alpha) + \cos(\beta)} \quad (25)$$

Fig. 10 Reflection and transmission on the surface of an infinite thick material



The refraction angle  $\beta$  can be calculated by

$$\cos(\beta) = \frac{\sqrt{N^2 - \sin^2(\alpha)}}{N} \quad (26)$$

With (21) and (22) we can write:

$$r_s = \frac{\cos(\alpha) - \sqrt{N^2 - \sin^2(\alpha)}}{\cos(\alpha) + \sqrt{N^2 - \sin^2(\alpha)}} \quad r_p = \frac{N^2 \cdot \cos(\alpha) - \sqrt{N^2 - \sin^2(\alpha)}}{N^2 \cdot \cos(\alpha) + \sqrt{N^2 - \sin^2(\alpha)}} \quad (27)$$

The dispersion is given by equations (2) and (3). The reflected THz signal is  $|r_s|$  or  $|r_p|$ .

### Example

An example for a measured reflection  $|r_s(f)|$  on a 7.7 mm thick glass plate is shown in figure 11 below. The absorption of such a thick glass sample is very strong in the THz spectral region so that a transmission measurement is not appropriate.

reflection 7.7 mm glass versus frequency f

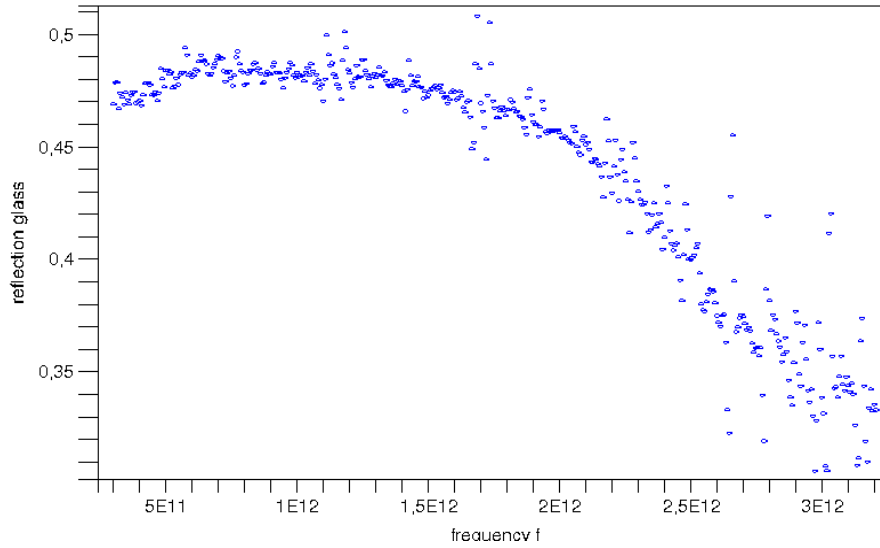


Fig. 11: Spectral reflection  $|r_S(f)|$  with perpendicular polarization of a 7.7 mm thick glass sample. The incidence angle is  $\alpha = 30^\circ$ .

In the measured spectral region up to 3.2 THz only one resonance absorption dip can be seen. Therefore the dielectric function can be formed with only one set of parameters  $\epsilon_\infty$ ,  $f_0$ ,  $f_P$  and  $\gamma$  in equation (9).

The needed start parameter set for a subsequent curve fitting can be found in two consecutive steps.

Step 1:

At first a dispersion free complex refractive index  $N = n + i \cdot k$  can be estimated in the following way:

- a) The imaginary part  $k$  of the refractive index is neglected for the lowest measured frequency  $f_L = 0.3$  THz. The real part  $n \sim N$  of the refractive index at  $f_L$  is calculated using equation (27) from the measured value  $|r_S(f_L)|$ :

$$n = \frac{\sqrt{1 - r_S \cdot (4 \cdot \sin^2(\alpha) - 2) + r_S^2}}{1 - r_S} \quad r_S > 0 \quad (28)$$

In case of the extraction of  $n$  from a measurement with parallel polarized THz light the formula is longer:

$$n = \frac{1}{2} \sqrt{\frac{2 \cdot (1 + r_P)^2 + 2 \cdot \sqrt{r_P^4 + 4 \cdot r_P^3 + 6 \cdot r_P^2 + 4 \cdot r_P + 1} + 4 \cdot \sin(\alpha)^4 \cdot (1 - r_P^2) - 4 \cdot \sin(\alpha)^2 \cdot (1 - r_P^2)}{(1 - r_P)^2 - \sin(\alpha)^2 \cdot (1 - r_P)^2}} \quad (29)$$

From figure 11 we get  $|r_S(f_L)| = 0.47$  at  $f_L = 0.3$  THz and  $\alpha = 30^\circ$ . Using (28) we can calculate  $n = 2.45$ .

