Photon–photon scattering in collisions of intense laser pulses

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Abstract. A scenario for measuring the predicted processes of vacuum elastic photon–photon scattering and four-wave mixing with intense modern lasers is investigated. The numbers of measurable scattered photons are calculated for the collision of two, Gaussian-focused, pulsed lasers. It is shown that a single intense 10 PW optical laser beam split into two counter-propagating pulses is sufficient for measuring the elastic process. Moreover, when these pulses are sub-cycle, by also considering the collision of two sech pulses, the results suggest that the frequency-shifting, four-wave mixing process should be measurable too.
Quantum electrodynamics (QED) is commonly regarded as a fantastically successful theory whose accuracy has been tested to one part in $10^{12}$ for free electrons [1] and one part in $10^9$ for bound electrons [2]. However, among its several predictions that are yet to be confirmed is the nature of electromagnetic interaction with the quantized vacuum. Already with the pioneering work of Sauter [3] and Heisenberg and Euler [4], it was clear that quantum mechanics predicts how particles traversing the classically empty space of the vacuum can interfere with ephemeral ‘virtual’ quantum states, whose lifetimes are of durations permitted by the uncertainty relation. Virtual electron–positron pairs can, in principle, be polarized by an external electromagnetic field, thus introducing nonlinearities into Maxwell’s equations, which break the familiar principle of superposition of electromagnetic waves in vacuum. Photons from multiple, vacuum-polarizing sources can then become coupled at the common point of interaction of polarized virtual pairs. One of the earliest mentions of the possibility for photon–photon scattering is that by Halpern [5] with the low-frequency limit calculated shortly thereafter by Euler and Kockel [6]. This process can be considered in terms of individual scattering events, such as the event of two photons scattering off a virtual pair [7] (a recent proposal for measuring this is given in [8]), or in the present case, in the low-frequency limit, in terms of classical fields modified to include quantum effects. The typical scale for such ‘refractive’ vacuum polarization effects, where no pair-creation takes place, was found to be given by the critical field strength required to ionize a virtual electron–positron pair, namely the pair-creation scale of $E_{cr} = m^2 c^3 / e\hbar = 1.3 \times 10^{16}$ V cm$^{-1}$ or an equivalent critical intensity of $I_{cr} = 2.3 \times 10^{29}$ W cm$^{-2}$, where $m$ and $e < 0$ are the mass and charge of an electron, respectively (for a review of strong-field QED, see, e.g., [9, 10]). With recent technological breakthroughs, the interest in measuring processes at these high intensities has been renewed. Indeed, photon–photon scattering in intense fields is predicted to manifest itself in a variety of
ways such as in a phase shift in intense laser beams crossing one another [11], in a frequency shift of a photon propagating in an intense laser [12], in polarization effects in crossing lasers such as vacuum birefringence and dichroism [13–16], in dispersion effects such as vacuum diffraction [17–19] and also in vacuum high harmonic generation [20]. Current experimental limits for photon scattering [21] and birefringence [22] are yet to reach those required by QED. Although the required intensities lie some seven orders of magnitude above the record high produced by a laser [23], recent progress at facilities such as the ongoing 10 PW upgrade to the Vulcan laser [24], as well as proposals for next-generation lasers HiPER and ELI aiming at three to four orders of magnitude less than critical, will put the experimental verification of these long-predicted nonlinear vacuum polarization effects finally within reach. This therefore motivates more realistic quantitative predictions.

In this paper, we focus on the phenomenon of photon–photon scattering in intense focused laser beams. We call this scattering ‘elastic’ when the spectrum of the diffracted probe pulse is equal to the incident spectrum and ‘inelastic’ when the spectrum of the diffracted probe pulse is different. Inelastic scattering will correspond to a shifting up or down of the frequencies of one pulse in multiples of frequencies of the other, more commonly known as ‘four-wave mixing’. When all external fields have the same frequency, four-wave mixing is then equivalent to lowest order vacuum high-harmonic generation. As an elastic process, the numbers of scattered photons have been calculated in the passage of one monochromatic Gaussian laser beam through another [25], as well as in the so-called single- and ‘double-slit’ setups [13, 17], where a probe Gaussian beam meets two other intense ones. Inelastic photon–photon scattering has been investigated theoretically as a four-wave mixing process using TE$_{10}$ and TE$_{01}$ modes in a superconducting cavity [26], in the collision of three, perpendicular, plane waves [27] and as generating odd harmonics involving a single, spatially focused monochromatic wave [28]. By incorporating both the pulsed and spatially focused nature of modern high-intensity laser beams, we perform a more accurate calculation of the signal of the elastic scattering process. We thereby investigate the robustness of the effect with a more detailed calculation than hitherto performed, including dependence on beam collision angle, impact parameter (lateral beam separation), longitudinal phase difference (through lag) and pulse duration (finite beam length). Inclusion of four-wave mixing terms with a pulsed setup allows us, moreover, to determine the possibility of measuring inelastic photon–photon scattering when a single 10 PW beam is split into two counter-propagating sub-cycle pulses. In what follows, we work in Gaussian cgs units (fine-structure constant $\alpha = e^2$), with $\hbar = c = 4\pi e_0 = 1$, unless explicit units denote otherwise.

