Boosting quantum vacuum signatures by coherent harmonic focusing

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We show that coherent harmonic focusing provides an efficient mechanism to boost all-optical signatures of quantum vacuum nonlinearity in the collision of high-intensity laser fields. Assuming two laser pulses of given parameters at our disposal, we demonstrate a substantial increase of the number of signal photons measurable in experiments where one of the pulses undergoes coherent harmonic focusing before it collides with the fundamental-frequency pulse. Imposing a quantitative criterion to discern the signal photons from the background of the driving laser photons as well as accounting for the finite purity of polarization filtering, the discernible signal photons are found to arise from manifestly inelastic interaction processes of the driving laser fields. Our results suggest that coherent harmonic focusing offers a promising new route to the first detection of signatures of quantum vacuum nonlinearities at the high-intensity frontier in the laboratory.

Introduction The quantum vacuum has remarkable properties. It is not trivial and inert, but amounts to a complex state whose properties are fully determined by quantum fluctuations. As these fluctuations comprise all existing particles, the quantum vacuum in principle even constitutes a portal to new physics beyond the Standard Model of particle physics. In order to obtain a measurable response, the quantum vacuum has to be probed by some external stimulus. A powerful means is provided by strong macroscopic electromagnetic fields which couple directly to the charged particle sector. Within the Standard Model, the leading effect arises from the effective coupling of the prescribed electric \( \vec{E} \) and magnetic \( \vec{B} \) fields via a virtual electron-positron pair. This process is governed by quantum electrodynamics (QED) and supplements Maxwell’s classical equations in vacuum with effective nonlinear couplings of the electromagnetic fields \( [1][4] \). For reviews emphasizing various theoretical aspects as well as prospects for the experimental detection of such effects, see \[5][13]\.

Up to now, deviations from Maxwell’s linear theory of electromagnetism in vacuo have never been directly observed for macroscopically controlled fields. This is because the effective self-interactions of the electromagnetic field are parametrically suppressed by powers of \( |\vec{E}|/E_{cr} \) and \( |\vec{B}|/B_{cr} \). Here, \( E_{cr} = \frac{m_e^2 c^3}{(e h)} \approx 1.3 \times 10^{18} \text{ V/m} \) and \( B_{cr} = E_{cr}/c \approx 4 \times 10^{9} \text{ T} \) are the critical electric and magnetic fields, respectively. The strongest macroscopic electromagnetic fields available in the laboratory are delivered by high-intensity laser systems reaching peak fields \( E \approx \mathcal{O}(10^{14}) \text{ V/m} \) and \( B \approx \mathcal{O}(10^{9}) \text{ T} \). While these fields clearly fulfill \( |\vec{E}| \ll E_{cr} \) and \( |\vec{B}| \ll B_{cr} \), they appear to be sufficient to facilitate a first detection of signatures of QED vacuum nonlinearities in macroscopic fields in a dedicated discovery experiment in the laboratory. The basic idea is to collide high-intensity laser pulses and to look for vacuum-fluctuation-induced modifications of their directional, spectral and polarization properties. These modifications are conveniently resolved in terms of signal photons. The most promising signature in experiment is provided by signal photons whose kinematics or polarization properties differ from the laser photons driving the effect, thereby allowing for a clear signal-to-background separation. For recent estimates of the prospective numbers of signal photons attainable in high-intensity laser pulse collisions, cf., e.g., \[10][26\]. The smallness of the signal in comparison to the huge number of laser photons makes its detection challenging. This is even true for the most advanced high-intensity laser facilities coming online now, such as CILEX \[27\], CoReLS \[28\], ELI \[29\] and SG-II \[30\].

In this letter, we show that the number of attainable and, in particular, discernible signal photons can be increased significantly for a given laser pulse energy put into the interaction volume. To this end, we rely on the mechanism of coherent harmonic focusing (CHF), pioneered by Refs. \[31][32\]. Our quantitative analysis relies on the novel numerical approach \[25\] allowing for first-principles simulations of photonic signatures of vacuum nonlinearities. We also provide analytical estimates based on a description of the driving laser fields as pulsed paraxial beams; cf. Ref. \[33\].

