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# Frequency resolved cross-correlation between optical and terahertz pulses: application to ultrashort laser pulse characterization

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**Abstract:** We have analyzed both theoretically and experimentally the spectrum of the optical pulses produced by the interaction of optical and THz pulses in a ZnTe crystal. Recorded as a function of the delay between the two pulses, the resulting spectrogram can be viewed as a frequency resolved cross-correlation between the optical and THz pulses making it possible to characterize the optical pulse.

**OCIS codes:** (190.7110) Ultrafast nonlinear optics; (320.7100) Ultrafast measurements; (300.6495) Spectroscopy, terahertz.

### **References and links**

- L. Duvillaret, S. Rialland, and J.-L. Coutaz, "Electro-optic sensors for electric field measurements. I. Theoretical comparison among different modulation techniques," J. Opt. Soc. Am. B 19(11), 2692–2703 (2002).
- L. Duvillaret, S. Rialland, and J.-L. Coutaz, "Electro-optic sensors for electric field measurements. II. Choice of the crystals and complete optimization of their orientation," J. Opt. Soc. Am. B 19(11), 2704–2715 (2002).
- G. Gallot and D. Grischkowsky, "Electro-optic detection of terahertz radiation," J. Opt. Soc. Am. B 16(8), 1204–1212 (1999).
- Y. Shen, T. Watanabe, D. A. Arena, C.-C. Kao, J. B. Murphy, T.Y. Tsang, X. J. Wang, and G. L. Carr, "Nonlinear cross-Phase modulation with intense single-cycle terahertz pulses," Phys. Rev. Lett. 99, 043901 (2007).
- Y. Shen, G. L. Carr, J. B. Murphy, T. Y. Tsang, X. Wang, and X. Yang "Spatiotemporal control of ultrashort laser pulses using intense single-cycle terahertz pulses," Phys. Rev. A 78, 043813 (2008).
- M. D. Thomson, M. Kre
  ß, T. Löffler, and H. G. Roskos, "Broadband THz emission from gas plasmas induced by femtosecond optical pulses: From fundamentals to applications," Laser Photonics Rev. 1(4), 349–368 (2007).
- S. Linden, H. Giessen, and J. Kuhl, "XFROG A new method for amplitude and phase characterization of weak ultrashort pulses," Phys. Stat. Sol. (b) 206(1), 119-124 (1998).
- M. Cornet, J. Degert, E. Abraham, and E. Freysz, "Terahertz Kerr effect in gallium phosphide crystal," J. Opt. Soc. Am. B 31(7), 1648–1652 (2014).
- 9. P. N. Butcher and D. Cotter, The Elements of Nonlinear Optics (Cambridge University, 1991).
- G. D. Boyd and M. A. Pollack, "Microwave nonlinearities in anisotropic dielectrics and their relation to optical and electro-optical nonlinearities," Phys. Rev. B 7(12), 5345–5359 (1973).
- 11. D. T. F. Marple, "Refractive index of ZnSe, ZnTe, and CdTe," J. Appl. Phys. 35(3), 539–542 (1964).
- Q. Wu and X.-C. Zhang, "Design and characterization of traveling-wave electrooptic terahertz sensors," IEEE J. Sel. Top. Quantum Electron. 2(3), 693–700 (1996).
- A. Nahata, A. S. Weling, and T. F. Heinz, "A wideband coherent terahertz spectroscopy system using optical rectification and electro-optic sampling," Appl. Phys. Lett. 69(16), 2321–2323 (1996).
- 14. M. Born and E. Wolf, Principles of optics (Cambridge University, 1999).
- 15. J.-C. Diels and W. Rudolph, Ultrashort Laser Pulse Phenomena (Academic Press, 2006).

