Paraxial Theory of Direct Electro-optic Sampling of the Quantum Vacuum

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Direct detection of vacuum fluctuations and analysis of subcycle quantum properties of the electric field are explored by a paraxial quantum theory of ultrafast electro optic sampling. The feasibility of such experiments is demonstrated by realistic calculations adopting a thin ZnTe electro optic crystal and stable few femtosecond laser pulses. We show that nonlinear mixing of a short near infrared probe pulse with the multiterahertz vacuum field leads to an increase of the signal variance with respect to the shot noise level.

Finite fluctuation amplitudes in the ground state of empty space represent the ultimate hallmark of the quantum nature of the electromagnetic radiation field. These vacuum fluctuations manifest themselves indirectly in a number of phenomena that are accessible to spectroscopy such as the spontaneous decay of excited atomic states as well as the Lamb shift [1] in atoms [2] and in quantum-mechanical electric circuits [3]. Access to the quantum aspects of electromagnetic radiation is provided by the analysis of photon correlation [4,5] or homodyning [6–11] measurements. However, these approaches require amplification of the quantum field under study to finite intensity and information is typically averaged over multiple optical cycles.

On the other side, precise determination of a voltage or electric field amplitude as a function of time represents a fundamental task in science and engineering. Optical techniques have to be applied when detecting electric fields oscillating in the terahertz (THz) range and above. Those approaches involve probing with ultrashort laser pulses of a temporal duration on the order of half an oscillation period at the highest frequencies under study. Far-infrared electric transients [12,13] may be characterized by photoconductive switching [14].

Electro-optic sampling in free space [15–17] allows field-resolved detection at high sensitivity in the entire far- and mid-infrared spectral range [18,19]. Direct studies of the complex-valued susceptibilities of materials and the elementary dynamics in condensed matter are performed with these methods [20,21]. The time integral of near-infrared to visible electric-field wave packets is accessible with attosecond streaking [22]. So far, all those techniques were restricted to the classical field amplitude. Very recently, direct access to the vacuum fluctuations of the multi-THz electric field has been established experimentally [23].

In this Letter, we demonstrate theoretically that the quantum properties of light may be accessed directly in the time domain, i.e., with subcycle temporal resolution. Our considerations are based on the realistic example of electro-optic detection with zinc-blende-type materials [24]. Even vacuum fluctuations may be sampled without amplification by broadband probing of electric field amplitudes in the multi-THz region with few-femtosecond laser pulses of moderate energy content.

We consider the geometry sketched in Fig. 1. An ultrashort near-infrared (NIR) wave packet with electric field $E_p$ propagates along the [110] axis of an electro-optic crystal (EOX) [24,25]. Its wave vector $k_p$ is perpendicular to the $z$ axis $e_z$ of the EOX. We select $E_p \parallel e_z$ [26]. In this configuration, the second-order nonlinear mixing of $E_p(t)$ and the incoming near-infrared (NIR) probe $E_{\text{NIR}}(t)$ allows for a wave front separation at the EOX surface followed by photodetection with balanced detectors.

FIG. 1 (color online). Setup and geometry for free space electro optic sampling. (a) The incoming near infrared (NIR) probe and multi THz signal fields mix in the electro optic crystal (EOX). The NIR (blue) spatial mode amplitude is depicted by the contour plot, whereas a THz (red) spatial mode is indicated by wave fronts. Bottom left corner: temporal profiles of the NIR intensity envelope $I_{\text{NIR}}(t)$ and a representative multi THz vacuum field $E_{\text{THz}}(t)$. After collimating with a lens ($L$), the modified NIR field is analyzed using a quarter wave plate ($\lambda/4)_\omega$, a Wollaston prism (WP), and balanced detectors ($D_1$, $D_2$) measuring the difference in photon flux of the split components. (b) Spatial directions determining the electro optic effect in a zinc blende type EOX and the following ellipsometry analysis.

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with an incident THz field \( \hat{E}_{\text{THz}}(t) \) induces nonlinear polarization in the EOX plane with the components [26]

\[
\begin{align*}
\frac{\partial E^{(2)}}{\partial t} & = -\varepsilon_0 d E^{(2)}(t) E_p(t), \\
\frac{\partial E_p}{\partial t} & = 0.
\end{align*}
\]

where \( \varepsilon_0 \) is the vacuum permittivity. The coupling constant \( d = -n^2 r_{41} \) can be determined from the electro-optic coefficient \( r_{41} \) and refractive index (RI) \( n \) at the center frequency \( \omega_c \) of \( E_p \) [32–34]. In general, both fields \( \hat{E}_{\text{THz}} \equiv \hat{E}_{\text{THz},x} \) and \( \hat{E}_p \) in Eq. (1) are quantized, whereas \( E_p = E_p(z) \) denotes the classical part of the probe field.