- b) The dispersion results in a maximum reflectance  $|r_S(f_{\max})| = 0.485$  at the frequency  $f_{\max} = 0.8$  THz. An estimated value for  $k$  we can calculate with  $n$  by using equation (28) again with the reflection value at  $f_{\max}$ . We interpret the difference between the refractive index calculated at  $f_{\max}$  and at  $f_L$  as  $k$ . This results in  $k = 2.54 - 2.45 = 0.09$ .

Step 2:

Using the estimated  $n$  and  $k$  values from step 1 the parameters  $\varepsilon_\infty$ ,  $f_0$ ,  $f_P$  and  $\gamma$  in equation (9) must be estimated to start the curve fit with equation (27). The start parameters can be found as follows:

- The value of  $\varepsilon_\infty$  must be between 1 and  $n^2$ . A good start value is  $\varepsilon_\infty = n$ . In this case some dispersion is included. In the example above it means  $\varepsilon_\infty = 2.45$ .
- The resonance frequency  $f_0$  must be between the frequency  $f_{\max} = 0.8$  THz for maximum reflectance and the frequency  $f_{\min} = 3.2$  THz for minimum reflectance. We estimate with

$$f_0 = f_{\max} + \frac{f_{\min} - f_{\max}}{2} \quad (30)$$

$$f_0 = 2 \text{ THz.}$$

- The damping constant  $\gamma$  can be estimated from the resonance width  $\gamma = f_{\min} - f_{\max} = 2.4$  THz.
- The plasma frequency  $f_P$  can be roughly estimated as follows:

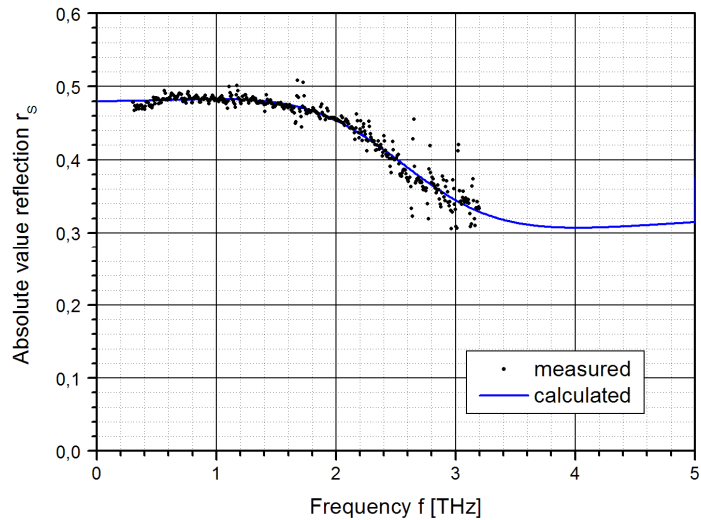
$$f_P \approx \sqrt{f_0 \cdot \gamma} \quad (31).$$

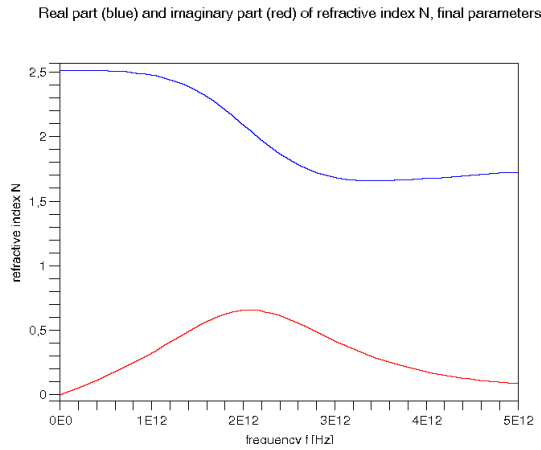
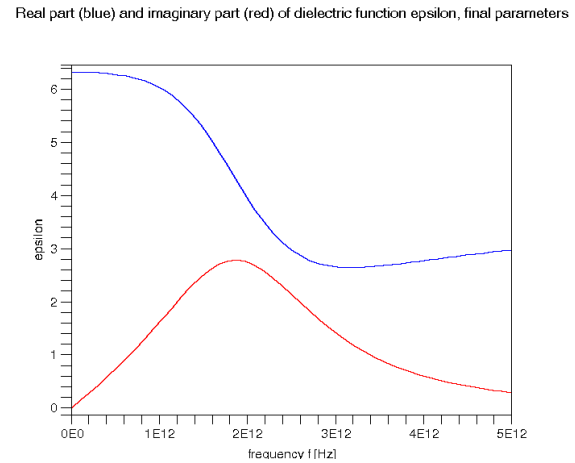
With the above estimated values for  $f_0$  and  $\gamma$  results  $f_P \approx 2.2$  THz.