### 1. Introduction

Nonlinear interaction of terahertz (THz) and ultrashort optical pulses in Zinc Blende crystals is widely used to perform electro-optic (EO) sampling of THz pulses [1, 2]: the THz pulse, due to the Pockels effect, induces a phase modulation of the optical pulse which is measured for different time delays  $\tau$  between the two pulses. In most experiments, the Pockels-induced phase is detected with photodiodes and very few attention is paid to the evolution of the spectrum of the probe pulse. Actually, according to Gallot and Grischkowsky [3], the spectrum of the optical pulse resulting from the interaction between the THz and optical fields in conventional EO sampling can be understood as the generation of phase-coherent sidebands, by sumand difference-frequency mixing (SFG-DFG), on the spectrum of the incident optical pulse. These sidebands can also be regarded as the result of a Pockels-induced cross-phase modulation (XPM). For intense THz pulses, it has been shown that higher order XPM, namely Kerrinduced, adds to this Pockels XPM to produce large modification of the optical spectrum [4, 5]. Unfortunately, in these experiments performed in a  $\langle 110 \rangle$  ZnTe crystal, the interaction of the THz wave, polarized along the  $\langle 001 \rangle$ -axis, with the probe pulse, cross-polarized with respect to the latter, leads to sidebands which are superimposed to the spectrum of the incident optical pulse. As a consequence, and unless very intense THz fields are applied, one can hardly infer from such an experiment what is actually the spectral content of the sidebands resulting from XPM, whatever its origin. From this vantage point, spectrally resolved EO sampling with such a geometry leads to a loss of information concerning the sampled THz wave since the original spectral content of the incident probe pulse overwhelms the information related to the interaction of the optical and THz pulses. So, one needs a specific detection scheme which makes it possible to get rid of the spectrum of the probe pulse and preserve the sidebands due to Pockels XPM.

In the present paper, we report on the spectral analysis of the optical sidebands produced by the interaction of optical and THz pulses in a ZnTe crystal. To this end, we make use of a special geometry of the polarization states of both beams which results in an amplitude modulation of the optical pulse, unlike in [4, 5], making it possible to separate the optical pulse generated by the Pockels XPM from the incident pulse. Measured as a function of the delay  $\tau$  it yields a spectrogram which can be viewed as a frequency resolved cross-correlation between the optical and THz pulses and enables to characterize the optical pulse. Hereafter we demonstrate that this technique can be easily applied to the measurement of the group-delay dispersion of an ultrashort optical pulse.

#### 2. Experimental setup



Fig. 1. (a) Experimental setup. BS: beamsplitter; HDPE: high density polyethylene. (b) Geometry of the experiment.  $(\hat{\mathbf{X}}, \hat{\mathbf{Y}}, \hat{\mathbf{Z}})$ : Cartesian frame of the crystal;  $(\hat{\mathbf{x}}, \hat{\mathbf{y}}, \hat{\mathbf{z}})$ : Cartesian frame of the laboratory.

The experiment is the following (Fig. 1(a)): an intense linearly polarized THz pulse spanning the 0.3-7 THz frequency range is emitted from air ionized by a two-color, namely 400 nm and 800 nm, femtosecond laser field [6]. This THz pump field is then collimated and focused onto a  $\langle 110 \rangle$  ZnTe crystal, with a thickness  $L = 300 \,\mu$ m, by two off-axis paraboloidal mirrors with a 150 mm-focal length. Inside the crystal, the  $\hat{y}$ -polarized THz beam, with an amplitude  $E_{THz} =$ 8 kV/cm, propagates collinearly with a weak 800 nm probe beam,  $\hat{z}$ -polarized thanks to a halfwave plate and a polarizer, the  $\hat{z}$ -axis coinciding with the  $\langle 001 \rangle$ -axis of the crystal (Fig. 1(b)). After the crystal, a polarizer transmits the component of the optical pulse generated along the  $\hat{y}$ -axis by the THz-induced Pockels effect. A delay stage makes it possible to delay the probe pulse with respect to the THz pump pulse. The temporal evolution of the optical pulse intensity transmitted by the polarizer is measured by a photodiode connected to a lock-in amplifier, whereas the temporal evolution of its spectrum is detected with a spectrometer. Finally, the THz intensity can be adjusted by a pair of high density polyethylene polarizers.