### 2. The scenario considered

In order to analyse the collision of two laser pulses, several collision parameters have been included. The envisaged scenario is shown in figure 1, in addition to which lateral and temporal centring and carrier envelope phase appear in the analytical setup. Spatial focusing and temporal pulse shape are present in taking the leading order spatial and temporal terms of the Gaussian beam solution to Maxwell’s equations (see, e.g., [29]). These approximations neglect terms of the order $O(w_{c,0}/y_{c})$ and $O(1/\omega_c \tau_c)$, respectively, where $c \in \{a, b\}$ is used throughout to denote a variable for the two beams a and b, the minimum beam waist is $w_{c,0}$, Rayleigh length $y_{c} = \omega_w^2/2$, beam frequency $\omega_c$ and full-width at half-maximum pulse duration $\tau_{FWHM}$, related to $\tau$ via $\tau \sqrt{2 \ln 2} = \tau_{FWHM}$. The condition $\omega_c \tau_c \gg 1$ limits the minimum pulse duration that can be consistently considered in our analysis. For the electric fields of the two beams
Figure 1. The envisaged experimental setup. \( \tau_{a,b} \) refer to the pulse durations in the Gaussian beam envelopes \( e^{-(u \pm y)^2/\tau_{a,b}^2} \), the \( E_b \) pulse is displaced from the \( y \)-axis by the coordinates \( x_0, z_0 \), both pulses have, in general, a carrier-envelope phase and the \( E_a \) pulse lags behind \( E_b \) by \( \Delta t \). The \( E_b \) field is incident on the detector.

We focus on the phenomenon of diffraction and specifically the detection of photons whose wavevectors differ significantly, either in orientation or in magnitude from those of the background lasers. As such, we envisage an array of photosensitive detectors being placed some

\[
E_a(x, y, z, t) = \tilde{\varepsilon}_a e^{\frac{-\tilde{x}_a^2}{w_{a,0}^2}} \sin \left[ \psi_a + \omega_a(t - \Delta t + \tilde{y}) - \eta_a(\tilde{y}) \right] f_a(t - \Delta t + \tilde{y}),
\]

\[
E_b(x, y, z, t) = \tilde{\varepsilon}_b e^{\frac{-\tilde{x}_b^2}{w_{b,0}^2}} \sin \left[ \psi_b + \omega_b(t - y) + \eta_b(y) \right] f_b(t - y),
\]

\[
\eta_c(y) = \tan^{-1} \left( \frac{y}{y_{r,c}} \right) - \frac{\omega_c y}{2 y^2 + y_{r,c}^2},
\]

where the coordinates \((\tilde{x}, \tilde{y}, \tilde{z})\) are the same as \((x, y, z)\) rotated anti-clockwise around the \( x \)-axis by an angle \( \theta \), with the polarization \( \tilde{\varepsilon}_a \) being similarly rotated so that \( \mathbf{k}_c \cdot \tilde{\varepsilon}_a = \mathbf{K}_c \cdot \tilde{\varepsilon}_a = 0 \) and \( |\tilde{\varepsilon}_c| = |\tilde{\varepsilon}_c| = 1 \), where \( \mathbf{k}_c \) is the beam wavevector, \( f_c \) describes the pulse shape with \( f_c(x) = e^{-(x/\tau_c)^2} \) being used, \( w_c \) is the beam waist \( w_c^2 = w_{c,0}^2(1 + (y/y_{r,c})^2) \) dependent on the transverse coordinate, \( \psi_c \) is a constant phase, \( \Delta t \) is the lag and \( \mathcal{E} \) is the field amplitude, which is defined by \( \int dt \int dx \int dz |E(x, y = 0, z, t)|^2/4\pi \) equalling the total beam energy or \( \mathcal{E}_c = 2\sqrt{2P_{c,0}/w_{c,0}} \), and \( P_{c,0} \) is the beam power, where we have already assumed that corrections to transversality \( \mathbf{k} \wedge \mathbf{E} = \mathbf{B} \) can be neglected, being as they are of the same order as neglected higher-order terms in the spatial Gaussian beam solution to Maxwell’s equations.

We focus on the phenomenon of diffraction and specifically the detection of photons whose wavevectors differ significantly, either in orientation or in magnitude from those of the background lasers. As such, we envisage an array of photosensitive detectors being placed some
distance away from the collision, \(y\), along the positive \(y\)-axis, much larger than the interaction volume. \(E_b\) is then incident on this detector.

