

Propagation of terahertz pulses in photoexcited media: Analytical theory for layered systems

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Optical pump–terahertz probe spectroscopy has become a widely used experimental tool for the investigation of the ultrafast far-infrared response of polar systems. In this paper the authors present an analytical method of calculating the propagation of ultrashort terahertz pulses in photoexcited media. The transient terahertz wave form transmitted through the sample is equal to a product of the incident terahertz field (at a mixed frequency), transient susceptibility, and a so called transfer function which depends on the properties of the sample in equilibrium. The form of the transfer function is derived for general layered systems and for specific cases including one-dimensional photonic crystals, thin films, and bulk samples. Simplified expressions directly applicable to the analysis of the experimental results related to the most common sample geometries are shown and discussed. © 2007 American Institute of Physics. [DOI: 10.1063/1.2748402]

I. INTRODUCTION

The time-domain terahertz spectroscopy uses broadband picosecond pulses of far infrared radiation with frequencies most often in the range of 0.1–3 THz. The pulses are detected using gating techniques which make it possible to resolve the temporal profile of their electric field with a subpicosecond resolution. Optical pump–terahertz probe (OPTP) experiments represent a powerful experimental method in which these terahertz pulses are used to probe changes of the far-infrared susceptibility (or conductivity) spectrum initiated by an optical excitation event. They provide access to subpicosecond, picosecond, and nanosecond dynamics of far-infrared polar response in a large variety of physical and chemical systems (see, e.g., Ref. 1 for a review).

In principle, the time resolution in the OPTP experiments is not limited by the terahertz pulse length (typically >1 ps); rather, it is limited by the bandwidth of the gated detection process which yields a subpicosecond resolution (≈ 0.2 – 0.4 ps). If an investigated system undergoes a change of its state on a subpicosecond time scale, its dynamical response must involve frequency components overlapping with the terahertz spectral range. This leads to a frequency mixing which distorts the transient terahertz wave form.^{2,3}

Any single measurement uses three pulses: optical pump, terahertz probe, and a gating detection pulse. The intervals between them correspond to two time variables which can be alternatively transformed to frequencies. The most complete experimental approach consists in the acquisition of a two-dimensional (2D) data grid in the time domain, i.e.,

in a measurement of a series of transient terahertz wave forms at closely spaced delays between the optical pump and terahertz probe. The key task is then to relate the observed ultrafast evolution of the raw terahertz signal to the underlying sample dynamics described by a transient susceptibility or conductivity.^{2–10} The first theoretical work which showed some hints on the correct OPTP measurement protocol and the data analysis was published by Kindt and Schmuttenmaer.² This concept was further developed and supplied by experimental results in Refs. 4 and 5. The same authors developed an approach based on numerical finite-difference time-domain calculations.^{6,7}

Subsequently, we have developed a general analytical frequency-domain methodology of the OPTP experiments. In Refs. 3 and 8 we have proposed a framework for the transformation of the raw terahertz data needed to obtain the transient response function of a photoexcited sample; in Ref. 9 we have demonstrated the power of our approach in experiments with a number of different physical systems from ultrafast semiconductors to solutions.

The main aim of this paper is to complete some gaps we have still left in our approach. More specifically, in Ref. 3 we have shown that the Maxwell equations describing the OPTP experiment can be analytically solved in a 2D Fourier (frequency) space. In such a case, the transient terahertz field ΔE (i.e., the change of the terahertz field due to the photoexcitation) can be written as the product of the incident terahertz wave E_{inc} , the transient susceptibility $\Delta\chi$ (or conductivity $\Delta\sigma$) (Ref. 8) of the photoexcited sample, and a transfer function Ξ which depends solely on the ground state properties of the sample,

$$\Delta E = \Xi E_{\text{inc}} \Delta\chi. \quad (1)$$

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The analysis of OPTP experiments then relies on a construction of the transfer function Ξ for the given sample and experimental geometry. In Ref. 3 we derived a general formula for Ξ in a single homogeneous layer and discussed how to extract correctly the transient susceptibility from the experimental data. In the current paper we discuss important special cases of the sample geometry where the function Ξ can be substantially simplified to render the interpretation of the data to be more transparent and easier. At the same time we generalize our approach to more complex structures including two-component samples and general layered structures.

The paper is organized as follows: Sec. II shortly summarizes our approach and shows the basic equations and their solutions in a general form. Section III is devoted to thin films and Sec. IV to bulk samples; issues related to the finite length of the optical pump pulses are addressed in Sec. V. Finally, in Sec. VI we generalize our approach to layered structures (one-dimensional photonic crystals). The paper can serve either as a quick reference providing the formulas which apply to the most common experimental situations (these are summarized in Fig. 2) or as a general recipe allowing one to derive the transfer function Ξ for specific sample geometries.

II. BASIC EQUATIONS AND QUANTITIES

A. Wave equation and simplifying conditions

We describe the problem using the two proper time variables of Representation II as introduced in Ref. 3: τ related to the gated detection of terahertz pulses (i.e., the real time with respect to the terahertz wave form profile) and the pump-probe delay τ_p (describing the time evolution of the excited sample). These variables are controlled experimentally by two independent delay lines. The related frequency-domain variables are ω and ω_p . In the frequency domain (ω, ω_p) the generation and propagation of the transient terahertz field $\Delta E(\omega, \omega_p; z)$ in the sample along its normal (z axis) is described by the following general wave equation with a source term [cf. Eq. (16) in Ref. 3]:

$$\frac{d^2 \Delta E}{dz^2} + n(\omega)^2 k_0^2 \Delta E = U(\omega, \omega_p; z). \quad (2)$$

Here $n(\omega)$ is the complex refractive index of the sample at terahertz frequencies in the ground state and $k_0 = \omega/c$ is the wave vector of the terahertz wave in vacuum. The right-hand side $U(\omega, \omega_p; z)$ describes the transient polarization (or transient current) in the sample. It is induced by the local terahertz field E_{THz} if the transient susceptibility $\tilde{\chi}$ (or conductivity $\tilde{\sigma}$) does not vanish,

$$U(\omega, \omega_p; z) = -k_0^2 \Delta \tilde{\chi}(\omega, \omega_p; z) E_{\text{THz}}(\omega - \omega_p; z). \quad (3)$$

The frequency mixing in terms of $\omega - \omega_p$ in this expression clearly demonstrates that new terahertz spectral components can appear in the photoexcited sample. We point out that for slowly evolving photoexcited systems the transient susceptibility differs from zero at a close vicinity of $\omega_p = 0$ and vanishes rapidly at higher values of ω_p . In this case the frequency mixing can be neglected, i.e., $E_{\text{THz}}(\omega - \omega_p; z)$ can be

replaced by $E_{\text{THz}}(\omega; z)$ in Eq. (3), and the data can be analyzed by using a simple quasi-steady-state approach based on one-dimensional Fourier transformations of the transient and reference wave forms for several pump-probe delays.^{3,8} In contrast, for samples exhibiting an ultrafast decay of the transient response (as compared to the terahertz pulse length) the frequency mixing has to be taken into account.

