A new time-domain method for subcycle metrology of quantum electric fields using a combination of a third order nonlinear optical process and homodyne detection with a local oscillator (LO) field is proposed and analyzed. The new method enables isolation of intrinsically weak quantum noise contribution by subtraction of the shot noise of the LO on a pulse-by-pulse basis. Together with the centro-symmetric character of the nonlinearity, this method unlocks novel opportunities toward terahertz and mid-infrared quantum field metrologies.

1. Introduction

Homodyne detection (HD)\cite{Gundogdu2011} is a central technique of signal analysis in quantum optics. A quantum field under study undergoes on a photodiode with a strong classical mode analysis in quantum optics. A quantum field under study interacts with a signal field with a high center frequency $\Omega$ (e.g., THz or MIR) and a probe pulse $E_P$ at a high center frequency $\omega_0$ (e.g., near-infrared, NIR) and bandwidth $\Delta \omega$.

Einstein’s mass–energy relation $E = mc^2$ implies that co-propagating, optical and infrared photons with energy $\hbar \omega_0$ and $\hbar \omega_P$ (e.g., THz or mid-infrared) can be converted into each other by a nonlinear, frequency-doubling process.

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\[ E_P \times E_\omega \propto E_{\omega_0} \]

The result is that $E_\omega$ and $E_P$ (which are in phase at $\tau = 0$, $\tau$ being the time delay between $E_\omega$ and $E_P$) present an interference pattern with a period $T$. Because the bandwidths $\Delta \Omega$ and $\Delta \omega$ are much smaller than $\omega_0$, the resulting interference is exploited for the HD of quantum contributions to the signal variance, based on a shot-by-shot carrier-envelope phase (CEP) modulation on $E_\omega$ of a free-running frequency comb\cite{Seletskiy2017}. Furthermore, dipole inactivity of optical phonons in common inversion-symmetric materials can be found under https://doi.org/10.1002/1por.202200706.

Recent demonstrations ported EOS to the quantum regime, showing direct sampling of a quantum vacuum field\cite{Moskalenko2019a, Moskalenko2019b} and even measuring the spatio-temporal correlations\cite{Moskalenko2019c} and causal structure of the electromagnetic ground state\cite{Moskalenko2019d}. Such developments promise direct routes toward MIR and THz quantum sensing technologies, while the time-domain character motivates new metrology protocols\cite{Seletskiy2015, Seletskiy2016, Seletskiy2017} as well as a path toward experimental quantum electro-dynamics in space-time\cite{Moskalenko2019a, Moskalenko2019b, Moskalenko2019c, Moskalenko2019d}.

In this paper, we propose a new scheme for time-domain metrology of quantum signals based on the third-order ($\chi^{(3)}$) nonlinear interaction between quantum $E_\omega$ and classical $E_P$, admitting full access to the term carrying interference of $E_{\omega_0}$ and the signal fields\cite{Gundogdu2011, Gundogdu2012, Gundogdu2013, Gundogdu2014}. In the four-wave mixing, the THz-induced second-harmonic (TFISH) signal arises from the nonlinear mixing product $E_P \times E_\omega \times E_T$, at frequencies $2\omega_0 \pm \Omega$, which can be superimposed in a background-free manner with the $E_{\omega_0}$ centered at $2\omega_0$. We show that this freedom opens an elegant opportunity for a direct self-referenced measurement of the quantum contribution to the signal variance, based on a shot-by-shot carrier-envelope phase (CEP) modulation on $E_\omega$ of a free-running frequency comb\cite{Seletskiy2017}. Furthermore, dipole inactivity of optical phonons in common inversion-symmetric materials

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positions the $\chi^{(3)}$-based scheme for efficient field-resolved detection in the 5–15 THz band, which is generally problematic for a host of EOS detection crystals. Finally, the new scheme does not require analysis of the polarization state of the probe, relaxing constraints on broadband polarization optics.

