Noncollinear electro-optic detection of terahertz waves: Advantages and limitations
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I. INTRODUCTION

Free space electro-optic sampling is an established technique to measure the temporal waveforms of terahertz pulses, which is widely used in terahertz time-domain spectroscopy (THz-TDS) and terahertz imaging. In the conventional electro-optic sampling scheme, a femtosecond laser pulse (probe pulse) propagates collinearly with terahertz pulse in an electro-optic crystal and experiences a terahertz-field-induced modulation of polarization due to the Pockels effect. By measuring the polarization change as a function of time delay between the pulses, the time-dependence of the terahertz electric field can be mapped point-by-point. For efficient sampling, the near-infrared group velocity should match the terahertz phase velocity. For a given wavelength $\lambda$ of the probe pulse, the velocity matching condition can be fulfilled only in a specific crystal. For example, ZnTe is widely used with Ti:sapphire lasers ($\lambda \approx 800 \text{ nm}$). For femtosecond Er-doped fiber lasers ($\lambda \approx 1.55 \mu \text{m}$), which are appropriate for compact terahertz spectrometers, there are no crystals providing collinear velocity matching.

In Ref. 16, a more versatile noncollinear scheme of electro-optic sampling was proposed. In this scheme, a tightly focused probe beam propagates in an electro-optic crystal at a certain angle to the loosely focused terahertz beam. The angle between the beams is chosen in such a way as to ensure synchronous propagation of the probe pulse and a terahertz wavefront in the direction normal to the wavefront. Thus, the probe pulse surfs a certain wavefront (slipping in the tangential direction) and accumulates a polarization change. Physically, the noncollinear detection scheme is, in fact, the inverted Cherenkov radiation scheme, and the noncollinearity angle equals the Cherenkov angle $\cos \beta = n_g/n_{THz}$, where $n_g$ is the optical group refractive index and $n_{THz}$ is the terahertz phase refractive index.

In Ref. 16, the noncollinear detection scheme was experimentally demonstrated with a LiNbO$_3$ crystal, which is characterized by a large optical-terahertz collinear velocity mismatch ($n_g \approx 2.25$ at $\lambda = 800 \text{ nm}$ and $n_{THz} \approx 5$). To provide a large Cherenkov angle ($\beta \approx 63^\circ$) between the terahertz and optical beams, terahertz radiation was introduced into the LiNbO$_3$ crystal through a Si prism attached to a lateral surface of the crystal. Simultaneously, such a scheme allows one to circumvent the effect of strong terahertz absorption in LiNbO$_3$ by positioning the optical beam along and close to the lateral surface of the crystal. Later, the noncollinear detection scheme was implemented with a GaAs crystal and an Er-doped fiber laser ($\lambda = 1.55 \mu \text{m}$).
angle ($\beta_{th} \approx 12^\circ - 15^\circ$), the detection scheme was simplified by omitting the coupling Si prism and introducing both the terahertz and probe beams through the front crystal face at normal and oblique incidence, respectively. Recently, noncollinear electro-optic sampling at two different wavelengths ($\lambda = 800$ nm and 1.55 $\mu$m) was performed in the same structure consisting of a LiNbO$_3$ plate and a Si prism coupler. By using the configuration with the probe beam propagating along the optical axis of the LiNbO$_3$ plate, the detrimental effect of strong intrinsic birefringence of LiNbO$_3$ was avoided without any additional optical elements, as it was in Ref. 16.

In addition to workability at various laser wavelengths with no need to match the crystal, the noncollinear detection scheme has the advantage of capability to operate with centimeter-thick crystals.17,18 This allows one to use sampling time windows longer than a hundred of picoseconds and, as a result, to achieve the spectral resolution of the detection as high as a few GHz.

A theory of noncollinear terahertz detection was only developed for the nonellipsometric method based on measuring the modulation of the probe beam intensity, rather than polarization.19,20 In this method, the intensity modulation is achieved by modulating the probe beam intensity, rather than polarization.19,20 In this method, the intensity modulation is achieved by an accurate angular separation of the contributions to the probe beam intensity from difference-frequency generation (DFG) and sum-frequency generation (SFG) processes.19,21 Such separation is not required for the ellipsometric detection scheme.