#### 3. Experimental results

We first measured the intensity,  $S_1$ , of the optical pulse transmitted by the polarizer after the ZnTe crystal as a function of the delay  $\tau$ . The result is displayed on Fig. 2(a), together with the intensity of the THz wave measured by conventional EO sampling, i.e. measurement of the Pockels-induced phase modulation by ellipsometry [3]. Both intensity profiles are in good agreement except near the zeros of the THz wave, due to both the lack of phase sensitivity of our detection scheme and the convolution of the THz wave with a probe pulse intensity profile of finite duration. Figure 2(b) confirms the quadratic evolution of the peak value of  $S_1$  with respect to the THz electric field strength,  $E_{THz}$ , as expected from the use of a quadratic detector, indicating thus that the THz wave is weak enough to modulate the optical pulse spectrum only through the Pockels effect.

Figure 3(a) displays the evolution of the spectrum of the  $\hat{y}$ -polarized optical pulse versus  $\tau$ , recorded for a pulse compression optimizing the peak of the THz pulse. To reduce the subsequent probe pulse temporal broadening we took care of having the same dispersive media (*i.e.* polarizer and half-wave plate) on the 1.5 mJ beam used to generate the THz and on the probe beam (not shown in Fig. 1). As can be seen, the spectrogram of Fig. 3(a) closely resembles the



Fig. 2. (a)  $S_1$ : intensity profile of the Pockels-induced optical pulse;  $S_{EO}$ : intensity profile of the THz wave measured by conventional EO sampling. (b) Peak value of the Pockels-induced optical pulse with respect to the THz electric field strength  $E_{THz}$ .



Fig. 3. (a) Spectrogram of the Pockels-induced optical pulse obtained with a pulse compression optimizing the peak of the THz pulse. (b) Spectrogram of the Pockels-induced optical pulse recorded when a 3 mm thick window of ZnSe is inserted on the path of the incident optical pulse. The dashed white lines indicate the overall slope of the spectrogram in the time delay – angular frequency plane.

trace given by the well-known Cross-correlation Frequency-Resolved Optical Gating (XFROG) characterization technique [7]. However, here, one pulse is in the optical range, whereas the other one is in the THz range. Hereafter, we demonstrate more precisely the connection between the spectrogram of Fig. 3(a) and the XFROG technique.

# 4. Spectrally resolved EO sampling in a $\langle 110 \rangle$ ZnTe crystal

Let us write

$$\mathbf{E}(t,x) = \frac{1}{2}\hat{\mathbf{E}}(t,x)\exp\left[i(k_{\rm pr}x - \omega_{\rm pr}t)\right] + \text{c.c.}$$

the electric field of the probe pulse, and

$$\mathbf{P}^{(2)}(t,x) = \frac{1}{2}\hat{\mathbf{P}}^{(2)}(t,x)\exp\left[i(k_{\rm pr}x - \omega_{\rm pr}t)\right] + {\rm c.c.}$$

the second-order polarization of the crystal, assuming the THz pulse is weak enough to avoid higher order nonlinear phenomena like Kerr effect [8]. Here,  $\omega_{pr}$  and  $k_{pr}$  stand for the angular frequency and the wave vector of the probe beam. Then, in the frame of the laboratory, within the slowly varying amplitude approximation, the probe wave equation writes [9]:

$$\frac{\partial \hat{E}_{\mu}(t',x)}{\partial x} = i \frac{\pi}{\lambda_{\rm pr} n_{\rm pr} \varepsilon_0} \hat{P}_{\mu}^{(2)}(t',x), \tag{1}$$

where  $t' = t - x/v_g$ ,  $n_{\rm pr}$  is the refractive index of ZnTe at the probe wavelength  $\lambda_{\rm pr}$ ,  $\varepsilon_0$  is the permittivity of vacuum,  $v_g$  is the group velocity of the probe pulse, and  $\mu = y, z$ .