A curve fit using formulas (2), (3) and (27) with the measured reflection  $|r_S(f)|$  values results in the fitted parameters  $\varepsilon_\infty=3.48$ ,  $f_0=2.20$  THz,  $f_P=3.71$  THz and  $\gamma=2.43$  THz. The calculated functions  $|r_S(f)|$ ,  $N(f)$  and  $\varepsilon(f)$  are shown in figures 12 and 13.

Fig 12:

Calculated reflection  $|r_S(f)|$  of a 7.7 mm thick glass plate with the dispersion parameters  $\varepsilon_\infty = 3.48$ ,  $f_0 = 2.20$  THz,  $f_P = 3.71$  THz,  $\gamma = 2.43$  THz after curve fitting using equations (2), (3) and (27).



Fig. 13a Calculated function  $N(f)$ Fig. 13b Calculated dielectric function  $\epsilon(f)$ 

## 2.4 Reflection on a thin solid plate

In case of a reflection measurement on a thin solid sample the multiple reflections on the front and back side of the sample must be taken into account. This results in typical Fabry-Perot interference patterns. Figure 14 shows these multiple reflections.

The total reflection  $r$  is the sum of the partial reflections  $r_i$ , which can be calculated for perpendicular (index  $s$ ) and parallel polarization (index  $p$ ) as follows:

$$r_{0,S} = \frac{\cos(\alpha) - \sqrt{N^2 - \sin^2(\alpha)}}{\cos(\alpha) + \sqrt{N^2 - \sin^2(\alpha)}} \quad r_{0,P} = \frac{N^2 \cdot \cos(\alpha) - \sqrt{N^2 - \sin^2(\alpha)}}{N^2 \cdot \cos(\alpha) + \sqrt{N^2 - \sin^2(\alpha)}} \quad (32)$$

$$r_1 = t_{f-} \cdot t_d \cdot r_b \cdot t_d \cdot t_{f+} = t_{f-} \cdot t_d^2 \cdot r_b \cdot t_{f+} \quad (33)$$

$$r_2 = t_{f-} \cdot t_d \cdot r_b \cdot t_d \cdot r_{f-} \cdot t_d \cdot r_b \cdot t_d \cdot t_{f+} = t_{f-} \cdot t_d^4 \cdot r_{f-} \cdot r_b^2 \cdot t_{f+} \quad (34)$$

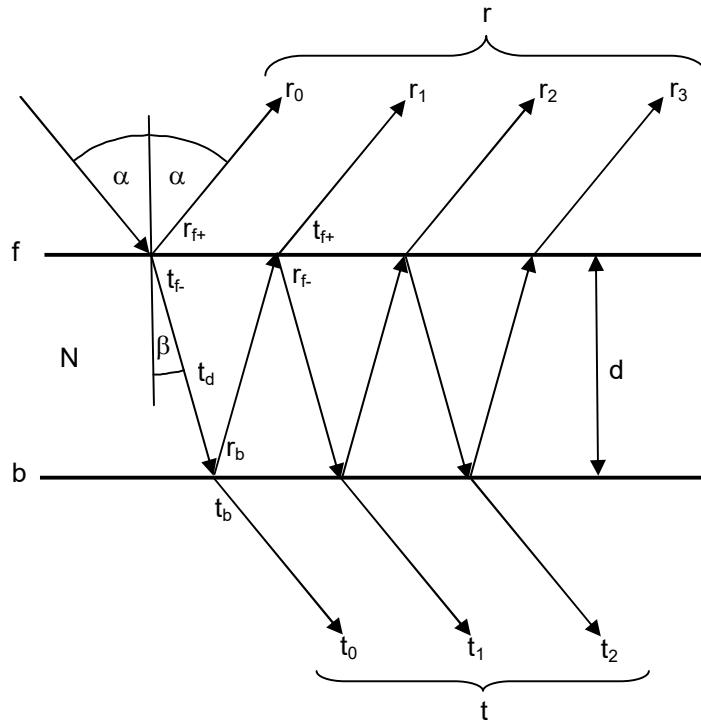


Fig. 14: Reflection  $r$  and transmission  $t$  of a thin solid plate with thickness  $d$  and refractive index  $N$