The local terahertz field E_{THz} is expressed in our model as a sum of a primary field E which would propagate through the sample in equilibrium and the transient field ΔE induced in the photoexcited sample: $E_{\text{THz}} = E + \Delta E$. The main approximation of our approach consists in assuming that $\Delta E \ll E$. We then replace $E_{\text{THz}}(\omega - \omega_p; z)$ by $E(\omega - \omega_p; z)$ in Eq. (3); Eq. (2) is then linearized and can be solved analytically. The quantity E_{inc} of Eq. (1) is the amplitude of the terahertz wave incident on the sample (which can be directly measured). The equilibrium field $E(z)$ inside the sample can be easily connected to E_{inc} within the linear optics approach by using appropriate Fresnel formulas.

As a starting point, we describe a single homogeneous layer. The pump pulse propagates in this layer with a group velocity v_g and we assume a linear absorption process characterized by an absorption coefficient α . The magnitude of $\Delta \tilde{\chi}(\omega, \omega_p; z)$ is proportional to the density of photoexcited particles and we introduce a transient susceptibility per unit pump fluence¹¹ $\Delta \chi(\omega, \omega_p)$,

$$\Delta \tilde{\chi}(\omega, \omega_p; z) = F(\omega_p) \exp[-(i\omega_p/v_g + \alpha)z] \Delta \chi(\omega, \omega_p), \quad (4)$$

where $F(\omega_p)$ is the spectral density of the pump fluence entering the sample. Unless explicitly stated, we will consider throughout this paper that the pump pulse is infinitely short in the time domain and we set $F=1$ for simplicity.

Under all these conditions, the expression for U reads [cf. Eq. (16) in Ref. 3]

$$U = -k_0^2 \exp[-i(\omega_p/v_g - i\alpha)z] E(\omega - \omega_p; z) \Delta \chi(\omega, \omega_p). \quad (5)$$

In the derivation, we have implicitly neglected a possible reflection of the pump pulse at the output face of the sample. This approximation is not fulfilled for any sample, nevertheless we deem that it is a very reasonable approximation for a large number of experimental situations. Its validity can be violated in the case when the thickness of the sample is smaller than the penetration depth of the optical radiation. In the case of thin samples we do take into account the multiple reflections of the terahertz pulse. By contrast, the difference between the refractive indices of materials in the optical range is usually much smaller than that in the terahertz spectral range and the optical power reflection coefficient is typically smaller than 10% in the majority of cases of interest. It means that second and higher-order internal reflections of the pump beam inside the sample can be usually safely neglected and the magnitude of a single reflection at the output side of the sample is to be discussed in some particular cases. We will come back to this issue further in the paper for some particular sample geometries considered here.

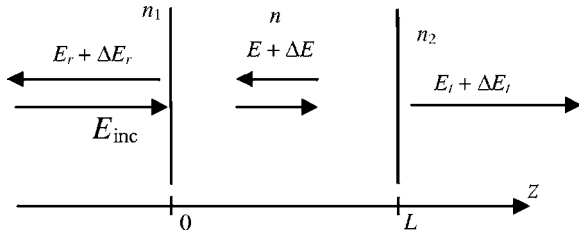


FIG. 1. Scheme of the fields in the sample in the photoexcited state. E_{inc} , E_r , and E_t are the incident, reflected, and transmitted fields, respectively, relative to the sample ground state. ΔE_r and ΔE_i are the photoinduced transient waves propagating in the backward and forward directions, respectively.

We can express Eq. (5) in terms of either the transient susceptibility $\Delta\chi$ or the transient conductivity $\Delta\sigma$ which are related together by

$$\Delta\chi(\omega, \omega_p) = \frac{\Delta\sigma(\omega, \omega_p)}{i\omega\epsilon_0}. \quad (6)$$

We think that the interpretation and modeling of experimental results is more straightforward and appropriate in terms of the transient conductivity for the majority of systems. However, as indicated by Eq. (6) the choice of the description of the photoexcitation and probing processes in terms of either $\Delta\chi$ and $\Delta\sigma$ is quite arbitrary and we have chosen to use $\Delta\chi$ in the subsequent paragraphs for the simple reason to abbreviate the notation.

B. Notation convention

To abbreviate and simplify the expressions we derive throughout this paper we introduce the following notation. All the variables and parameters which are evaluated at frequency ω will be denoted by lowercase letters, while the values of these parameters at the mixed frequency $\omega - \omega_p$ are denoted by uppercase letters, e.g., the refractive index of the sample is denoted by $n \equiv n(\omega)$ and $N \equiv n(\omega - \omega_p)$; the (possibly complex) refractive indices of media 1 and 2 which are adjacent to the sample (see Fig. 1) are denoted as $n_i \equiv n_i(\omega)$ and $N_i \equiv n_i(\omega - \omega_p)$, $i=1,2$. Media 1 and 2 can correspond, e.g., to air, cuvette material, film substrate, etc. In any of these cases we assume that they are thick such that the echoes corresponding to multiple internal reflections inside these media can be resolved in the time-domain scan and cut away.

A complete list of the symbols and expressions used in this paper is given below (note that this notation slightly differs from the one introduced in Ref. 3). The wave vectors in the sample k , K and in the adjacent media k_i , K_i read

$$k = n \frac{\omega}{c}, \quad K = N \frac{\omega - \omega_p}{c}, \quad (7)$$

$$k_i = n_i \frac{\omega}{c}, \quad K_i = N_i \frac{\omega - \omega_p}{c} \quad (i=1,2); \quad (8)$$

in addition, we define

$$K_F = K + \frac{\omega_p}{v_g} - i\alpha, \quad (9)$$

$$K_B = -K + \frac{\omega_p}{v_g} - i\alpha. \quad (10)$$

The amplitude reflection (r_i, R_i) and transmission (t_i, T_i) coefficients read

$$r_i = \frac{n - n_i}{n + n_i}, \quad R_i = \frac{N - N_i}{N + N_i} \quad (i=1,2), \quad (11)$$

$$t_1 = \frac{2n_1}{n + n_1}, \quad T_1 = \frac{2N_1}{N + N_1}, \quad (12)$$

$$t_2 = \frac{2n}{n + n_2}, \quad T_2 = \frac{2N}{N + N_2}, \quad (13)$$

$$t_{12} = \frac{2n_1}{n_1 + n_2}, \quad T_{12} = \frac{2N_1}{N_1 + N_2}. \quad (14)$$

This notation allows us to skip the frequency arguments in the subsequent complicated equations. However, notice that this does not bring any additional restriction nor approximation on the results derived in this paper: All refractive indices can be complex and frequency dependent.