The effect of quantum mechanical fluctuations of the probe field on \( \hat{E}^{(2)} \), assuming a sufficiently large \( E_p \).

The nonlinear polarization \( \hat{E}^{(2)} \) represents a source in the inhomogeneous wave equation describing the evolution of the electric field \( \hat{E} \) in the EOX. The fields \( \hat{E} = \hat{E}, \hat{E}^{(2)} \) propagating in the forward direction \( r_{\parallel} \) (see Fig. 1) can be decomposed as \( \hat{F}(r,t) = \int d\Omega \hat{F}(\Omega) e^{i(\Omega r_{\parallel})} \), where \( \Omega = o m_{\Omega} c_0 \), and \( n_\Omega \) is the velocity of light and the frequency-dependent RI of the EOX, respectively. Using the paraxial approximation [35, 36], the inhomogeneous wave equation reads

\[
\left[ \Delta + 2i k_o \frac{\partial}{\partial r_{\parallel}} \right] \hat{E}(r;\omega) = -\frac{\alpha_p^2}{\varepsilon_0 c_0^2} \hat{E}^{(2)}(r;\omega),
\]

where \( r = (r_x, r_z) \) and \( \Delta = (\partial^2/\partial r_x^2) + (\partial^2/\partial r_z^2) \). From Eq. (1) we obtain \( \hat{P}^{(2)}(r;\omega) = -\alpha_p^2 \varepsilon_0 c_0^2 \hat{E}^{(2)}(r;\omega) \). Inserting Eq. (6) into Eq. (7) and neglecting the second-order terms in \( \hat{E}^{(2)} \) as well as the mixed terms depending linearly both on \( \hat{E}^{(2)} \) and on \( \hat{E}_{\text{THz}} \) (contained in \( \hat{\phi} \)) [42] and \( \hat{E}_{\text{THz}} \) (contained in \( \hat{\phi} \)) [42], the total detected quantum signal becomes
\[ \hat{S} = \hat{\mathcal{N}}_s - \hat{\mathcal{N}}_c = \hat{S}_{\text{EO}} + \hat{S}_{\text{SN}}. \] (8)

Here, the electro-optic signal (EOS) \( \hat{S}_{\text{EO}} \) is

\[ \hat{S}_{\text{EO}} = C \int d^2r_1 \int_0^\infty \frac{d\omega}{\hbar} \frac{n(\omega)}{\hbar} |E_p(\mathbf{r})|^2 \langle \phi(\mathbf{r}) + \text{H.c.} \rangle \] (9)

and the shot noise (SN) contribution \( \hat{S}_{\text{SN}} \) reads

\[ \hat{S}_{\text{SN}} = C \int d^2r_1 \int_0^\infty \frac{d\omega}{\hbar} \frac{\delta(\omega)}{\hbar} |E_p(\mathbf{r})|^2 \delta^e(\mathbf{r}) + \text{H.c.}. \] (9)

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denotes the Hermitian conjugate and \( \delta^e(\mathbf{r}) = e^{i\pi/4} |\delta^e(\mathbf{r}) + i\delta^e(\mathbf{r})|/\sqrt{2} \) is the circular component of the vacuum contribution of the probe field [43].

Summing up the signals from both detectors, the expectation value of the number of detected photons per probe pulse \( N = \langle \hat{N}_s + \hat{N}_c \rangle = \langle 4\pi a_\omega c_\omega / \hbar \rangle \int_0^\infty \frac{d\omega}{\hbar} |\eta(\omega)/\omega| \times |\alpha_p(\omega)|^2 \rangle \) results.

Using Eqs. (3) and (5) in Eq. (9), we obtain

\[ \hat{S}_{\text{EO}} = \frac{d\eta(\omega)}{c_\omega n} \int d^2r_1 \frac{g_0^0(r_1)}{\hbar} \int_0^\infty d\Omega \frac{E_{\text{Thz}}(r_1; \Omega) R(\Omega)}{-\hbar \Omega}. \] (10)

We have introduced the response function \( R(\Omega) = \text{sinc}((\Omega/2c_\omega)(\eta_\Omega - n_\Omega))/\Omega \) in Eq. (10) is given by [26]

\[ \frac{E_{\text{Thz}}(r_1; \Omega)}{\hbar} = -i \sum_{l,p} \sqrt{\frac{\hbar}{4\pi e_\omega c_\omega n_\Omega}} \tilde{a}_{s,l,p}(\Omega) g_{l,p}^0(r_1) \] (11)

for \( \Omega > 0 \). \( \tilde{a}_{s,l,p}(\Omega) \) annihilates a photon with frequency \( \Omega \) orbital quantum numbers \( l, p \), and polarization \( e_s \). We have introduced the transverse mode functions \( g_{l,p}(r_1) \) \( L\mathcal{G}_\Omega(l, r_1; \hbar_k = 0; \Omega) \).