$$r = r_0 + \sum_{k=0}^{\infty} t_{f-} \cdot t_{f+} \cdot t_d^2 \cdot r_b \cdot (r_b \cdot r_{f-} \cdot t_d^2)^k = r_0 + \frac{t_{f-} \cdot t_{f+} \cdot t_d^2 \cdot r_b}{1 - r_b \cdot r_{f-} \cdot t_d^2} \quad (35)$$

If we consider the refractive index outside the solid plate as 1, then the transmission and reflection coefficients in equation (35) for perpendicular and parallel polarization can be written as follows:

$$t_{f-,S} = \frac{2 \cdot \cos(\alpha)}{\cos(\alpha) + \sqrt{N^2 - \sin^2(\alpha)}} \quad t_{f-,P} = \frac{2 \cdot N \cdot \cos(\alpha)}{N^2 \cdot \cos(\alpha) + \sqrt{N^2 - \sin^2(\alpha)}} \quad (36)$$

$$t_{f+,S} = \frac{2 \cdot \sqrt{N^2 - \sin^2(\alpha)}}{\cos(\alpha) + \sqrt{N^2 - \sin^2(\alpha)}} \quad t_{f+,P} = \frac{2 \cdot N \cdot \sqrt{N^2 - \sin^2(\alpha)}}{N^2 \cdot \cos(\alpha) + \sqrt{N^2 - \sin^2(\alpha)}} \quad (37)$$

$$r_{b,S} = r_{f-,S} = \frac{\sqrt{N^2 - \sin^2(\alpha)} - \cos(\alpha)}{\cos(\alpha) + \sqrt{N^2 - \sin^2(\alpha)}} \quad r_{b,P} = r_{f-,P} = \frac{\sqrt{N^2 - \sin^2(\alpha)} - N^2 \cdot \cos(\alpha)}{N^2 \cdot \cos(\alpha) + \sqrt{N^2 - \sin^2(\alpha)}} \quad (38)$$

$$t_d = e^{i \cdot \Phi} = e^{i \cdot 2 \cdot \pi \cdot d \cdot f \cdot \sqrt{N^2 - \sin^2(\alpha)} / c} \quad (39)$$

An approximation of  $t_d$  in case of  $k \ll n$  is:

$$t_d = e^{i \cdot \Phi} = e^{\left( \frac{2 \cdot \pi \cdot d \cdot f \cdot \sqrt{n^2 - \sin^2(\alpha)}}{c} \cdot i \right)} \cdot e^{\left( \frac{-2 \cdot \pi \cdot d \cdot f \cdot n \cdot k}{c \cdot \sqrt{n^2 - \sin^2(\alpha)}} \right)} \quad \text{for } k \ll n \quad (40)$$

The measured THz pulse in the time domain can be Fourier transformed and then compared with the absolute spectral reflection coefficient  $|r(f)|$  in equation (35).



### Example

An example for a measured reflection  $|r_s(f)|$  on a 1 mm thick glass plate is shown in figure 15 below.

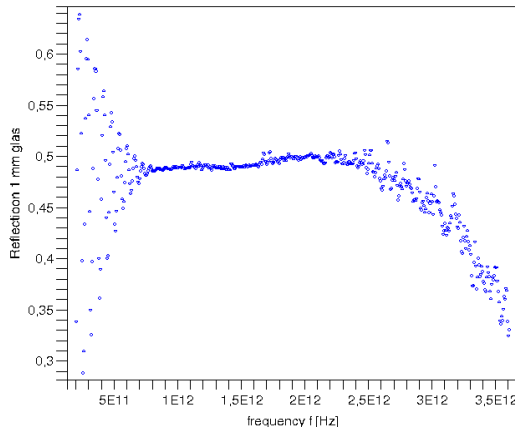


Fig. 15a Measured spectral reflection  $|r_s(f)|$  on a 1 mm thick glass plate. The incidence angle is  $\alpha=30^\circ$

