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Simple and distortion-free optical sampling of terahertz pulses via heterodyne detection schemes

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We present and demonstrate experimentally two simple and distortion-free methods to sample terahertz pulse in zinc-blende-type crystals like ZnTe or GaP. They are based on an optical heterodyne detection scheme which makes it possible to measure the terahertz field by detecting the optical probe beam or its second harmonic. Both are compared to conventional electro-optic sampling and give results in good agreement with the latter.

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

1. INTRODUCTION

Electro optic (EO) sampling, hereafter reported as conventional EO sampling (CEOS), is a widely used technique for the measurement of the electric field of a terahertz (THz) pulse with subpicosecond time resolution [1 3]: in a zinc blende crystal, the polarization state of a linearly polarized ultrashort optical pulse is modulated by the THz pulse through the Pockels effect. The corresponding phase retardation $\Gamma$, proportional to THz electric field amplitude $E_{\text{THz}}$, is then measured by ellipsometry for different time delays $\tau$ between the two pulses. The ellipsometer, consisting of a $\lambda/4$ plate followed by a suitably oriented Wollaston polarizer, converts the phase modulation into an amplitude modulation of the optical probe pulse measured by a balanced photodetector. Different alternatives to this technique have been proposed, but they suffer from some detection nonlinearities [4,5]. Even though these nonlinearities can be corrected, they increase the complexity of the measurement process with respect to conventional EO. Very recently, new methods based on a technique commonly used in optically heterodyne detected optical Kerr effect spectroscopy have been introduced [6,7]. These distortion free techniques are based on two measurements at opposite optical biases near the zero transmission point in a crossed polarizers detection geometry. They lead to an increase, by 1 order of magnitude, of the signal enhancement factor, defined as the ratio of the normalized detected balanced difference signal to $\Gamma$.

Hereafter, we propose two novel and simple schemes, based on a special geometry of the polarization states of the optical and THz beams. The first one leads to an amplitude modulation of the optical pulse at the output of the zinc blende crystal and makes it possible to separate the optical pulse generated by the Pockels effect, hereafter called the signal pulse, from the incident pulse [8]. Thanks to an optical heterodyne detection of these signal pulses, one can perform a distortion free sampling of THz pulses within a single measurement. In the second one, we take advantage of this geometry where the Pockels nonlinearity couples to the intrinsic second order nonlinearity of the EO crystal, leading to the generation of a second harmonic signal [the Pockels induced second harmonic (PISH) effect], which can be used, through an optical heterodyne detection, to sample terahertz pulses [9]. Since the latter detection scheme, which shows some similarities with the first one, offers interesting perspectives for the sampling of THz pulses generated by near infrared pulses, we will present it here with more details than in [9].

This paper is organized as follows. In Section 2, we present the experimental setup of our optically heterodyne detected EO sampling and derive the expression of the detected signal. The latter expression is then analyzed and confronted with the experiment. Next, in Section 3, we give a comprehensive analysis of the optically heterodyne detected PISH signal and compare it, experimentally, with conventional EO sampling. Finally, in Section 4, we compare both detection schemes.

2. OPTICALLY HETERODYNE-DETECTED ELECTRO-OPTIC SAMPLING

A. Experimental Setup

The experiment is the following [Fig. 1(a)]: a linearly polarized THz pulse is emitted from air ionized by a two color, namely
400 and 800 nm, femtosecond laser field [10]. This THz pump field is then collimated and focused onto a (110) EO crystal (ZnTe or GaP), with a thickness \( L \) (\( L = 300 \mu \text{m} \) for ZnTe and 200 \( \mu \text{m} \) for GaP), by two off axis paraboloidal mirrors with a 150 mm focal length. Inside the crystal, the \( \hat{y} \) polarized THz beam propagates collinearly with a weak 800 nm probe beam, initially \( \hat{z} \) polarized by a half wave plate and a polarizer, the \( \hat{z} \) axis coinciding with the \( (001) \) axis of the crystal [Fig. 1(b)]. After the crystal, the transmitted optical probe beam is split by a 50/50 nonpolarizing beam splitter. Both beams are then sent through a quarter wave plate and a polarizer, and their intensity is recorded by photodiodes. The fast axis of the quarter wave plate in front of the photodiode PD1 (resp. PD2) is oriented at 50\(^\circ\). The axes of the polarizers are set along the \( \hat{y} \) axis, so that the incident probe beam is blocked in the absence of THz and \( \beta = 0^\circ \).