### 3. Derivation of scattered field

When external electromagnetic fields that polarize the vacuum comprise photons with energies much less than the electron mass \((\omega \ll m)\), their evolution can be well approximated by an effective description in which the vacuum fermion dynamics has been integrated out and only photon degrees of freedom remain. The Euler–Heisenberg Lagrangian [4] is an effective Lagrangian which includes such fermion dynamics to one-loop order. When the field strength is much less than critical \((E \ll E_{cr})\), the Euler–Heisenberg Lagrangian can be well approximated by its weak-field expansion, which, neglecting derivative terms, is

\[
\mathcal{L} = \frac{1}{8\pi} (E^2 - B^2) + \frac{1}{360\pi^2 E_{cr}^2} [(E^2 - B^2)^2 + 7(E \cdot B)^2] + \frac{1}{E_{cr}^4} O((E^2 - B^2)^3) + \frac{1}{E_{cr}^4} O((E \cdot B)^3).
\]  

(4)

The weak-field expansion equation (4) is depicted in figure 2 and can be understood as coupling the flux of electromagnetic fields from different sources with one another. It can also be shown [30] that the leading order expansion of the QED four-photon ‘box diagram’ (see figure 3) in \(\omega/m\) leads to the same Lagrangian as equation (4), allowing one to interpret the nonlinear interaction in terms of photons. Extremizing equation (4) with respect to the vector potential returns the wave equation for \(E\) and \(B\) fields modified by the one-loop, weak-field vacuum current, \(J_{vac}\):

\[
\nabla^2 E - \partial_t^2 E = 4\pi J_{vac},
\]

(5)

\[
J_{vac} = \left[ \nabla \wedge \partial_t \mathbf{M} - \nabla (\nabla \cdot \mathbf{P}) + \partial_t^2 \mathbf{P} \right].
\]

(6)
where \( \mathbf{P} = \frac{2e}{\hbar} \mathbf{E} - \frac{1}{4\pi} \mathbf{E}, \mathbf{M} = \frac{2e}{\hbar} \mathbf{B} + \frac{1}{4\pi} \mathbf{B} \) and

\[
\mathbf{P} = \frac{\alpha^2}{180\pi^2 m^4} \left[ 2(E^2 - B^2) \mathbf{E} + 7(\mathbf{E} \cdot \mathbf{B}) \mathbf{B} \right],
\]

\[
\mathbf{M} = -\frac{\alpha^2}{180\pi^2 m^4} \left[ 2(E^2 - B^2) \mathbf{B} - 7(\mathbf{E} \cdot \mathbf{B}) \mathbf{E} \right].
\]

Using the beam transversality, \( \mathbf{k}_c \land \mathbf{E}_c = \mathbf{B}_c \), \( \mathbf{P} \) and \( \mathbf{M} \) can be written entirely in terms of the electric or magnetic field. One can then write the vacuum polarization as a series \( P_i = \chi^{(1)}_{ij} E_j + \chi^{(3)}_{ijkl} E_j E_k E_l + \cdots + \chi^{(2n+1)}_{ijkl\ldots m} E_j \cdots E_m + \cdots \), where electric susceptibilities \( \chi \) only occur at odd orders due to charge-conjugation symmetry (Furry’s theorem). Therefore, four-, six-, eight-, etc wave mixing can, in principle, occur, although each extra order will be suppressed by a factor \( \alpha(E/E_{cr})^2 \).

An iterative approach can be used to solve equation (5), which, since \( J^{(0)} \propto [\alpha(E^{(0)}/E_{cr})^2]E^{(0)} \) and \( \alpha(E/E_{cr})^2 \ll 1 \), can be understood as perturbative:

\[
E^{(n+1)}(\mathbf{x}, t) = E^{(n)}(\mathbf{x}, t) + \int d^3x' \frac{J^{(n)}(E^{(n)} \ldots , E^{(0)}))_{t=t_{ret}}}{|\mathbf{x} - \mathbf{x}'|},
\]

where \( E^{(n)} \) is the \( n \)th-order perturbative solution of equation (5), \( E^{(0)} \) is the zero-field vacuum solution, obeyed by the Gaussian beams in vacuum, approximated by \( \mathbf{E}_a \) and \( \mathbf{E}_b \), \( J^{(n)} \) is the \( n \)th iteration of the current occurring on the right-hand side of the wave equation, \( (\mathbf{x}, t) \) are the coordinates in the detector plane and \( t_{ret} = t - |\mathbf{x} - \mathbf{x}'| \) is the retarded time. By making the approximation that

\[
E^{(n)}_d(\mathbf{x}, t) = \int d^3x' \frac{J^{(n)}(E^{(n)}(\mathbf{x}'), \ldots , E^{(0)}(\mathbf{x}'))_{t=t_{ret}}}{|\mathbf{x} - \mathbf{x}'|} \ll E^{(0)}(\mathbf{x}, t),
\]

for all \( n \), the resultant electric field can be well approximated by \( E = E^{(1)} = E^{(0)} + E_d, E_d = E^{(0)}_d \), i.e. the zero-field vacuum solution plus the lowest order ‘diffracted field’, \( E_d \).