References \[31][32\] demonstrated that CHF can pave the way towards extreme intensities. They showed that the reflection of a relativistically intense laser pulse from the oscillating boundary of an overdense plasma produces a harmonic spectrum with the spectrum intensity scaling as \( I_n \sim n^{-5/2} \), where \( n \geq 1 \) labels the \( n \)th harmonic \[31\]. These harmonics can be focused coherently down to a spot size of about \( \lambda/n \) using a concave plasma surface of appropriate curvature, where the wavelength of the initial pulse is given by \( \lambda \) \[32\]. While an improved description
of the process resulted in a slight revision of the power as \(5/2 \to 8/3\) \[31\], in this letter we stick to the original prediction of \[31\].

As a concrete example, we employ CHF to boost photonic signatures of QED vacuum nonlinearity in the head-on collision of two high-intensity laser fields of given parameters. For definiteness, we assume the initial laser pulses to agree in both wavelength \(\lambda\) and pulse duration \(\tau\). One comprises an energy \(W\) and is focused to a beam waist of \(w_0 = \lambda\). The other is reflected at a concave overdense plasma surface, effectively partitioning the laser pulse energy – which after the reflection process is also assumed to be given by \(W\) – as \(W = \sum_{n=1}^{n_{\text{max}}} W_n\) into the individual harmonics. Here, \(W_n = W n^{-5/2}/H_{n_{\text{max}}^{5/2}}\) is the energy put into the \(n\)th harmonic, \(n_{\text{max}}\) is the harmonic cutoff and \(H_{n_{\text{max}}^{5/2}} = \sum_{n=1}^{n_{\text{max}}} 1/n^q\) is a generalized harmonic number. The plasma surface focuses the \(n\)th harmonic to a waist of \(w_{0,n} = \lambda/n\), such that the electric peak field amplitude of the \(n\)th harmonic scales as \(E_{0,n} \sim \sqrt{W_n/\pi w_{0,n}^2} \sim n^{-1/4}\). This CHF pulse collides head-on with the fundamental-frequency pulse at zero impact parameter and temporal offset, i.e., both pulses are focused to the same point and reach their peak amplitude at the same time.

**Formalism**  The amplitude for emission of a single signal photon with wave vector \(\vec{k}\) and polarization \(p\) from the electromagnetized QED vacuum reads \[33\]

\[
S_{(p)}(\vec{k}) = \langle \gamma_p(\vec{k})|\Gamma_{\text{int}}[A(x),a(x)]|0\rangle. 
\]

(1)

Here, \(|\gamma_p(\vec{k})\rangle \equiv \hat{a}_{k,p}^\dagger|0\rangle\) denotes the single signal photon state and \(\Gamma_{\text{int}}[A(x),a(x)]\) encodes the vacuum-fluctuation-mediated interactions of the operator-valued signal photon field \(a(x)\) \[33\] with the driving macroscopic electromagnetic field \(A(x); \quad F^{\mu \nu} = \partial^\mu A^\nu - \partial^\nu A^\mu\). The latter is treated as a classical background \[36\]. For driving fields of frequencies \(\omega \ll \frac{m^2}{\hbar}\), these effective interactions are governed by the one-loop Heisenberg-Euler effective Lagrangian \(\mathcal{L}_{\text{HE}}^{1\text{-loop}}\) \[2\], implying

\[
\Gamma_{\text{int}}[A(x),a(x)] \simeq \int d^4x \, \hat{a}_{k,p}^\dagger (x) \, j_{\mu}(x),
\]

(2)

where \(j_{\mu}(x) = 2 \partial^\nu \frac{\partial \mathcal{L}_{\text{HE}}^{1\text{-loop}}}{\partial \partial A^\nu}\) sources the signal photons. The above validity criterion is met for present and near-future high-intensity lasers of optical to x-ray frequencies.

In the Heaviside-Lorentz System and units \(c = \hbar = 1\) adopted throughout this letter, the leading contribution of \(\mathcal{L}_{\text{HE}}^{1\text{-loop}}\) in the weak-field limit reads \[11\] \[2\]

\[
\mathcal{L}_{\text{HE}}^{1\text{-loop}} \simeq \frac{m_e^2}{8\pi} \frac{1}{45} \left( \frac{e}{m_e} \right)^4 [B^2 - \overline{E}^2]^2 + 7(B \cdot \overline{E})^2].
\]

(3)

Equation \(2\) is valid for \(|\vec{E}| \ll E_{\text{cr}}\) and \(|\vec{B}| \ll B_{\text{cr}}\) and should allow for the reliable study of all-optical signatures of QED vacuum nonlinearity driven by high-intensity lasers with an accuracy on the one percent level \[25\].