However, when the probe and THz pulses temporally overlap in the crystal, their interactions within the crystal generates a weak optical signal polarized along the \( \hat{y} \) axis [8]. The latter is therefore transmitted by the polarizers. The THz beam is chopped and the signal, \( \Delta I(\Gamma, \beta) = I_1(\Gamma, \beta) - I_2(\Gamma, \beta) \), measured by the two photodiodes versus the time delay \( \tau \) between the probe and THz pulses, is recorded by a lock in amplifier.

### B. Electro-Optic Sampling by Optical Heterodyne Detection

Let us see what is the expression of the signal \( \Delta I(\Gamma, \beta) \) provided by the setup of Fig. 1. In the \( (\hat{y}, \hat{z}) \) basis, the electric field of the incident probe beam writes

\[
\mathbf{E} = E_0 \begin{bmatrix} 0 \\ 1 \end{bmatrix}.
\]

At the output of the EO crystal, after interaction with the THz pulse, this field is given by [8]

\[
\mathbf{E} = \mathbf{E}_0 \left[ \begin{array}{c} \sin \left( \frac{x}{2} \right) \\ \cos \left( \frac{x}{2} \right) \end{array} \right].
\]

Now, if one considers the modulation depth \( M(\Gamma, \beta) = \frac{I_1(\Gamma, \beta)}{I_1(0, \beta)} \) of the signal detected by PD1 [6], it is then possible to express the enhancement factor \( g \) and the distortion parameter \( d \) associated with our method. They are related to the modulation depth, in the weak field limit, by

\[
M(\Gamma, \beta) = g \Gamma + d(\Gamma^2 + O(\Gamma^3)).
\]

From Eqs. (4) and (5), one has

\[
g = \frac{1}{\sin(2\beta)},
\]

\[
d = \frac{1}{2} \cos^2(2\beta).
\]

Equation (8) agrees with the expression of the enhancement factor found for EO sampling expressed in the terminology of optical heterodyne detection [6], where the heterodyning of the signal is performed in front of the EO crystal. On the other hand, the distortion parameter we found is multiplied by a factor \( \cos^2(2\beta) \) compared to [6], meaning that our detection scheme is less sensitive to distortion as \( \beta \) increases. However, since the enhancement factor is better for small values of \( \beta \), \( d \approx 1/2 \) over the optimal operating range of our detection scheme.

Finally, this distortion is removed by measuring the subtraction of the signals detected by both photodiodes,

\[
\Delta I(\Gamma, \beta) = \frac{I_0}{2} \sin(2\beta) \sin \Gamma,
\]

whose enhancement factor is \( 2g \).

\[
\mathbf{E} = E_0 \left[ \begin{array}{c} i \sin(\Gamma/2) \\ \cos(\Gamma/2) \end{array} \right],
\]

with

\[
\Gamma = \frac{2\pi L}{\lambda} n_p r_{41} E_{THz}.
\]
C. Results and Discussion

Figure 2 displays the THz electric fields and their spectra, detected with our setup when ZnTe [Figs. 2(a) and 2(b)] or GaP [Figs. 2(c) and 2(d)] crystals are used, for values of the angle $\beta$ ranging from 5° to 29°. For comparison, all these fields are normalized to one and superimposed to the normalized THz fields obtained in [7].

Indeed, the smaller the heterodyning angle $\beta$, the better is the enhancement factor $2g$ of EO sampling by optical heterodyne detection. For instance, for $\beta = 5^\circ$, the enhancement factor is $2g \approx 1/\beta \sim 10$, a value identical to the enhancement factor obtained in [7].

3. TERAHERTZ PULSES SAMPLING THROUGH OPTICALLY HETERODYNE-DETECTED PISH SIGNAL

In a recent paper [9], we have shown that it was possible to detect THz pulses via the detection of the second harmonic (SH) of an optical pulse induced through its interaction with an intense THz pulse in a zinc blende crystal (the so called PISH effect). Briefly, with the geometry of Fig. 1, an optical signal pulse is induced by the THz pulse through the Pockels effect. For the sake of simplicity, assuming monochromatic plane waves at angular frequencies $\omega$ in the optical range and $\Omega$ in the THz range, the electric field of the signal is $E_{\text{sig}} = E_y^{(\omega=\Omega)} \approx E_y^{\omega}$, since $\Omega \ll \omega$. This field then couples to the fundamental electric field, $E_y^{\omega}$, through the second order nonlinearity of the crystal, $\chi^{(2)}_{XYZ}$, to generate a second harmonic signal with an electric field given by [9]:

$$E^{(12)}_{\text{PISH}} \propto \chi^{(2)}_{XYZ} r_{11}(E_z^{\omega})^2 E_{\text{THz}} L/\lambda.$$  

Since the PISH signal is proportional to the THz electric field, it has been used to detect the latter. However, such a detection requires to heterodyne the PISH signal in order to avoid losing the phase of the THz waveform. This was done by inserting a quarter wave plate on the optical beam pathway in front of the crystal.