By substituting \( E^{(0)} = \mathbf{E}_a + \mathbf{E}_b \) into equations (6) and (9), and by enforcing the assumption that the dimensions of the interaction volume are much smaller than the typical detector coordinates, following similar steps to [13, 14, 17], one arrives at

\[
E_d(x, t) = \frac{\alpha^2}{360\pi^2 m^4 r_a^2} \left[ E_a^2 \mathbf{v}_1 \sum_{j=1}^{6} \mathcal{V}_j + E_b^2 \mathbf{v}_2 \sum_{j=7}^{12} \mathcal{V}_j \right],
\]

\[
\mathbf{E}_d(x, \omega) = \frac{\alpha^2 \omega^2 r_a}{360\pi^2 m^4} \left[ E_a^2 \mathbf{v}_1 \sum_{j=1}^{6} \mathcal{V}_j + E_b^2 \mathbf{v}_2 \sum_{j=7}^{12} \mathcal{V}_j \right]
\]

where \( \mathcal{V}_j \) and \( \mathbf{V}_j \) are integrals over the interaction volume, given in equations (A.3) and (A.4), \( r = |x| \) is the detector point distance and \( \mathbf{v}_{1,2} \) are the diffracted field polarization vectors given in equations (A.1) and (A.2). Splitting the plane-wave part of the input fields \( \mathbf{E}_a, \mathbf{E}_b \) into positive and negative frequencies, the 12 terms in equations (11) and (12) are produced, corresponding to the six possible orientations of the currents connected by the effective vertex in figure 2. As the interaction contains terms of the order \( O[(\mathbf{E}_a + \mathbf{E}_b)^3] \), and as the purely cubic terms \( E_{a,b}^3 \) necessarily disappear (both electromagnetic invariants are zero for the individual Gaussian beams, transverse in this approximation), for an incident current of frequency \( \omega_0 \), the resultant
signal can have a frequency $\omega_b$, $\omega_b \pm 2\omega_a$, $2\omega_b \pm \omega_a$, corresponding to the two beams’ elastic and inelastic components, respectively. The diffracted field polarization vectors $\mathbf{v}_{1,2}$ appear as geometrical factors and from their definition in equations (A.1) and (A.2), one can see that on the detector ($y > 0$), the $\omega_a$ and $\omega_a \pm 2\omega_b$ signals from pulse $a$ are strongly suppressed, as would be expected as the $E_a$ pulse travels from the interaction region away from the detector. So, in general, direct back-scattering of photons is suppressed. After a further analytical integration in $x$, the remaining two-dimensional integrals from equations (A.3) and (A.4) were then evaluated numerically in C++, partly using the GSL library [31].

One can interpret the classical field incident on the detector as being composed of a total number of photons $N_i$ by dividing its total energy by the photon energy so that $N_i = \int_{-\infty}^{\infty} d\omega dx dz \tilde{I}_i(\omega, r)/|\omega|$, where the total spectral density $\tilde{I}_i(\omega, r) = |\tilde{E}_i(\omega, r)|^2/8\pi^2 = |\tilde{E}_b(\omega, r) + \tilde{E}_a(\omega, r)|^2/8\pi^2 (\tilde{E}_a(\omega, r)$ is taken to be zero on the detector in the current setup) and where $y$ is taken large enough that the surface perpendicular to the Poynting vector can be well approximated as being flat. We will therefore occasionally refer to the ‘photon’ picture and mean this interpretation. Although the spectral density extends to negative frequencies, it is consistent to interpret the differential number of photons as this divided by the absolute frequency because the total energy is the integration over all frequencies and all energy is carried by positive-frequency photons (see also [32] on this point). We then calculate the number of ‘accessible’ photons that fall on the detector plane, by integrating over the annulus that satisfies $N_i(x, z) - 100 N_b(x, z) > 0$ for $N_i(x, z) = \int_{-\infty}^{\infty} d\omega \tilde{I}_i(\omega, r)/|\omega|$, $i \in \{a, b, d, t\}$.

4. Elastic photon–photon scattering

Current and next-generation high-intensity lasers will typically produce pulses with many optical cycles and so unless some special resonance condition is fulfilled, one would expect the elastic cross-section, where incident and outgoing spectra have the same form, to be the largest. By ‘elastic’, we are therefore referring to terms in $E_d$ with equal incoming and outgoing frequencies, the quantum version being

$$\gamma(\omega_1, k_1, \varepsilon_1) + \gamma(\omega_2, k_2, \varepsilon_2) \rightarrow \gamma(\omega_1', k_1', \varepsilon_1') + \gamma(\omega_2', k_2', \varepsilon_2'),$$

(13)

where $\gamma$ represents a photon, $\omega_i$ its frequency, $k_i$ its wavevector and $\varepsilon_i$ its polarization. Photons scattering in equation (13) can take their parameters from any of the photons in either beam. The interaction between the beams contains terms, in the elastic case, containing $E_d^2 E_b$ for an intense pulse $a$ and probe pulse $b$. The larger the spatial integral of this term can be made and the larger the temporal integral of the mod-square of this spatial integral, the larger the number of diffracted photons. Since pulse $a$ is typically many-cycle, the interaction acquires a constant term over the volume of its square, indicating that the number of elastically diffracted photons should, in general, increase with the spatio-temporal overlap of the beams.

