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RESEARCH ARTICLE

A direct method to calculate second-order two-dimensional terahertz spectroscopy in frequency-domain based on classical theory

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Abstract Previous theoretical researches on the two-dimensional terahertz spectroscopy (2DTS), which are conducted via inefficiently time-consuming numerical simulation, deal with only single-mode system. To overcome the limitations, we derive a classical-theory-based analytical solution which is applicable to multi-modes system. Three typical weak sources of nonlinearities are introduced. The findings suggest that the analytical results correspond well with those obtained by the traditional numerical simulation. Thus the study provides a more efficient and practical method to directly calculate 2DTS, and, in a broader sense, sheds new light on the theory of 2DTS.

Keywords two-dimensional spectroscopy, terahertz, classical method

1 Introduction

Two-dimensional (2D) spectroscopy is an important tool to study the dynamics of vibrational excitation of matter [1]. It provides richer information than one-dimensional (1D) spectroscopy by which it is difficult to obtain the nonlinear information of materials. In 1D time-resolved spectroscopy, the signal emitted from the interaction of a single light pulse with materials is measured in time-domain, and the sample's intrinsic absorption spectrum can be acquired via Fourier transform (FT). If another pulse with an interval T to the former one is also incident on the sample, a nonlinear signal is emitted, which is determined as a function of detection time t and interval T. If we scan t and T, the 2D-FT of the signal yields two frequency dimensions, detection frequency ω_t and excitation frequency ω_T [2], thus the spectroscopy is a 2D one.

2D spectroscopy has been widely applied in infrared region to study inhomogeneous distribution of resonance frequencies, anharmonicity of intra- and intermolecular vibrational modes, and the coupling between different modes, among other things [1,3–9]. Because of the advent of intense terahertz (THz) pulse sources [10–12], two-dimensional terahertz spectroscopy (2DTS) is now available, revealing more nonlinear characteristics of materials in THz region [1,3,4,13–15].

The physical model of 2DTS can be briefly explained as follows. As is shown in Fig. 1, two successive light pulses E_1 and E_2 are collinearly incident on samples at time $t = -T$ and $t = 0$ respectively, thus inducing second-order nonlinear polarization $P_{\mathrm{NL}}(t,T)$ which in turn leads to a nonlinear electric field $E_{\mathrm{NL}}(t,T)$. Using 2D-FT in t and T dimensions, we can acquire the corresponding spectrum in frequency-domain as a function of detection frequency ω_t and excitation frequency ω_T.

In experiments, the interval T between E_1 and E_2 can be easily controlled by an optical delay line. For each certain interval T, the nonlinear signal E_{NL} is measured as a function of real time t, thus the measurements for different T yield a 2D function $E_{\mathrm{NL}}(t,T)$. After 2D-FT, the experimental 2DTS is obtained [4,14,15].

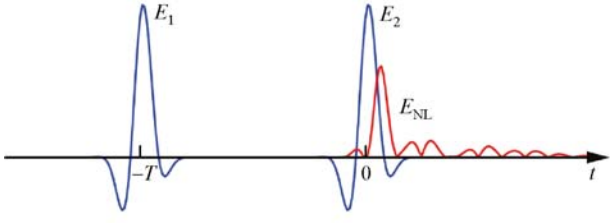

Fig. 1 Timing schematic diagram of 2DTS

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To distinguish the contributions from different sources of nonlinearities, one needs to perform theoretical calculation to fit the measured results. In THz region, the THz photon energy $\hbar\omega$ is so low that $\hbar\omega \ll k_B T_s$ (k_B is the Boltzmann constant and T_s is the temperature of the sample). In another word, 2DTS system has a classical nature. Therefore, current theoretical analysis on 2DTS has been majorly based on Lorentz model in the framework of classical theory [1,16–18]. Despite their significance, these researches are narrow-ranged as they deal with only single-mode systems, and the derivation results are given as nonlinear polarization $P_{NL}(t,T)$ in terms of multiple integral. Specifically, there are three limitations: first, the multiple integral is time consuming; secondly, numerical 2D-FT needs to be performed to transform the results into frequency-domain; and thirdly, excessive effort is required to obtain the nonlinear electric field.