Assuming that we are far from resonance, in the frame of the crystal the second-order polarization corresponding to the interaction of the optical probe and THz pulses through the Pockels effect is given by [9]:

$$\hat{\mathbf{P}}^{(2)}(t',x) = 2\varepsilon_0 \boldsymbol{\chi}^{(2)} : \hat{\mathbf{E}}(t',x) \mathbf{E}_{\text{THz}}\left(t' + \frac{x}{v_g}, x\right),$$

where  $\chi^{(2)}$  is the second-order nonlinear susceptibility tensor of the crystal. The latter is related to the common electro-optic tensor **r** by  $\chi_{ijk}^{(2)} = -\frac{1}{2}n_i^2n_j^2r_{ijk}$ , where i, j, k = X, Y, Z, and n is the refractive index in the optical range [10]. For an isotropic crystal like ZnTe,  $n_i = n_{\rm pr} \forall i$ , so that this relation reduces to  $\chi^{(2)} = -\frac{1}{2}n_{\rm pr}^4$ **r**, with  $r_{41} = r_{ijk}$  ( $i \neq j \neq k$ ) the only nonzero element of the electro-optic tensor.

In the frame of the laboratory, the THz and optical electric fields write  $\mathbf{E}_{\text{THz}}(t,x) = E_{\text{THz}}(t,x)\mathbf{\hat{y}}$  and  $\mathbf{\hat{E}}(t,0) = \hat{E}_z(t,0)\mathbf{\hat{z}}$ , respectively, so that  $\mathbf{\hat{P}}^{(2)}$  becomes:

$$\hat{P}_{\mu}^{(2)}(t',x) = \varepsilon_0 n_{\rm pr}^4 r_{41} \hat{E}_z(t',x) E_{\rm THz} \left(t' + \frac{x}{v_g},x\right) \delta_{\mu,y} \quad (\mu = x, y, z).$$
<sup>(2)</sup>

Thus, with the peculiar geometry considered here, the only nonzero component of  $\hat{\mathbf{P}}^{(2)}$  is along the y-axis. As a consequence, to the first order, the interaction of the optical probe pulse and the THz wave via the Pockels effect gives rise to an optical pulse at angular frequency  $\omega_{\rm pr}$ , cross-polarized with respect to the incident optical pulse, so that, at a point x within the crystal, the optical pulse envelope now writes  $\hat{\mathbf{E}}(t,z) = \hat{E}_y(t,x)\hat{\mathbf{y}} + \hat{E}_z(t,x)\hat{\mathbf{z}} = \hat{\mathbf{E}}_y(t,x) + \hat{\mathbf{E}}_z(t,x)$ . However, this new component can also couple to the THz wave and, in turn, impacts on the propagation of  $\hat{\mathbf{E}}_z(t,x)$  through to the second-order nonlinear polarization

$$\hat{\mathbf{P}}^{\prime(2)}(t',x) = 2\varepsilon_0 \mathbf{\chi}^{(2)} : \hat{\mathbf{E}}_{\mathbf{y}}(t',x) \mathbf{E}_{\mathrm{THz}}\left(t' + \frac{x}{v_g}, x\right),$$

which, in the frame of the laboratory, writes

$$\hat{P'}_{\mu}^{(2)}(t',x) = \varepsilon_0 n_{\rm pr}^4 r_{41} \hat{E}_y(t',x) E_{\rm THz} \left( t' + \frac{x}{v_g}, x \right) \delta_{\mu,z} \quad (\mu = x, y, z).$$
(3)