Upon insertion of Eq. \(2\) into Eq. \(1\), the signal photon emission amplitude can be expressed as

\[
S_{(p)}(\vec{k}) = \frac{e_{\mu}^{(p)}(\vec{k})}{\sqrt{2k_0}} \int d^4x \, e^{ikx} \, j_{\mu}(x) \bigg|_{k_0 = |\vec{k}|},
\]

(4)

where \(e_{\mu}^{(p)}(\vec{k}) = (0, \vec{e}_{(p)}(\vec{k}))\), fulfilling \(\vec{k} \cdot \vec{e}_{(p)}(\vec{k}) = 0\), is the polarization vector of the signal photon state \(|\gamma_p(\vec{k})\rangle\).

The unit vectors perpendicular to \(\vec{k} = k\hat{z}\), with \(\vec{e}_{\perp}(\vec{k}) = (\cos \varphi \sin \vartheta, \sin \varphi \sin \vartheta, \cos \vartheta)\) and \(k \geq 0\), can be parameterized by an angle \(\beta\),

\[
\vec{e}_\perp(\beta) = e_1(\vec{k}) \cos \beta + e_2(\vec{k}) \sin \beta,
\]

(5)

where \(e_1(\vec{k}) = \vec{k}_1, e_{\perp}(\vec{k}) = \vec{k}_2, \vec{e}_2(\vec{k}) = \vec{k}_3\). We use Eq. \(4\) to define two linearly independent vectors \(\vec{e}_{(p)}(\vec{k}) = e_{(p)}(\vec{k})\), with \(\beta_p = \beta_0 + \frac{\pi}{2}(p-1), \quad p \in \{1,2\}\), and a suitably chosen \(\beta_0\), to span the transverse polarizations of signal photons of wave vector \(\vec{k}\).

With these definitions, Eq. \(4\) yields

\[
S_{(p)}(\vec{k}) = i \sqrt{\frac{k}{2}} \int d^4x \, e^{i(k \cdot x - k t)} \times [e_{\perp}(\beta_p) \cdot \vec{B} - e_{\perp}(\beta_p + \frac{\pi}{2}) \cdot \vec{M}].
\]

(6)

where the polarization \(\vec{B}\) and magnetization \(\vec{M}\) of the quantum vacuum are defined as \[37\]

\[
\vec{B} = \frac{\partial \mathcal{L}_{\text{HE}}^{1\text{-loop}}}{\partial E} \quad \text{and} \quad \vec{M} = -\frac{\partial \mathcal{L}_{\text{HE}}^{1\text{-loop}}}{\partial B}.
\]

(7)

The differential number of signal photons of polarization \(p\) is related to the modulus square of Eq. \(6\) and reads

\[
d^3N_{(p)}(\vec{k}) = \frac{d^3k}{(2\pi)^3} |S_{(p)}(\vec{k})|^2.
\]

(8)

For a polarization insensitive measurement we have

\[
d^3N = \sum_{p=1}^2 d^3N_{(p)}.
\]

**Field configuration**  To describe the electromagnetic fields of a focused laser pulse (wavelength \(\lambda\), energy \(W\), duration \(\tau\), waist \(w_0 = \lambda\)), we employ the spectral pulse model \[38\], detailed in Sec. III D of Ref. \[25\]. These fields fulfill Maxwell’s equations in vacuum exactly and are conveniently represented in terms of a complex vector potential in radiation gauge,

\[
\vec{A}(x) = \int \frac{d^3k}{(2\pi)^3} e^{i(k \cdot x - k t)} \sum_{q=1}^2 \hat{e}_{(q)}(\vec{k}) a_q(\vec{k}),
\]