To overcome the limitations, we propose a classical theory based analytical method which not only can be used to calculate 2DTS directly and efficiently, but also is applicable to multi-mode system.

2 Theoretical derivation

The derivation of our analytical solution is explained as follows.

Assuming E_1 and E_2 are parallel, the total incident field is the sum of two pulses:

$$E(t,T) = E_2(t) + E_1(t+T). \tag{1}$$

The system is regarded as summation of a series of oscillators described by Lorentz model. For a N-mode system, the linear equation of motion of the ith vibrational mode is

$$\frac{\mathrm{d}^2 r_i}{\mathrm{d}t^2} + \gamma_i \frac{\mathrm{d}r_i}{\mathrm{d}t} + \omega_i^2 r_i = \frac{1}{m_i} q_i E(t,T), \tag{2}$$

where r_i, γ_i, m_i, ω_i and q_i are respectively the vibrational coordinate, damping constant, mass, resonance frequency and effective charge of the ith mode. The solution of this equation is

$$r_i(t,T) = q_i S_i(t) * E(t,T), \tag{3}$$

where $S_i(t)$ is

$$S_i(t) = \frac{1}{m_i \varepsilon_i} \mathrm{e}^{-\frac{1}{2}\gamma_i t} \sin(\varepsilon_i t),\ \varepsilon_i = \sqrt{\omega_i^2 - \gamma_i^2/4},\ (t>0), \tag{4}$$

and $*$ is convolution in t dimension. The FT of $S_i(t)$ is

$$\tilde{S}_i(\omega_t) = \frac{1}{m_i} \frac{1}{\omega_i^2 - \mathrm{j}\gamma_i \omega_t - \omega_t^2}, \tag{5}$$

where j is the imaginary unit.

The induced polarization can be expressed as

$$P(t) = \rho \sum_{i=1}^{N} p_i = \rho \sum_{i=1}^{N} q_i r_i, \tag{6}$$

where ρ is the density of the molecule and p_i is the effective dipole moment of the ith mode.

According to the perturbation method, r_i and P are expanded in the order of E as

$$r_i = r_i^{(1)} + r_i^{(2)},\ P = P^{(1)} + P^{(2)},\ E = E^{(1)} + E^{(2)}, \tag{7}$$

where $r_i^{(1)}$ and $P^{(1)}$ are the linear terms, while $r_i^{(2)}$ and $P^{(2)}$ are the second-order nonlinear terms, and $r_i^{(n)}$, $P^{(n)} \propto (E^{(1)})^n$. The electric fields are determined by

$$\left[\frac{\partial^2}{\partial z^2} + \mu_0 \omega_t^2 \left(\varepsilon_0 + \rho \sum_{i=1}^{N} q_i \tilde{S}_i(\omega_t)\right)\right] \tilde{E}^{(1)}(z,\omega_t) = 0, \tag{8}$$

$$\left[\frac{\partial^2}{\partial z^2} + \mu_0 \omega_t^2 \left(\varepsilon_0 + \rho \sum_{i=1}^{N} q_i \tilde{S}_i(\omega_t)\right)\right] \tilde{E}^{(2)}(z,\omega_t,\omega_T)$$

$$= -\mu_0 \omega_t^2 \tilde{P}^{(2)}(z,\omega_t,\omega_T), \tag{9}$$

where ε_0, μ_0 and z are respectively the permittivity of vacuum, permeability of vacuum and propagation distance in the medium.

In general, there are three typical second-order nonlinearities: anharmonicity (AH), nonlinear coupling (NC), and nonlinear damping (ND) in 2DTS [17]. AH corresponds to the nonlinearity rising from vibrational coordinate dependence of the resonance frequency.

