Interpretation of ultrafast pump-probe terahertz experiments in the time domain: How to exploit two-dimensional correlations

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Optical-pump-terahertz-probe spectroscopy has the potential to distinguish between several classes of carrier motion, among others high-frequency oscillatory motion, free motion, and quasifree motion within bounded domains. We present a simplified formalism, applicable to thin samples, which allows identification of these three classes of photoexcited species on basis of the two-dimensional correlations in the time-domain experimental data.

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I. INTRODUCTION

Time-resolved THz spectroscopy[1–14] is rapidly imposing as a technique to determine the mobility of optically excited carriers in a wide variety of materials. Even though the technique has ultrashort capability (100-fs pulsed lasers have been commercially available for decades), it is mostly used as a method to determine conductivities on the subnanosecond time scale. In the few cases in which subpicosecond dynamics are addressed, the theoretical framework is either too complex[15] or insufficient [in the sense that the two-dimensional (2D)-correlated data are described in terms of uncorrelated 1D stacks[8,9,16–24]] to provide a satisfactory description of the measured data. There is one notable exception: Beard and Schmuttenmaer[25] were able to give a beautiful simulation of their measurements on a dye (tetra-butyl-naphtalocyanine, also known as TBNC) in toluene, a simulation that was later fine-tuned by Nemec and co-workers.[26]

It is remarkable that up to date hardly any 2D contour plots have been published in the literature, and on those rare occasions the meanders observed at (sub)picosecond pump-probe delays are left without explanation.[17,27] Numerical simulations of the full Maxwell equations have the drawback of being very time consuming, and they are useful only if one knows the nature of the primary photoproducts beforehand. Yet even for model systems like bulk GaAs, with enormous signal levels, the primary photoproducts remain elusive: Beard and co-workers write that they were not able to account for differences between simulation and experiment in the first picosecond after excitation.[15]

An important step forward was made by Nemec, Kadlec, and Kuzel in 2002,[28] who derived an analytical solution in the perturbative limit. This solution has the advantage of simplification without loss of applicability (in the large majority of experiments one is specifically interested in the perturbative regime), and calculations need much less computing time.[26,29] However, the major issue, that of the immediate assessment of the primary photoproducts, remained unsolved.

In this paper we elaborate on previous work[30,31] by deriving a simple expression in the thin-film limit. It shows that a differential representation of the 2D time-domain data unveils the nature of the primary photoproducts. This new representation paves the way to exploiting the full time-resolving capability of pump-probe terahertz spectroscopy, and effectively assessing the nature of the primary photoproducts: specifically, we address the differentiation between excitons, free charges in unbounded media, and quasifree charges in bounded media (like in polymers, nanorods, or crystalline islands).

II. THEORETICAL FRAMEWORK

In an optical-pump terahertz-probe experiment a single laser beam is divided into three parts: (i) a pump beam, used to photoexcite the sample; (ii) a generation beam, to generate a THz pulse in either a nonlinear crystal[32] or a biased emitter;[33] and (iii) a detection beam, to detect the time-dependent waveform of the transmitted THz pulse by electro-optical sampling[32] or using a semiconductor antenna.[33] These three beams can be delayed separately, though only two of the three delays are independent as far as the experimental results are concerned.

The main approximations we make in our analysis is in assuming a perturbative response of the system to the pump pulse (this corresponds to neglecting the interaction between the pump-induced polarizabilities), and an ultrathin sample, thereby disregarding the exponential decay of the excitation density (Lambert-Beer) and corresponding diffusion effects.[34–37] Other assumptions made in the analysis are that the medium is isotropic and homogeneous, with a material response depending on frequency only. In a previous publication it was shown that for the above assumptions the differential electric field $\Delta E(t,\tau)$ relates to the driving field $E_0(t)$ as follows:[31]

$$\Delta E(t,\tau) = -\beta \frac{q^2\rho_0}{m}\theta(\tau) \int_0^\tau dt' E_0(t-t')\Delta\dot{G}(t')e^{-\Gamma(\tau-t')} \qquad \beta \equiv \frac{d}{2n\varepsilon_0 c} \tag{1}$$

with $\Delta G \equiv G_{es} - G_{gs}$ the difference between the excited and ground-state Green functions; $\theta(\tau)$ the Heaviside function; Γ, q, ρ_0, m the carrier's reciprocal lifetime, charge, mass, and number density; d, ε_0, c, n the sample thickness, the vacuum permittivity, the speed of light, and the sample's index of refraction in the terahertz window, respectively. The two temporal variables determine the relative delays for the detection, pump, and generation paths, L_d, L_p, L_g respectively,

according to $t \equiv (L_d - L_g)/c$ and $\tau \equiv (L_d - L_p)/c$.[26,29,30] The couple (t,τ) is the natural choice for experiments, as it exploits the fact that for negative τ the experimental data vanish. From a theoretical point of view it is not the natural choice, because the time-dependent solution to Maxwell's equations is obtained when varying the detection delay with fixed pump and generation delays.