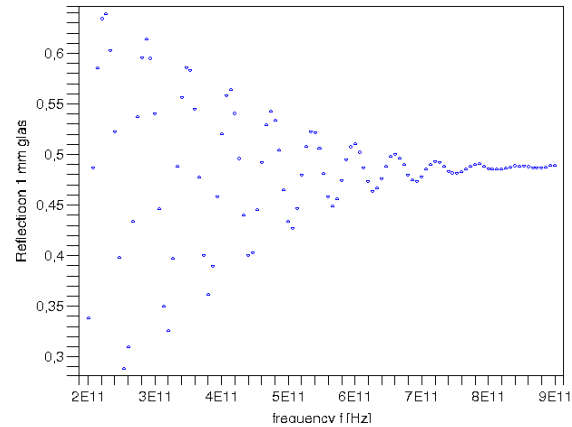


Fig. 15b Low frequency part of measured spectral reflection  $|r_s(f)|$  on a 1 mm thick glass plate.

In the measured spectral region up to 3.7 THz only one resonance absorption dip can be seen. Therefore the dielectric function can be formed with only one set of parameters  $\epsilon_\infty$ ,  $f_0$ ,  $f_p$  and  $\gamma$  in equation (9).

The needed start parameter set for a subsequent curve fitting can be found in two consecutive steps.

#### Step 1:

At first a dispersion free complex refractive index  $N = n + i \cdot k$  can be estimated in the following way:

- a) The real part  $n$  of the refractive index  $N$  can be estimated from the interference pattern in the low frequency region, which is shown in fig. 15b. These interferences are only possible if the material absorption is low, so that the THz pulse undergoes several traces between the boundaries of the glass plate. We can determine  $n$  from these interferences by neglecting  $k$  using equation (40) with the following formula:

$$n = \sqrt{\left(\frac{c}{2 \cdot d \cdot \Delta f}\right)^2 + \sin^2(\alpha)} \quad (41)$$

One period  $\Delta\Phi=2\cdot\pi$  in fig. 15b is equivalent to a frequency period  $\Delta f = 61.6$  GHz. With  $d=1$  mm and  $\alpha=30^\circ$  we can calculate using equation (41)  $n= 2.49$ .

Because the measured spectral reflectance  $|r_s(f)|$  is not constant at higher frequencies  $> 1$  THz there must be a significant dispersion of the refractive index  $N(f)$ . A starting value for  $k$  can be estimated in two different ways

- b) From the frequency dependent decrease of the interference pattern a start value for  $k$  can be estimated in the following way:

We consider only the maxima of the interferences at frequencies, where the real part of the phase  $\Phi$  in equation (39) is equal to  $(1+2\cdot m)\cdot\pi$  ( $m=0,1,2,\dots$ ) and we neglect the imaginary part  $k$  of the refractive index  $N$  in the transmission coefficients  $t_f$ ,  $t_{f+}$  and in the reflection coefficients  $r_b=r_f$  in equations (36) – (38). Then the influence of  $k$  on the reflectance  $|r_s(f)|$  is only described with the transmission  $t_d$  in equation (40) and we get

$$|r_m| = \frac{\cos(\alpha) - \sqrt{N^2 - \sin^2(\alpha)}}{\cos(\alpha) + \sqrt{N^2 - \sin^2(\alpha)}} \frac{4 \cdot \cos(\alpha) \cdot \sqrt{n^2 - \sin^2(\alpha)} \cdot (\sqrt{n^2 - \sin^2(\alpha)} - \cos(\alpha)) \cdot e^{\left(\frac{-4 \cdot \pi \cdot d \cdot f \cdot n \cdot k}{c \cdot \sqrt{n^2 - \sin^2(\alpha)}}\right)}}{\left(\sqrt{n^2 - \sin^2(\alpha)} + \cos(\alpha)\right)^3 \cdot \left(1 + \frac{\left(\sqrt{n^2 - \sin^2(\alpha)} - \cos(\alpha)\right)^2 \cdot e^{\left(\frac{-4 \cdot \pi \cdot d \cdot f \cdot n \cdot k}{c \cdot \sqrt{n^2 - \sin^2(\alpha)}}\right)}}{\left(\sqrt{n^2 - \sin^2(\alpha)} + \cos(\alpha)\right)^2}\right)} \quad (42)$$

