Temporally resolved electro-optic effect

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The electro-optic effect between an ultrafast optical probe pulse and an ultrashort terahertz pulse is shown to depend on the time derivatives of the product of the probe and terahertz electric fields. Application of this theory to temporally resolved single-shot terahertz detection techniques, where the electro-optic effect is temporally localized within an optical probe pulse, shows that the description presented here differs fundamentally and verifiably from that commonly used in literature.

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Single-shot electro-optic (EO) terahertz (THz) detection techniques are of growing interest, and, for example, have been establishing themselves at the forefront of ultrafast relativistic electron-beam diagnostics. Techniques for the single-shot EO detection of the relativistic Coulomb field of ultrashort electron bunches have been developed and demonstrated at several laboratories, such as the FELIX FEL facility in the Netherlands,[1,2] the Stanford Linear Accelerator Center[3] and Brookhaven National Laboratory[4] in the United States, and the VUV FEL facility at DESY in Germany.[5] EO detection is now widely considered to be one of the most promising techniques for ultrafast electron-beam diagnostics.

In this Letter we develop the theory of the EO effect to describe intrapulse temporal modulations in arbitrary ultrafast pulses, as required for single-shot THz detection techniques. In previous experiments and analyses of the EO effect where the time-localized changes to the optical probe were sought, the EO effect has been described as an additional optical field generated according to the product of the optical and THz fields,[6–9] $E_{\mathrm{opt}}^{\mathrm{out}}(t)=E_{\mathrm{opt}}^{\mathrm{in}}(t)[1+aE_{\mathrm{THz}}(t)]$, where a is a constant. In this Letter we show that, contrary to this, the EO-generated optical field is in fact proportional to the time derivative of the fields, $E_{\mathrm{opt}}^{\mathrm{out}}(t)=E_{\mathrm{opt}}^{\mathrm{in}}(t)+a\,\mathrm{d}/\mathrm{d}t[E_{\mathrm{opt}}^{\mathrm{in}}(t)\cdot E_{\mathrm{THz}}(t)]$.

Following the approach of Gallot and Grischkowsky,[10] the EO effect is derived from the coherent addition of fields generated through sum- and difference-frequency mixing of THz and optical fields. With this approach, Gallot and Grischkowsky derived a frequency-dependent EO phase change, which they then used to derive the change in integrated probe intensity, for a specified time delay between optical and THz pulses. In contrast, here we obtain the electric field as a function of time within the optical pulse, necessary for application to single-shot THz detection techniques.

The two-photon polarization for THz and optical frequency mixing is written in an arbitrary axis frame as $P_i=\epsilon_0\Sigma_j\Sigma_k\chi_{ijk}^{(2)}E_j^{\mathrm{THz}}E_k^{\mathrm{opt}}$, where the subscripts denote the geometrical components. Following a coordinate transformation to a principal axis frame of reference in which the induced polarization and input optical field are parallel, we have $P_i=\epsilon_0\chi_{\mathrm{eff},i}^{(2)}E^{\mathrm{THz}}E_i^{\mathrm{opt}}$, where E^{THz} without subscript refers to the THz field magnitude rather than any particular geometrical component. In this principal axis frame we can examine the EO effect in a single-axis component with no loss of generality; in the following, a single principal axis is considered, and the corresponding component subscript is omitted.

For sum-frequency generation[11] the THz, optical probe, and sum-frequency electric fields are defined in the frequency-domain terms of an envelope $A(z,\omega)$ and a fast oscillating factor; for the THz fields we have $\tilde{E}_{\mathrm{THz}}(z,\omega)=A_{\mathrm{THz}}(z,\omega)\exp(i\Re e\{k_{\mathrm{THz}}\}z)$, while the optical input and sum frequency fields are similarly denoted with the subscripts opt and sum, respectively. k is the wavenumber of the respective fields. The difference frequency field can be obtained from the Hermitian property of the fields, and so is not initially considered explicitly. Applying the slowly varying envelope approximation to the optical field envelopes, $A_{\mathrm{opt}},A_{\mathrm{sum}}$, the solution to the wave equation $(\partial/\partial z+k^{\mathrm{I}})A_{\mathrm{sum}}(z,\omega_{\mathrm{sum}})=\exp[-ik^R(\omega_{\mathrm{sum}})]\times i\mu_0\omega_{\mathrm{sum}}^2\times P(\omega_{\mathrm{opt}},\omega_{\mathrm{THz}})/2k_{\mathrm{opt}}^R(\omega_{\mathrm{sum}})$ is[10,11]

$$A_{\mathrm{sum}}(z,\omega_{\mathrm{sum}})=\frac{i\omega_{\mathrm{sum}}^2\chi_{\mathrm{eff}}^{(2)}(\omega_{\mathrm{sum}};\omega_{\mathrm{opt}},\omega_{\mathrm{THz}})}{2c^2k_{\mathrm{opt}}^{\mathrm{R}}(\omega_{\mathrm{sum}})}\times\left[\frac{\exp(i\Delta kz)-1}{i\Delta k}\right]\exp[-k_{\mathrm{opt}}^{\mathrm{I}}(\omega_{\mathrm{sum}})z]\times A_{\mathrm{THz}}(0,\omega_{\mathrm{THz}})A_{\mathrm{opt}}(0,\omega_{\mathrm{opt}}), \tag{1}$$