Finally, according to Eqs. (1), (2) and (3), the probe pulse envelope,  $\hat{\mathbf{E}}(t,z) = \hat{E}_y(t,x)\hat{\mathbf{y}} + \hat{E}_z(t,x)\hat{\mathbf{z}}$ , obeys to the following coupled wave equations:

$$\frac{\partial \hat{E}_{y}(t',x)}{\partial x} = i\kappa \hat{E}_{z}(t',x) E_{\text{THz}}\left(t' + \frac{x}{v_{g}},x\right),\tag{4a}$$

$$\frac{\partial \hat{E}_z(t',x)}{\partial x} = i\kappa \hat{E}_y(t',x) E_{\text{THz}}\left(t' + \frac{x}{v_g}, x\right),\tag{4b}$$

where  $\kappa = \pi n_{\rm pr}^3 r_{41} / \lambda_{\rm pr}$ .

Solving Eq. (4) requires to solve also the wave equation for the THz field. However, in the weak field regime, one can consider that the THz wave is left unaltered by the optical pulse during its propagation. Finally, the THz wave is just affected by dispersion/absorption in ZnTe, an effect which can be readily calculated as shown later. In order to integrate Eq. (4), let us

introduce the new fields  $\hat{E}_{\pm}(t',x) = \hat{E}_y(t',x) \pm \hat{E}_z(t',x)$ , with  $\hat{E}_{\pm}(t',0) = \pm \hat{E}_z(t',0)$ . Then, Eq. (4) lead to the following uncoupled wave equations

$$\frac{\partial \hat{E}_{\pm}(t',x)}{\partial x} = \pm i\kappa \hat{E}_{\pm}(t',x) E_{\mathrm{THz}}\left(t'+\frac{x}{v_g},x\right),$$

whose integration is straightforward and finally leads to

$$\hat{E}_{y}(t',L) = i\hat{E}_{z}(t',0)\sin\left[\kappa\int_{0}^{L}E_{\text{THz}}\left(t'+\frac{x}{v_{g}}+\tau,x\right)\mathrm{d}x\right],$$

$$\hat{E}_{z}(t',L) = \hat{E}_{z}(t',0)\cos\left[\kappa\int_{0}^{L}E_{\text{THz}}\left(t'+\frac{x}{v_{g}}+\tau,x\right)\mathrm{d}x\right],$$

where we have taken into account the fact that the probe pulse is delayed by an amount of time  $\tau$  with respect to the THz wave. Since in our experiment  $\kappa \int_0^L E_{\text{THz}} (t' + x/v_g + \tau, x) dx \ll 1$ , one gets  $\hat{E}_z(t', L) \simeq \hat{E}_z(t', 0)$  and

$$\hat{E}_{y}(t',L) \simeq i\kappa \hat{E}_{z}(t',0) \int_{0}^{L} E_{\text{THz}}\left(t' + \frac{x}{v_{g}} + \tau, x\right) \mathrm{d}x.$$
(5)

There is a one to one correspondence between the field given by Eq. (5) and the optical pulse generated through sum- and difference-frequency by interaction of the probe pulse with the THz wave. Consequently, the Fourier transform of Eq. (5), denoted  $\hat{E}_y(\omega, L)$ , fully describes the optical sidebands mentioned in [3]. However, with the geometry of our experiment, these sidebands can be detected independently from the spectrum of the incident optical pulse,  $\hat{E}_z(\omega, 0)$ , since these two spectra are cross-polarized.

Let us now calculate  $\hat{E}_y(\omega, L)$  whose squared modulus corresponds to the spectrogram of Fig. 3. In order to account for dispersion/absorption in the THz range, one needs, first, to express Eq. (5) as a function of the spectral amplitudes  $E_{\text{THz}}(\Omega, 0)$  of the input THz pulses, defined as the Fourier transform of  $E_{\text{THz}}(t', 0)$ . We have:

$$E_{\rm THz}\left(t'+\frac{x}{v_g}+\tau,x\right) = \int_{-\infty}^{+\infty} \frac{\mathrm{d}\Omega}{2\pi} e^{-i\Omega t'} E_{\rm THz}(\Omega,0) e^{i\Omega\left[\tilde{n}(\Omega)-n_g\right]x/c} e^{-i\Omega\tau},\tag{6}$$

where  $\tilde{n}(\Omega) = n(\Omega) + i\kappa(\Omega)$  is the complex refractive index of the crystal in the THz range, and  $n_g = n_g(\omega_{\rm pr})$  is the group index for the optical pulse. In Eq. (6), the phase factor  $\Omega \tilde{n}(\Omega) x/c$ simply corresponds to the phase accumulated by the THz pulse in the course of its propagation within the crystal. Inserting Eq. (6) into Eq. (5), and expressing  $\hat{E}_z(t', 0)$  as the Fourier transform of  $\hat{E}_z(\omega, 0)$  then leads to

$$\hat{E}_{y}(\omega,L) = \int_{-\infty}^{+\infty} dt' e^{i\omega t'} \hat{E}_{y}(t',L),$$

$$= i\kappa \int_{0}^{L} dx \int_{-\infty}^{+\infty} \frac{d\Omega}{2\pi} \hat{E}_{z}(\omega-\Omega,0) E_{\text{THz}}(\Omega,0) e^{i\frac{\Omega x}{c} \left[\tilde{n}(\Omega) - n_{g}\right] - i\Omega\tau}.$$
(7)

Assuming that the input optical pulse may have some residual group-delay dispersion (GDD),  $\phi_{in}^{(2)}$ ,  $\hat{E}_z(\omega, 0)$  then writes:  $\hat{E}_z(\omega, 0) = \hat{E}_z(\omega) \exp[i\phi_{in}^{(2)}\omega^2/2)]$ , where  $\hat{E}_z(\omega)$  can be determined from the input pulse spectrum. However, Eqs. (1) and (4) neglect the dispersion within the crystal. To the first order, the latter can be accounted for by replacing, in Eq. (7),  $\hat{E}_z(\omega, 0)$ by  $\hat{E}_z(\omega, x) = \hat{E}_z(\omega, 0) \exp[i\phi(\omega, x)]$ , with  $\phi(\omega, x) = k_{pr}''\omega^2 x/2$ , where  $k_{pr}'' = (d^2k/d\omega^2)_{\omega_{pr}}$ ,  $k(\omega) = \omega n(\omega)/c$ , and  $n(\omega)$  is the refractive index of ZnTe in the optical range, taken from [11]. All calculations done, these substitutions lead to:

$$\hat{E}_{y}(\omega,L) = i\kappa L \int_{-\infty}^{+\infty} \frac{\mathrm{d}\Omega}{2\pi} \operatorname{sinc}\left(\frac{\Delta k'L}{2}\right) \hat{E}_{z}(\omega-\Omega,0) E_{\mathrm{THz}}(\Omega,0) e^{i\left[\frac{1}{2}\phi_{\mathrm{in}}^{(2)}(\omega-\Omega)^{2}-\Omega\tau+\frac{1}{2}\Delta k'L\right]}, \quad (8)$$

with sinc  $(x) = \sin(x)/x$  and  $\Delta k' = \Delta \tilde{k} + \frac{1}{2}k''_{\text{pr}}(\omega - \Omega)^2$ , where  $\Delta \tilde{k} = \Omega \left[\tilde{n}(\Omega) - n_g\right]/c$ . The latter equation can be written as follows:

$$\hat{E}_{y}(\omega,L) = i\kappa L \int_{-\infty}^{+\infty} \frac{\mathrm{d}\Omega}{2\pi} \operatorname{sinc}\left(\frac{\Delta k'L}{2}\right) \hat{E}_{z}(\omega-\Omega,0) E_{\mathrm{THz}}(\Omega,0) e^{-\frac{1}{2c}\kappa(\Omega)\Omega L} \times e^{i\varphi(\omega,\Omega)}, \quad (9)$$

with

$$\varphi(\omega,\Omega) = \frac{1}{2}\phi_{\rm in}^{(2)}(\omega-\Omega)^2 - \Omega\tau + \frac{1}{4}\phi_{\rm ZnTe}^{(2)}(\omega-\Omega)^2 + \frac{1}{2}\Delta k(\Omega)L.$$
 (10)