2. Generation of the TFISH and LO Fields

Figure 1 shows the schematic of the proposed metrology scheme. It consists of an interferometer with two paths. The upper path combines $E_r$, the THz signal to be characterized, and one part of the NIR probe pulse $E_p' = \sqrt{R} E_p$, both passing through a $\chi^{(3)}$ nonlinear crystal to generate the TFISH signal $E_{2P}$. In the lower path, the second part of the probe $\sqrt{1-R} E_p$ undergoes a broadband second harmonic generation (SHG) process in a $\chi^{(2)}$ nonlinear crystal, to generate the LO pulse $E_{LO}$ centered at a frequency $2\omega_o$. $E_{LO}$ is mixed with $E_{LO}$ at a beamsplitter, and analyzed by the balanced HD technique. The measurement system is based on a free-running frequency comb, whereas an external electro-optic modulator (EOM) provides an opportunity to control the carrier-envelope phase shift $\Delta \phi_{CEP}$ of the LO field on a pulse-by-pulse basis. This feature is exploited for the self-referenced detection, where the difference in the variance sampled by two adjacent LO pulses isolates the quantum contribution (Figure 1c), as detailed below.

The total electric field passing through the $\chi^{(3)}$ crystal, $E$, can be represented as a sum: $E = E_p' + E_{2P} + E_T$. When $E_T$ corresponds to a quantum signal, its properties together with those of $E_{2P}$ and $E_p'$ should be described in terms of operators denoted as $\hat{E}_T$, $\hat{E}_{2P}$ and $\hat{E}$, respectively. Since the probe is a strong coherent field, it is sufficient here to describe it classically using the corresponding mean value $\langle \hat{E}_p' \rangle \equiv E_p'$. Thus, the generation of $E_{2P}$ is described by the third order nonlinear polarization

$$p_{2P} = 3 \varepsilon_0 \chi^{(3)} R \hat{E}_p (t) \otimes (\hat{E}_p' + \hat{E}_T) + \text{H.c.}$$

where we have decomposed $E_p (t)$ into its positive $E_p^+$ ($E_p^+$) and negative $E_p^-$ ($E_p^-$) frequency parts, $\varepsilon_0$ is the vacuum permittivity. We have assumed that the interaction can be well described by an effective $\chi^{(3)}$ constant, neglecting its frequency dependence. Further, we consider such a crystal orientation that it is sufficient to include only one linear polarization component for each of the involved fields. We also ignore any other third order nonlinear polarization terms since they are comparably much weaker, due to the fact that $E_p'$ is a strong coherent field and $E_T$ is a weak quantum signal, or the corresponding generated field contributions are removed by an appropriate bandpass filter before the final beamsplitter. Under these general assumptions, we can describe the electromagnetic wave propagation in the $\chi^{(3)}$ crystal using the inhomogeneous wave equation within the slowly varying amplitude approximation (SVA) for plane waves. We decompose all fields, $X = \hat{E}_p, \hat{E}_T, \hat{E}_{2P}, \hat{p}_{NL}$, into forward-propagating plane waves as $X(z,t) = \int d\omega X(z,\omega) e^{i(\omega t - \omega z)} d\omega$ + H.c. using a convention $X(z,\omega) \equiv 0$ for $\omega < 0$, $X^{(+)}(z,\omega) \equiv X(z,\omega)$, and $X^{(-)}(z,\omega) \equiv \{X(z,\omega)\}^*$, which has to be taken into account for convolutions we define below. Then solving the propagation equation for the TFISH under the assumptions of small crystal thickness d and negligible depletion of the probe, we obtain (cf. Supporting Information):

$$\hat{E}_{2P}(\omega) = i d A \varepsilon_0 C(\omega) \{ R + (\hat{E}_p^+ + \hat{E}_T^+) \} (\omega) + \hat{E}_p(\omega)$$

where $\hat{E}_p(\omega)$ is the frequency-dependent refractive index and $\varepsilon_0$
is the speed of light in vacuum. We decomposed \( E_p = A_p f(\omega) \) into its (real) amplitude \( A_p \) and (generally complex) normalized frequency distribution \( f(\omega) \), defining a gating function \( R(\omega) = (f * f)(\omega)e^{i\omega\tau} \). The time \( \tau \) corresponds to the center of the probe pulse relative to the incoming THz field (Figure 1a).