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where the difference in wave vectors is denoted $\Delta k = k_{\mathrm{opt}}(\omega_{\mathrm{opt}}) + k_{\mathrm{THz}}(\omega_{\mathrm{THz}}) - k_{\mathrm{opt}}(\omega_{\mathrm{sum}})$, and $k^{\mathrm{R}}, k^{\mathrm{I}}$ are the real and the imaginary parts of k. Energy conservation requires that for the sum-frequency generation $\omega_{\mathrm{sum}} = \omega_{\mathrm{opt}} + \omega_{\mathrm{THz}}$, which allows elimination of the input optical field in Eq. (1); in the following we write $\chi^{(2)}_{\mathrm{eff}}(\omega_{\mathrm{sum}};\omega_{\mathrm{opt}},\omega_{\mathrm{THz}}) = \chi^{(2)}_{\mathrm{eff}}(\omega_{\mathrm{sum}},\omega_{\mathrm{THz}})$. Similarly Δk can be considered as a function of only ω_{sum} and ω_{THz}, which we denote with $\Delta k \equiv \Delta k(\omega_{\mathrm{sum}},\omega_{\mathrm{THz}}) = k_{\mathrm{opt}}(\omega_{\mathrm{sum}} - \omega_{\mathrm{THz}}) + k_{\mathrm{THz}}(\omega_{\mathrm{THz}}) - k_{\mathrm{opt}}(\omega_{\mathrm{sum}})$. For additional clarity, the THz frequencies are denoted $\Omega \equiv \omega_{\mathrm{THz}}$ in the following equations. While Eq. (1) was derived from the perspective of sum-frequency generation, the more general result encompassing both sum- and difference-frequency generation is obtained by allowing negative THz frequencies.[12] In extending the THz range to negative frequencies, it is also necessary for the wavenumbers to be related by $k(-\omega) = -k^{*}(\omega)$ so that the wave propagation direction is fixed. With this extension, integrating Eq. (1) over the THz spectrum, and recognizing that at $z=0$ the envelopes $A(0,\omega_{\mathrm{opt}})$ and fields $\tilde{E}(0,\omega_{\mathrm{opt}})$ are identical by definition, the total sum- and difference-frequency field at frequency ω is

$$\tilde{E}_{\mathrm{sum}}(z,\omega) = \frac{i\omega^2}{2c^2 k^{\mathrm{R}}_{\mathrm{opt}}(\omega)} \exp[ik_{\mathrm{opt}}(\omega)z] \times \int_{-\infty}^{\infty} \mathrm{d}\Omega \left\{ \chi^{(2)}_{\mathrm{eff}}(\omega,\Omega) \left[\frac{\exp(i\Delta k(\omega,\Omega)z) - 1}{i\Delta k(\omega,\Omega)} \right] \times \tilde{E}_{\mathrm{THz}}(0,\Omega)\tilde{E}_{\mathrm{opt}}(0,\omega-\Omega) \right\}, \tag{2}$$

where the subscript sum in $\tilde{E}_{\mathrm{sum}}$ indicates the sum over positive (sum-frequency generation) and negative (difference-frequency generation) THz frequencies. We note that Eq. (2) is equivalent to that given by Gallot and Grischkowsky,[10] with the exception that they repeat the above calculation for negative THz frequencies and include this as a distinct "difference frequency" generation contribution. Their result also differs from Eq. (2) with only the real part of $\chi^{(2)}$ considered to play a role; while such a conclusion can be obtained if the medium is assumed to be lossless, here we make no such assumption.