In [17], photon–photon scattering was calculated for the case of two co-propagating focused intense Gaussian beams counter-propagating against another probe, Gaussian beam—the so-called ‘matterless double-slit’ configuration. When the separation between the two intense beams is set to zero (a limit explicitly calculated in [33]), the calculation provides a test of the formulae derived here. When we set $x_0 = \zeta_0 = \Delta t = 0$ and take the limit $\tau_{a,b} \rightarrow \infty$ in equation (11), with $\theta = 0$, we confirm that the analytical expressions agree. Moreover, in [13], a similar double-slit configuration was calculated with the probe beam meeting perpendicularly two intense beams that counter-propagate. Again, the analytical expression of
this is reproduced here when the two intense beams co-propagate, their separation is set to zero and the aforementioned limits in the present setup are taken with $\theta = \pi/2$. As a numerical test of our expressions, we can compare the diffracted field intensities $I_d(t, r) = |\mathbf{E}_d(t, r)|^2/4\pi$ for $I_d(0, r)$ and compare them with the head-on single-slit results of [33], which used the parameters $\lambda_a = 0.8 \mu m$, $\lambda_b = 0.527 \mu m$, $w_{a,0} = 0.8 \mu m$, $w_{b,0} = 290 \mu m$, $P_a = 50$ PW and $P_b = 20$ TW. In order to obtain agreement in the diffracted field intensity between the present values and the literature values, $\tau_{a,b}$ had to be set to around $10^4$ fs, which is unexpectedly large compared to the pulse durations considered in the reference ($\tau_a = 30$ fs, $\tau_b = 100$ fs). We will elaborate on the non-trivial dependence of $I_d$ on pulse duration in appendix B, where it is explained why most of the difference between these photon distributions already disappears for a pulse duration $\tau = 10^3$ fs. When the number of accessible photons was calculated for the present, pulsed system with $\theta = 0.1$ and the same durations as suggested in [33], the number of diffracted photons $N_d$ also fell from the estimated value of around 36 to around 0.4.

The present treatment allows for the two lasers to be equally strong and we consider the more experimentally accessible situation of having a single laser, split into two colliding pulses, both focused to ultra-high intensities. If we use $B_j$ and $\Gamma_j$ to denote the powers of $E_a$ and $E_b$ that occur in the diffracted field equations (11) and (12), then $N_d$ scales with $\varepsilon_a^{2B_j}E_b^{2\Gamma_j}$, and if we keep the power of the laser constant ($E_c = 2\sqrt{2}P_{c,0}/w_{c,0}$), for each term, the optimal division of the total power between the beams is

$$\frac{P_{a,0}}{P_{b,0}} = \frac{B_j}{\Gamma_j}, \quad P_{a,0} = \frac{B_j}{B_j + \Gamma_j} P_{t,0},$$

for total power $P_{t,0} = P_{a,0} + P_{b,0}$. In other words, to optimize for a near on-axis diffraction experiment, as investigated here, if pulse b is the probe, the $E_a^2E_b$ term dominates in the diffracted field, and the optimal amplitude of the signal is reached when we set $P_{b,0} = P_{a,0}/2 = P_{t,0}/3$.

For base parameters similar to that of the Vulcan laser [24], $\lambda_a = \lambda_b = 0.91 \mu m$, $\tau_a = \tau_b = 30$ fs, $P_a = 5$ PW, $P_b = 5$ PW, with $w_{a,0} = 0.91 \mu m$, $w_{b,0} = 100 \mu m$, $\hat{\mathbf{E}}_a = \hat{\mathbf{E}}_b = \hat{\mathbf{x}}$, a summary of the dependence of $N_d$ on several variables is given in figure 4. We will comment on the plots sequentially, in which solid lines represent what one could intuitively expect, as explained in the following. Starting from the right-hand side of the first plot and moving in the direction of falling $w_{b,0}$, we see that $N_d(w_{b,0})$ scales approximately as $\propto w_{b,0}^{-2}$, indicated by the solid line. Since $N_d$ for such a setup is proportional to $E_b^2$ and since this is inversely proportional to the area of focusing, the dependence on $\propto w_{b,0}^{-2}$ is as expected. Deviation occurs when a maximum is reached (see, e.g., [13] for details) beyond which $N_d(w_{b,0})$ falls rapidly as the background from $\mathbf{E}_b$ gradually covers the entire detector, leaving no signal. The dependence on beam separation $N_d(x_0)$ is also intuitive and is seen to have excellent agreement with a Gaussian, normalized in height, with a width of $w_{b,0}/2$ (a fit of $\exp(-2x_{\hat{z}}^2/w_{b,0}^2)$ is plotted in the figure). Simply by integrating the transverse Gaussian distributions of the two beams and then squaring ($N_d \propto |\mathbf{E}_d|^2$), one arrives at this dependence. The third plot of $N_d(\lambda)$ ($\lambda = \lambda_a = \lambda_b$) is a log–log plot where the dependence begins for small $\lambda$ as $N_d \approx \lambda^{-3.5}$ but then for larger values tends to $N_d \approx \lambda^{-3.5}$. This is shown by all the points lying between these two solid lines. Since the power of each beam is inversely proportional to wavelength, and since the $N_d \propto P_{a,0}^2P_{b,0}$, one