In this paper, we develop a theory of noncollinear ellipsometric electro-optic sampling detection of terahertz waves. The theory generalizes the approach developed in Ref. 2 for the collinear detection geometry.

II. THEORETICAL FORMALISM

Let a probe optical pulse propagate in an electro-optic crystal in the $+x$-direction with the optical group velocity $V_g = c/n_0$, where $c$ is the speed of light (Fig. 1). The probe beam is assumed to be Gaussian with its waist at $x = 0$ and linearly polarized along the $y$ axis. The terahertz beam is assumed to be collimated with a uniform field distribution across the beam. It propagates at angle $\beta$ to the probe beam. To comply with the detection geometries in Refs. 17 and 18, the terahertz beam polarization is parallel to the polarization of the probe beam (the $y$ axis). The probe beam experiences nonlinear interaction with the terahertz field while propagating through the region $0 < x < L$. In Fig. 1, the interaction length $L$ is determined by the crossing length of the probe and terahertz beams. In practice, $L$ can be limited by the crystal thickness, rather than the crossing length. This, however, does not alter the consideration. It should be also emphasized that in the scheme with a LiNbO$_3$ crystal,16 the terahertz beam is strongly attenuated rather than the crossing length. This, however, does not alter the consideration.

The electric field of the probe pulse at $x = 0$ can be written as

$$E_{opt}^0(y, z, t) = F(t)e^{-i(\omega t + z^2)/r^2},$$

where $F(t)$ is the temporal waveform and $r_0$ is the probe beam waist radius. By taking Fourier transforms with respect to time $t$ and transverse coordinates $y, z$, the electric field of the probe pulse can be written in the plane wave approximation20

$$E^0_{opt}(y, z, \omega) = \int_{-\infty}^{\infty} dk_x dk_y \tilde{F}(\omega) G(k_x, k_y) e^{-i\omega t + i k_x y + i k_y z},$$

where $\tilde{F}(\omega)$ and $G(k_x, k_y) = r_0^2 \exp\left[-(k_x^2 + k_y^2) r_0^2/4\right]/(4\pi)$ are Fourier spectral amplitudes. We assume that the weak nonlinear interaction with the terahertz field does not affect the main $y$-polarization component of the optical field. Therefore, its Fourier amplitude (with respect to time $t$) at an arbitrary position $x$ can be written as

$$E^x_{opt}(y, z, \omega) = \tilde{F}(\omega) \int_{-\infty}^{\infty} dk_x dk_y \tilde{G}(k_x, k_y) e^{ik_x x + ik_y y + i\omega z},$$

where $k_x = k \sqrt{1 - (k_y^2 + k_z^2)/k^2}$ is the wave vector component in the $x$-direction, $k = \omega n_{opt}(\omega)/c$ is the wavenumber at frequency $\omega$, and $n_{opt}(\omega)$ is the optical phase refractive index of the crystal.

The electric field of the terahertz pulse in the interaction region $0 < x < L$ can be written in the plane wave approximation22 as

$$E^\text{THz}_y(x, z, t + \tau) = \int_{-\infty}^{\infty} d\Omega \tilde{E}^\text{THz}_y(x, z, \Omega, \tau)e^{-i\Omega t},$$

with

$$\tilde{E}^\text{THz}_y(x, z, \Omega, \tau) = \tilde{\Lambda}(\Omega)e^{-i\Omega t + i k_x x + i k_y y},$$

where $\tau$ is the time delay between the terahertz and optical pulses.
$\hat{A}(\Omega)$ is the spectral amplitude, which determines the temporal waveform of the terahertz pulse, and $K_x = (\Omega/c)\eta_{THz} \cos \beta$, $K_z = (\Omega/c)\eta_{THz} \sin \beta$ are the $x, z$ components of the terahertz wave vector in the crystal.