To proceed further, three closely related approximations in the material optical properties are invoked, while maintaining the generality at THz frequencies: (i) a frequency independent *optical* absorption coefficient, such that $k^{I}_{\mathrm{opt}}(\omega_{\mathrm{opt}}) = k^{I}_{\mathrm{opt}}(\omega) \equiv \beta$, where β is constant; (ii) zero *optical* group-velocity dispersion, such that $k^{R}_{\mathrm{opt}}(\omega_{\mathrm{opt}}) = (\omega_{\mathrm{opt}}/c)n_{\mathrm{opt}}$, $k^{R}_{\mathrm{opt}}(\omega) = (\omega/c)n_{\mathrm{opt}}$, where n_{opt} is constant; and (iii) that $\chi^{(2)}$ is independent of the optical frequency, such that $\chi^{(2)}(\omega,\Omega) \equiv \chi^{(2)}_{\mathrm{eff}}(\Omega)$. In the first two approximations it is implicit that the sum and difference frequency falls within the optical spectral region. The third approximation actually follows as a natural consequence of the first approximations, through the separability of $\chi^{(2)}(\omega,\Omega) \sim f(\omega)g(\Omega)$, and the proportionality between the functions $f(\omega), g(\Omega)$ and the EO material permittivity.[13]

In addressing phase matching, Δk is expanded in a Taylor series about the frequency ω; $\Delta k(\omega,\Omega) = k_{\mathrm{opt}}(\omega) - (\partial k_{\mathrm{opt}}/\partial\omega')|_{\omega}\Omega + k_{\mathrm{THz}}(\Omega) - k_{\mathrm{opt}}(\omega)$. Identifying $(\partial k_{\mathrm{opt}}/\partial\omega')|_{\omega}$ with the inverse of the optical group velocity $n_g(\omega)/c$, and utilizing the zero group-velocity dispersion approximation with $n_g(\omega) = n_g^{\mathrm{opt}}$ where n_g^{opt} is a constant, we obtain a wavevector mismatch dependent only on the THz frequencies, $\Delta k(\Omega) = k_{\mathrm{THz}}(\Omega) - n_g^{\mathrm{opt}}\Omega/c$.

Therefore, using only the three specified approximations on the optical properties, the complex spectrum of the sum- and difference-frequency fields becomes

$$\tilde{E}_{\mathrm{sum}}(z,\omega) = \frac{z}{cn_{\mathrm{opt}}} \exp\left(i\omega\frac{n_{\mathrm{opt}}z}{c} - \beta z \right) i\omega \int_{-\infty}^{\infty} \mathrm{d}\Omega \times \chi^{(2)}_{\mathrm{eff}}(\Omega)\zeta(\Omega)\tilde{E}_{\mathrm{THz}}(0,\Omega)\tilde{E}_{\mathrm{opt}}(0,\omega-\Omega), \tag{3}$$

$$\text{where } \zeta(\Omega) \equiv \left\{ \frac{\exp[i\Delta k(\Omega)z] - 1}{i\Delta k(\Omega)z} \right\}. \tag{4}$$

The frequency mixed fields in the time domain are obtained from the Fourier transformation of the fields given in Eq. (3), $E_{\mathrm{sum}}(z,t) \equiv FT\{\tilde{E}_{\mathrm{sum}}(z,\omega)\}$; recognizing the Fourier transform of the convolution of $[(\chi^{(2)}_{\mathrm{eff}}\zeta\tilde{E}_{\mathrm{THz}}) * \tilde{E}_{\mathrm{opt}}](\omega)$, together with the identification of $FT\{-i\omega\tilde{G}(\omega)\} = \mathrm{d}G(t)/\mathrm{d}t$, where $G(t) \equiv FT\{\tilde{G}(\omega)\}$, it can be concluded that

$$E_{\mathrm{sum}}(z,t) = -\frac{z}{cn_{\mathrm{opt}}} \exp(-\beta z) \times \frac{\mathrm{d}}{\mathrm{d}t}\{[\chi^{(2)}_t(t-\tau) * \zeta_t(t-\tau) * E_{\mathrm{THz}}(0,t-\tau)] \times E_{\mathrm{opt}}(0,t-\tau)\}, \tag{5}$$

where $\tau \equiv n_{\mathrm{opt}}z/c$. In Eq. (5), $\zeta_t(t)$ and $\chi^{(2)}_t(t)$ are the Fourier transforms of $\zeta(\Omega)$ and $\chi^{(2)}_{\mathrm{eff}}(\Omega)$, respectively. For compactness, in the following we write an effective observable THz field as $E^{\mathrm{eff}}_{\mathrm{THz}}(0,t) \equiv \chi^{(2)}_t(t) * \zeta_t(t) * E_{\mathrm{THz}}(0,t)$. In many practical situations, for example, where a suitably thin EO material is used and the THz spectrum is below 2 THz, $\chi^{(2)}_{\mathrm{eff}}(\Omega)\zeta(\Omega)$ may be sufficiently flat over the THz spectral range that the time domain response can be approximated by a delta function, $\chi^{(2)}_t(t) * \zeta_t(t) \to \eta\delta(t)$, where η is constant, and hence $E^{\mathrm{eff}}_{\mathrm{THz}} = \eta E_{\mathrm{THz}}$.