3 The field strengths in [17, 33] were calculated using a conservative form of the beam intensity with power per unit area for an area $\pi w_{c,0}^2$, rather than the $\pi w_{c,0}^2/2$ which is manifest from an integration of the intensity of a Gaussian beam over the transverse plane.
Figure 4. Dependence of the number of measurable elastically diffracted photons $N_d$ on various parameters, where parameters held constant take the values $\lambda_a = \lambda_b = 0.91 \mu m, w_{a,0} = 0.91 \mu m, w_{b,0} = 100 \mu m, \tau_a = \tau_b = 30 \text{ fs}, P_a = 5 \text{ PW}, P_b = 5 \text{ PW}, \hat{\varepsilon}_a = \hat{\varepsilon}_b = \hat{x}$. 

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would expect at least a dependence of \( N_d(\lambda) \sim \lambda^{-3} \). In contrast, the dependence of \( N_d(\tau) \) can be straightforwardly derived. For \( \tau_a = \tau_b = \tau \), one notes that when \( \tau \ll w_{b,0}, y_{t,b} \), the interaction volume in the beam propagation direction is governed by the Gaussian pulse shape. Further noting that \( N_d \) essentially involves a double integration on the longitudinal beam coordinate (by taking the mod-squared), as well as an integral over \( t \), the dependence \( N_d(\tau) \propto \tau^3 \) appears, which shows excellent agreement for small \( \tau \) with the full numerical integration, displayed by the log–log plot of \( N_d(\tau) \) in the fourth figure. The larger \( \tau \) is for \( \tau > w_{b,0} \), the more the decay along the beam propagation axis is described by focusing rather than pulse terms. For large enough \( \tau \), \( I_d \) depends only on focusing terms and since the yield \( N_d \) is acquired from an integration over time, we have \( N_d(\tau) \propto \tau \) and this transition can be seen by the second, linear, fit line for large \( \tau \) in the figure. An estimation of the dependence of \( N_d(\theta) \propto (1 + \cos \theta)^2 \) on beam intersection angle comes from the geometrical factor in \( v_1 \propto (1 + \cos \theta) \), which must be squared and gives the approximate agreement shown in the fifth plot. For small angles, \( N_d(\theta) \propto 1 - \theta^2/2 \), making the dependence relatively weak for near head-on collisions (\( N_d \) remains at 90\% of its value up to \( \theta \approx \pi/7 \)). The final plot of \( N_d(\Delta \tau) \) closely resembles a Gaussian with width \( 9\tau \) and so for this setup \( N_d \) is relatively insensitive to lag.

To optimize the number of diffracted photons, we mention that by choosing the laser pulses’ polarization to be perpendicular, all the results in figure 4 will be increased by a factor \( 49/16 \approx 3 \). Another strategy could be to use higher harmonics of the probe laser. If the same parameters as in figure 2 are used, for a collision angle of \( \theta = 0.1 \), assuming a 40\% reduction in energy due to generating the second harmonic, \( N_d \approx 4 \). If this process could be repeated to generate the fourth harmonic, with a 16\% reduction, \( N_d \approx 13 \). As previously argued in [17], such numbers of scattered photons should allow detection in experiment. A discussion of sources of background noise and when they can be effectively neglected is given in [17, 33].

5. Inelastic photon–photon scattering (four-wave mixing)

Due to its nonlinear dependence on the source fields, the diffracted field can contain frequencies that are different from those of the sources. We label this part of the diffracted field as ‘inelastic’, which in other applications can be produced by combining three distinct frequencies to produce a fourth—hence the term ‘four-wave mixing’. The quantized analogue of one of these processes is given by

\[
\gamma(\omega_1, k_1, \varepsilon_1) + \gamma(\omega_2, k_2, \varepsilon_2) \rightarrow \gamma(\omega_3, k_3, \varepsilon_3) + \gamma(\omega_4, k_4, \varepsilon_4),
\]