To find the terahertz-field-induced change of the probe beam polarization, we need to calculate the $z$-component of the optical field (at frequency $\omega_0$), which is orthogonal to the main polarization component given by Eq. (3). The orthogonal field component is generated by the $z$-component of the second-order nonlinear polarization at the optical frequency $\omega$,

$$\tilde{P}_z^{NL}(x, y, z, \omega, \tau) = \int_{\infty}^{\infty} dy \tilde{E}_z^{NL}(x, y, z, \omega, \tau),$$

(6)

where $\chi^{(2)}$ is a component of the nonlinear susceptibility tensor (it is determined by the crystal symmetry and experiment geometry) and $\varepsilon_0$ is the vacuum permittivity. Equation (6) accounts for the processes of sum-frequency generation (SFG) and difference-frequency generation (DFG), which correspond to $\Omega > 0$ and $\Omega < 0$, respectively (for $\omega > 0$).

By substituting Eqs. (3) and (5) into Eq. (6), the nonlinear polarization can be represented in the form (Appendix),

$$\tilde{P}_z^{NL}(x, y, z, \omega, \tau) = \int_{\infty}^{\infty} dy \tilde{E}_z^{NL}(x, y, z, \omega, \tau),$$

(7)

with

$$\tilde{P}_z^{NL}(x, k_x, k_z, \omega, \tau) = 2\varepsilon_0 \chi^{(2)} \int_{\infty}^{\infty} d\Omega \Phi(\omega - \Omega) \hat{A}(\Omega) \exp\left(\frac{k_0^2}{2k^2} + i\frac{k}{\Omega}(1 - k_0^2)\right) \tilde{P}_z^{NL}(x, k_x, k_z, \omega, \tau),$$

(8)

where $k_0^2 = k(\omega - \Omega) \left(1 - \left(k_y^2 + (k_z - K_z)^2\right)/k^2(\omega - \Omega) + K_z^2\right)$ and $k(\omega - \Omega)$ is the optical wavenumber taken at frequency $\omega - \Omega$.

Starting from the Maxwell equations with the nonlinear polarization $\tilde{P}_z^{NL}(x, k_x, k_z, \omega, \tau)$ included as a source, one can obtain the following equation for the optical field transform

$$E_z^{opt}(x, k_x, k_z, \omega, \tau),$$

where

$$\frac{\partial^2 E_z^{opt}}{\partial x^2} + k_z^2 E_z^{opt} = -\mu_0 \omega^2 \left(1 - \frac{k_z^2}{k^2}\right) P_z^{NL}(x, k_x, k_z, \omega, \tau),$$

(9)

where $\mu_0$ is the vacuum permeability. By using the slowly varying envelope approximation, the solution of Eq. (9) at $x \geq L$ can be written as

$$E_z^{opt}(x, k_x, k_z, \omega, \tau) = \int_{-\infty}^{\infty} dx' E_z^{opt}(x, k_x, k_z, \omega, \tau) = \frac{i\mu_0 \omega^2}{2k^2} \int_{0}^{L} P_z^{NL}(x, k_x, k_z, \omega, \tau) dx'.$$

(10)

Substitution of Eq. (8) into Eq. (10) gives

$$E_z^{opt}(x, k_x, k_z, \omega, \tau) = \frac{\chi^{(2)} \omega^2 L}{k^2 c^2} \exp\left(\frac{k}{\Omega}(1 - k_0^2)\right) \int_{-\infty}^{\infty} d\Omega \Phi(\omega - \Omega) \hat{A}(\Omega) \exp\left(\frac{k_0^2}{2k^2} - \frac{k_0^2}{k^2}\right) \tilde{P}_z^{NL}(x, k_x, k_z, \omega, \tau),$$

(11)

where $\Delta k_x = k_0^{NL} - k_x$ is the wave number mismatch.

To proceed, we use the paraxial approximation for the probe beam ($k_x \ll k_z$), inequality $K_x \ll k_z$, and the expansion $k(\omega - \Omega) \approx k(\omega) - \Omega/V_x$. This allows us to make the following approximations in the phase terms in Eq. (11):

$$k_x \approx k - \frac{k_x^2}{2k} - \frac{k_0^2}{2k} + \frac{(k_x - K_x)^2}{2k}.$$  

(12)

In the amplitude factors in Eq. (11), we make more rough approximations,

$$k_x \approx k, \quad 1 - \frac{k_x^2}{k^2} \approx 1, \quad \sin\left(\frac{\Delta k_x L}{2}\right) \approx \sin\left(\frac{K_x - \Omega}{V_x}\right).$$