Using this relation  $k$  can be estimated from a maximum value of  $|r_m(f)|$  with the following formula:

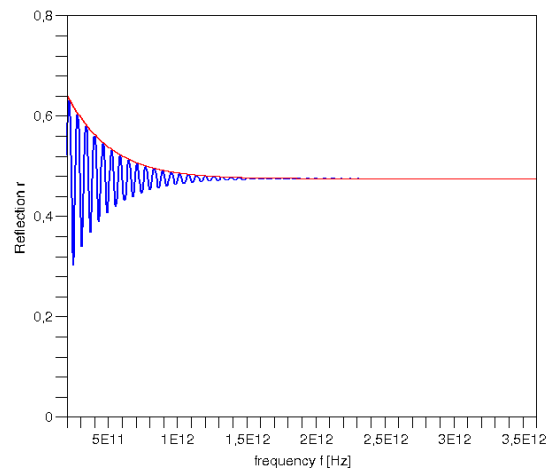
$$k = - \frac{\ln \left( \frac{\cos^2(\alpha) - r_m \cdot \sin^2(\alpha) - n^2 + \sin^2(\alpha) + r_m \cdot n^2 + 2 \cdot \sqrt{n^2 - \sin^2(\alpha)} \cdot r_m \cdot \cos(\alpha) + r_m \cdot \cos^2(\alpha)}{r_m \cdot \sin^2(\alpha) - r_m \cdot n^2 + n^2 - \sin^2(\alpha) + 2 \cdot \sqrt{n^2 - \sin^2(\alpha)} \cdot r_m \cdot \cos(\alpha) - \cos^2(\alpha) - r_m \cdot \cos^2(\alpha)} \right) \cdot c \cdot \sqrt{n^2 - \sin^2(\alpha)}}{4 \cdot \pi \cdot d \cdot f \cdot n} \quad (43)$$

From measurement with  $r_m=0.543$  at  $f=0.473$  THz,  $\alpha=30^\circ$ ,  $d=1$ mm and  $n=2.49$  we get  $k=0.0817$ .

- c) The dispersion results in a maximum reflectance  $|r_s(f_{\max})| = 0.495$  at the frequency  $f_{\max} = 2$  THz. For an estimated value of  $k$  we can calculate  $n$  by using equation (28) with the reflection value at  $f_{\max}$ . We interpret the difference between the refractive index calculated at  $f_{\max}$  and from the interferences in the low frequency region as  $k$ . This results in  $k = 2.61 - 2.49 = 0.11$ .

The result of the calculated spectral reflection with dispersion free refractive index  $N=2.49 + i \cdot 0.0817$  using equation (34) and (41) is shown in figure 16.

Fig. 16: Calculated spectral reflectance of a glass plate with the parameters  $d=1$ mm,  $\alpha=30^\circ$ ,  $N=2.49 + i \cdot 0.0817$ , perpendicular polarization using equation (35) (blue) and (42) (red).



Step 2:

Using the estimated  $n$  and  $k$  values from step 1 the parameters  $\varepsilon_\infty$ ,  $f_0$ ,  $f_P$  and  $\gamma$  in equation (9) must be estimated to start the curve fit with equation (34). The start parameters can be found as follows:

- The value of  $\varepsilon_\infty$  is between 1 and  $n^2$ . A good start value is  $\varepsilon_\infty = n$ . In this case some dispersion is included. In the example above it means  $\varepsilon_\infty = 2.49$ .
- The resonance frequency  $f_0$  must be between the frequency  $f_{\max} = 2$  THz for maximum reflectance and the frequency  $f_{\min} = 3.7$  THz for minimum reflectance. We estimate with

$$f_0 = f_{max} + \frac{f_{min} - f_{max}}{2} \quad (45)$$

$f_0 = 2.85$  THz.

- The damping constant  $\gamma$  can be estimated from the resonance width  $\gamma = f_{min} - f_{max} = 1.7$  THz.
- The plasma frequency  $f_P$  must be chosen in such a way, that the right value for  $n$  results in eq. (9). This condition can be fulfilled with equation (23) above.

For the 1 mm thick glass sample with start values for  $n = \epsilon_\infty = 2.49$ ,  $k = 0.0817$ ,  $f_0 = 2.35$  THz, as above and  $f = f_L = 0.473$  THz the start value for  $f_P$  can be calculated with (23) as  $f_P = 5.43$  THz.