Recently, to circumvent some limitations of lithium niobate or ZnTe (two or three photon absorption in the spectral range of the pump, absorption in the THz range, phase mismatch), new materials has been developed to generate intense THz pulses by optical rectification: orientation patterned GaAs [12] or stilbazolium salt single crystals [13], for example.

Fig. 2. EO sampling by optical heterodyne detection performed in ZnTe [(a) and (b)] and GaP [(c) and (d)]. (a),(c) Normalized THz electric field, for different $\beta$ angles, versus the time delay $\tau$ compared to the field measured with a CEOS setup (dotted line). (c),(d) Corresponding spectra.
They share the common characteristic of being pumped in the near infrared, and may require some frequency doubling of the pump pulse for further characterization of the emitted THz wave by EO sampling [13]. One can avoid the insertion of a nonlinear crystal for SH generation of the pump beam prior to EO sampling, or the use of a THz Michelson interferometer to retrieve the THz waveform by first order autocorrelation [14], by resorting to the heterodyne detection of the THz wave via THz field induced second harmonic generation in a plasma [15] or in a suitable EO crystal as we did.

Consequently, since THz detection through optically heterodyne detected PISH signal deserves some attention, we are going to present it in detail compared to the elementary treatment of [9] where, for instance, no derivation of the analytical expression for the measured signal or evaluation of the enhancement factor was given.

A. Experimental Setup and Analytical Expression of the Optically Heterodyne-Detected PISH Signal

The experimental setup is the same as in [9]. THz pulses are generated in LiNbO₃ by optical rectification of tilted pulse front, 800 nm, femtosecond pulses, and the EO crystal is a 300 μm thick ZnTe. A quarter wave plate, with its fast axis oriented at an angle \( \beta \) with respect to the \( \hat{z} \) axis, is inserted on the optical probe beam pathway in front of the EO crystal (Fig. 4). The THz beam is modulated by a mechanical chopper. The THz induced SH signal, filtered by a 400 nm bandpass filter and measured by a photomultiplier tube with a monochromator, is recorded by a lock in amplifier as a function of the time delay \( \tau \) between the probe and THz pulses. Note that, contrary to usual heterodyne detection schemes, this method does not require any polarizer prior to the detector.

Without any loss of generality, we model the probe and THz beams by monochromatic nondepleted waves [9]:

\[ E_\ell(t, x) = \frac{1}{2} E_\omega^\ell(x) \exp\left[ i(\omega t - k_\ell x) \right] + c.c. \quad (\ell = y, z) \quad (11) \]

and

\[ E_{THz}(t, x) = \frac{1}{2} E_{THz} \exp\left[ i(\Omega t - k_{THz} x) \right] \hat{y} + c.c., \quad (12) \]

where \( k_\omega = \omega n_\omega/c \), \( k_{THz} = \Omega n_{THz}/c \), and \( n_\omega \) (resp. \( n_{THz} \)) is the refractive index of the crystal at the probe (resp. THz) frequency. At the entrance of the crystal, for a positive heterodyning angle \( +\beta \), the probe beam amplitude writes

\[ \begin{bmatrix} E_\alpha^\prime(\beta, 0) \\ E_\alpha^\prime(\beta, 0) \end{bmatrix} = E_0 \begin{bmatrix} \frac{1}{2} \sin(2\beta) \\ (1 + i)\sin^2 \beta \end{bmatrix} \quad (13) \]

The (110) cut ZnTe crystal, due to the applied THz field, is birefringent with the following principal axes [16]: \( \hat{e}_1 = \hat{x}, \hat{e}_2 = (\hat{y} + \hat{z})/\sqrt{2}, \) and \( \hat{e}_3 = (\hat{y} + \hat{z})/\sqrt{2} \), whose refractive

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**Fig. 3.** EO sampling by optical heterodyne detection performed in ZnTe [(a) and (b)] and GaP [(c) and (d)]. (a),(c) THz electric field, for different \( \beta \) angles, versus the time delay \( \tau \). (c),(d) Evolution of the amplitude of the signal displayed in (a) and (c) with respect to the heterodyning angle \( \beta \). The solid line curve corresponds to a fit of the data with the function \( \sin(2\beta) \).