(13)

Using all these approximations, we perform the inverse Fourier transform of Eq. (11) with respect to $K_x$ and arrive at

$$e^{\frac{iL^2(\omega - \Omega)}{c n_{opt}} \frac{d}{dx} \exp\left[-\frac{k}{\Omega}(1 - k_0^2)\right] \int_{-\infty}^{\infty} d\Omega \Phi(\omega - \Omega) \hat{A}(\Omega) \exp\left(-\frac{k_0^2}{2k^2}\right) \tilde{P}_z^{NL}(x, k_x, k_z, \omega, \tau) \exp\left(\frac{k_0^2}{2k^2} + i\frac{k}{\Omega}(1 - k_0^2)\right) \left(1 - \frac{L}{2R}\right) \exp\left[-\frac{i\Omega}{2V_x}\right].$$

(14)

where $x > L$ do not play a substantial role in the ellipsometric detection, unlike the nonellipsometric method.

The main $y$-polarization component of the probe beam (at frequency $\omega$) propagates according to Eq. (3) and, under the paraxial approximation, can be presented at $x = L$ as a standard
Gaussian beam,

\[
\tilde{E}_y^{\text{opt}}(L, y, z, \omega) = \tilde{F}(\omega) \frac{r_0}{r} \exp \left( -\frac{y^2 + z^2}{2r^2} \right) \times \exp \left[ ikL - i\varphi + i \frac{k(y^2 + z^2)}{2R} \right].
\]  

(15)

Due to nonlinear optical generation of the z-polarization component, Eq. (14), the probe beam polarization changes from linear at the crystal input to slightly elliptical at the crystal output. To measure this ellipticity, a balanced ellipsometric scheme is typically used (Fig. 2). In this scheme, the slightly elliptical (almost linear) probe beam polarization at the crystal output is transformed into two orthogonal (y- and z-) polarization components, which are sent to a balanced photodetector (BP). The detector measures the intensity difference between the two orthogonal components.

By using the Jones matrix for a quarter-wave plate with its fast and slow axes at 45°, a Wollaston prism (WP) splits the probe beam into two orthogonal \( y \) and \( z \) polarization components, which are sent to a balanced photodetector (BP). The detector measures the intensity difference between the two orthogonal components.

The energies of the beams equal

\[
W_{yz}(\tau) = \int_{-\infty}^{\infty} dy dz \Phi_{yz}(y, z, \tau),
\]  

(19)

and the photodetector output signal is proportional to their relative difference \( S(\tau) = [W_{yz}(\tau) - W_{yz}(\tau)']/W_0 \), where \( W_0 \) is the energy of the incident probe pulse (Eq. (1)).

By substituting Eqs. (15) and (14) as \( \tilde{E}_{yz}^{\text{in}} \) to Eq. (16), then substituting Eq. (16) to Eq. (18), and performing the integration in Eq. (19), we finally obtain

\[
S(\tau) = 2\pi^2 \varepsilon_0 \varepsilon_0 \gamma L \frac{\gamma^{2c}}{2} W_0^{-1} \int_{-\infty}^{\infty} d\Omega T(\Omega) A(\Omega) e^{-\Omega^2},
\]  

(20)

where \( T(\Omega) = C(\Omega) D(\Omega)/H(\Omega) \) with

\[
C(\Omega) = \int_{-\infty}^{\infty} d\omega \Phi(\omega - \Omega) \Phi(-\omega),
\]  

(21a)

\[
D(\Omega) = \text{sinc}[\Omega L(n_{THz} \cos \beta - n_0)](2c) \times e^{i\Omega(n_{THz} \cos \beta - n_0)(2c)},
\]  

(21b)

\[
H(\Omega) = e^{-\Omega^2/\Omega_c^2},
\]  

(21c)

\[
\Omega_c = \frac{2\sqrt{2c}}{r_0 n_{THz} \sin \beta} \sqrt{\frac{1 + \gamma}{\gamma/4 + (1 + \gamma/2)^{-1}}},
\]  

(21d)

\( \gamma = L^2/x_0^2 \). In Eq. (20), the factor \( \omega \) was approximated by the optical carrier frequency \( \omega_0 \) and pulled out of the integral. This approximation is reasonable for the typical experimental situation in which the optical bandwidth of the probe pulse is small compared to the optical carrier frequency.