(9)

with spectral amplitudes \(a_q(\vec{k})\) encoding the spatio-temporal field structure. The associated real-valued electric and magnetic fields are given by \(\vec{E}(x) = \Re\{-\partial_t \vec{A}(x)\}\)
and $\vec{B}(x) = \mathfrak{R}(\nabla \times \vec{A}(x))$. For a laser pulse which propagates in $\pm \vec{k}$ direction, and is polarized along $\vec{e}_\pm$ in the focus at $\vec{x} = 0$, where the peak field is reached at $t = 0$, the spectral amplitudes are given by

$$a_q(\vec{k}) \rightarrow a_q^+(\vec{k}) = \pm \frac{(2\pi)^3}{ik} \vec{e}_\pm \cdot \vec{e}_q(\vec{k}) \Theta(\pm k_\parallel) \frac{k_\parallel}{k} \sqrt{W_T \lambda e^{-(\frac{x}{\lambda})^2} - (\frac{x}{\lambda})^2 [(k - \omega(\lambda))^2]}.$$  

(10)

Here, $\Theta(.)$ denotes the Heaviside function, $\omega(\lambda) = \frac{2\pi}{\lambda}$ the laser photon energy, $k_\parallel = \vec{k} \cdot \vec{n}$ the momentum component along $\vec{n}$, and $k_\perp = \sqrt{k^2 - k_\parallel^2} \geq 0$. The amplitudes have been constructed such that the zeroth-order paraxial Gaussian beam is reproduced for weak focusing and long pulse durations.

To model our scenario of a fundamental-frequency laser pulse (propagation direction $\vec{n}$, polarization $\vec{e}_+$ in the focus) colliding head-on with a CHF pulse (polarization $\vec{e}_-$ in the focus) containing $n_{\text{max}}$ harmonics, we choose the spectral amplitudes in Eq. (9) as

$$a_q(\vec{k}) \rightarrow a_q^+(\vec{k}) + \sum_{n=1}^{n_{\text{max}}} \sqrt{W_n/W} a_q^-(\vec{k}) |_{\lambda \rightarrow \frac{\lambda}{n}}.$$  

(11)

Subsequently, we refer to the laser pulse propagating in $\pm \vec{k}$ direction as “$\pm$” pulse; the “$+$” (“$-$”) pulse is the fundamental-frequency (CHF) pulse.

Instead, it is also determined by the wavelength and given by $\tau_{CHF} \approx \lambda/n_{max}$ [32]; cf. Fig. 1.

Resorting to a paraxial beam model, the far-field angular decay of the driving laser photons $N^\pm(n)$ in the $n$th mode is approximately given by

$$\frac{dN^\pm(n)}{d\vartheta^\pm} \approx 4\pi^2 W_n n_\omega e^{-2(\pi \vartheta^\pm)^2}.$$  

(12)

Here, $\vartheta^\pm$ is the polar angle measured relative to the beam axis $\pm \vec{n}$. For the fundamental-frequency “+” pulse we have $n = 1$. On the other hand, the “$-$” pulse consists of $n_{\text{max}}$ modes, i.e., $dN^- = \sum_{n=1}^{n_{\text{max}}} dN^-(n)$. The associated total numbers of driving laser photons are $N^+ \approx W_\omega$ and $N^- \approx N^+ H_{n_{\text{max}}}^{(7/2)}/H_{n_{\text{max}}}^{(5/2)} \leq N^+$. All modes are diffraction limited and thus characterized by the same radial beam divergence $\theta = \frac{\pi}{\omega}$.

Results

In the remainder, we use the following parameters: $\lambda = 800$ nm, $\tau = 5$ fs and $W = 25$ J. Both pulses are linearly polarized; the angle between their polarization vectors in the focus $\phi = \angle(\vec{e}_+, \vec{e}_-)$ is kept as a free parameter. The value of $\tau = 5$ fs is chosen mainly for numerical convenience, as it allows us to scale $n_{\text{max}}$ up to 12. Such small pulse durations have so far been achieved only at sub-Joule pulse energies [40]; state-of-the-art high-intensity laser pulses of tens of Joules featuring pulse durations $\gtrsim 20$ fs [29]. We have explicitly confirmed for $n_{\text{max}} = 6$ and ELI-NP [29] laser parameters ($\lambda = 800$ nm, $\tau = 20$ fs, $W = 200$ J) that the effects detailed below also persist for longer pulse durations; cf. the Supplementary Material.