**Fig. 4.** Experimental setup. PMT, photomultiplier tube; (\( \hat{x}, \hat{y}, \hat{z} \)), Cartesian frame of the laboratory.
indices are \( n_1 = n_{ao} \), \( n_2 = n_w + \frac{1}{2} n_{ao} r_{41} E_{THz} \), and \( n_3 = n_w + \frac{1}{2} n_{ao} r_{41} E_{THz} \), respectively. As a consequence, in the frame of the crystal [see Fig. 1(b)], the probe field amplitude at each point \( x \) of the crystal is given by

\[
E_{\chi}^{\omega}(\beta, x) = E_{\chi}^{\omega}(\beta, x),
\]

where

\[
f_{\beta} = \frac{1 + i}{2} \sin(2\beta) + (1 + i)\sin^2 \beta, \quad i,
\]

and \( \Gamma_x = 2\pi x n_{ao}^3 r_{41} E_{THz}/\lambda \). These equations make it possible to compute the second order polarization radiating the THz induced field at frequency \( 2\omega \). Its amplitude is given by [17]

\[
\mathcal{P}_{\chi}^{2\omega}(\beta, x) = \frac{1}{2} e_{\chi}^{(2)}(\beta) E_k^{\omega} E_k^{\omega} E_k^{\omega},
\]

where \( i \neq j \neq k = X, Y, Z \) and \( \chi_{XYZ}^{(2)} = \chi_{YXZ}^{(2)} = \chi_{XZY}^{(2)} = \chi_{YYX}^{(2)}. \) In the frame of the laboratory, it writes

\[
\mathcal{P}_{\chi}^{2\omega}(\beta, x) = 0,
\]

and acts as a source term for the nonlinear wave equation of the SH field amplitude. Within the slowly varying amplitude approximation, this equation is

\[
\left( \frac{\partial}{\partial x} + \alpha_{2\omega} \right) E_{\chi}^{\omega} = \frac{i}{2} \omega c_{n_{ao} r_{41}} \mathcal{P}^{(2)}_{\chi} \exp(i\hbar \Delta k) \quad (\ell = x, y),
\]

where \( \alpha_{2\omega} \) is the absorption coefficient at \( 2\omega \) and \( \Delta k = 2\omega (n_w n_{ao} r_{41}/\hbar) \) is the phase mismatch between the fundamental and SH fields. Since, in our experiment, \( \alpha_{2\omega} \Delta k \gg \Gamma_x/\hbar \), one gets, at the output of the crystal:

\[
E_{\chi}^{\omega}(\beta, L) = i \frac{1}{4} \left[ f_{\beta}^2 e^{i\delta} g_{\beta}^2 e^{-i\delta} \right],
\]

where \( A = \alpha \chi^{(2)} E_{\chi}^{\omega} D^{(k)} / \left[ 2 c n_{ao} r_{41} (i\Delta k + \alpha_{2\omega}) \right] \).

The expansion of \( f_{\beta}, g_{\beta} \), and \( E_{\chi}^{\omega}(\beta, L) \), assuming that \( \Gamma, \beta \ll 1 \), makes it possible to calculate the SH intensity for an heterodyning angle \( +\beta \), \( I_{2\omega}(\Gamma, \beta) \propto |E_{2\omega}^{\omega}(\beta, L)|^2 + |E_{2\omega}^{\omega}(\beta, L)|^2 \). Neglecting irrelevant terms of order 3 and more in \( \Gamma \) and \( \beta \), one has

\[
I_{2\omega}(\Gamma, \beta) \propto \frac{|A|^2}{4} \left[ 2 \sin^2(2\beta) + \sin^2 \Gamma + \sin(2\beta) \sin(2\Gamma) \right]
\]

\[
\frac{3}{2} \sin^2(2\beta) \sin^2 \Gamma.
\]

The first term in Eq. (26) corresponds to the intensity of the SH local oscillator,

\[
I_{LO}^{2\omega} = I_{2\omega}(0, \beta) \propto \frac{|A|^2}{2} \sin^2(2\beta),
\]

whereas the intensity of the SH homodyne signal, \( I_{sig}^{2\omega} \), is given by

\[
I_{sig}^{2\omega} = I_{2\omega}(\Gamma, 0) \propto \frac{|A|^2}{4} \sin^2 \Gamma.
\]

In the terminology of optical heterodyne detection, one has to consider the modulation depth, \( M(\Gamma, \beta) = |I_{2\omega}(\Gamma, \beta)/I_{LO}^{2\omega}| \), of the PISH signal detected for a positive heterodyning angle \( +\beta \). As \( \Gamma, \beta \to 0 \), we found that

\[
M(\Gamma, \beta) = g\Gamma + d(\Gamma)^2 + O(\Gamma^3),
\]

with \( g = 1/\sin(2\beta) \) and \( d = 1/2 \), results identical to the one found for EO sampling in this context [6].