Here,  $\phi_{ZnTe}^{(2)} = k_{pr}''L$  is the GDD resulting from the propagation of the optical pulse through ZnTe, and  $\Delta k(\Omega) = \Omega [n(\Omega) - n_g]/c$  corresponds to the wave vector mismatch between the optical and THz pulses. Thus,  $\hat{E}_y(\omega, L)$  corresponds to the integral of a rapidly oscillating term,  $\exp[i\varphi(\omega,\Omega)]$ , multiplied by a slowly varying amplitude term. Note that the latter spans the 0 - 4 THz range due to both the cardinal sine function and the absorption in ZnTe [12]. Moreover, within this spectral range, the phase matching condition is fulfilled in ZnTe [13]. As a consequence, in Eq. (10), one has  $\Delta k(\Omega)L \simeq 0$ .

Let us now discuss about the differences between our spectrally resolved EO sampling and the XFROG technique. In the latter, one needs two pulses, one of which is well characterized to serve as a reference, with the same order of duration. Here, we have a femtosecond pulse interacting with a picosecond THz wave. The THz wave could be seen as the reference since it can be fully characterized through conventional EO sampling. But this is precluded by its too long duration which blurs any temporal information (amplitude and phase) concerning the ultrashort optical pulse. From this point of view, the spectrally resolved EO sampling is not equivalent to the XFROG technique, and cannot be used to reconstruct the optical pulse (amplitude and phase) knowing the electric field of the THz wave. However, it would be wrong to conclude that no information concerning the spectral phase of the optical pulse can be inferred from the spectrogram of Fig. 3. Indeed, Eq. (8) indicates that a photon generated by Pockels XPM at a frequency  $\omega_{\rm pr} + \omega$  results from a spectral interference, over the whole spectrum of the THz pulse, between a photon in the optical range, at frequency  $\omega_{pr} + \omega - \Omega$ , and a photon in the THz range at frequency  $\Omega$ . So, the spectral components shown in the spectrogram of Fig. 3 are sensitive to the relative spectral phase between the incident optical pulse and the THz wave. Upon the 0 – 4 THz range transmitted by the ZnTe crystal, this relative phase is given by  $\varphi(\omega, \Omega)$ , with  $\Delta k(\Omega) L \simeq 0$ . Since the phase matching condition in ZnTe cancels out the dependence of  $\varphi(\omega, \Omega)$  with respect to the dispersion in the THz spectral range (term  $\Omega n(\Omega)/c$  in  $\Delta k(\Omega)$ ), it means that the spectral content of the spectrogram is only sensitive to the dispersion in the optical range  $(\phi_{in}^{(2)}, \phi_{ZnTe}^{(2)})$ , that is to the spectral phase of the optical probe pulse, as we will see in the next section.

### 5. Discussion

We are now ready to see what kind of information about the probe pulse one can infer from the spectrogram of Fig. 3(a). Actually, since the latter is slightly tilted with respect to the angular frequency axis, we can already say that the probe pulse is not Fourier-transform-limited. It is