In contrast to the probe beam, the waist size \( w_0' \) characterizing these mode functions is a free parameter of the expansion (11). Inserting Eq. (11) into Eq. (10) and selecting \( w_0' = w_0/\sqrt{2} \), we can evaluate the spatial integral using \( \int d^2r_1 \frac{g_0^0(r_1)}{\hbar} g_{l,p}^0(r_1) \) \( = (1/\sqrt{\pi w_0^2}) \delta_{l,p} \delta_{l,p,0} \). Then we obtain from Eq. (10)

\[ \hat{S}_{\text{EO}} = -i \sqrt{B} \int_0^\infty d\Omega \frac{\hbar}{\eta_\Omega} [\tilde{a}_{s,0,0}(\Omega) R(\Omega) - \text{H.c.}] \] (12)

where \( B = (d^2N a_\omega^2 \hbar)/(4\pi^2 e_\omega c_\omega n_\Omega w_0^2) \).

As an input, we now consider a THz quantum field with no coherent (classical) contribution: \( \langle \hat{E}_{\text{Thz}} \rangle = 0 \), e.g., a bare multi-THz vacuum. Then \( \langle \hat{S} \rangle = 0 \) since \( \langle \hat{S}_{\text{SN}} \rangle = 0 \) and \( \phi \) in Eq. (9) depends linearly on \( E_{\text{Thz}} \), thus also \( \langle \hat{S}_{\text{EO}} \rangle = 0 \). However, the variance of the signal does not vanish. If the range of detected THz frequencies, determined by \( R(\Omega) \), does not overlap with the frequency content of the probe beam, the signal variance \( \langle \hat{S}^2 \rangle - \langle \hat{S} \rangle^2 \) can be written as \( \langle \hat{S}^2 \rangle = \langle \hat{S}_{\text{EO}}^2 \rangle + \langle \hat{S}_{\text{SN}}^2 \rangle \). Calculating the SN contribution using the paraxial quantization [38], we obtain the expected result \( \langle \hat{S}_{\text{SN}}^2 \rangle = N \).

Evaluating \( \langle \hat{S}_{\text{EO}}^2 \rangle \) for the multi-THz vacuum yields

\[ \langle \hat{S}_{\text{EO}}^2 \rangle = N^2 \left( \frac{1}{c_\omega} \frac{\hbar}{\pi^2 e_\omega c_\omega n_\Omega w_0^2} \right) \int_0^\infty d\Omega \frac{\hbar}{\eta_\Omega} |R(\Omega)|^2, \] (13)

where we have used \( \langle \hat{a}_{l,p,0}(\Omega) \hat{a}_{l,p,0}^\dagger(\Omega) \rangle = \delta(\Omega - \Omega) \). Note that the expectation values of all other possible quadratic combinations of \( \hat{a}_{l,p,0} \) and \( \hat{a}_{l',p,0} \) vanish. The first two factors on the right-hand side of Eq. (13) determine the sampling efficiency. The fundamental physics is contained in the third factor representing the variance of the multi-terahertz vacuum field \( \langle \Delta E^2 \rangle = \hbar/e_\omega (\Delta \lambda \Delta \Omega \Delta \Delta t) \). The transverse area \( \Delta x \Delta y \) is set by the cross section of the sampling mode which is proportional to \( w_0^2 \). The ratio of \( c_\omega \) to the integral containing the response function \( R(\Omega) \) determines the longitudinal cross-sectional area. It corresponds to the effective spatial length \( \Delta x \Delta t \) times the temporal duration \( \Delta t \) of the sampling pulse which become modified by the phase-matching conditions and renormalized due to the refractive index \( n_\Omega \) inside the EOX. Consequently, \( \langle \hat{S}_{\text{EO}}^2 \rangle \) may be modulated in an experiment by lateral or transverse expansion of the four-dimensional space-time volume over which the probe pulse averages while keeping \( \langle \hat{S}_{\text{SN}}^2 \rangle \) exactly constant [23].