where again \( \gamma \) represents a photon, \( \omega \), its frequency, \( k \), its wavevector and \( \varepsilon \), its polarization and the photons can take their parameters from any of those offered by the beams. There are several possible four-wave mixing processes, and depending on the directions of their momenta, the number of the four photons occurring on either side of equation (15) could be different. Here we investigate the feasibility of accessing this process using two laser beams. Since the diffracted field is produced in phase with the disturbance, there exists a condition that the phases must agree at the point of interaction. As the external fields have approximately a simple plane-wave dependence on the phase, this provides a resonance condition \( k = k_1 + k_2 + k_3 \) for one generated field’s four-vector \( k \) and three driving field’s four-vectors \( k_1, k_2 \) and \( k_3 \). Let us consider different components of this condition, with \( \omega_{c,i}, i \in \{1, 2\} \) representing the frequencies of two possible wavevectors supplied by pulses a and b in the interaction \( E_a^2 E_b \). Then for the 12 terms occurring
in the diffracted field, we have
\[ \omega = \text{sgn}(\beta_j)(\omega_{a,1} + \delta_{[\beta_j]}^2 \omega_{a,2}) + \text{sgn}(\gamma_j)(\omega_{b,1} + \delta_{[\gamma_j]}^2 \omega_{b,2}), \]  
\[ \omega \frac{y}{r} = \text{sgn}(\beta_j)[\omega_{a,1} \cos \theta_{a,1} + \delta_{[\beta_j]}^2 \omega_{a,2} \cos \theta_{a,2}] + \text{sgn}(\gamma_j)[\omega_{b,1} \cos \theta_{b,1} + \delta_{[\gamma_j]}^2 \omega_{b,2} \cos \theta_{b,2}], \]  
\[ \omega \frac{\rho}{r} = \text{sgn}(\beta_j)[\omega_{a,1} \sin \theta_{a,1} + \delta_{[\beta_j]}^2 \omega_{a,2} \sin \theta_{a,2}] + \text{sgn}(\gamma_j)[\omega_{b,1} \sin \theta_{b,1} + \delta_{[\gamma_j]}^2 \omega_{b,2} \sin \theta_{b,2}], \]  
where \( \omega \) is the frequency generated, \( j \) runs from 1 to 12, \( \beta_j, \gamma_j \in \{−2, −1, 0, 1, 2\} \) are defined in table A.1, of appendix A and define the possible combinations of frequencies involved, \( \theta_{c,i} \) are the angles the photons make with the positive \( y \)-direction and \( \text{sgn}(x) \) returns the sign of \( x \) with \( \text{sgn}(0) = 0 \). Therefore, the detection coordinate and harmonic order are already linked at this stage. It turns out to be difficult to satisfy these conditions simultaneously with just two laser beams and a fixed observation angle. For example, if we take \( \beta_j = 2, \gamma_j = 1 \), with \( \omega_{a,1} = \omega_{a,2} = \omega_a, \omega_{b,1} = \omega_{b,2} = \omega_b \) for simplicity and a more-or-less head-on collision of the lasers, so \( \theta_{a,[1,2]} \) is approximately equal to \( \pi - \theta_{b,[1,2]} \) and \( \theta_{b,[1,2]} \) is small, then, to first order, from equations (16) and (17) we have \( \omega = 2\omega_a + \omega_b \) and \( \omega y/r \approx −2\omega_a + \omega_b \). Since \( y/r \approx 1 \) on the detector, the contribution from this term can therefore only be satisfied by a small range of frequencies around \( \omega_a = 0 \), which are not typically populated in the spectrum of beam a. The energy-momentum condition equations (16)–(18) can be most easily seen occurring in the exponent of the integral \( \hat{V}_j \) in equation (A.4), where they appear as frequencies of plane waves to be integrated over in \( y, z \), becoming Gaussian-like after integration. The larger the deviation from these conditions, the higher the frequency of oscillation to be integrated over, the more exponentially small the resulting amplitude, typical of evanescent waves.