linearly chirped due to some small positive GDD after compression of the laser output to optimize the THz generation and propagation through the ZnTe crystal. From Eqs. (9) and (10), one can deduce the GDD of the probe pulse from the inverse of the "slope" of the spectrogram. Indeed, as already mentioned, Eq. (9) indicates that  $\hat{E}_y(\omega, L)$  corresponds to the integral of a rapidly oscillating term,  $\exp[i\varphi(\omega,\Omega)]$ , multiplied by a slowly varying amplitude term. According to the stationary phase approximation [14], the only significant nonzero contributions to  $\hat{E}_y(\omega, L)$  then occur for angular frequencies  $\omega$  for which the phase  $\varphi(\omega,\Omega)$  is stationary. Consequently, the spectrogram will have significant values around angular frequencies such that  $\partial \varphi/\partial \Omega = 0$ , that is for  $\omega = \omega_{\rm pr} + \Omega - \tau / \left(\phi_{\rm in}^{(2)} + \phi_{\rm ZnTe}^{(2)}/2\right) \simeq \omega_{\rm pr} - \tau / \left(\phi_{\rm in}^{(2)} + \phi_{\rm ZnTe}^{(2)}/2\right)$ , since  $\omega_{\rm pr} \gg \Omega$ . From the straight line superimposed to the spectrogram of Fig. 3(a), we obtained  $\phi_{\rm in}^{(2)} + \phi_{\rm ZnTe}^{(2)}/2 = -\Delta \tau / \Delta \omega = 1210 \, {\rm fs}^2$ . From [11] and [15], one gets  $\phi_{\rm ZnTe}^{(2)} = 640 \, {\rm fs}^2$ , leading to  $\phi_{\rm in}^{(2)} = 890 \, {\rm fs}^2$ . This value is in good agreement with the one deduced from an autocorrelation of the probe pulse performed in front of the ZnTe, which gives  $\phi_{\rm in}^{(2)} = 860 \, {\rm fs}^2$ .

At first glance, our determination of  $\phi_{in}^{(2)}$  is rather rough due to the uncertainty on the position of the straight line used to quantify the tilt of the spectrogram. Moreover, the latter is also slightly curved owing to the presence of some cubic dispersion in the optical pulse which is not accounted for by the straight line "fit" used here. However, despite its simplicity, our method gives rather good values of GDD as confirmed by the following experiment: we inserted on the path of the probe beam, in a region where there is no overlap between optical and THz beams, a 3 mm thick window of ZnSe. The latter adds to  $\phi_{in}^{(2)}$  a GDD  $\phi_{ZnSe}^{(2)} = 3096 \text{ fs}^2$ , as calculated from the refractive index given in [11]. The spectrogram resulting from the interaction of this probe pulse with the THz wave in ZnTe is shown in Fig. 3(b). Due to the probe pulse broadening, the amplitude of the recorded signal decreases, leading to a slight degradation of the spectral sensitivity of the spectrogram that  $\phi_{in}^{(2)} + \phi_{ZnSe}^{(2)} + \phi_{ZnTe}^{(2)}/2 = 4250 \text{ fs}^2$ , leading to  $\phi_{ZnSe}^{(2)} = 3040 \text{ fs}^2$ , in good agreement with the value given above. From repeated experiments, we estimated the uncertainty on the value of  $\phi_{i}^{(2)}$  to be about 7%.

# 6. Conclusion

We have measured and modeled the evolution of the spectrum of the optical pulse generated during EO sampling in ZnTe of a THz wave by an optical pulse versus the time delay between them. We have also demonstrated that this spectrogram can be used to measure the GDD of the optical pulse. This characterization technique strongly depends on the EO crystal used, since it requires that the phase matching condition is fulfilled upon the spectral range transmitted by the crystal. Note that this method could be potentially extended to the determination of higher orders of dispersion in the optical range, provided that the spectrogram is fitted with more elaborate functions than a straight line. From a practical point of view, our experimental setup is also suited for the characterization of THz pulses by detecting the optical pulse generated via the Pockels effect by ellipsometry. Therefore, just by adding a spectrometer to a conventional THz time-domain spectroscopy (THz-TDS) experiment, one can measure the phase and amplitude of both optical and THz pulses used to perform THz-TDS spectroscopy. As a consequence, one does not need another setup, such as FROG or SPIDER, to characterize the optical pulse.

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