C. Solutions

Depending on the thickness of the sample and on the temporal length of the measured terahertz wave form, the right-hand side of Eq. (2) should be properly constructed and appropriate boundary conditions should be imposed. For optically thick samples, for which the pulse echoes coming from the internal (Fabry-Pérot) reflections on the sample input and output faces are well separated in time from each other, a temporal windowing procedure can be applied to the raw time-domain data, i.e., the measured wave forms can be cut at a suitable time τ to remove the signal coming from these Fabry-Pérot reflections while keeping the one corresponding to the direct pass through the sample. This procedure usually simplifies the data analysis.¹² If, by contrast, the sample is optically thin, such that the individual Fabry-Pérot echoes cannot be separated in time, the whole wave form should be measured and analyzed accordingly. In this last case the right-hand side of Eq. (2) reads

$$U = -k_0^2 \Delta\chi(\omega, \omega_p) E_{\text{inc}} T_1 A [\exp(-iK_F z) + R_2 \exp(-2iKL) \exp(-iK_B z)], \quad (15)$$

where

$$A = \frac{1}{1 - R_1 R_2 \exp(-2iKL)}, \quad (16)$$

and where $E_{\text{inc}} \equiv E_{\text{inc}}(\omega - \omega_p)$, as shown in Fig. 1, denotes the incident terahertz field in medium 1 at the interface with the sample (i.e., at $z=0^-$). The term T_1 in expression (15) accounts for the transmission losses at the input interface of the sample while A accounts for the internal reflections of the primary terahertz field E in the sample,

$$E(\omega - \omega_p; z) = E_{\text{inc}} T_1 A [\exp(-iKz) + R_2 \exp(-iKL) \exp(-iK(L-z))]. \quad (17)$$

The solution of the wave equation [Eq. (2)] in the sample with the right-hand side of Eq. (15) reads

$$\Delta E(z) = \delta \exp(-ikz) + \gamma \exp(ikz) - k_0^2 \Delta \chi E_{\text{inc}} T_1 A \times \left[\frac{\exp(-iK_F z)}{k^2 - K_F^2} + R_2 \exp(-2iKL) \frac{\exp(-iK_B z)}{k^2 - K_B^2} \right]. \quad (18)$$

The transient magnetic field then can be calculated using $\Delta H = i/(\mu_0 \omega) d\Delta E/dz$. The coefficients γ and δ are evaluated using the continuity conditions of the fields at the sample boundaries (i.e., at $z=0$ and $z=L$). The primary fields E_{inc} , E_r , and E_t (see Fig. 1 for the symbol definition) fulfill the usual Fresnel equation; the boundary conditions for the transient fields then read

$$\Delta E_r = \Delta E(z=0), \quad -k_1 \Delta E_r = i \frac{d\Delta E(z=0)}{dz} \quad (19)$$

$$\Delta E_t = \Delta E(z=L), \quad k_2 \Delta E_t = i \frac{d\Delta E(z=L)}{dz}. \quad (20)$$

Introducing Eq. (18) into Eqs. (19) and (20) one finally finds the following solution of the wave equation with the boundary conditions:

$$\Delta E_t(\omega, \omega_p) = \Xi E_{\text{inc}}(\omega - \omega_p) \Delta \chi(\omega, \omega_p), \quad (21)$$

where the transfer function Ξ only depends on the equilibrium sample properties,

$$\Xi = -\frac{k_0^2 T_1}{k_2 + k} \exp(-ikL) A a \left\{ r_1 \frac{1 - \exp[-i(K_F + k)L]}{K_F + k} + \frac{1 - \exp[-i(K_F - k)L]}{K_F - k} + \left[r_1 \frac{1 - \exp[-i(K_B + k)L]}{K_B + k} + \frac{1 - \exp[-i(K_B - k)L]}{K_B - k} \right] R_2 \exp(-2iKL) \right\}. \quad (22)$$

The factor

$$a = \frac{1}{1 - r_1 r_2 \exp(-2ikL)} \quad (23)$$

represents the multiple reflections of the transient wave in the sample. Equation (22) is equivalent to Eq. (A4) in Ref. 3 in a slightly different notation. [Note also that there is a misprint in Ref. 3: The first sign in the second line of Eq. (A4) must be “+” instead of “×”.]

For optically thick samples, the temporal windowing can be applied to the time-domain data. The phase of the terahertz signal coming directly to the detector (without any internal reflections in the sample) should be close to $-ikL$. The “windowed” transfer function relative to a direct (single) pass through the sample can be easily obtained from Eq. (22) if all the terms with the phase larger than or equal to $-2iKL$ or $-2iKL$ (which, in fact, represent the terahertz field coming into the detector after the signal window) are omitted. One finds

$$\Xi = -\frac{k_0^2 T_1}{k_2 + k} \exp(-ikL) \left\{ \frac{r_1}{K_F + k} + \frac{1 - \exp[-i(K_F - k)L]}{K_F - k} - R_2 \frac{\exp[-i(K_B - k + 2K)L]}{K_B - k} \right\}. \quad (24)$$

This equation can be directly compared to Eqs. (30) and (31b) in Ref. 3. Note, however, that the last term in the above equation [Eq. (24)] is usually small and that it was neglected in Ref. 3.

Equations (22) and (24) for the transfer function are the basic equations of this paper. They are quite general and can be directly used for the evaluation of OPTP data; however, they are rather complex. Much simpler expressions can be found in some special but frequent experimental cases (see Fig. 2), and the investigation of these special cases is the subject of the subsequent sections.

III. THIN FILMS

A. Single-component systems

For thin films, the interference of all the Fabry-Pérot reflections should always be taken into account, which means that the transfer function defined by Eq. (22) is of interest here. We consider sufficiently thin films where $kL \ll 1$, and consequently also $KL \ll 1$ [i.e., thinner than the wavelength of terahertz radiation in the sample; see also Figs. 2(a)–2(c)] for all the available terahertz spectral components: the arguments $-ikL$ and $-iKL$ of the exponentials in (22) can be developed into power expansions. As the upper limit of the terahertz pulse bandwidth in OPTP experiments is typically at about 2.5 or 3 THz, about micrometer-sized or thinner films can be described within this approximation.

Note that, e.g., for a strongly photoexcited thin semiconductor films, a condition $n_{\text{exc}} \omega L / c \ll 1$ may not be satisfied in the excited state due to a high density of free carriers causing the refractive index n_{exc} to be high. However, the value of n_{exc} is accounted for by $\Delta \chi$ in our model. It means that in this case the conditions $kL \ll 1$ and $\Delta E \ll E$ are still the measure of applicability of the thin film approximation.

First we study the case of a strong or a medium optical absorption of the film, i.e., $\alpha L \geq 1$ ($\alpha \geq k$) as shown in Figs. 2(a) and 2(b). In this case the developments of the exponential terms to the lowest order yield, e.g.,

$$a(\omega) \approx \frac{t_{12}}{t_1 t_2}, \quad (25)$$

and

$$\frac{1 - \exp[-i(K_F - k)L]}{K_F - k} \approx \frac{1 - \exp(-\alpha L)}{-i\alpha}. \quad (26)$$

The other terms in Eq. (22) are developed similarly. Finally, we obtain

$$\Xi \approx \frac{T_{12}}{n_1 + n_2} \left(-\frac{ik_0}{\alpha} \right) [1 - \exp(-\alpha L)]. \quad (27)$$

For a strong optical absorption of the film [$\alpha L \gg 1$, Fig. 2(a)] one finds