III. ANALYSIS AND DISCUSSION

By analyzing Eq. (20), one can see that \( T(\Omega) \) has a meaning of a nonlinear transfer function. It is a product of three frequency dependent factors, which act as spectral filters on the terahertz
spectral amplitude $\tilde{A}(\Omega)$. The factor $C(\Omega)$ is not specific for the noncollinear detection scheme under investigation. It enters the transfer function of the conventional collinear scheme as well. The factor is the optical spectral autocorrelation function, which can be also presented in the form

$$C(\Omega) = \frac{1}{2\pi} \int dt F^2(t) e^{-i\Omega t}. \quad (22)$$

For a Gaussian pulse $F(t) = F_0 \exp(-t^2/T_{\text{opt}}^2)\cos(\omega_0 t + \varphi)$ and $F(\omega) = F_0 T_{\text{opt}}(4\sqrt{\pi})^{-1/2} \sum \exp[-(\omega + \omega_0)^2 T_{\text{opt}}^2/4 + i\varphi]$, one can evaluate integrals in Eqs. (21a) or (22) and obtain

$$C(\Omega) = \frac{F_0^2 T_{\text{opt}}^2}{4\pi^2} e^{-\Omega^2 T_{\text{opt}}^2/8}. \quad (23)$$

Equation (23) operates as a low-pass filter with a cut-off frequency $\sim T_{\text{opt}}^{-1}$. For typical experimental conditions of THz-TDS with $T_{\text{opt}} \leq 100 fs$, this cut-off frequency is usually much larger than the terahertz bandwidth and, therefore, the factor $C(\Omega)$ has little effect on the detection accuracy.

The phase-matching factor $D(\Omega)$ differs from its counterpart for the collinear scheme$^7$ by the presence of $\cos \beta$. This difference reflects the main advantage of the noncollinear scheme, i.e., its capability to provide optical-terahertz velocity matching, even in materials with large collinear mismatch, by choosing properly the noncollinearity angle $\beta$. For perfect noncollinear matching, i.e., $n_{THz} \cos \beta - n_2 = 0$ (or $\beta = \beta_{\text{THz}}$), one has $D(\Omega) = 1$.

The main limitation on the detection accuracy comes from the factor $H(\Omega)$ [Eq. (21c)], which operates similarly to $C(\Omega)$ as a low-pass filter but with the cut-off frequency $\Omega_c$, determined mainly by the probe beam radius $r_0$ and crossing angle $\beta$, rather than the probe pulse duration in $C(\Omega)$. The diffraction broadening of the probe beam is included into $H(\Omega)$ through the parameter $\gamma$.

The solid lines in Fig. 3(a) show the dependence of the cut-off frequency $\Omega_c$ on the probe beam radius $r_0$ for the experimental conditions of Ref. 17, i.e., for a GaAs crystal and a fiber laser ($\lambda = 1.55 \mu m$). For convenience, the values of the commonly used full width at half maximum (FWHM) of the probe beam $r_{\text{FWHM}} = \sqrt{2 \ln 2} r_0 \approx 1.2 r_0$ are also shown. For calculating $\Omega_c$, the following parameters were used: $\beta = 45^\circ$, $n_{\text{opt}} = 3.38$, $n_{\text{THz}} = 3.63$, and three values of $L$, i.e., 3, 5, and 10 mm. The large interaction lengths $L$ are ensured by the small crossing angle $\beta$ of the terahertz and optical beams. Indeed, the crossing length of the beams equals $D_{\text{THz}}/\sin \beta$, where $D_{\text{THz}}$ is the width of the terahertz beam. For $\beta = 45^\circ$, it gives $L \approx 4.1 D_{\text{THz}}$. For typical $D_{\text{THz}} \sim 0.5-3 \mu m$, we obtain $L \sim 2-12 \mu m$.