As will be demonstrated below, to a very good approximation the signal photons $N^\pm(\varphi)$ emitted into the “$+$” half-space, characterized by wave vectors $\vec{k}$ fulfilling $\pm \vec{k} \cdot \vec{\hat{n}} > 0$, can be interpreted as arising from the “$+$” pulse and being quasi-elastically scattered off the “$+$” pulse; cf. also Ref. [25]. Manifestly inelastic scattering processes characterized by an energy transfer of order $\omega$ are generically suppressed in comparison to the elastic contributions [32, 33, 34]. The study of photon scattering in the head-on collision of two linearly polarized paraxial beams [22] then suggests that an angle of $|\phi| = \frac{\pi}{2}$ between the polarization vectors $\vec{e}_\pm$ maximizes the signal photon number $N$ attainable in a polarization insensitive measurement. By contrast, the number $N_\perp$ of signal photons scattered into a perpendicularly polarized ($\perp$) mode is expected to become maximum for an angle of $|\phi| = \frac{\pi}{2} \mod \tau$. We have explicitly confirmed this behavior in our simulations (see Fig. 4 in the Supplementary Material) and stick to these optimal choices of the relative polarization alignments when providing results for $N$ and $N_\perp$ in the remainder.

Aiming at the analysis of the $\perp$ signal photons emitted into the “$+$” half-space, we ensure from the outset that the polarization basis is chosen such that $\vec{e}_{(1)}(\vec{k}) \cdot \vec{e}_\pm = 0$, respectively. This can be achieved by adjusting $\beta_0$.

![FIG. 1. Characteristics of the CHF pulse with total energy $W = 25$ J, envelope $\tau = 5$ fs and fundamental wavelength $\lambda = 800$ nm.](image-url)
accordingly as a function of $k$. In turn, $c_{(1)}(\hat{k})$ spans the $\perp$ mode and $N_{\perp}^{+} = 0$. The signal photons of polarization vector $\hat{e}_{(1)}(\hat{k})$ emitted into the $\perp$ half-space constitute $N_{\perp}^{+}$.

Figure 2 depicts the attainable numbers of signal photons $N^{+}$ and $N_{\perp}^{+}$ as a function of $n_{\max}$. Assuming the effective waist, pulse duration and Rayleigh range of the CHF pulse made up of $n_{\max}$ harmonics to scale as $w_{\text{CHF}} = w_{0}/n_{\max}$ and $\tau_{\text{CHF}} \sim z_{R\text{,CHF}} \sim 1/n_{\max}$, the simulation results are remarkably well described by

$$N^{+}_{(p)}(n_{\max}) = c^{+}_{(p)} n_{\max} \frac{1}{(1 + 2n_{\max}^{2})^{\frac{H_{n_{\max}}(1/4)}{H_{n_{\max}}(5/2)}}},$$

with polarization dependent numerical constants $c^{+}_{(p)}$. This expression only accounts for quasi-elastically scattered signal photons, and follows from Eqs. (7) and (10) of Ref. [32] upon identification of the probe with the fundamental frequency beam and the pump with the CHF pulse of duration $\tau_{\text{CHF}} \ll \tau$. The CHF peak field energy per spot size is determined as $\sqrt{W_{\text{CHF}}/w_{\text{CHF}}^{2}} = \sum_{n=1}^{n_{\max}} \sqrt{W_{n}/w_{0,n}^{2}}$. Equation (13) implies that the CHF process can increase the signal photon numbers $N^{+}_{(p)}$ at most by a factor of $\frac{N^{+}_{(p)}(n_{\max} = 1)}{N^{+}_{(p)}(n_{\max} = 1)} \sim \frac{128}{27\zeta(2)} \approx 2.6$ with respect to the collision of two fundamental-frequency pulses. Asymptotically, the increase of the CHF peak field with $n_{\max}$ is compensated by a decrease of the effective focusing volume.