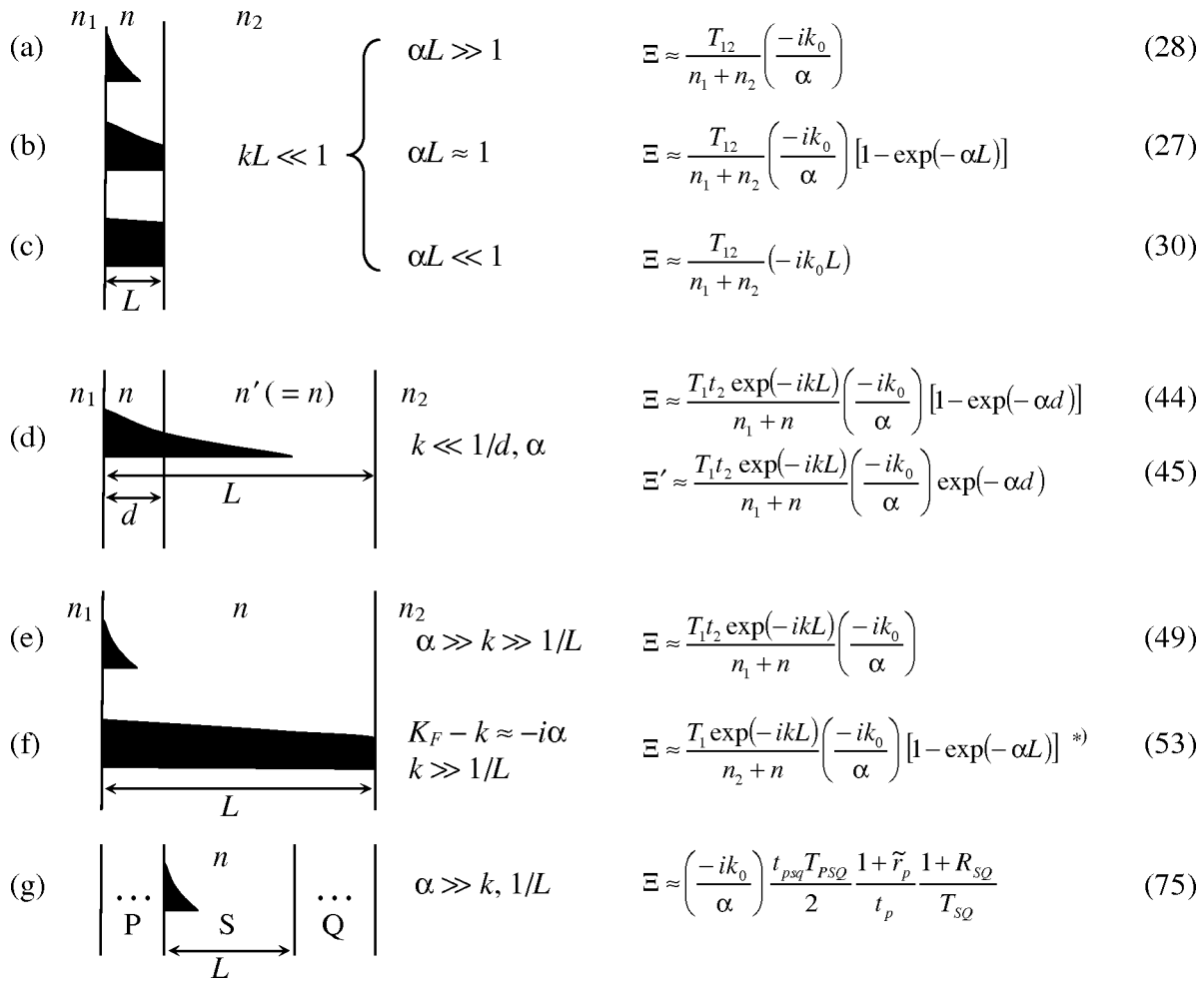


FIG. 2. Special sample geometries discussed in this paper. The black areas indicate the density of photoexcited particles interacting with the terahertz field. [(a)–(c)] Thin film (self-standing or on a substrate): photoexcitation of the thin film only; (d) thin film on a substrate: photoexcitation of the sample and of a part of the substrate; (e) thick sample: surface photoexcitation; (f) thick sample: bulk photoexcitation, quasi-phase matched case; (g) layered sample (photonic structure): parts P and Q are general layered structures and the part S is a homogeneous sample layer. Right panel: summary of the transfer functions found for these special geometries and the corresponding equation numbers in the text; see also Eq. (52) for a slightly more general form of this transfer function.

$$\Xi \approx \frac{T_{12}}{n_1 + n_2} \left(\frac{-ik_0}{\alpha} \right). \quad (28)$$

In the remaining case of a weak optical absorption of the thin film ($\alpha L \ll 1$), we obtain a quasiuniform excitation of the sample [Fig. 2(c)] and we get instead of Eq. (26)

$$\frac{1 - \exp[-i(K_F - k)L]}{K_F - k} \approx iL. \quad (29)$$

The resulting expression then reads

$$\Xi \approx \frac{T_{12}}{n_1 + n_2} (-ik_0 L). \quad (30)$$

Note that in this particular case of a very thin film, $E_t = T_{12} E_{\text{inc}}$; Eqs. (6), (21), and (30) when put together yield

$$\Delta E_t(\omega, \omega_p) = - \frac{L}{\epsilon_0 c (n_1 + n_2)} \Delta \sigma(\omega, \omega_p) E_t(\omega - \omega_p); \quad (31)$$

this equation is equivalent to Eq. (A5) in Ref. 10 which was derived directly for this very specific case.

The expressions for thin films presented in this paragraph are important especially for the time-resolved spec-

troscopy of semiconductor thin films deposited on optically transparent substrates. Typical examples of the application are silicon thin films (radiation damaged silicon on sapphire,^{9,13} microcrystalline and amorphous silicon,^{14,15} superconductors,^{16,17} etc.). By changing the wavelength of the optical pump pulse, one can achieve the three experimental situations depicted in Figs. 2(a)–2(c). Let us consider a 1 μm silicon layer as a typical thin film sample. For an excitation at 800 nm, where the light penetration depth is about 10 μm , one obtains a quasihomogeneous carrier density throughout the film: The experiment is described by Eq. (30). In contrast, for a pump wavelength of 400 nm the radiation penetration depth reaches only about 100 nm: The experiment is then described by Eq. (28).

The problem of the nonvanishing reflection of the optical excitation pulse at the output face of the thin film concerns only the cases (b) and (c) as shown in Fig. 2. Two issues should be considered: the time resolution of the pump-probe experiment and the magnitude of the transient terahertz signal. Concerning the time resolution, the time of a round trip of the optical radiation in a 1 μm thick thin film is smaller than 25 fs for typical optical refractive indices (≤ 3.5), i.e.,

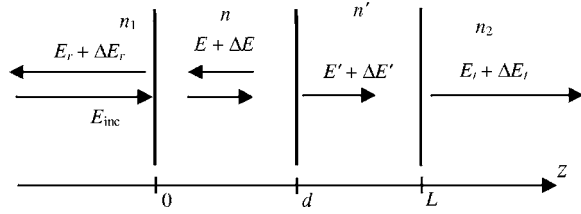


FIG. 3. Scheme of the fields in the photoexcited sample consisting of a thin film on a substrate with a nonvanishing terahertz response in the excited state (the ratio of thicknesses of the film and substrate is not in scale).

the time broadening of the excitation event is negligible for thin films. As for the signal magnitude, notice that the physical meaning of the square bracket in Eq. (27) is the fraction of the incident pulse fluence absorbed in the sample. One can then correct this value to obtain the true fraction of the absorbed fluence taking into account the pump beam reflection at the back side of the thin film.