Mathematically, AH is introduced by the addition of a third-order term to the harmonic potential function of the vibrational mode as

$$V(r) = \sum_{i=1}^{N} \frac{1}{2} m_i \omega_i^2 r_i^2 + \frac{1}{3!} \sum_{i,j,k=1}^{N} B_{ijk} r_i r_j r_k, \tag{10}$$

where $B_{ijk} = \partial^3 V(r)/\partial r_i \partial r_j \partial r_k$ is the coupling parameter. The $r_i r_j$ term does not exist since it can be removed by simply changing the set of modes [19]. Thus, the $\omega_i^2 r_i$ term in Eq. (2) becomes

$$\omega_i^2 r_i \rightarrow \frac{1}{m_i} \frac{\partial}{\partial r_i} V = \omega_i^2 r_i + \frac{1}{2m_i} \sum_{j,k=1}^{N} B_{ijk} r_j r_k. \tag{11}$$

By selecting terms proportional to the same order of E, we obtain the equations for them as

$$\frac{\mathrm{d}^2 r_i^{(1)}}{\mathrm{d}t^2} + \gamma_i \frac{\mathrm{d}r_i^{(1)}}{\mathrm{d}t} + \omega_i^2 r_i^{(1)} = \frac{1}{m_i} q_i E^{(1)}(t,T), \tag{12}$$

$$\frac{\mathrm{d}^2 r_i^{(2)}}{\mathrm{d}t^2} + \gamma_i \frac{\mathrm{d}r_i^{(2)}}{\mathrm{d}t} + \omega_i^2 r_i^{(2)} = \frac{1}{2m_i} \sum_{j,k=1}^{N} B_{ijk} r_j^{(1)} r_k^{(1)}. \tag{13}$$

By solving Eqs. (12) and (13) with only cross terms of E_1 and E_2 kept, we achieve the solution of the second-order polarization in time-domain as

$$P^{(2)}_{\mathrm{AH}}(t,T) = -\rho\sum_{i,j,k=1}^{N} B_{ijk}q_iq_jq_kS_i(t)$$

$$\{[S_j(t)*E_2(t)][S_k(t)*E_1(t+T)]\}. \quad (14)$$

This expression agrees with those in Refs. [1,16–18] when $N=1$. According to Eqs. (8) and (9), the induced second-order electric field is

$$\tilde{E}^{(2)}_{\mathrm{AH}}(z,\omega_t,\omega_T)$$

$$= -\tilde{R}^{(2)}\sum_{i,j,k=1}^{N} B_{ijk}q_iq_jq_k\tilde{S}_i(\omega_t)\tilde{S}_j(\omega_t-\omega_T)\tilde{S}_k(\omega_T), \quad (15)$$

where

$$\tilde{R}^{(2)} = \frac{\rho\mu_0\omega_t^2}{\Delta k^{(2)}[\Delta k^{(2)}+2k(\omega_t)]}\tilde{E}_2(\omega_t-\omega_T)\tilde{E}_1(\omega_T)$$

$$(\mathrm{e}^{\mathrm{j}\Delta k^{(2)}z}-1)\mathrm{e}^{\mathrm{j}k(\omega_t)z}, \quad (16)$$

$$\Delta k^{(2)} = k(\omega_t-\omega_T)+k(\omega_T)-k(\omega_t), \quad (17)$$

$$k(\omega_t) = \sqrt{\mu_0\omega_t^2\left(\varepsilon_0+\rho\sum_{i=1}^{N}q_i\tilde{S}_i(\omega_t)\right)}. \quad (18)$$

For NC, the dipole moment is expressed as

$$p = \sum_{i=1}^{N}q_ir_i+\frac{1}{2}\sum_{i,j=1}^{N}q_{ij}r_ir_j, \quad (19)$$

where $q_{ij} = -\partial^2 pE/\partial r_i\partial r_j$ is the coupling parameter. Thus, the q_iE/m_i term in Eq. (2) becomes

$$\frac{1}{m_i}q_iE\rightarrow\frac{1}{m_i}\frac{\partial}{\partial r_i}pE = \frac{1}{m_i}q_iE+\frac{1}{m_i}\sum_{j=1}^{N}q_{ij}r_j^{(1)}E. \quad (20)$$