Having determined the temporal domain form of the optical field generated through frequency mixing with the THz field, in the small-signal limit where the field strength of this generated field is small compared with that of the input optical field, it follows that the total field can be written as the sum of the input optical field and the optical sum- and difference-frequency field,

$$\tilde{E}_{\mathrm{total}}(z,\omega) = \tilde{E}_{\mathrm{opt}}(z,\omega) - i\omega B \exp(i\omega\tau)[\tilde{E}^{\mathrm{eff}}_{\mathrm{THz}} * \tilde{E}_{\mathrm{opt}}](0,\omega), \quad (6)$$

$$E_{\mathrm{total}}(z,t) = E_{\mathrm{opt}}(z,t) + B\frac{\mathrm{d}}{\mathrm{d}t}[E^{\mathrm{eff}}_{\mathrm{THz}}(0,t-\tau)E_{\mathrm{opt}}(0,t-\tau)], \quad (7)$$

where $B \equiv -z/cn_{\mathrm{opt}} \exp(-\beta z)$.

The equivalent frequency-domain and time-domain expressions of Eqs. (6) and (7) are quite general and are applicable to temporally localized modulations within an optical pulse, such as those used for single-shot THz measurements. They also describe the common effects of EO phase retardation in a Pockels cell and of EO frequency shifting. For a constant dc THz field $E^{\mathrm{eff}}_{\mathrm{THz}}$, and a monochromatic optical input $E_{\mathrm{opt}}(t) = E^{(0)}_{\mathrm{opt}} \cos(\omega_0 t)$, the time-domain expression leads to $E_{\mathrm{total}}(t) \approx E^{(0)}_{\mathrm{opt}}\{\cos(\omega_0 t) - \omega_0 B E^{\mathrm{eff}}_{\mathrm{THz}} \sin(\omega_0 t)\} \approx E^{(0)}_{\mathrm{opt}} \cos(\omega_0 t + \phi)$, where the retardation phase shift is $\phi = \omega_0 B E^{\mathrm{eff}}_{\mathrm{THz}}$. In the case of monochromatic THz and optical fields, frequency sidebands are generated, as is most easily seen in the frequency-domain expression; the convolution in Eq. (6) will result in fields at the sum and difference frequencies $\omega_{\mathrm{opt}} \pm \Omega$.

We note that the common description for the EO effect within chirped optical pulses[6–9] $E^{\mathrm{out}}_{\mathrm{opt}}(t) = E^{\mathrm{in}}_{\mathrm{opt}}(t)[1 + a \cdot E_{\mathrm{THz}}(t)]$ is neither compatible with the result presented in Eq. (7) nor capable of producing a retardation phase shift as described above; that earlier description must be considered incorrect. In looking for experimental evidence of the time-derivative description presented here, we note that the time derivative introduces a $\pi/2$ phase shift between input and EO generated fields. The time derivative is therefore observable in the relative phase of the input and EO generated fields. Jamison *et al.*[8] have reported spectral interference observations for chirped pulses modulated by unipolar THz pulses that are capable of distinguishing this relative phase. Their experiments were explained through the incorrect $E^{\mathrm{out}}_{\mathrm{opt}}(t) = E^{\mathrm{in}}_{\mathrm{opt}}(t)[1 + a \cdot E_{\mathrm{THz}}(t)]$ description of the EO effect. However, a reanalysis of their experiment shows an incompatibility between experimental observations and that description of the EO effect, while in contrast producing excellent agreement when using the time-derivative description given here; briefly, the effect of a quarter-wave plate, introducing a further $\pi/2$ phase shift, was not accounted for in their analysis and led to the misleading apparent agreement in experiment and (incorrect) theory.

For very short THz pulses, or for optical probe pulses only a few cycles long, experimental conditions that are increasingly encountered in both electron beam diagnostics and in pump–probe THz spectroscopy,[14] the derivative $\mathrm{d}/\mathrm{d}t[E^{\mathrm{in}}_{\mathrm{opt}}(t)E_{\mathrm{THz}}(t)]$ in Eq. (7) will contain nonnegligible contributions from the derivatives of the THz field and of the optical pulse envelope. These contributions will remain in phase with the input field, producing an amplitude modulation proportional to the THz field strength, whereas the derivative of the optical carrier field will be $\pi/2$ out of phase with the input optical field, producing a net phase shift.

In conclusion, a time-domain description of the EO effect has been derived. The dependence on the time derivatives of the fields differs fundamentally from previous descriptions and is necessary for consistency with experimental observations. New amplitude modulation contributions are shown to arise for very short THz pulses or few-cycle optical pulses.

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