THz Pulse Duration Influence on High Energy Level Excitation Due to Cascade Mechanism

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Abstract. We study influence of an incident broadband THz pulse duration on the spectral features of a signal transmitted through/reflected from a substance covered by a disordered structure by means of computer simulation. It is well-known that under real conditions, the results of a standard THz TDS undergo various factors. For example, a substance under investigation can be put into a bulk medium with ordinary properties. This often results in the distortion of the reflected/transmitted pulse spectra and hence, one may reveal additional absorption frequencies which can be thought as belonging to a dangerous substance. An issue from this situation may be a substance emission spectrum using. As we showed the emission frequencies appear due to the cascade mechanism of higher energy level excitation. In this paper we study the incident THz pulse duration influence on the emission frequencies manifestation.

1. Introduction
At present, the development of computer and optical technology offers novel opportunities for the creation of new sensors and systems providing a sufficient accuracy for practice in infrared (IR) and terahertz (THz) ranges of frequencies. Their usage is promising way since it provides a visualization and spectral analysis of an object under investigation without contact with it (so called, nondestructive control), and therefore, devices based on using the IR and THz ranges of frequencies can be applied for quality control in various industries and for security applications [1]. It is well-known that an IR spectroscopy has been till now for the quality control of meat products [2, 3], milk [4], other food [5] and for a control of human health. It is also employed in other areas, for example to analyze the composition of the air [6], analysis of composite materials [7]. Because the characteristic frequencies of various explosives and dangerous substances lie in the THz and IR regions of the spectrum [8–14], then this opens many opportunities for new security systems developing.

There are examples of practical realization of Raman spectroscopy devices measuring IR spectra of substances concealed by covers [15–17]. However, an improvement of devices for the nondestructive detection of substances is of great importance. In connection with that the THz radiation high penetration ability through clothes and ordinary materials is worth attention [18, 19] because the THz range of frequencies allows us to perform measurements through comparably dense or thick covers. Nevertheless, it is desirable to extend distances between an object under investigation and measurement device.

To overcome the crucial difficulties of a remote substance identification it is urgent to solve some problems. One of them arises from a fact that the THz radiation wavelength lies in
the range of 0.3-3 mm. However, the macroscopic peculiarities of the sample structure, such as density fluctuations or granule sizes, belongs also to this range. This leads to a strong modulation of the THz pulse interaction with the sample. Moreover, an object surface roughness can result in non-uniform scattering depending on the radiation wavelength [20,21]. One more difficulty is the presence of the object dielectric permittivity. For example, there are layers (or domains) with different densities and, consequently, different permittivities in the object under investigation. This can result, obviously, in frequency-dependent reflection from the whole object. Let us note that a paper, which often covering the sample under investigation, can act as disordered structure [22]. Therefore, the covering distorts THz spectra, when a remote measurement is made to provide the identification and detection of substance [23].

Early, we have demonstrated the existence of the cascade mechanism of high energy level excitation at a THz pulse action on substance and this leads to the higher frequency appearance in the spectrum of a pulse transmitted through or reflected from a substance [24, 25]. These emission frequencies gives us a more reliable information about the chemical structure of the irradiated object. Therefore, a detailed analysis of the various factors, which influences on the emission frequency brightness, is of the great interest. In this paper we numerically investigate an influence of one key factor - the pulse duration - on the cascade mechanism of high energy level excitation for a medium with multiple transition frequencies.

2. Problem statement

We carry out computer simulation of the THz pulse propagation in a finite medium layer possessing non-linear non-instantaneous response. Initially, the pulse is located in a vacuum, then it falls on the left boundary of the medium, transmits through it, and goes out of the medium in a vacuum. This process is accompanied by a partial reflection of the pulse from the medium boundaries, and a part of the THz pulse appears to be reflected in the opposite direction. Below we discuss the spectra of the pulses transmitted through the medium as well as reflected from it. To describe an electromagnetic pulse propagation in a multilevel medium we use the multilevel Maxwell-Bloch system of equations in dimensionless form:

\[
\frac{\partial H}{\partial t} = -\frac{\partial E}{\partial z}, \tag{1}
\]

\[
\frac{\partial D}{\partial t} = -\frac{\partial H}{\partial z}, \tag{2}
\]

\[
D = E + 4\pi P, \tag{3}
\]

\[
\frac{\partial \rho_{mn}}{\partial t} = -\left(\gamma_{mn} + i\omega_{mn}\right) + i\alpha E \sum_{q} (d_{mq}\rho_{qn} - \rho_{mq}d_{qn}), \tag{4}
\]

\[
\frac{\partial \rho_{mm}}{\partial t} = -\sum_{q} (W_{mq}\rho_{mm} - W_{qm}\rho_{qq}) + i\alpha E \sum_{q} (d_{mq}\rho_{qm} - \rho_{mq}d_{qm}), \tag{5}
\]

\[
P = \sum_{mn} (d_{mn}\rho_{mn}). \tag{6}
\]