Finally, the heterodyned PISH signal is

\[
\Delta I_{2\omega}(\Gamma, \beta) = I_{2\omega}(\Gamma, \beta) - I_{2\omega}(\Gamma, \beta),
\]

\[
\propto \frac{|A|^2}{2} \sin(2\beta) \sin(2\Gamma),
\]

\[
\Delta I_{2\omega}(\Gamma, \beta) \propto 2 \frac{L}{\lambda} \beta \chi^{(2)}(r_{41}) \Gamma_{THz} E_{THz}.
\]

As expected, the heterodyned PISH signal is proportional to the THz electric field and makes it possible to measure the latter. Note that, in the equation given in [9] for \( \Delta I_{2\omega}(\Gamma, \beta) \), the square power law dependence regarding \( I_{2\omega}, \chi^{(2)} \), and \( r_{41} \) is missing.

B. Results and Discussion

Figure 5(a) displays the PISH intensities, \( I_{2\omega}(\Gamma, \beta) \), measured for \( \beta = \pm 10^\circ \). Both signals oscillate around zero thanks to the mechanical chopper and the lock in amplifier, which make it possible to get rid of the contribution of the local oscillator, \( I_{LO}^{2\omega} \). Note also that, by adding these two signals, one would get the homodyne signal, \( I_{sig}^{2\omega} \). On the other hand, their subtraction gives a signal directly proportional to the THz electric field, as evidenced by Fig. 5(b) where \( \Delta I_{2\omega}(\Gamma, \beta) = I_{2\omega}(\Gamma, \beta) - I_{2\omega}(\Gamma, \beta) \) is compared to the THz waveform measured by CEOS. As can be seen, the agreement between our technique and CEOS is excellent. This is also evidenced by the corresponding spectra displayed in Fig. 5(c). The signal to noise ratio was found to be \( \sim 10^3 \) in both cases. Note that, contrary to Section 2, these experiments were not performed in dry air, explaining why the measured THz waveforms and spectra are less clean than those of Fig. 2.

4. COMPARISON OF THE TWO DETECTION SCHEMES

First, the two detection schemes presented above offer the same performances in terms of enhancement factor and distortion parameter. So, the choice of one technique rather than the other will depend on the user’s aim. If one intends to measure THz pulses generated by amplified Ti:sapphire laser systems, the recommended scheme is the optically heterodyne detected electro optic sampling (Section 2) for the following reasons: first of all, this setup is very close to the widespread CEOS setup.
usually dedicated to this task; it makes it possible to acquire the THz waveforms in one scan, limiting the distortions that may arise from any drift of the experimental setup between two consecutive scans for two different heterodyning angles; finally, it does not require the use of a photomultiplier tube, which is much more expensive and fragile than a photodiode. However, this scheme, like the second one, requires good polarizing optics (polarizers, wave plates). Furthermore, these optics must be identical (i.e., from the same manufacturer) on both arms after the EO crystal in order to reduce as much as possible the influence of any intrinsic birefringence. As already mentioned in Section 3, the second detection scheme is interesting if one intends to characterize THz pulses from a setup pumped by near infrared laser pulses. It is a good alternative to much more complex detection techniques, like THz field induced second harmonic generation in a plasma or the use of a Michelson interferometer, since it is very close to the setup of CEOS and requires very few modifications of the latter to be implemented in the laboratory.

5. CONCLUSION

Two simple and distortion free methods to sample THz pulses in zinc blende type crystals, based on an optical heterodyne detection scheme, have been presented and demonstrated experimentally. The first one makes it possible to measure, in a single measurement and without distortion, a THz waveform by detecting an optical probe beam. Regarding the second one, its single measurement and without distortion, a THz waveform by experimentally. The first one makes it possible to measure, in a detection scheme, have been presented and demonstrated.

Fig. 5. (a) PISH signals for two opposite heterodyning angles: $\beta = 10^\circ$ and $\beta = 10^\circ$. (b) EO signal measured with a CEOS setup (dashed red curve) and heterodyned PISH signal difference, $\Delta I_{\text{meas}}$, for $\beta = 10^\circ$ (black curve). All the signals are plotted as a function of the time delay, $\tau$, between the probe and THz pulses. (c) Corresponding spectra.

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