Concerning the quasihomogeneous excitation of a weakly absorbing thin film described by Eq. (30), one should simply take into account the fact that the true density of excited particles is appropriately higher than that estimated from a single pass of the optical pump beam.

B. Two-component systems

In some cases the semiconductor thin films are grown on substrates which show an appreciable terahertz signal in the photoexcited state [see Fig. 2(d)]. Typical examples of such systems are ultrafast semiconductors such as proton-bombarded InP (Ref. 18) or low-temperature grown GaAs layer (LT-GaAs) on a GaAs single crystal substrate. For a micrometer thick LT-GaAs layer, 18% of free carriers are generated in the bulk GaAs substrate if an optical excitation at 800 nm is used.¹⁹ The carrier dynamics in the layer and in the substrate are described by distinct transient conductivity (or susceptibility) functions. We treat such a sample as a two-component system—we consider both the film susceptibility $\Delta\chi(\omega, \omega_p)$ and that of the substrate $\Delta\chi'(\omega, \omega_p)$ to describe the response of the whole sample. The aim of this paragraph is to derive the appropriate transfer functions Ξ and Ξ' for such a system.

We adopt a notation in which all the quantities relative to the substrate are primed (see Fig. 3). The wave equation [Eq. (2)] is valid for the substrate provided that the refractive index n at the left-hand side is replaced by n' and the right-hand side U is properly constructed. We denote by d the thickness of the thin film.

The Fabry-Pérot reflections inside the thin film should be taken into account. In contrast, we assume that the substrate is thick enough to make it possible to eliminate the Fabry-Pérot reflections inside the substrate by a proper choice of the time window in the windowing procedure; i.e., only the direct pass through the substrate is considered as shown in Fig. 3. The calculation of the right-hand side of Eq. (2) is then straightforward. It reads inside the film

$$U = -k_0^2 \Delta\chi(\omega, \omega_p) E_{\text{inc}} T_1 A' [\exp(-iK_F z) + R' \exp(-2iKd) \exp(-iK_B z)], \quad (32)$$

and in the substrate (where the signal coming from the backward propagating wave is eliminated by the temporal windowing)

$$U' = -k_0^2 \Delta\chi'(\omega, \omega_p) E_{\text{inc}} T_1 T' A' \exp(-iK_F d) \times \exp[-iK'_F(z-d)]. \quad (33)$$

All the primed quantities are defined analogously to those introduced in Eqs. (7)–(14) and (16),

$$T' = \frac{2N}{N+N'}, \quad (34)$$

$$R' = \frac{N-N'}{N+N'}, \quad (35)$$

$$A' = \frac{1}{1-R_1 R' \exp(-2iKd)}, \quad (36)$$

$$K'_F = K' + \frac{\omega_p}{v'_g} - i\alpha', \quad (37)$$

$$K'_B = -K' + \frac{\omega_p}{v'_g} - i\alpha'. \quad (38)$$

The solutions of the wave equation [Eq. (2)] then read

$$\Delta E(z) = \delta \exp(-ikz) + \gamma \exp(ikz) - k_0^2 \Delta\chi E_{\text{inc}} T_1 A' \times \left[\frac{\exp(-iK_F z)}{k^2 - K_F^2} + R' \exp(-2iKL) \frac{\exp(-iK_B z)}{k^2 - K_B^2} \right], \quad (39)$$

$$\Delta E'(z) = \delta' \exp(-ik'z) + \gamma' \exp(ik'z) - k_0^2 \Delta\chi' E_{\text{inc}} T_1 T' A' \times \exp[-i(K_F - K'_F)d] \frac{\exp(-iK'_F z)}{k'^2 - K_F'^2}. \quad (40)$$

The continuity conditions of the electric and magnetic fields at the boundaries $z=0$, $z=d$, and $z=L$ yield the constants δ , δ' , γ , and γ' and, finally, the output transient terahertz field ΔE_t . The expression for ΔE_t is rather complex in the general case, therefore we present it here only for a special case of a substrate which shows the same equilibrium properties as the thin film, i.e., $n=n'$, $K_F=K'_F$, etc. Such a requirement is fulfilled in LT-GaAs/GaAs structure⁵ but also in other ultrafast semiconducting structures where a surface layer with an ultrafast response is created by ion bombardment.^{18,20} In this case $R'=0$, $T'=1$, and $A'=1$. We also assume that the excitation beam is fully absorbed in the thick substrate ($\alpha L \gg 1$). Note, however, that the dynamical properties of the film and substrate in the photoexcited state remain different: $\Delta\chi \neq \Delta\chi'$. One finds finally

$$\Delta E_t(\omega, \omega_p) = [\Xi \Delta\chi(\omega, \omega_p) + \Xi' \Delta\chi'(\omega, \omega_p)] E_{\text{inc}}(\omega - \omega_p), \quad (41)$$

where

$$\Xi = -\frac{k_0^2 T_1}{k_2 + k} \exp(-ikL) \left[\frac{r_1}{K_F + k} + \frac{1}{K_F - k} \right] - \Xi', \quad (42)$$

$$\Xi' = -\frac{k_0^2 T_1}{k_2 + k} \exp(-ikL) \left[\frac{r_1 \exp(-ikd)}{K_F + k} + \frac{\exp(ikd)}{K_F - k} \right] \times \exp(-iK_F d). \quad (43)$$

As we deal with a thin film sample we can assume $k_0 d \ll 1$. We may also assume $ad \approx 1$; for $ad \gg 1$ no carriers are created in the substrate and we get a situation equivalent to that shown in Fig. 2(e), for $ad \ll 1$ the signal from the thin film would be entirely overwhelmed by that of the substrate. Using these approximations we find in the lowest-order development

$$\Xi \approx \frac{T_1 t_2 \exp(-ikL)}{n_1 + n} \left(\frac{-ik_0}{\alpha} \right) [1 - \exp(-\alpha d)], \quad (44)$$

$$\Xi' \approx \frac{T_1 t_2 \exp(-ikL)}{n_1 + n} \left(\frac{-ik_0}{\alpha} \right) \exp(-\alpha d), \quad (45)$$

which means that the magnitudes of the signals coming from the film and from the substrate are directly proportional to the optical energy absorbed in the respective parts of the sample.

Note that our treatment can be generalized for multicomponent layered samples for which one finds the following general expression:

$$\Delta E_t(\omega, \omega_p) = \left[\sum_i \Xi^{(i)} \Delta \chi^{(i)}(\omega, \omega_p) \right] E_{\text{inc}}(\omega - \omega_p), \quad (46)$$

where $\Delta \chi^{(i)}(\omega, \omega_p)$ are the transient susceptibilities of the individual components of the sample. The transfer functions $\Xi^{(i)}$ then should be evaluated using the form of the right-hand side of the wave equation and the continuity conditions appropriate for the particular sample geometry.