Here, \(E\) and \(H\) are the electric and magnetic field strength correspondingly. \(D\) is the electric field induction, \(\rho_{mn}\) is a density matrix element. Diagonal elements of the density matrix characterize the number of medium molecules belonging to the corresponding energy level. Non-diagonal elements describe correlations between different energy levels and define the polarization of the medium (6). \(\omega_{mn}\) is the matrix of frequencies corresponding to the energy level transition between \(m\) and \(n\) levels. \(d_{mn}\) is the dipole moment of the corresponding transition. \(\gamma_{mn}\) is the relaxation rate of a non-diagonal matrix element, which defines the medium polarization relaxation. This parameter is varied below with the aim to study its influence on the cascade
mechanism of molecule high energy level excitation. \( W_{mn} \) is the population relaxation rate. Typically \( W_{mn} \) is several orders of magnitude smaller than \( \gamma_{mn} \) and thus, below we consider it to be equal to 0. \( \alpha \) is a dimensionless constant, which characterizes the electromagnetic field-matter interaction (that is a medium polarizability) and it depends on the dimensional physical parameters of both the pulse and the medium:

\[
\alpha = \frac{4\pi M d_0^2}{\hbar \omega_0}.
\]  

In (7) \( M \) is the concentration of molecules in the medium, \( d_0 \) and \( \omega_0 \) are the characteristic values of the dipole moment and the carrier frequency of a wave packet measured in Debye and THz units correspondingly. The choice of the parameter values will be discussed below.

The pulse propagation through the vacuum is described by the classic Maxwell equations (1-3 with \( P = 0 \)).

The incident pulse shape is taken as a Gaussian one with cosine filling:

\[
E(z, t = 0) = H(z, t = 0) = E_0 \exp \left( \frac{-(z-z_0)^2}{\tau_p^2} \right) \cos(\omega_p(z-z_0)),
\]

which falls on the medium. Here \( E_0 \) is the initial field amplitude, \( z_0 \) is the pulse centre position, which is chosen so that the whole pulse is located in vacuum initially, \( \tau_p \) is the pulse duration (equal to its length in dimensionless units), \( \omega_p \) is the carrier frequency of the wave packet. Such initial conditions describe the pulse propagation in the positive direction of \( z \)-axis. The length of the pulse is chosen so that it contains about 5 or 6 oscillation periods in it.

Molecules of the medium are initially in the ground state:

\[
\rho_{11} = 1; \quad \rho_{mn} = 0, (mn) \neq (11).
\]

These conditions correspond to the absolute zero of temperature. However, for investigation of the cascade mechanism appearance we consider this case corresponding to ”pure conditions” of its observation. At the room temperature the effect will become more pronounced, but the interpretation of our results would be much more complicated because of a number of possible energy level transitions.

### 3. Computer simulation results and discussion

We consider the medium with six energy levels, three of which have similar transition frequencies from the ground level of molecules. The incident pulse duration varies in such a way that the pulse spectra corresponding to various pulse durations differ noticeably.

The computer simulation dimensionless parameters are given in the table 1. These parameters do not associate with a particular substance but they are similar to characteristic values commonly used in the THz range of frequencies. Namely, they correspond to a pulse with the carrier cyclic frequency of 0.87 THz, duration of about 6 ps and peak intensity about of \( 10^6 \text{W/cm}^2 \) propagating through a thin ( 2 mm) medium layer with transition frequencies up to 2 THz.

The incident pulses and their spectra are depicted in the figures 1 and 2. The frequencies of energy level transitions are marked in the same pictures. It can be seen that the spectra corresponding to different pulse durations have various amplitudes at the transition frequencies, most notably, at the frequencies marked 1-2, 1-3, 1-4.

The chosen parameters correspond to negligible reflection from the medium. Therefore, the effective dielectric permittivity is close to that of the vacuum. Thus, the reflected signal consists only of the medium re-emission signal. Obviously, this situation corresponds to our
Table 1. The parameters of the numerical simulations.

| Parameter | Value |
|-----------|-------|
| $\omega_{mn}$ | $(0 -4 -4.5 -5.5 -11 -15)\begin{pmatrix} 4 & 0 & -0.5 & -1.5 & -7 & -11 \\ 4.5 & 0.5 & 0 & -1 & -6.5 & -10.5 \\ 5.5 & 1.5 & 1 & 0 & -5.5 & -9.5 \\ 11 & 7 & 6.5 & 5.5 & 0 & -4 \\ 15 & 11 & 10.5 & 9.5 & 4 & 0 \end{pmatrix}$ |
| $d_{mn}$ | 0.0075 $\begin{pmatrix} 0 & 1 & 1 & 1 & 1 & 1 \\ 1 & 0 & 1 & 1 & 5 & 5 \\ 1 & 1 & 0 & 1 & 5 & 5 \\ 1 & 1 & 1 & 0 & 5 & 5 \\ 1 & 5 & 5 & 5 & 0 & 25 \\ 1 & 5 & 5 & 5 & 25 & 0 \end{pmatrix}$ |
| $\gamma_{mn}$ | $\frac{\omega_{mn}}{50}$ |
| $L_s$ | 6 |
| $\alpha$ | 67.54 |
| $E_0$ | 0.05 |
| $\omega_p$ | 5 |
| $\tau_p$ | 1.5 1.8 2.7 4.5 |

Figure 1. Incident pulses with different durations.