By using perturbation method, we obtain the second-order polarization in time-domain as

$$P^{(2)}_{\mathrm{NC}}(t,T) = \rho\sum_{i,j=1}^{N}q_{ij}q_iq_jS_i(t)*\left\{\begin{array}{l}[S_j(t)*E_2(t)]E_1(t+T)\\+[S_j(t)*E_1(t+T)]E_2(t)\end{array}\right\}$$

$$+\rho\sum_{i,j=1}^{N}q_{ij}q_iq_j[S_i(t)*E_2(t)][S_j(t)*E_1(t+T)]. \quad (21)$$

This expression matches that in Refs. [1,16–18] when $N=1$. The induced second-order electric field is

$$\tilde{E}^{(2)}_{\mathrm{NC}}(z,\omega_t,\omega_T)$$

$$= \tilde{R}^{(2)}\sum_{i,j=1}^{N}q_{ij}q_iq_j\left\{\begin{array}{l}\tilde{S}_i(\omega_t)[\tilde{S}_j(\omega_t-\omega_T)+\tilde{S}_j(\omega_T)]\\+\tilde{S}_i(\omega_t-\omega_T)\tilde{S}_j(\omega_T)\end{array}\right\}. \quad (22)$$

ND is introduced by adding a second-order term to the damping as

$$\gamma_i\frac{\mathrm{d}r_i}{\mathrm{d}t}\rightarrow\gamma_i\frac{\mathrm{d}r_i}{\mathrm{d}t}+\sum_{j,k=1}^{N}\gamma_{ij}^{k}\frac{\mathrm{d}r_k}{\mathrm{d}t}r_j, \quad (23)$$

where γ_{ij}^{k} is the nonlinear damping parameter. Through derivation, the polarization is

$$P^{(2)}_{\mathrm{ND}}(t,T) = -\rho\sum_{i,j,k=1}^{N}\gamma_{ij}^{k}q_iq_jq_k$$

$$S_i(t)\left\{\begin{array}{l}[S_j(t)*E_2(t)]\left[\dfrac{\mathrm{d}S_k(t)}{\mathrm{d}t}*E_1(t+T)\right]\\+[S_j(t)*E_1(t+T)]\left[\dfrac{\mathrm{d}S_k(t)}{\mathrm{d}t}*E_1(t)\right]\end{array}\right\}. \quad (24)$$

And the induced second-order electric field is

$$\tilde{E}^{(2)}_{\mathrm{ND}}(z,\omega_t,\omega_T)$$

$$= -\tilde{R}^{(2)}\sum_{i,j,k=1}^{N}\gamma_{ij}^{k}q_iq_jq_k\tilde{S}_i(\omega_t)\left[\begin{array}{l}\tilde{S}_j(\omega_t-\omega_T)\tilde{S}_k^{d}(\omega_T)\\+\tilde{S}_j^{d}(\omega_t-\omega_T)\tilde{S}_k(\omega_T)\end{array}\right], \quad (25)$$

where

$$\tilde{S}_i^{d}(\omega_t) = \left(\varepsilon_i\mathrm{e}^{-\mathrm{j}\pi\omega_t/2\varepsilon_i}-\frac{\gamma_i}{2}\right)\tilde{S}_i(\omega_t). \quad (26)$$

3 Results

To demonstrate the solution, a 3-mode system is considered, the parameters of which are set as

$$\begin{aligned}&\omega_1 = 0.9\times2\pi\ \mathrm{THz},\\&\omega_2 = 1.2\times2\pi\ \mathrm{THz},\\&\omega_3 = 1.7\times2\pi\ \mathrm{THz},\\&\gamma_i = 0.2\omega_i,\ i=1,2,3,\\&m_1 = m_2 = m_3,\\&q_1 = q_2 = q_3.\end{aligned} \quad (27)$$