IV. BULK SAMPLES

The thickness of the bulk samples (slabs) is assumed to be high enough to enable the temporal windowing procedure. The time distance of individual terahertz echoes $2nL/c$ should exceed the period $1/\nu_{\text{min}}$ of the minimum useful frequency within the bandwidth of the terahertz pulse. This means $kL > \pi$, hence $k_0 L \gg 1$. Since the signal corresponding to the direct pass of the terahertz pulse is separated from the other Fabry-Pérot reflections, the transfer function defined by Eq. (24) is to be applied to the experimental data.

A. Surface excitation

This case concerns highly absorbing samples in the optical range such that the pump pulse reaches a thin surface layer only [see Fig. 2(e)]. We meet this situation typically in thick semiconductor wafers (GaAs, InP, etc.). The condition of a strong optical absorption $\alpha L \gg 1$ implies for the transfer function [Eq. (24)]

$$\Xi \approx -\frac{k_0^2 T_1}{k_2 + k} \exp(-ikL) \left[\frac{r_1}{K_F + k} + \frac{1}{K_F - k} \right], \quad (47)$$

which finally yields

$$\Xi \approx -k_0 T_1 t_2 \exp(-ikL) \frac{K_F + k_1}{(k + k_1)(K_F^2 - k^2)}. \quad (48)$$

If, in addition, we assume that the optical penetration depth is much smaller than the terahertz wavelength ($\alpha \gg k_0$) formula (48) can be further simplified,

$$\Xi \approx \frac{T_1 t_2 \exp(-ikL)}{n_1 + n} \left(\frac{-ik_0}{\alpha} \right). \quad (49)$$

This last approximation is valid for the experiments where the optical penetration depth is of the order of a few microns or less which is true for the interband transitions in direct gap semiconductors.

Note also that, as expected, Eq. (49) is equivalent to Eq. (44) in the strong absorption limit ($\alpha d \gg 1$) where all the optical power is absorbed in the first layer of a two-component sample.

B. Quasi-phase-matched case

We have stressed in Ref. 3 the importance of the phase matching condition for thick samples with low optical absorption. In such a sample the density of optical excitation is nonzero over the whole thickness of the sample or over its appreciable part. However, a high transient terahertz signal can be obtained only if the group velocity of the optical pulse in the sample is close to the phase velocity of the terahertz radiation. This is expressed mathematically by the equation: $K_F - k \approx -i\alpha$. Under this simplifying condition,

$$K_F + k \approx -i\alpha + 2k, \quad (50)$$

$$K_B - k \approx -i\alpha - 2K, \quad (51)$$

and the transfer function [Eq. (24)] becomes

$$\Xi \approx \frac{k_0 T_1 \exp(-ikL)}{n + n_2} \left[\frac{1 - \exp(-\alpha L)}{i\alpha} + \frac{r_1}{i\alpha - 2k} - \frac{R_2 \exp(-\alpha L)}{i\alpha + 2K} \right]. \quad (52)$$

The first term in the square brackets of Eq. (52) represents the quasi-phase-matched contribution, while the second and the third ones come from terms for which the phase matching cannot be achieved. Consequently, their relative importance increases for samples with stronger optical absorption (where the quasi-phase-matched term is diminished due to α). In contrast, if the optical absorption is low, $1/\alpha \gtrsim 500 \mu\text{m}$, the phase-matched term clearly dominates. In this case we obtain

$$\Xi \approx \frac{T_1 \exp(-ikL)}{n + n_2} \left(\frac{-ik_0}{\alpha} \right) [1 - \exp(-\alpha L)]. \quad (53)$$

If, in addition, $\alpha L < 1$, we can develop the exponential term into series to get

$$\Xi \approx \frac{T_1 \exp(-ikL)}{n+n_2}(-ik_0L). \quad (54)$$

From this expression the phase-matched character of the generation of the transient terahertz field is apparent through the linear thickness dependence.

It is important to note that the part of the optical pump pulse which is reflected at the output face of the sample cannot give rise to phase-matched terms. These terms have similar form to the second and third terms in the square brackets of Eq. (52), but they are even smaller as they are proportional to $R_{\text{opt}} \exp(-\alpha L)$ (where R_{opt} is the optical power reflection coefficient of the interface between the sample and medium 2).

V. PUMP PULSE WITH A FINITE LENGTH AND CONTINUOUS PUMPING

Our model is also able to describe the terahertz signal from a sample which was excited by an arbitrarily long optical pump pulse. The key is Eq. (4) where the Fourier transformation $F(\omega_p)$ of the pump pulse time-domain intensity profile can be introduced. This spectral function then enters, as a multiplicative factor, the expressions for the transfer function Ξ that we have derived up to now. The immediate consequence of this additional contribution is the spectral filtering of the response in ω_p : The factor reduces the available spectral range where the transient susceptibility can be experimentally determined.

A similar approach can be also applied to the case when the experimental data were taken in a non-collinear geometry, i.e., when the optical pump beam forms a nonzero angle with the terahertz beam. Simple geometrical considerations show that the different spatial parts of the pump beam reach the input surface of the sample within the finite input aperture at different times. This makes the excitation event spread in time, which can be taken into account in the first approximation by an effective lengthening of the pump pulse, hence by an introduction of an appropriate spectral distribution $F(\omega_p)$ in the expressions for Ξ .

It turns out that our model can be easily generalized to account for the case where the terahertz signal transmitted through the sample is measured under the regime of a continuous pumping (or possibly of a continuous influence of an external physical parameter such as bias electric field, magnetic field, etc.). In such experiments, one wishes to evaluate small modulations of the terahertz properties of the sample which are induced by the chosen external parameter. An experiment of this type has been reported recently;²¹ in that paper we determined the electric field induced changes in the dielectric properties of a strontium titanate thin film. A low-frequency (quasistatic) ac bias voltage (synchronizing at the same time the lock-in detection) was applied to the sample and the changes in the terahertz transmission of the sample were measured.

Such an experiment is also described by Eq. (21) and, depending on the sample geometry (thin film versus thick slab), by Eq. (22) or (24) where $\omega_p=0$ [i.e., $F(\omega_p)=\delta(\omega_p)$]. If

the sample excitation is homogenous, one also sets $\alpha \rightarrow 0$. For a thick sample where time windowing can be performed, we obtain

$$\Xi(\omega_p=0, \alpha=0) = -\frac{t_1 \exp(-ikL)}{n_2+n} \left[\frac{r_1+r_2}{2n} + ik_0L \right]. \quad (55)$$

Taking into account the relation $\Delta\chi=2n\Delta n$ and introducing Eq. (55) into Eq. (21) we finally find

$$\begin{aligned} \Delta t &\equiv \frac{\Delta E(\omega, 0)}{E_{\text{inc}}(\omega)} \\ &= t_1 t_2 \exp(-ikL) \left[\frac{1}{n} - \frac{1}{n+n_1} - \frac{1}{n+n_2} - ik_0L \right] \Delta n. \end{aligned} \quad (56)$$

It can be easily verified that this equation is reduced to

$$\Delta t = \frac{dt}{dn} \Delta n, \quad (57)$$

where

$$t = t_1 t_2 \exp(-ikL) \quad (58)$$

is the transmission of the slab in the ground state (without excitation).