A curve fit using formulas (2), (3) and (35) with the measured reflection  $|r_S(f)|$  values results in the fitted parameters  $\epsilon_\infty = 2.93$ ,  $f_0 = 2.66$  THz,  $f_P = 4.58$  THz,  $\gamma = 2.14$  THz. To get a better coincidence between the measured and calculated values the glass thickness must be corrected from 1 mm to 0.983 mm. The calculated functions  $|r_S(f)|$ ,  $N(f)$  and  $\epsilon(f)$  are shown in figures 17 and 18.

It is clearly seen in fig. 16, that the imaginary part  $k$  of the refractive index cannot be described using a fixed value as is assumed above in a first approximation. The reason is the large absorption at the resonance frequency  $f_0 = 2.66$  THz.

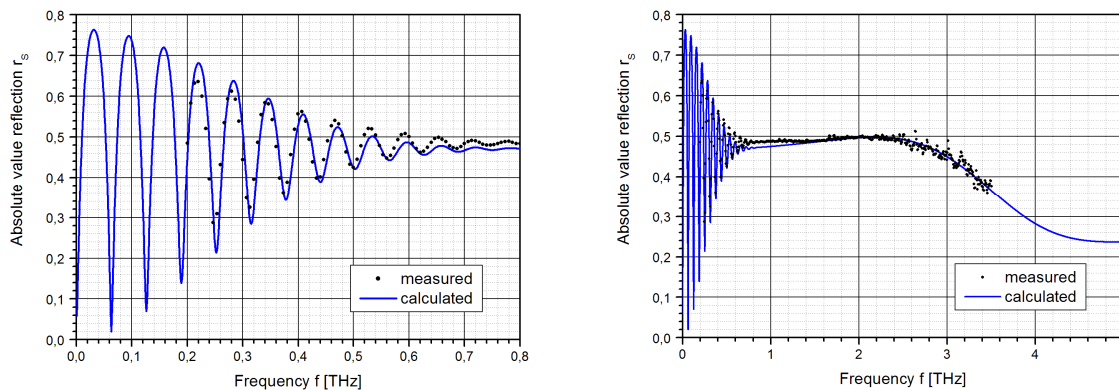


Fig 17: Calculated reflection  $|r_S(f)|$  of a 1 mm thick glass plate with the dispersion parameters  $\epsilon_\infty = 2.93$ ,  $f_0 = 2.66$  THz,  $f_P = 4.58$  THz,  $\gamma = 2.14$  THz after curve fitting using equations (2), (3) and (35).

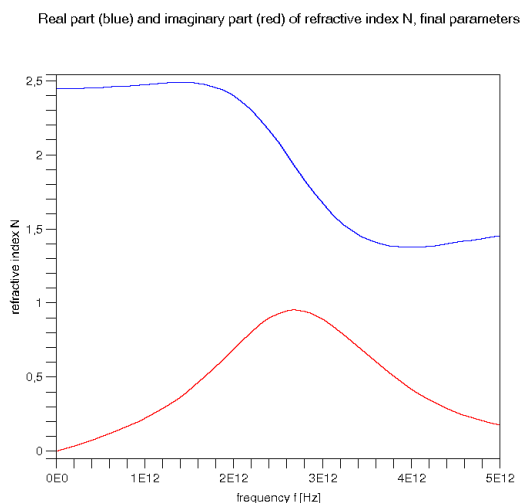


Fig. 18a Calculated function  $N(f)$

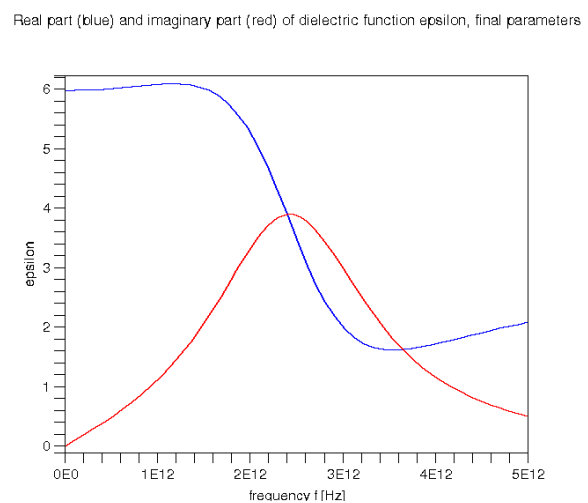


Fig. 18b Calculated dielectric function  $\epsilon(f)$