We choose a typical THz shape pulse as incident pulses,

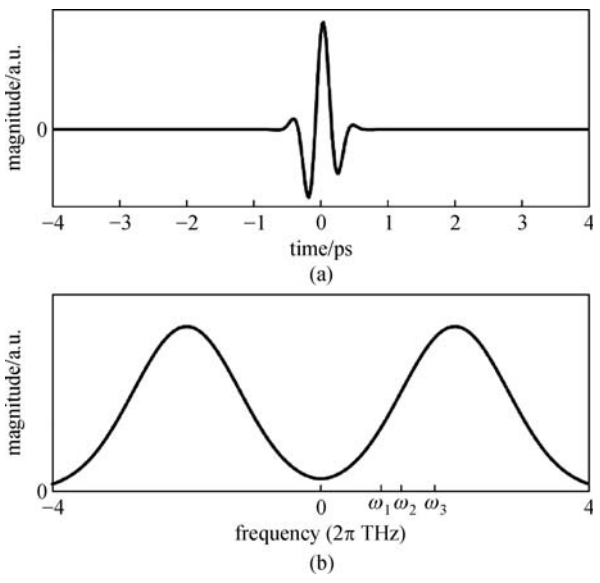

Fig. 2 THz pulse used in calculation shown in time- (a) and frequency- (b) domain

as shown in Fig. 2, and assume $E_1(t)$ and $E_2(t)$ are identical.

Given that the purpose of this section is to validate the results, only three cases are considered: 1) all nonlinear parameters are zero except $B_{123} = B_{132} = ... = B_{321}$ (AH), 2) all nonlinear parameters are zero except $q_{12} = q_{21}$ (NC), 3) all nonlinear parameters are zero except $\gamma^1_{23} = \gamma^1_{32}$ (ND). The nonlinear electric field $\tilde{E}^{(2)}(\omega_t,\omega_T)$ is calculated by two methods, i.e., our analytical solution and the traditional simulation method of which the process is: 1) the polarization $P^{(2)}(t,T)$ is calculated in time-domain through numerical integral according to Eqs. (14), (21) and (24) respectively; 2) numerical 2D-FT is performed so that the polarization in frequency-domain $\tilde{P}^{(2)}(\omega_t,\omega_T)$ is obtained; 3) the electric field $\tilde{E}^{(2)}(\omega_t,\omega_T)$ is numerically calculated according to Eq. (8). Figure 3 illustrates the calculated absolute value of nonlinear electric fields with normalized magnitude. Six sub-graphs show the electric fields corresponding to three different sources of nonlinearities and the two calculation methods. Frequency $\omega_i + \omega_j$ and $\omega_i - \omega_j$ are expressed as $\Omega^{ij} = \omega_i + \omega_j$ and $\Omega^i_j = \omega_i - \omega_j$ in these sub-graphs.

4 Discussion

Obviously, the analytical results agree well with the simulations. For AH (Figs. 3(a) and 3(d)), peaks appear at $(\omega_t,\omega_T) = \pm(\omega_i, \pm\omega_j), \pm(\omega_i,\omega_i \pm \omega_j), \pm(\omega_i \pm \omega_j,\omega_i)$, where $i,j = 1,2,3 \quad (i \neq j)$, as predicted in Eq. (15). These peaks correspond to the coupling between mode 1, 2, and 3 through the anharmonicity of potential. For NC (Figs. 3(b) and 3(e)), peaks appear at $(\omega_t,\omega_T) = \pm(\omega_i, \pm\omega_j)$, $\pm(\omega_i,\omega_i \pm \omega_j)$, $\pm(\omega_i \pm \omega_j,\omega_i)$, where $i,j = 1,2 \quad (i \neq j)$, as predicted in Eq. (22). These peaks correspond to the coupling between mode 1 and 2 through the nonlinearity of dipole moment. For ND (Figs. 3(c) and 3(f)), peaks appear at $(\omega_t,\omega_T) = \pm(\omega_i, \pm\omega_j), \pm(\omega_i,\omega_i \pm \omega_j), \pm(\omega_i \pm \omega_3,\omega_i)$, $\pm(\omega_3 - \omega_i,\omega_3)$, where $i = 1,2$, $j = 1,2,3 \quad (i \neq j)$, as predicted in Eq. (25). These peaks correspond to the coupling between the vibrational coordinate of mode 1, 2 and the