Finally, \( E_\text{LO}(\omega) \) is the co-propagating background vacuum field, existing even in the absence of the probe field. As we will see below, this contribution can be directly characterized in the current setup by studying the signal variance when \( \phi\text{CEP} = 0 \) (cf. Figure 1c) is induced on the LO field. In the above derivation, we have neglected the back-action effect of the \( \chi^{(3)} \) interaction on the co-propagating THz field \( \hat{E}_T \). That is appropriate if the first term on the right-hand side of Equation (2) represents a small correction to the second term and is in accordance with our evaluation of the variance signals.

### 3. Homodyne Detection

As described, HD is enabled by letting \( \hat{E}_\text{LO} \) interfere with the LO field \( E_{\text{LO}} \) which is produced via the SHG in the \( \chi^{(3)} \) crystal. The SHG process can be realized with a high conversion efficiency \( \eta_p \), and the resulting mode \( f_{\text{LO}}(\omega) \) of the LO still closely resembles the mode of the TFISH field. For the details of a possible realization, see Supporting Information. Since the LO represents a strong coherent field, we can describe it classically for the homodyning part of the setup. At the beamsplitter before the detectors, we can express it in the frequency domain as \( E_{\text{LO}}(\omega) = A e^{-i\phi} e^{-i\Omega t} f_{\text{LO}}(\omega) \), where \( A = \sqrt{\eta_p} \sqrt{1 - RA_p} \) is the amplitude and \( \phi \) is a variable CEP induced by the EOM. An elegant alternative realization of such phase shift between the two arms of the setup can be provided by a dual frequency comb.\(^{[36]}\)

We assume the LO has effectively the same time delay \( \tau \) as the TFISH field because of the equal optical path lengths corresponding to the upper and lower arms of the setup. To assure that this is completely fulfilled in experiment, an additional tuning can be introduced by inserting an auxiliary delay line.

The operator for the signal we are interested in is given by the number of photons of balanced photodetectors, \( N_S \rightarrow \hat{N}_S \equiv \hat{S}_\text{hom} \), with

\[
\hat{S}_\text{hom} = C A \int_0^\infty d\omega_2 \frac{d\omega_1}{h\omega_2} \eta(\omega_1) \eta(\omega_2) e^{i\phi} f_{\text{LO}}(\omega_1) \hat{E}_{\text{THz}}(\omega_2) + \text{H.c.} \tag{3}
\]

where \( C = 4\pi c\epsilon_0 F \), \( F \) is the effective detection area and \( \eta(\omega) \) is the frequency-dependent quantum efficiency of the photodetectors. Using the decomposition of \( \hat{E}_\text{LO} \) given by Equation (2), we can write \( \hat{S}_\text{hom} \) as \( \hat{S}_\text{hom} = \hat{S} + \hat{S}_B \), with \( \hat{S} \) corresponding to the background vacuum signal, and

\[
\hat{S}(\tau) = C' e^{-i\Omega \tau} \int_0^\infty d\omega_1 \frac{d\omega_2}{h\omega_2} \eta(\omega_2) f_{\text{LO}}(\omega_1) e^{i\phi} \hat{E}(\omega_2) + \text{H.c.} \tag{4}
\]

where \( C' = 6\pi \epsilon_0 \Omega F \sqrt{\eta_p} (1 - RA_p)/h \) and the resulting gating function, limiting the effective integration range in Equation (4) to THz frequencies, is given by

\[
G_\text{LO}(\Omega) = \sqrt{\eta_p} f_{\text{LO}}(\omega) \frac{f(\omega) e^{i\omega\tau}}{\sqrt{\pi}}
\]

\[
G_\text{LO}(\Omega) = \int_0^\infty d\omega \eta(\omega) f_{\text{LO}}(\omega) \frac{f(\omega) e^{i\omega\tau}}{\sqrt{\pi}} \tag{5}
\]