An analogous relation can be found also for thin films; we obtain in the static limit

$$\Xi(\omega_p=0, \alpha=0) \Delta\chi(\omega) = \frac{dt}{dn} \Delta n, \quad (59)$$

where

$$t = \frac{t_1 t_2 \exp(-ikL)}{1 - r_1 r_2 \exp(-2ikL)}. \quad (60)$$

VI. ONE-DIMENSIONAL PHOTONIC STRUCTURES

Recently a modulator based on the photoexcitation of a semiconductor wafer inserted inside a one-dimensional photonic crystal has been reported.²² In that work the terahertz transmittance of the element was controlled by optical pulses generating free carriers at the surface of a GaAs defect layer enclosed in the middle of a Bragg-type layered structure.

As a rule layered structures have quite complicated dispersion and transmission properties. Notably, they may exhibit forbidden bands, where they totally reflect the incident radiation, and very narrow defect modes, which are characterized by a high transmission and a strong localization of the electromagnetic radiation in the vicinity of the defect layer.^{23–25} They are then usually designed to operate in this specific narrow spectral region and also their time response is of interest only in this particular range. The description in the frequency space is then highly appropriate.

The windowing procedure is not applicable for photonic crystals since the pulses partially reflected at various interfaces of the structure usually overlap in time. Hence we should take into account all the reflections of the terahertz wave within the structure. These add up to an output terahertz pulse which is substantially stretched in time compared to the input one; this effect is correlated with the presence of sharp features in the transmission spectra of the photonic

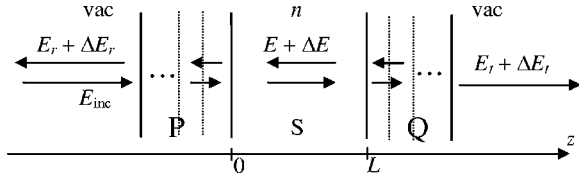


FIG. 4. Scheme of the fields in the photoexcited layered structure consisting of three blocks: P , S , and Q . Blocks P and Q are general layered structures which are optically transparent and/or insensitive to photoexcitation; these blocks surround the sample layer S which is the only part exhibiting a photoinduced terahertz response. The origin of coordinates is chosen at the input face of the sample layer for convenience.

structure. Consequently, such a structure makes the data very sensitive to the frequency mixing as it was pointed out in the text below Eq. (3), unless the frequencies involved in the dynamical response of the system are small compared to the width of the sharp features in the transmission spectra of the structure. In other words, the quasi-steady-state approximation of the data analysis can be used only if the response of the photoexcited sample is slower than the length of the terahertz pulse stretched by the structure which can easily exceed tens or even hundreds of picoseconds.²² The quasi-steady-state approximation is not able to describe correctly faster dynamics; our theory then appears to be highly appropriate to provide a background for the interpretation of the time response of these fast devices.

At first, it is necessary to calculate the distribution of the electromagnetic field in the structures in equilibrium; this allows one to construct properly the right-hand side of the wave equation [Eq. (2)].

For the description of the equilibrium properties of the layered structures we use the transfer matrix formalism.^{26,27} Let us consider a stack of homogeneous layers with their normals parallel to the z axis. The transfer matrix Γ_j links the tangential components of the electric and magnetic fields at input and output interfaces of the j th layer,²⁷

$$\begin{pmatrix} E_{j-1,j} \\ \eta_0 H_{j-1,j} \end{pmatrix} = \Gamma_j \begin{pmatrix} E_{j,j+1} \\ \eta_0 H_{j,j+1} \end{pmatrix}, \quad (61)$$

where for the case of the normal incidence, which is treated in this paper, the transfer matrices Γ_j read

$$\Gamma_j = \begin{pmatrix} \cos(k_j d_j) & i \sin(k_j d_j) / n_j \\ i n_j \sin(k_j d_j) & \cos(k_j d_j) \end{pmatrix}, \quad (62)$$

where d_j is the layer thickness, n_j is its complex refractive index, $k_j = \omega n_j / c$ is the z component of the wave vector inside the layer, and $\eta_0 = \sqrt{\mu_0 / \epsilon_0}$ is the vacuum wave impedance. The transfer matrix of the whole layered structure is obtained as a product of transfer matrices of individual layers.

Let us now consider the structure schematically shown in Fig. 4. It consists of three blocks denoted by the letters P , S , and Q . The blocks P and Q consist of optically transparent media; they are general layered structures which do not exhibit any photoinduced terahertz response while S is a single homogeneous layer which exhibits a transient terahertz response upon photoexcitation. Therefore we call S a sample layer. We introduce a notation similar to that defined by Eqs.

(7)–(14): The transfer matrices and their elements evaluated at frequency ω are denoted by lower-case letters (transfer matrices p , s , and q and matrix elements p_{ij} , s_{ij} , and q_{ij}), while by using upper-case letters we refer to the transfer matrices (P , S , Q) or the matrix elements (P_{ij} , S_{ij} , Q_{ij}) evaluated at frequency $\omega - \omega_p$.

The transmission and reflection coefficients at terahertz frequency ω of any general layered structure surrounded by air read

$$t_m = \frac{2}{m_{11} + m_{12} + m_{21} + m_{22}}, \quad (63)$$

$$r_m = \frac{m_{11} + m_{12} - m_{21} - m_{22}}{m_{11} + m_{12} + m_{21} + m_{22}}, \quad (64)$$

where m_{ij} are the transfer matrix components of the structure. Analogous expressions can be also written for the transmission and reflection coefficients T_M and R_M calculated at frequency $\omega - \omega_p$.

The electric field distribution in the sample layer which is to be substituted to the right-hand side [Eq. (5)] of the wave equation reads

$$E(\omega - \omega_p; z) = E(\omega - \omega_p; 0) \cos(Kz) - i \frac{\eta_0 H(\omega - \omega_p; 0)}{N} \sin(Kz), \quad (65)$$

where the fields at the input face of the sample layer can be calculated using the transfer matrix formalism as follows:

$$\begin{pmatrix} E(0) \\ \eta_0 H(0) \end{pmatrix} = SQ \begin{pmatrix} E_t \\ E_t \end{pmatrix} = SQ \begin{pmatrix} T_{PSQ} E_{\text{inc}} \\ T_{PSQ} E_{\text{inc}} \end{pmatrix}; \quad (66)$$

T_{PSQ} is the transmission coefficient of the whole PSQ structure. The substitution into Eq. (65) yields

$$E(z) = \left[\exp(-iKz) \frac{1 + R_0 R_{SQ}}{T_0 T_{SQ}} + \exp(+iKz) \frac{R_0 + R_{SQ}}{T_0 T_{SQ}} \right] T_{PSQ} E_{\text{inc}}, \quad (67)$$

where $R_0 = (N-1)/(N+1)$ and $T_0 = 2N/(N+1)$.