In Fig. 3(a), as the probe beam radius decreases the cut-off frequency at first increases, reaching a maximum at $r_0 \approx 15-25 \mu m$ (depending on $L$) and then drops to zero. The drop can be attributed to a significant diffraction broadening of the probe beam at the interaction distance $L$. Indeed, by analyzing Eq. (21d) as a function of $r_0$, we find that the function has a maximum at $\gamma = 4$, i.e., when the confocal parameter $2x_R$ equals $L$: $2x_R = L$. A further decrease of $r_0$ (and, therefore, $x_R$) increases the effect of diffraction.

For the scheme with GaAs, the found optimal values of $r_0$ are too small for practical use due to the geometry of oblique incidence of the probe beam on the crystal surface. Indeed, by using Snell’s law $\sin \alpha = n_{\text{opt}} \sin \beta$, we obtain the incidence angle $\alpha \approx 55^\circ$ corresponding to $\beta \approx 14^\circ$. For such angles, the probe beam width inside the crystal exceeds the incident beam width by a factor of $\cos 14^\circ/\cos 55^\circ \approx 1.7$. Thus, the realistic values of $r_0$ begin from 30 to 40 $\mu m$, where the solid curves for different $L$ practically coincide [Fig. 3(a)].

The dotted lines in Fig. 3(a) show the dependence of $\Omega_c$ on $r_0$ for the experimental conditions of Ref. 18, i.e., for a LiNbO$_3$ crystal and a Ti:sapphire laser ($\lambda = 800 nm$). In this case, the following parameters were used: $\beta = 65^\circ$, $n_{\text{opt}} = 2.23$, $n_{\text{THz}} = 5$, and two values of $L$, i.e., 3 and 5 mm. Due to a large crossing angle $\beta$, the interaction length $L$ practically coincides with the width of the terahertz beam in the crystal, i.e., $L \approx 1.1 D_{\text{THz}}$. In the geometry with the terahertz beam introduced into the crystal through its lateral surface$^{18}$, $L$ exceeds the terahertz beam width in vacuum by a factor of $1/\cos 41^\circ \approx 1.33$.

Despite the substantial difference in the parameters between the solid and dotted curves in Fig. 3(a), the curves for the same values of $L$ (3 and 5 mm) have almost coinciding maximum positions at $r_0 \approx 15-20 \mu m$. This can be explained by close values of the Rayleigh length for the two experiments. Indeed, although the wavelength $\lambda = 800 nm$ is 1.9 times shorter than $\lambda = 1.55 \mu m$, the refractive index $n_{\text{opt}} = 2.23$ of LiNbO$_3$ at 800 nm is 1.5 times smaller than $n_{\text{opt}} = 3.38$ of GaAs at 1.55 $\mu m$. The small values of $r_0$ are realistic for the conditions of the experiment$^{18}$ due to the normal incidence of the probe beam on the crystal surface.

In general, the dotted curves are lower than the solid ones in Fig. 3(a), i.e., the detection bandwidth is substantially wider for the scheme with GaAs than for the one with LiNbO$_3$. Mathematically,
this is explained by larger \(\sin \beta\) in the denominator of Eq. (21d) for LiNbO\(_3\) (\(\approx 0.9\)) than for GaAs (\(\approx 0.2\)).

Physically, the \(\beta\)-dependent filtering effect, which is inherent to the noncollinear detection geometry, is explained by a tilt of the terahertz wavefronts with respect to the pancake-shaped probe pulse [Fig. 3(b)]. Due to the tilt, different transverse parts of the probe pulse interact with different phases of the terahertz field. This reduces the total modulation of the probe beam polarization. In particular, one can expect a zero modulation when the opposite edges of the probe pulse interact with the terahertz wavefronts separated by one terahertz wavelength \(\lambda_{THz}\), i.e., for \(2r_0 \sin \beta = \lambda_{THz}\).

Expressing \(\lambda_{THz}\) as \(\lambda_{THz} = 2\pi c / \Omega_{THz}\), the above-written condition determines the cut-off frequency as

\[
\Omega_c \sim \frac{\pi c}{r_0 \sin \beta}.
\]

The estimate given by Eq. (24) agrees well with accurate Eq. (21d) in the case when diffraction is negligible \((\gamma \ll 1)\).