Figure 2. The incident pulse spectra. Vertical lines mark the transition frequencies between energy levels of the medium.

aim. Nevertheless, it would be possible to add a non-resonant permittivity to our model. But in this case the computation result interpretation will become complicate. Hence, we pay our attention to the signal which transmit through the medium layer and then it propagates in positive direction of z - axis.

In the figure 3 the spectra of the reflected signals are shown. We see that the reflected pulse spectrum possesses maxima with highly varied amplitudes in different ranges of frequencies. Therefore, the figure 4 depicts the spectra in the central area of the spectrum $\omega = [3, 7]$. In this area of the spectrum the incident pulse spectra are localized. As a result of the pulse interaction
with the medium, the spectra are transformed in comparison with their initial shapes, namely minima (absorption lines) appear. In spectroscopy the appearance of these lines can be an argument for determining the chemical structure and classifying the substance.

The pulses of all durations exhibit these minima at the frequencies of 4.5 and 5.5, but the maximal one ($\tau_p = 4.5$) has the spectrum which is so narrow, that it only bends and no minima are observed in it. Since the spectra of the shorter pulses are broad enough, the absorption line at the frequency 4 is present in each of them.

As a result of the electromagnetic field energy absorption, the medium is excited, its molecules transit to higher energy levels from the ground one due to the cascade mechanism of their excitation. From higher energy levels molecules transit to lower ones. This process is accompanied by the emission of radiation at a corresponding frequency. In a multilevel medium there is a lot of possible transition series. Thus, the medium can re-emit the absorbed energy at frequencies different from those they were absorbed.

The figure 5 depicts the pulse spectra in the range $[0, 3]$ where such spectral intensity maxima are located. It is worth noticing that the dependence of maximal spectral intensities on the
duration of the incident pulse has a non-monotonous character. Namely, for the energy level transitions $2\rightarrow 3$ and $2\rightarrow 4$ we see the highest spectral intensity if the pulse with the shortest duration falls on a medium. However, the spectral intensity corresponding to transition $3\rightarrow 4$ achieves a maximum intensity if the pulse duration is equal to 2.7 dimensionless units. The fact is remarkable that the propagation of the pulse with maximal duration results in a spectrum with almost negligible peak $2\rightarrow 4$.

![Figure 5. The spectra of transmitted pulses. Low frequency range. Vertical dotted lines mark transition frequencies in the medium.](image)

Let us discuss the spectral line appearance in the range of frequencies which are higher than those belonging to the incident pulse spectrum. During the interaction process, molecules of the medium are excited to upper energy levels, and then they can relax down to neighboring energy levels and emits the radiation as discussed before. After that they can relax to the ground energy level and emit at the same frequencies at which the THz pulse energy absorption takes place (Fig. 3). On the other hand, the few-cycle pulse is characterized by its broad spectrum and, moreover, the energy levels in the THz frequency range are located quite densely, having transition frequencies rather close to each other. Thus, the absorption of THz photon sequence is not impossible in the medium under consideration. This is why we name this absorption mechanism as the cascade mechanism of high energy level excitation.

In the figure 6 the transmitted pulse spectra are shown in the range of frequencies $[9\rightarrow 17]$. The influence of the incident pulse duration is obvious. Between the frequencies 9 and 12 a pair of overlapping spectral maxima is noticeable. It can be seen that the maximal spectral intensity for these maxima changes with the incident pulse duration. It should be also emphasized that the rightmost of those peaks overlaps two different frequencies corresponding to two energy level transitions, namely, $(10.5$ and $11)$. Therefore, spectrum shape is dependent on the incident pulse duration. For the longer pulses the peak tends to be centred at the frequency 10.5. For shorter ones the peak center is closer to 11.

The last peak, corresponding to the transition between the energy levels 6 and 1, changes its height depending on the pulse duration as well, but due to its separate location, its shape is mostly unaffected.

**4. Conclusion**

We have studied the influence of the THz pulse duration on the pulse interaction with the multilevel medium layer. We demonstrated a significant influence of the pulse duration on the transmitted signal spectrum. Medium emission at higher and lower frequencies are the most
remarkable. Depending on the pulse duration the spectral lines can change its intensity or disappear altogether. Therefore, this dependence is not monotonic. Thus, the pulse must be taken into account in spectroscopic experiments in order to provide the correct characterization of the studied medium.

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