We have introduced \( \omega_{\text{cut}} \) to represent an optional upper spectral limit for the collected NIR photons, as discussed below. Deriving Equation (4), we switched the order of integrations between the integral of Equation (3) and that of the convolution coming there from Equation (2). Casting the electric field operator in terms of creation \( a_i(\Omega) \) and annihilation \( a_i(\Omega) \) operators, leading to \( \hat{E}(\Omega) = -i\sqrt{\frac{\pi}{2\Omega}} C N \hat{a}_i(\Omega) \) for \( \Omega > 0 \), the TFISH induced contribution to the homodyne signal can be rearranged as

\[
\hat{S}(\tau) = -iC' \sqrt{\frac{C}{\pi}} \int_0^\infty d\Omega \frac{\sqrt{\Omega}}{\sqrt{\pi\Omega}} G_\text{LO}(\Omega) e^{-i\omega\tau} \hat{a}_i(\Omega) + \text{H.c.} \tag{6}
\]

In order to obtain the quantum statistics of the operator \( \hat{S}(\tau) \) at each time delay \( \tau \) from the general Equations (6) or (4) and thus to get access to the time-resolved properties of the quantum field \( \hat{E}_T \), we need to assume a particular form of this field.

The simplest case for the consideration is provided by the bare THz vacuum field. In this case, the mean values of both contributions to \( \hat{S}_\text{hom} \) vanish. Further, since these contributions are determined by creation/annihilation operators stemming from different frequency ranges, they are uncorrelated. Thus, the total variance is given by the sum of the variance of the TFISH part,

\[
\langle \hat{S}_\text{Vac} \rangle = \sigma_{\text{Vac}}^2 = \langle \hat{S}(\tau) \rangle = C' \frac{C}{\pi} \int_0^\infty d\Omega \frac{\sqrt{\Omega}}{\sqrt{\pi\Omega}} |G_\text{LO}(\Omega)|^2 \tag{7}
\]

and of the variance originating from the background vacuum in the range of \( \omega_0 \) frequencies (here we assume \( \omega_{\text{cut}} = \infty) \),

\[
\langle \hat{S}_B^2 \rangle = \sigma_{\text{Vac}}^2 = A^2 C' \int_0^\infty d\omega \frac{d\omega_2}{h\omega_2} |f_{\text{LO}}(\omega_2)|^2 |\eta(\omega_2)|^2 \tag{8}
\]

Both variances are independent of the time delay \( \tau \). The TFISH part is determined by the properties of the gating function \( G_\text{LO}(\Omega) \), which follow from the relation between \( G_\text{LO}(\Omega) \) and \( G_\text{LO}(-\Omega) \) and can be influenced by the phase shift \( \phi \). When the temporal profiles of the TFISH and SHG signals coincide, we have \( G_\text{LO}(\Omega) = C(\Omega) \). Then \( G_\text{LO}(\Omega) \) is real and vanishes for \( \phi = 0 \). Its absolute value is maximized for \( \phi = \pm \pi/2, \) with \( |G_\text{LO}(\Omega)| = |G_\text{LO}(\Omega) + C(\Omega)| = 2|\text{Re}(G_\text{LO}(\Omega))| \), representing the optimal configuration for the sampling of the THz vacuum. With \( \hat{S}_\text{hom} = \hat{S} + \hat{S}_B \) for \( \phi = 0 \), the corresponding measurement outcomes can be used for the elimination of the background NIR vacuum contribution on the pulse-by-pulse basis (self-referencing), alternating the CEP by \( \pi/2 \) between the pulses (see Figure 1c).

In order to sample THz quantum fields beyond the bare vacuum, we need to get access to both generalized quadratures of the sampled field in the time domain.\(^{[14,25,28]}\) This is possible to achieve with a variety of the proposed setup, introducing an asymmetry between \( G_\text{LO}(\Omega) \) and \( G_\text{LO}(-\Omega) \) contributions to \( G_\text{LO}(\Omega) \). One of the easiest ways to realize this, suitable for our discussion here, is based on the cuts in the spectra of the detected photons that can be implemented via the corresponding frequency bandpass filters.\(^{[25,28]}\) Looking at the second line of Equation (5) one can anticipate that, for example, for a bandpass filter cutting the frequencies above the central frequency of the LO, \( G_\text{LO}(\Omega) \) dominates over \( G_\text{LO}(-\Omega) \) in terms of the absolute magnitude and \( G_\text{LO}(\Omega) \) becomes complex. In the easiest case, it can be written as
4. Results