To make the analysis more illustrative, we model the input single-cycle terahertz pulse [Eq. (4)] as

\[
E_{THz}^{in}(\xi) = \frac{\xi E_0}{t_{in}} e^{-\xi^2 / T_{in}^2}, \quad \Lambda(\Omega) = \frac{\Omega T_{in}^2 E_0}{4\pi} e^{-\Omega^2 T_{in}^2 / 4},
\]

where \(\xi = t + \tau - x(n_{THz}/c) \cos \beta + \tau(n_{THz}/c) \sin \beta\). The terahertz wave pulse and its amplitude spectrum are shown in Fig. 4. By substituting \(\Lambda(\Omega)\) from Eq. (25), \(C(\Omega)\) [Eq. (23)], \(D(\Omega) = 1, H(\Omega)\) [Eq. (21c)], and \(W_0 = \pi^{1/2}2^{-5/2}E_0c\tau_{opt}C_0r_0^2\tau_{opt}\) to Eq. (20), one can obtain an analytical formula for the output signal,

\[
S(\tau) = 2\xi^{(2)}E_0 \frac{\omega_0 L}{c\tau_{opt}} T_{in}^2 \frac{\tau}{T_{out}^2} e^{-\tau^2 / T_{out}^2},
\]

with

\[
T_{out} = \sqrt{T_{in}^2 + T_{opt}^2 / 2 + 4 / \Omega_c^2}.
\]

According to Eq. (26), the output signal is a single-cycle pulse, which is similar to the input terahertz waveform [Eq. (25)] but has a longer time constant \(T_{out}\) [Eq. (27)]. As discussed above, the term \(T_{opt}^2 / 2\) in Eq. (27) is negligible for a typical THz-TDS setup, and the constant time distortion is mainly due to the term \(4 \Omega_c^{-2}\).

In Eq. (26), both the terahertz field amplitude \(E_0\) and the interaction length \(L\) depend on the focusing conditions of the terahertz beam. For example, weaker focusing decreases \(E_0\) but increases the crossing length \(L\) (Fig. 1). From energy conservation, it follows that the product \(E_0 L\) and, therefore, the detection efficiency remains constant. This conclusion, however, is valid only as long as the crossing length is shorter than the crystal thickness. Otherwise, \(L\) will be limited by the crystal thickness. Thus, tighter focusing is advantageous for using thinner crystals. Additionally, reducing the interaction length \(L\) by tighter focusing suppresses the effect of the probe beam diffraction divergence, thus increasing the detection bandwidth.

Figures 4(a) and 4(b) show the output signals and their spectra for different radii (40, 80, and 120 \(\mu m\)) of the probe beam.
angle $\beta_{\text{Ch}} \approx 63^\circ$. Thus, the noncollinear detection scheme based on LiNbO$_3$ crystal is not applicable to detecting broadband terahertz pulses.

For the detection scheme with a GaAs crystal, the influence of terahertz dispersion, i.e., frequency dependence of $n_{\text{THz}}$ in Eq. (21b), may become significant for long interaction lengths. (For the scheme with a LiNbO$_3$ crystal, the influence of terahertz dispersion is negligible due to a short propagation distance of the terahertz pulse from the lateral crystal surface to the probe beam.) To estimate the effect of dispersion in GaAs, we approximate the function $n_{\text{THz}}(\Omega)$ by the Sellmeier equation given in Ref. 15 and plot the coherence length $L_{\text{coh}} = \pi c/[\Omega|n_{\text{THz}}(\Omega) \cos \beta - n_p|]$ and the modulus of the phase-matching factor $|D(\Omega)|$ as functions of terahertz frequency in Figs. 5(a) and 5(b). It is seen from Figs. 5(a) and 5(b) that one can efficiently control the effect of dispersion on the spectral uniformity and bandwidth of the detection by slightly varying the crossing angle $\beta$. In general, however, the effect of the low-pass factor $H(\Omega)$ is dominating over the phase-matching factor $|D(\Omega)|$ [Fig. 5(b)].

IV. CONCLUSION

To conclude, a theory of ellipsometric electro-optic sampling detection of terahertz waves by nonlinearly propagating femtosecond laser pulses in electro-optic crystals was developed. In the noncollinear geometry, the crossing of the terahertz and probe laser beams at the Cherenkov angle ensures optical-terahertz velocity matching for various types of crystals and laser wavelengths, unlike the conventional collinear detection scheme. This provides high versatility of the noncollinear scheme, its high efficiency, and high spectral resolution due to workability with thick crystals.