Equation (1) applies for a single carrier species, created instantaneously, and decaying exponentially in time. It can be generalized to include several carrier species, with an arbitrary temporal evolution of the population, by introducing the two-dimensional conductive response function $\Delta\sigma(t,\tau)$ and its Fourier transform $\Delta\sigma(\omega,\zeta)$:

$$\begin{aligned} \Delta E(t,\tau) &= -\beta \int_{-\infty}^{\infty} dt' E_0(t-t')\Delta\sigma(t',\tau-t'), \\ \Delta E(\omega,\zeta) &= -\beta E_0(\omega)\Delta\sigma(\omega+\zeta,\zeta). \end{aligned} \tag{2}$$

This generalization presents the differential field as a convolution in the time domain. Equation (2) reduces to Eq. (1) whenever the two-dimensional response assumes the special form

$$\Delta\sigma(t,\tau) = \frac{q^2\rho_0}{m}\theta(\tau)\Delta\dot{G}(t)e^{-\Gamma\tau}. \tag{3}$$

The generalized conductivity describes both Ohmian conduction (real part of the frequency-dependent conductivity) and dielectric effects (imaginary part), which have their origin in the current and polarization terms of the Maxwell equations.

The Fourier transforms of the measured differential and driving fields, $\Delta F(\omega,\zeta)$ and $F_0(\omega)$, can be obtained from the differential and driving fields in the sample by multiplication with the envelope functions of the pump (ξ_p) and detection (ξ_d) pulses, as follows:

$$\begin{aligned} \Delta F(\omega,\zeta) &= \Delta E(\omega,\zeta)\xi_d(\omega+\zeta)\xi_p(\zeta) \\ F_0(\omega) &= E_0(\omega)\xi_d(\omega). \end{aligned} \tag{4}$$

Note that for the measured differential field $\Delta F(\omega,\zeta)$ the argument of the detection envelope in frequency domain is not simply ω but $\omega+\zeta$, because of the fact that in time domain the detection convolution occurs along the diagonal, for which $\tau - t$ is constant (i.e., the generation delay line L_g is constant).[26,29,30] Equation (4) can further be generalized to include effects of detection efficiency[38] and beam propagation[39,40] by straightforward multiplication.

For a single excited species, decaying exponentially with rate Γ, the two-dimensional conductivity $\Delta\sigma(\omega,\zeta)$ can be written in terms of the one-dimensional steady-state conductivity $\Delta\sigma_{st}(\omega)$. Indeed, from Eq. (3) one obtains the identity

$$(\Gamma - i\zeta)\Delta\sigma(\omega,\zeta) = \Delta\sigma_{st}(\omega) \equiv -i\omega\frac{q^2\rho_0}{m}\Delta G(\omega). \tag{5}$$

Note that the dimensions of the two conductivities $\Delta\sigma(\omega,\varsigma)$ and $\Delta\sigma_{st}(\omega)$ differ; in time domain, however, the two conductivities have the same dimension (Ω^{-1} m^{-1} s^{-1} in SI units). Using Eqs. (4) and (5), a single species with infinite lifetime produces a differential field of the form

$$\begin{aligned} -i\zeta \lim_{\Gamma\to 0}\frac{\Delta F(\omega,\zeta)}{\xi_p(\zeta)} &= -\beta E_0(\omega)\Delta\bar{\sigma}_{st}(\omega+\zeta), \\ \Delta\bar{\sigma}_{st}(\omega) &\equiv \Delta\sigma_{st}(\omega)\xi_d(\omega), \end{aligned} \tag{6}$$

The bar over the steady-state conductivity stands for multiplication with the detection pulse in frequency domain, or equivalently, for convolution in the time domain. The above expression suggests that the experimental data be displayed, in time domain, as a temporal derivative along the vertical axis (pump delay). Moreover, Eq. (6) nicely illustrates that pump-probe terahertz spectroscopy is able to retrieve from the experimental data only the convolution (in time domain) of conductivity and detection envelope. Also note that the right-hand side of Eq. (6) contains the actual driving field $E_0(\omega)$ rather than the measured driving field $F_0(\omega)$. This implies that, for a single species of infinite lifetime, the time-derived experimental data, after deconvolution with the pump pulse, can be written as a simple product of the driving field and the convoluted steady-state response:

$$\lim_{\Gamma\to 0}\frac{\partial\Delta F(t,\tau)}{\partial\tau} \div \xi_p(\tau) = -\beta E_0(t-\tau)\Delta\bar{\sigma}_{st}(\tau). \tag{7}$$

The symbol $\otimes$ denotes a convolution, and $\div$ denotes a deconvolution. According to Eq. (7), in the special case of a single species with infinite lifetime and in the case of negligible pump duration, the time-shifted driving field $E_0(t)$ is displayed along any horizontal line (τ const), and the convoluted steady-state conductivity along any diagonal ($t-\tau$ const).