The right-hand side of the wave equation then has a form analogous to Eq. (15),

$$U = -k_0^2 \Delta \chi E_{\text{inc}} [X_F \exp(-iK_F z) + X_B \exp(-iK_B z)], \quad (68)$$

with

$$X_F = (1 + R_0 R_{SQ}) \frac{T_{PSQ}}{T_0 T_{SQ}}, \quad (69)$$

$$X_B = (R_0 + R_{SQ}) \frac{T_{PSQ}}{T_0 T_{SQ}}. \quad (70)$$

The transient field in the sample layer S is then described by an equation similar to Eq. (18),

$$\Delta E(z) = \delta \exp(-ikz) + \gamma \exp(ikz) - k_0^2 \Delta \chi E_{\text{inc}} \left[\frac{X_F \exp(-iK_F z)}{k^2 - K_F^2} + \frac{X_B \exp(-iK_B z)}{k^2 - K_B^2} \right]. \quad (71)$$

The boundary conditions for the transient field [cf. Eqs. (19) and (20) and Fig. 4]

$$\begin{pmatrix} \Delta E_r \\ -\Delta E_r \end{pmatrix} = P \begin{pmatrix} \Delta E(0) \\ \eta_0 \Delta H(0) \end{pmatrix}, \quad \begin{pmatrix} \Delta E(L) \\ \eta_0 \Delta H(L) \end{pmatrix} = Q \begin{pmatrix} \Delta E_t \\ \Delta E_t \end{pmatrix} \quad (72)$$

lead to Eq. (21), with an appropriate form of the transfer function Ξ describing the response of a photonic crystal containing a photoexcited layer. We write here the solution only for the particular case when $\alpha L \gg 1$ (the excitation pulse is completely absorbed within the sample layer), which is well fulfilled in the previously reported experiment.²² Under this assumption the transfer function Ξ takes the following form:

$$\Xi = k_0 \frac{t_{psq}}{2} \left[X_F \frac{k_0(p_{11} + p_{21}) + K_F(p_{12} + p_{22})}{k^2 - K_F^2} + X_B \frac{k_0(p_{11} + p_{21}) + K_B(p_{12} + p_{22})}{k^2 - K_B^2} \right]. \quad (73)$$

This expression can be further simplified assuming $\alpha \gg k_0$ and taking into account that

$$p_{12} + p_{22} = \frac{1 + \tilde{r}_p}{t_p}, \quad (74)$$

where \tilde{r}_p is the amplitude reflection coefficient of the block P standing alone in the air if the incident light impinges on the block from the right-hand side.²⁸ One finally finds

$$\Xi = \left(-\frac{ik_0}{\alpha} \right) \frac{t_{psq} T_{PSQ}}{2} \frac{1 + \tilde{r}_p}{t_p} \frac{1 + R_{SQ}}{T_{SQ}}. \quad (75)$$

In Ref. 22 we experimentally characterized the response of a photonic crystal inside its forbidden band close to a defect mode. The crystal consisted of parts P and Q of alternating layers with high and low refractive indices and of a defect S enclosed in the middle. In such a case the transmission function of the structure [$t_{psq}(\omega)$ or $T_{PSQ}(\omega - \omega_p)$] is very small within the forbidden band (say between some frequencies ω_1 and ω_2) with the exception of a quite narrow range of the defect mode (at ω_0). The existence of the forbidden band is related to the periodic character of the blocks P and Q while the appearance of the defect mode is connected to the presence of the defect layer S in the middle of the structure. In contrast, no defect mode appears in the forbidden band if the periodicity of the photonic crystal is not broken by the defect (i.e., if the defect layer is either absent or placed as an outer layer of the structure at any side).

We discuss the two right-most terms of Eq. (75): $(1 + \tilde{r}_p)/t_p$ and $(1 + R_{SQ})/T_{SQ}$. The field reflectance of the blocks P and SQ inside the forbidden band reaches values close either to -1 or 1 . In the former case the discussed terms are small and the transmittance of the structure at the defect mode frequency is rather insensitive to the photoexcitation.

In the latter case, these terms do not exhibit any sharp feature between ω_1 and ω_2 and show only a flat frequency dependence. Moreover, they reach quite high values of $\sim 2/t_p$ and $2/T_{SQ}$ ($t_p, T_{SQ} \ll 1$) accounting for the enhancement of the interaction due to the field localization close to the defect layer.²⁹ One can show that in this case the field distribution inside the structure shows antinodes at the interfaces PS and SQ .²² The high value of these terms also makes the parts of the spectra outside the band gap relatively unimportant.

The transient response of the element within the band gap is then proportional to

$$\Delta E_t(\omega, \omega_p) \propto -t_{psq} \Delta \sigma(\omega, \omega_p) E_t(\omega - \omega_p). \quad (76)$$

In this equation only small values of ω_p are considered such that $\omega - \omega_p$ stays within the forbidden band. It follows that the dynamical response of the whole photonic element is determined by the lifetime of the carriers in the semiconductor encoded into $\Delta \sigma$ and by the lifetime of the terahertz wave inside the structure introduced through the linewidth of the defect mode in t_{psq} . Note that the transmitted terahertz reference wave $E_t(\omega - \omega_p)$ shows a sharp peak at $\omega_p = \omega - \omega_0$ due to the equilibrium transmission properties of the photonic structure [$E_t(\omega - \omega_p) = E_{\text{inc}}(\omega - \omega_p) T_{PSQ}$]. It follows that (i) if the decay of the conductivity in the sample layer occurs on the picosecond or subpicosecond time scale, such that $\Delta \sigma(\omega, \omega_p)$ is a relatively broad function in ω_p , the transient response of the structure is anyway limited to small values of ω_p being controlled by the lifetime of the terahertz wave in the structure through T_{PSQ} ; (ii) the frequency mixing in the OPTP experiment can be neglected only if $\Delta \sigma(\omega, \omega_p)$ is a narrow function of ω_p compared to the linewidth of the defect mode at $\omega - \omega_p = \omega_0$ given by T_{PSQ} .

We discussed here merely the basic predictions for the transient properties of photonic structures. A more detailed analysis supplied with experimental results on particular structures will be published elsewhere.³⁰

The problem of multiple reflections of the optical pump pulse in the structure needs to be discussed and evaluated separately for any particular structure. However, in our case of an optically opaque sample layer there are no back reflections from the part Q . The block P is then usually required to be transparent for the optical radiation in order to ensure an efficient optical pump. The total optical power incident on the interface between the blocks P and S can be calculated by using the transfer matrix formalism applied in the optical spectral range. The issue of the time extent of the optical excitation event due to these reflections will not be of crucial importance in the majority of cases. This is because of an unavoidable increase of the response time of the photonic structure due to the large lifetime of terahertz photons inside the structure given by the form of t_{psq} .

VII. CONCLUSION

We have described the propagation of terahertz pulses in photoexcited layered media. The transient terahertz wave forms obtained in the transmission geometry are given by a product of the incident field (at a mixed frequency), transient susceptibility, or conductivity, and a transfer function de-

pending solely on the sample properties in the ground state and on the properties of surrounding media. The forms of the transfer functions for the most frequently encountered experimental cases are summarized in Fig. 2. Our treatment is also applicable to multicomponent samples (where different layers of the sample structure exhibit different transient responses) and for one-dimensional photonic structures.

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¹¹In some experimental cases including time-resolved spectroscopy of solutions or thin organic films it may be useful to define a transient susceptibility per unit excitation density as $\Delta\chi/\alpha$. Note also that the absorption

coefficient α appears in the denominator of the majority of expressions for Ξ in Fig. 2.

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