We showed that nonlinear ellipsometric detection scheme operates as a low-pass terahertz filter, unlike the nonlinear heterodyne detection scheme, which operates as a bandpass filter. The cut-off frequency of the noncollinear ellipsometric scheme is determined by the probe beam radius (including its diffraction broadening in the crystal) and the crossing (Cherenkov) angle of the terahertz and probe beams. The filtering effect can distort the detection signal by narrowing the signal’s spectrum and stretching correspondingly its waveform.

In the noncollinear scheme with a GaAs crystal and a fiber laser probe beam, the distortions of typical terahertz pulses (with the spectrum width $\lesssim 3$ THz) are shown to be insignificant for the probe beam (1/e) radius in the reasonable range $\approx 15-50 \mu m$ (outside the crystal). The insignificance of the distortions can be explained by a small ($\approx 12^\circ$–$15^\circ$ at the laser wavelength of 1.55 $\mu m$) crossing (Cherenkov) angle of the terahertz and probe beams in GaAs. One can expect the similar results for GaP (with $\approx 19^\circ$ Cherenkov angle at 1.55 $\mu m$) used instead of GaAs. Thus, the semiconductor crystals, such as GaAs and GaP, in conjunction with fiber lasers fit well the noncollinear detection scheme.

In the noncollinear scheme with a LiNbO$_3$ crystal and a Ti: sapphire (or fiber) laser probe beam, the distortions can be significant even for a short (3 mm long) interaction distance and optimal probe beam radius of $\approx 15 \mu m$. This can be explained by a large ($\approx 63^\circ$) Cherenkov angle in LiNbO$_3$. Thus, the LiNbO$_3$-based noncollinear scheme can only be used for the detection of terahertz pulses of relatively narrow ($\approx 2$ THz) bandwidth.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

M. A. Kurnikov: Conceptualization (supporting); Formal analysis (equal); Investigation (lead); Validation (lead); Visualization (lead); Writing – original draft (equal); Writing – review & editing (supporting). M. I. Bakunov: Conceptualization (lead); Formal analysis (equal); Funding acquisition (lead); Investigation (supporting); Methodology (lead); Project administration (lead); Supervision (lead); Validation (supporting); Visualization (supporting); Writing – original draft (equal); Writing – review & editing (lead).

DATA AVAILABILITY

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

APPENDIX: DERIVATION OF EQS. (7) AND (8)

By substituting Eqs. (3) and (5) into Eq. (6), the nonlinear polarization can be represented in the form

$$
I_{z}^{NL}(x, y, z, \omega, \tau) = 2e_0 \int_{\infty}^{\Omega_{\text{cutoff}}} d\Omega \tilde{F}(\omega - \Omega) \tilde{A}(\Omega) e^{-\frac{i\omega \tau}{\tau_0}} \times 
\int_{-\infty}^{\infty} dk_x dk_y G(k_x, k_y) e^{i(k_x x + k_y y + \Omega z)}. $$

(A1)
In Eq. (A1), we change variable \( k_z \) to \( k'_z = k_z + k_y \). This allows us to rewrite the last two integrals as

\[
\int_{-\infty}^{\infty} dk_y dk_z \tilde{G}(k_y, k_z) e^{i k_z (\omega - \Omega) x + i k_y y + i (k_z + k_y) z} = \int_{-\infty}^{\infty} dk_y dk'_z \tilde{G}(k_y, k'_z - k_z) e^{i k'_z (\omega - \Omega) x + i k_y y + i (k'_z - k_z) z},
\]

(A2)

where

\[
k'_z(\omega - \Omega) = k(\omega - \Omega) \left[ 1 - \frac{k_z^2 + (k'_z - k_z)^2}{k^2(\omega - \Omega)} \right]^{1/2}.\]

(A3)

By renaming \( k'_z \) as \( k_z \) and combining \( k'_z(\omega - \Omega) \) and \( K_z \) into \( K'_z = k'_z(\omega - \Omega) + K_z \), we arrive at Eqs. (7) and (8).

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