The ratio of the coefficients $c^{+}$ and $c^{+}_{(p)}$ extracted in Fig. 2 is $c^{+}/c^{+}_{(p)} \approx 23.0$, and thus roughly agrees with that found for counter-propagating paraxial beams $c^{+}/c^{+}_{\perp} = \frac{27\zeta(2)}{2\pi} \approx 21.8$ [12]. The accurate description of $N^{+}_{(p)}$ by Eq. (13) suggests that Eq. (4) of Ref. [33] can serve as an analytical estimate for the angular decay of $N^{+}_{(p)}$. With the above assumptions, we obtain

$$\frac{dN^{+}_{(p)}(n_{\max}, n_{\max})}{d\vartheta^{+} d\vartheta^{+}} \approx \frac{4\pi^{2} c^{+}_{(p)}}{n_{\max}(1 + 2n_{\max}^{2})^{\frac{H_{n_{\max}}(1/4)}{H_{n_{\max}}(5/2)}}} \times \frac{1 + 2n_{\max}^{2}}{1 + 8n_{\max}^{2}} e^{-\frac{2\pi(\vartheta^{+})^{2} + 2\pi n_{\max}^{2}}{1 + 8n_{\max}^{2}},}$$

which is fully characterized by $c^{+}_{(p)}$ and $n_{\max}$. This provides us with an estimate for the radial divergence of the signal photons emitted into the $\perp$ half-space,

$$\theta^{+}_{\text{sig}}(n_{\max}) \approx \theta \sqrt{\frac{1 + 8n_{\max}^{2}}{1 + 2n_{\max}^{2}}} \frac{n_{\max} \rightarrow 1}{\rightarrow 2\theta}.$$
The driving laser photons fulfill the criterion \( d^3N_\perp/d\omega dk > \mathcal{P} d^3N/d\omega dk \) and the situation is different. In this case, the number of discernible signal photons is substantially reduced in comparison to \( N_{\text{ideal}} \), even for an ambitious polarization purity as small as \( \mathcal{P} = 10^{-10} \), yielding \( N_{\perp, \text{dis}} \approx 10.44 \) perpendicularly polarized discernible signal photons at \( \approx 2\omega \). The corresponding results qualitatively agree with those depicted in Fig. 3 upon identifying \( \mathcal{N} \to \mathcal{P} \mathcal{N} \) and \( N_{\text{dis}} \to N_{\perp, \text{dis}} \).

Finally, it is instructive to compare the above CHF results with the numbers of discernible signal photons attainable in the collision of two fundamental-frequency pulses of the same energy \( (n_{\text{max}} = 1) \), yielding \( N_{\perp, \text{dis}} \approx 2.15 \times 10^{-6} \), \( N_{\text{ideal}} \approx 57.93 \) and \( N_{\text{real}} \approx 6.59 \times 10^{-3} \). In particular the results for \( N_{\text{dis}} \) and \( N_{\perp, \text{dis}} \) underpin the substantial enhancement of several orders of magnitude in the number of discernible signal photons relevant for experiment achieved by CHF.

### Conclusions

We have demonstrated in an idealized setup that coherent harmonic focusing can substantially increase the number of discernible signal photons in the collision of high-intensity laser pulses for a given energy put into the interaction volume. We are confident that our findings will pave the way for many further theoretical ideas and proposals as well as dedicated experimental campaigns aiming at the first verification of quantum vacuum nonlinearity using coherent harmonic focusing and replications based on conventional higher-harmonic generation techniques.

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SUPPLEMENTARY MATERIAL

Here we provide some additional material underpinning the arguments given in the main text. Figure 4 confirms that the total number of signal photons attainable in a polarization insensitive measurement \( N \) and the number of signal photons scattered into a perpendicularly polarized mode \( N_\perp \) reach their maxima for different choices of the relative angle between the polarization vectors of the driving laser pulses in the focus \( \phi = \angle (\vec{e}_+, \vec{e}_-) \). As detailed in the main text, considerations based on an analysis of the head-on collision of two paraxial laser fields predict \( N \) (\( N_\perp \)) to be at a maximum for \( |\phi| = \frac{\pi}{4} \) (\( |\phi| = \frac{\pi}{2} \mod \pi \)). In the angle interval \( \phi \in [0^\circ \ldots 90^\circ] \) considered in Fig. 4, the corresponding angle is \( \phi = 90^\circ \) (\( \phi = 45^\circ \)) for \( N \) (\( N_\perp \)). Our simulation data presented in Fig. 4 are perfectly compatible with these predictions.