In practice the conditions for Eq. (7) are not realized, as the photoexcited species have finite lifetimes and, possibly, a finite rate of ingrowth. Yet one may still obtain a good impression of the steady-state conductivity by observing the τ derivative of the two-dimensional experimental data along the 45° diagonal. In the case of finite population lifetimes and multiple species the generalized two-dimensional conductivity must be retrieved from the data upon using Eqs. (2) and (4), instead of Eq. (7). Due to the intrinsic limitation of the laser pulse duration, the retrieved experimental quantity is not the conductivity itself, but rather its convolution with the laser pulse:

$$\begin{aligned} \Delta\bar{\sigma}_{st}(t) &\equiv \Delta\sigma_{st}(t)\otimes\xi_d(t), \\ \Delta\bar{\sigma}(t,\tau) &\equiv \Delta\sigma(t,\tau)\otimes\otimes\xi_d(t)\xi_p(\tau). \end{aligned} \tag{8}$$

The one-dimensional convoluted conductivity $\Delta\bar{\sigma}_{st}(t)$ is derived from Eq. (7), and the two-dimensional convoluted conductivity $\Delta\bar{\sigma}(t,\tau)$ follows upon Fourier back-transforming the field ratio $\Delta F(\omega-\zeta,\zeta)/E_0(\omega-\zeta)$ [cf. Eqs. (2) and (4)].

III. ESTIMATION AND RETRIEVAL OF THE CONVOLUTED CONDUCTIVITY

Equations (5) and (8) determine the relation between the electronic response and the convoluted conductivity. For the three basic types of response considered in this paper (Drudean free motion, oscillatory motion of a polarizable exciton, and quasifree motion on a bounded domain), the Green functions have the following form:

$$\begin{aligned} G_{\text{Drude}}(t) &= \frac{1-e^{-\gamma t}}{\gamma}\theta(t), \\ G_{\text{exciton}}(t) &= \frac{\sin(\Omega t)}{\Omega}e^{-\gamma t/2}\theta(t), \\ G_{\text{domain}}(t) &= 8\frac{mD}{k_BT}\sum_{k=0}^{\infty} c_k^{-2}e^{-(c_k/a)^2 Dt}\theta(t). \end{aligned} \tag{9}$$

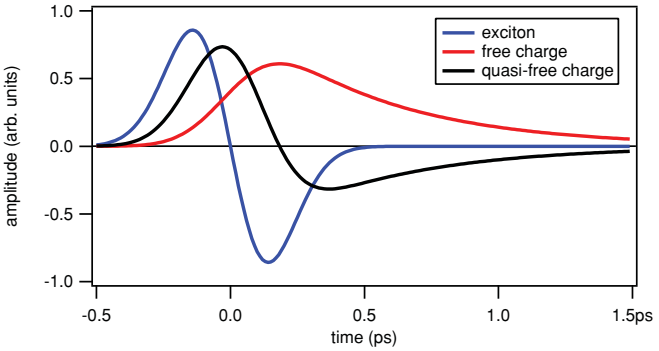

FIG. 1. (Color online) Convoluted stead-state conductivity $\Delta\bar{\sigma}_{st}(t)$ for three different species: free charges, tightly bound excitons, and quasifree charges on a bounded domain. Although the amplitudes of the three traces are chosen arbitrarily, the temporal succession of the peaks is fully determined by the Green functions. The excitonic curve is the derivative of the laser pulse (assumed Gaussian). For the scattering rate of the free charge $\gamma = 2$ ps^{-1} was taken; $D/a^2 = 0.2$ ps^{-1} for the quasifree charge.