To illustrate the results, we assume the following realistic specifications of the sampling few-femtosecond NIR laser pulse: center frequency 255 THz, spectral bandwidth 150 THz with rectangular spectral shape and flat phase, leading to \( \omega_p = 247 \text{ THz} \), and waist size \( w_0 = 3 \mu m \) [44]. We consider a \( l = 7 \mu m \) thick ZnTe EOX with \( r_{41} = 4 \text{ pm/V} \) [45,46], \( n = 2.76, n_s = 2.9, \) and \( n_s \) varying only slightly (from 2.55 to 2.59) for the relevant THz frequencies [26]. The resulting integrand function entering Eq. (13) is shown in Fig. 2(a) (for details, see Ref. [26]). Diffraction effects are taken into account by excluding wavelengths \( \lambda < 2n_s \) w_0 > w_0.

Based on this input, we calculate the dependence of the rms value of the signal \( \Delta S = \langle \hat{S}^2 \rangle^{1/2} \) on the average number \( N \) of photons in the sampling NIR pulse, as shown in Fig. 2(b) on a double-logarithmic scale. Above a certain \( N \), the EOS contribution of the multi-THz vacuum changes the typical SN scaling. The relative increase of the rms value of the signal with respect to the SN level, \( \Delta S \Delta S_{\text{SN}}/\Delta S_{\text{SN}} \), is depicted in Fig. 2(c) for moderate \( N \) and with linear scaling. For even higher \( N \), the vacuum
contribution starts to dominate so that the dependence saturates to the constant EOS level [Fig. 2(b)]. Subtracting the SN contribution from the total signal variance, the bare EOS variance induced by the sampled quantum field can be analyzed.

To elaborate on this point, we apply our theory to a multi-THz vacuum which is squeezed in an interval around a center frequency $\Omega_c$. The corresponding state of light is generated by the continuum squeezing operator [47–49] acting on the multi-THz pure vacuum (PV) state considered above.

The frequency-dependent squeezing parameter $\xi$ of the electric fields of an input state satisfies the condition $\xi^2 \equiv \xi^2_{\Omega_c} = \xi_{\Omega_c}$. We assume that all spatial and polarization modes are squeezed equally. In this case, the EOS can be obtained from Eq. (12) by differentiating it from the trivial shot noise of the high-frequency gating pulse. The crucial aspects are a strong quantum fluctuations of the multi-THz vacuum electric contrast, electro-optic sampling provides a true subcycle resolution of the probed multi-THz electric field. Moreover, registration of photons is transferred into the NIR, circumventing the lack of efficient single-photon detectors in the multi-THz frequency range. Most importantly, the multi-THz quantum field may be studied without the necessity to reduce or amplify its photon content—even if it remains in its ground state. For a detailed discussion, see Ref. [26].

In conclusion, we theoretically clarify the contribution of the quantum fluctuations of the multi-THz vacuum electric field to the signal in ultrabroadband electro-optic sampling by differentiating it from the trivial shot noise of the high-frequency gating pulse. The crucial aspects are a strong localization of the sampling beam in space and time as it passes the nonlinear crystal, a large second-order nonlinear coefficient and proper phase matching that might be further optimized selecting an even more appropriate material than the thin piece of ZnTe we have considered as an example [23].

For a multi-THz squeezed vacuum, the possibility to trace the

FIG. 3 (color online). (a) Frequency dependence of the squeeze factor $r$. Squeezing correlates $\Omega$ and $2\Omega_c - \Omega$ modes (as indicated by arrows). (b) Error contour in the complex amplitude plane for PV (gray circle) and SV with $\theta = 0$ ($\theta = \pi$) [red (green) ellipse with reduced uncertainty in the phase (amplitude) quadrature $\theta$ ($X$)]. (c) Normalized (with respect to the constant PV level, solid black line) EOS variance in dependence on the time delay $\tau$ of the probe NIR pulse for SV with $\theta = 0$ ($\theta = \pi$) [solid red line (dashed green line)].
oscillations of the EOS variance with the time delay of the probe pulse is predicted. Positions occur where the noise remains significantly below the level of unsqueezed vacuum. The same formalism can be applied for the analysis of more complex quantum fields in a time-resolved and nondestructive manner. Experimental implementation of these ideas might open up a new chapter of quantum optics operating predominantly in the time domain and with subcycle access to the quantum state of electromagnetic radiation.

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[40] See Ref. [41] for a case with a simpler transverse mode structure.
[42] The mixed terms were also neglected already in Eq. (1). The second order terms in $\delta E^s \langle \delta E^s \delta E^s \rangle$ do not contribute to the expectation value of the signal, neither to its variance or any higher moments.
[43] The phase shift is of no physical importance for the vacuum field contribution.