Let us first illustrate how the proposed scheme for sampling of quantum fields operates in the case when we apply it to the bare THz vacuum, using typical experimental parameters. We assume that the probe is Gaussian, has central frequency $\omega_0$ and spectral width $\sigma$. The LO field has central frequency $2\omega_0$ and spectral width $\sqrt{2}\sigma$. Ignoring dispersion and assuming a flat response for the detector crystal, that is, $\eta(\omega_0) = n$ and $\eta(\omega_0) = \eta$ as well as no spectral cuts for the detected photons, meaning $\omega_{cut} = \infty$ in Equation (5), gives $G_\phi(\Omega) = \eta \exp\left(-\Omega^2/8\sigma^2\right)/2\sqrt{2}\pi\sigma$. The TFISH contribution to the variance is maximized for $\phi = -\pi/2$, with $G_\phi(\Omega) = 2G_\phi(\Omega)$ here. We get then

$$\langle S^2 \rangle_{\text{vac}} = \frac{9d^2\eta^2\eta_0^2(1 - R)R^2\pi^2\sigma(\chi^{(2)}/\omega_0^2k^4)}{8\epsilon_0^2c^2\eta_0^2\epsilon_0^2k^2}N_p^3$$

where $N_p = 4\xi^2nC^2f_0\pi\omega_0\omega_0^2 \approx \frac{\Omega^2nC^2}{2\epsilon_0^2\omega_0^2}$ is the average number of photons in the probe. Under the same conditions, we obtain

$$\langle S^2 \rangle_\text{cut} \approx \frac{n^2\eta^2}{2\pi\sqrt{2}\sigma\omega_0} = \eta^2\eta_0(1 - R)N_p$$

Figure 2a shows $\langle S^2 \rangle_{\text{vac}}$, $\langle S^2 \rangle_\text{cut}$ and the total variance $\langle S^2 \rangle_{\text{total}}$ as functions of the number of photons in the probe pulse, contrasted with an estimation of the back-action efficiency of the $\chi^{(3)}$ process in the perturbative regime (see Supporting Information for details). We choose the parameters considering a probe laser with 1.55 μm central wavelength and 6 fs pulse duration and a thin 12 μm slab of diamond for the $\chi^{(3)}$ crystal. The variance of the TFISH part surpasses the variance due to the background vacuum at around $N_p = 3.7 \times 10^{11}$ photons per pulse. However, the back-action becomes significant around the same photon number, thus invalidating the perturbative approach. To prevent this, $N_p$ can be chosen at a lower value, implementing a weak measurement. To achieve the required signal-to-noise ratio, the measurement results can be then averaged over a sufficient number of probe pulses, as in refs. [20, 22, 27].

As an illustration of the scheme going beyond sampling of the THz quantum vacuum, Figure 2b shows the variance signals for a broadband cat state, as a function of the time delay of the probe. The utilized cat state is defined as $|\text{cat}\rangle = \frac{1}{\sqrt{2}}(|\alpha_1\rangle + |\alpha_2\rangle)$, where $\{|\alpha_i\rangle\}$ represents a continuous multimode coherent state with the spectral amplitudes $\alpha_i$ corresponding to a classical few-cycle THz pulse (see p. 85 of ref. [38]). To access both quadratures, we performed the frequency cut on the spectral content of the photons collected by the photodetectors, determining the gating function, Equation (5), by choosing $\omega_{cut} = 2\omega_0$. Further details on the calculation are given in Supporting Information.
5. Conclusion

We have developed a new scheme for sampling of quantum fields in the time domain and illustrated it by calculating the electric-field variance and its dynamics for the quantum vacuum and a pulsed broadband cat state, respectively. The scheme is feasible for typical experimental parameters and has a number of intrinsic advantages, such as automatic subtraction of the contaminating shot-noise by the shot-by-shot self-referencing. High sensitivity in the 5–15 THz provides the ability to study low-energy quantum dynamics in condensed matter, while frequency filtering of homodyne signal gives access to both generalized electric-field quadratures, en route toward subcycle quantum tomography. Finally, lifting the reliance on high-precision polarization optics holds promise for future imaging and microscopy applications involving THz and MIR quantum fields.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

balanced homodyne detection, cat states, electro-optic sampling, quantum metrology, quantum vacuum, ultrabroadband

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