Here, γ represents the scattering rate; $\Omega = \sqrt{\omega_{res}^2 - (\gamma/2)^2}$ is the underdamped excitonic oscillation frequency; $k_B T$ is the product of Boltzmann constant and temperature; a is the linear size of the bounded domain; $c_k \equiv (2k+1)\pi$; and D is the diffusion coefficient in an unbounded medium. The Green function for free motion on a bounded domain was derived assuming equal probability of excitation throughout the domain, by applying Kubo's formalism to classical diffusive motion (see the Appendix). In the high-frequency limit the pump-induced field is proportional to the derivative of the driving field, as can be appreciated in measurements of exciton polarizability.[41] This behavior corresponds with the theoretical fact that, for $\omega_{res} \to \infty$ and finite scattering rate γ, the excitonic Green function approaches a δ function in time (see the Appendix).

In Fig. 1 the one-dimensional convoluted conductivity $\Delta\bar{\sigma}_{st}(t)$ is shown corresponding to the electronic response for three different species: a free (Drude) electron, a polarizable exciton, and a free charge on a bounded domain. The conductivities were calculated combining Eqs. (5), (8), and (9). For excitons and quasifree charges on a bounded domain the integral over the full curve vanishes, implying no net displacement of the charge after passage of the THz driving field.

The signatures presented in Fig. 1 allow for straightforward identification of a large majority of the observed photoexcited species. As an example we show a theoretical prediction of the measured differential field $\Delta F(t,\tau)$ for a purely excitonic signal, as would be measured typically for photoexcited nanocrystals of CdSe.[41] Note that the signatures associated with the exciton and the quasifree charge are both single cycle, differing only by the relative amplitude of the two extrema (symmetric for the exciton, asymmetric for the quasifree charge). A more reliable criterion for distinguishing between the two species might therefore be found in the fact that the excitonic signature vanishes at zero time delay, while the quasifree signature is close to peaking. Furthermore, it is important to realize that any frequency dependence in

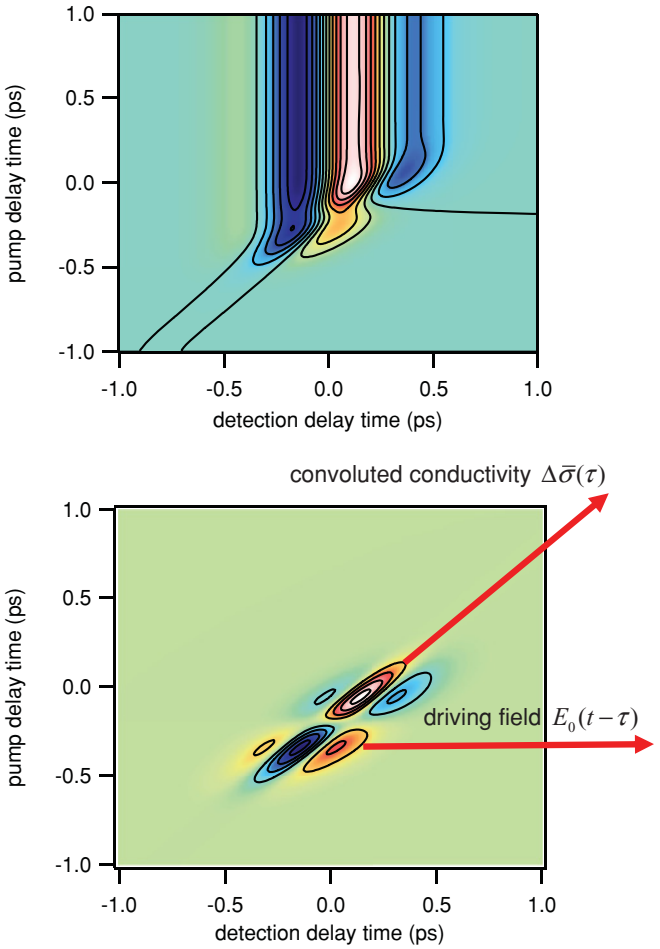

FIG. 2. (Color online) Theoretical prediction of the convoluted differential field $\Delta F(t,\tau)$ (upper panel) and its vertical temporal derivative $\partial\Delta F(t,\tau)/\partial\tau$ (lower panel) for the case of a purely excitonic response. In both figures, detection time t runs along the horizontal axis, and pump time τ runs along the vertical axis.

beam propagation[39,40] or detection efficiency[38] distorts the signatures shown; specifically, in a standard experimental configuration one may expect low frequencies to be filtered out.

Figure 2 shows both the measured differential field and its derivative to time. In the latter representation, the driving field shows up along the horizontal axis. The convoluted conductivity must be read off along the diagonal, as the driving field does not vary for constant $t - \tau$ [cf. Eq. (7)]. For arbitrary forms of the two-dimensional conductivity $\Delta\bar{\sigma}(t,\tau)$ the vertical derivative of the differential field $\partial\Delta F(t,\tau)/\partial\tau$ still is useful for estimating the nature and population dynamics of the photoexcited species.