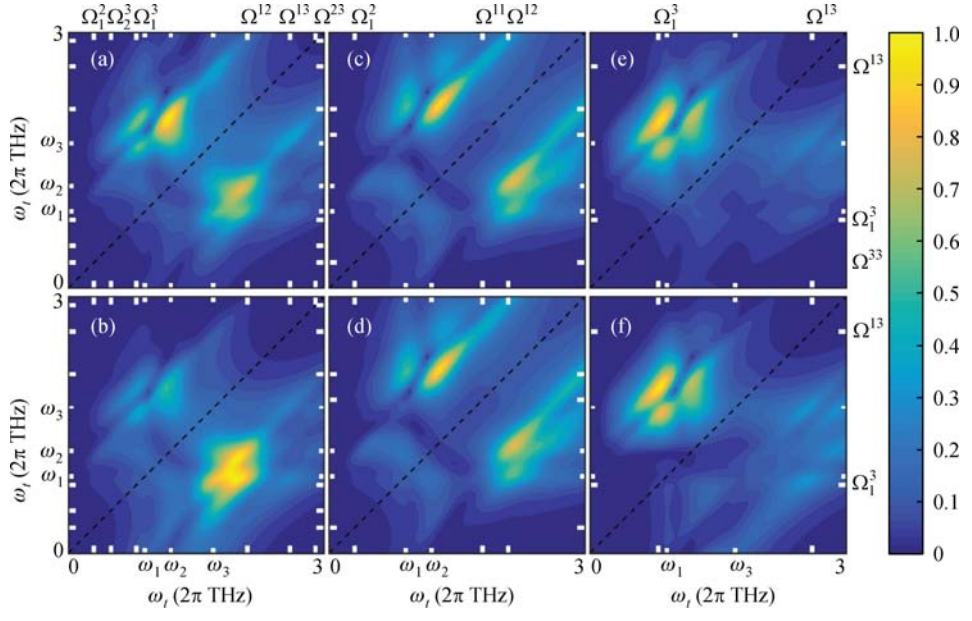

Fig. 3 Normalized 2D diagrams corresponding to (a) AH, (b) NC, and (c) ND obtained by analytical calculation, and those corresponding to (d) AH, (e) NC, and (f) ND obtained by numerical simulation. Only the first quartile is presented to display more details

velocity of mode 3 through the nonlinearity of damping.

However, the calculation amount of the analytical solution is much smaller than numerical simulation, since the former only involves multiplication and addition, while the later involves integral. The ratio of the calculation amounts of these two methods is nearly $1 : 0.19F_{\mathrm{w}}/\Delta f$ (F_{w} is frequency window and Δf is the spectral resolution). For experiments the frequency window may be over 30 THz [4], and if the spectral resolution is 0.1 THz, the calculation amount of numerical simulation is 57 times larger than that of analytical calculation.

5 Conclusions

Using multi-oscillator model involving three sources of weak nonlinearities, we have achieved a classical-theory-based analytical solution for second-order 2DTS. By calculating the spectrum of a 3-mode system, we have verified that our analytical calculations are consistent with the traditional numerical simulation results. Compared to former theoretical works which only dealt with single-mode systems, our solution has two major advantages: first, it is more wide-ranging in term of application as it can be applied to multi-modes system; secondly, it has much lower computational cost as it directly gives the second-order transmission field instead of polarization and no numerical integral needs to be performed. To conclude, this new analytical approach offers a more efficient and practical method to directly obtain second-order 2DTS in replacement of complex simulation. In a broader sense, our study sheds new light on the theory of 2DTS, which may enable promising future applications.

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