![Graphs showing numbers of signal photons](image)

**FIG. 4.** Numbers of signal photons \( N \) attainable in a polarization insensitive measurement (left) and numbers of signal photons \( N_\perp \) scattered into a perpendicularly polarized mode (right) for different values of \( n_{\text{max}} \) as a function of the relative angle between the polarization vectors of the driving laser pulses in the focus \( \phi = \angle (\vec{e}_+, \vec{e}_-) \). The panels in the upper (lower) line show results for signal photons emitted into the “−” (“+”) direction. The number of attainable signal photons \( N \) (\( N_\perp \)) is at a maximum for an relative angle of \( \phi = 90^\circ \) (\( \phi = 45^\circ \)) between the polarization vectors \( \vec{e}_\pm \). The lines are least squares fits of the functions \( N^\pm (\phi) = A(133 - 60 \cos(2\phi)) \) and \( N^\pm_\perp (\phi) = B(133 - 60 \cos(2\phi)) + C \sin^2(2\phi) \), with fitting coefficients \( A \), \( B \) and \( C \), to the simulation data points (filled circles). The first fitting function is modeled after the analytical result for \( N(\phi) \) in the head-on collision of two paraxial beams [42]. The latter amounts to a combination of the former and the corresponding paraxial result for \( N_\perp (\phi) \) [42]. For non-paraxial beams all signal photons \( N \) generically exhibit a non-vanishing overlap with the \( \perp \) mode \( N_\perp \), motivating this combination. This contribution is also essential in accounting for the asymmetry of the simulation data for \( \phi \approx \frac{\pi}{2} \mod \pi \).

Figure 5 exemplifies the angular decay of the signal photons as inferred from our simulation in comparison to the respective analytical estimates [14] and [16]. The corresponding curves are in good agreement.

Finally, in Fig. 6 we depict the spectra of the driving laser photons \( \mathcal{N} \) and signal photons attainable in a polarization insensitive measurement \( N \) for the same scenario as discussed in the main text, but assuming ELI-NP [29] parameters (\( \lambda = 800 \) nm, \( \tau = 20 \) fs, \( W = 200 \) J) for the driving laser fields and \( n_{\text{max}} = 6 \). This results in \( N_{\text{dis}} \approx 314 \) discernible signal photons attainable in a polarization insensitive measurement. Figure 6 is qualitatively very similar to Fig. 3 in the main body of this letter. However, in Fig. 6 the frequency spread of both the various modes constituting the driving laser pulses and the signal photons is substantially smaller. The reason for this is the larger pulse envelope for the ELI-NP scenario, which is a factor of 4 larger than the one considered in the main body of this letter.
FIG. 5. Angular decay of the signal photons emitted in “−” (left plot) and “+” (right plot) directions. More specifically, here we compare numerical simulation data for the angular decay of $N^+(n_{\text{max}} = 12)$ and $N^-(n_{\text{max}} = 12, n = 5)$ with the respective analytical estimates \[14\] and \[16\]. The radial divergences extracted by fitting Gaussian curves to the simulation data, $\theta_{\text{sig}}(n = 5) \simeq \theta$ and $\theta_{\text{sig}}(n_{\text{max}} = 12) \simeq 20\theta$, are in good agreement with the analytical estimates \[15\] and \[17\].

FIG. 6. Spectra of the driving laser photons $\mathcal{N}$ and signal photons attainable in a polarization insensitive measurement $N$ for ELI-NP \[29\] laser parameters ($\lambda = 800$ nm, $\tau = 20$ fs, $W = 200$ J) and $n_{\text{max}} = 6$. The white dashed circles indicate lines of constant photon energy $k = n\omega$ with $n \in \mathbb{N}$. Note that different color scales are used in the top, middle and bottom panels. For comparison, we also depict analytical estimates for the radial divergences: in the top panel $\theta = 1/\pi$ is the radial divergence of a diffraction limited Gaussian beam. The radial divergences highlighted in the middle panel are determined from Eqs. \[15\] and \[17\]. The bottom panels focus on the spectral domain where the differential number of signal photons surpasses the differential number of driving laser photons. Here, we confront the spectrum of the driving laser photons (left) with the filtered signal photon spectrum fulfilling the criterion $d^3N/d^3k > d^3\mathcal{N}/d^3k$ (right) adopting the same linear color scale. Integrating the latter, we obtain $N_{\text{dis}} \approx 314$ discernible signal photons per shot at $\approx 2\omega$. 