In this paper we have presented a theoretical framework for optical-pump terahertz-probe spectroscopy on thin samples. From the relation of the measured differential field to the generalized conductivity it was deduced that the character of the observed species (free charge, tightly bound exciton, and quasifree charge on a bounded domain) is readily appreciated from the experimental data when displayed as a temporal derivative to the pump-probe delay time. The signatures

of these three species are distinct enough to grant their identification on the basis of the experimental data.

APPENDIX: GREEN FUNCTION FOR MOTION ON A BOUNDED DOMAIN

The mean-square displacement of a carrier, initially localized with equal probability at an arbitrary point on a linear chain of length a, has the following time dependence:[42,43]

$$\langle \Delta x^2(t) \rangle = a^2 \theta(t) \left\{ \frac{1}{6} - 16 \sum_{k=0}^{\infty} c_k^{-4} e^{-(c_k/a)^2 Dt} \right\}, \qquad c_k = (2k+1)\pi, \tag{A1}$$

where D stands for the diffusion constant in a polymer of infinite length; it is related to the dc-mobility by the Einstein relation $\mu_{\rm dc} = eD/k_BT$. The Kubo formalism, valid in the perturbative regime (i.e., for low driving fields), relates the mobility to the Fourier transform of the velocity autocorrelation function,[44,45] which may be expressed as the second time derivative of the average displacement:[46,47]

$$\mu(\omega) = -\frac{q}{2k_BT} \int_0^{\infty} dt e^{i\omega t} \frac{d^2}{dt^2} \langle \Delta x^2(t) \rangle \tag{A2}$$

with e, k_B, T the elementary charge, Boltzmann's constant, and temperature, respectively. In Eq. (A2) the high-temperature limit was used for the universal prefactor;[44,45] care should be taken, however, as the usual approximation $\frac{1}{2}\hbar\omega \coth(\frac{1}{2}\hbar\omega/k_BT) \approx k_BT$ is on the verge of its validity, since for 1 THz and room temperature $k_BT \approx 6\hbar\omega$.

On the other hand, from Eq. (5), the mobility is proportional to the Green function in frequency domain:

$$\mu_{st}(\omega) = \frac{\sigma_{st}(\omega)}{q\rho_0} = -i\omega \frac{q}{m} G(\omega). \tag{A3}$$

From Eqs. (A1)–(A3) it follows that the Green function for diffusive motion along a linear chain is proportional to the temporal derivative of the mean-square displacement:

$$G_{\rm diffusion}(t) = \frac{m}{2k_BT} \frac{\partial}{\partial t} \left\langle \Delta x^2(t) \right\rangle = 8 \frac{mD}{k_BT} \theta(t) \sum_{k=0}^{\infty} c_k^{-2} e^{-(c_k/a)^2 Dt}. \tag{A4}$$

Although this Green function implies an infinite acceleration at time $t = 0$, it is in most cases appropriate for our goal, due to the fact that the physically impossible discontinuity in the particle's position at zero time is smeared out (and thereby rendered continuous) by the convolution with the detection pulse. The diffusive Green function of Eq. (A4) explains why the pump-induced field leads with respect to the Terahertz waveform for experiments performed on semiconductor polymers,[11,17,48] the lead angle increasing with D/a^2. However, it fails to reproduce, in the limit of infinite chain length $a \to \infty$, the Drude Green function for a free charge [Eq. (9)]; this is due to the fact that the diffusion equation allows for infinitely fast mass displacements, faster than the natural limit established by the electron's inertia. A primitive but effective way to correct for the unphysical acceleration implied in the diffusive response is to limit the latter as follows:

$$G_{\rm domain}(t) \equiv \min \left[\frac{1 - e^{-\gamma_D t}}{\gamma_D}, G_{\rm diffusion}(t) \right], \tag{A5}$$

where "min" selects the smallest of the two arguments. Due to this limitation the Green function for diffusion on a bounded domain $G_{\rm domain}(t)$ starts out like a Drude response with scattering rate $\gamma_D = k_BT/mD$, until at some critical time (when the Drude response equals the diffusive response), the diffusive response $G_{\rm diffusion}(t)$ takes over. Equation (A5) displays the correct limits for both infinitely long and infinitely short chain lengths:

$$\lim_{a\to\infty} G_{\rm domain}(t) = G_{\rm Drude}(t), \qquad 12\gamma_D D \lim_{a\to 0} \frac{G_{\rm domain}(t)}{a^2} = \gamma \lim_{\omega_{res}\to\infty} \omega_{res} G_{\rm exciton}(t) = \delta(t). \tag{A6}$$

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