We investigated the ansatz that for short enough pulses, the bandwidth of the two lasers becomes wide enough that equations (16)–(18) can be fulfilled simultaneously for a measurable number of photons. Essentially, for this four-wave interaction, three different photon energies can be supplied by two lasers. To make this statement explicit, instead of using a temporal envelope to describe the pulse, we can consider building the pulses in the frequency domain:
\[ E'_n(x, t) = \int_{−\infty}^{\infty} d\omega_n E'^{\text{mono}}_n(x, t, \omega_n) g(\omega_n, \omega_n, 0), \]  
where \( E'^{\text{mono}}_n(x, t, \omega_n) \) is the electric field of a monochromatic Gaussian beam, frequency \( \omega_n \) and \( g(\omega_n, \omega_n, 0) \) is the spectral density of the pulse \( E'_n(x, t) \), with peak frequency \( \omega_n, 0 \). Then due to our interaction being cubic in the fields in the form of \( E_a^B E_b^\delta \), integration over this current in the frequency domain, equation (A.4), would include three extra integrations over frequency \( \int d\omega_1 d\omega_2 d\omega_3 g(\omega_1, \omega_1, 0) g(\omega_2, \omega_2, 0) g(\omega_3, \delta_{[\beta]}^2 \omega_1, 0 + \delta_{[\gamma]}^2 \omega_2, 0) \delta(\omega - \omega_j) \), where \( \omega_j \) is the sum of central frequencies occurring in a given term in the diffracted field (defined in appendix A) and where the final delta function appears explicitly from an integration over \( t \). Here it is apparent that due to the finite bandwidth, in general, three different energies enter the effective vertex in figure 2 from the two lasers. Moreover, if the spectrum is taken to be Gaussian \( g(\omega_n, \omega_n, 0) = \exp[−(\omega_n - \omega_n, 0)^2 \tau_n^2/4] \tau_n/2\sqrt{\pi} \), we have, setting \( \theta = 0 \),
\[ E'_n(x, t) = \hat{E}_n \int_{−\infty}^{\infty} d\omega_n E_n,0 e^{−\frac{c^2}{w(x)^2} \frac{1}{2} (\omega_n - \omega_n, 0)^2} \frac{\sin \left[ \omega_n (y − t) + \tan^{-1} \left( \frac{2y}{\omega_n w_n, 0} - \frac{2\omega_n y (x^2 + z^2)}{4y^2 + \omega_n^2 w_n, 0,0 y^2} \right) \right]}{\sqrt{1 + (y/y_0, t)^2}} \]  
\[ = E'^{\text{mono}}_n(x, t, \omega_n, \omega_n) e^{−\left( \frac{c^2}{w(x)^2} \right)^2} + \text{h.o.t.}, \]
where the remaining higher-order terms are of the same order as those neglected in the Gaussian beam solution. Therefore, the use of a Gaussian temporal envelope in $E_a$ and $E_b$ (equations (1) and (2)) is equivalent to integrating over three different photon frequencies of the approximately Gaussian spectra of the external fields in the interaction.

When the collision of the beams is on-axis and the diffraction angle is small, the number of diffracted photons can be approximated analytically. Since the calculation of the diffracted field containing mixed frequencies involves non-trivial numerical integration of highly oscillating functions, these analytical results can be used as a test of the numerical method. The excellent agreement between numerics and analytics is shown in figure C.1 and explained in appendix C, in part corroborating our numerical approach. We can see from equation (C.1) that the spectral density for the generated mixed frequencies has a different shape compared to the background, namely with a minimum at $\omega = 0$ and two maxima, whose positions at the centre of the detector are $\omega^\pm = (\omega_b/2)(1 \pm 12/((\omega_b \tau)^2)^{1/2})$. Using a spectral filter, for short enough pulses, this could, in principle, be used to separate the different inelastic scattering signals from each other and the elastically scattered and background photons on the detector.

The pulse duration of each laser plays an important role in four-wave mixing. By choosing a temporal profile for the beam that is Gaussian, we already have implicitly the lower bound $\tau \gg 1/\omega$. As the pulse duration and longitudinal coordinate are linked, a natural upper bound is also formed for our calculation in the assumption that the diffracted field is smaller than the vacuum polarizing fields equation (10). Assuming that scattered photons arriving at a point on the detector are generated at the centre of the beams’ intersection, the integration is exclusively over regions in which the polarizing beams are more intense than the diffracted field when $\tau \ll 2w_{b,0}y/\rho$, giving $1/\omega \ll \tau \ll 2w_{b,0}y/\rho$. The lower bound limits our ability to assess the importance of the inelastic process. We require a large bandwidth $\Delta\omega/\omega$ for these mixed-frequency photons to be on-shell, but from the bandwidth theorem, $\Delta\omega/\omega \sim 1/\omega \tau \ll 1$ from our limitation on $\tau$. As a consequence, with a two-beam setup, spectrally separating off the inelastic signal would be experimentally challenging, as this signal is generated when the bandwidth of the elastic background overlaps these mixed frequencies. More promising seems to be to observe the change in $N_d$ due to inelastic scattering becoming significant as $\tau$ is reduced. In figure 5, we plot this ratio $(N_t - N_e)/N_e$ against $\tau_a$, where $N_e$ is the number of photons scattered when only the elastic terms are included in equation (12). The results suggest that for short enough pulse durations, the inelastic process can influence the total number of measured photons substantially. In figure 5 the proportion reaches over 20%, for a minimum pulse duration of $\tau = 1$ fs, equivalent to $\omega_b \tau_a \approx 2$. In addition, although the pulse durations are short, assuming again 40% attenuation each time a second harmonic is generated from the probe, the total number of diffracted photons ranges from 1 to 4 (at $\tau_a = 1$, 2 fs, respectively). Although the analysis is limited by how small $\tau_a$ can be consistently made, these results lend support to the ansatz that two laser beams with a large bandwidth, especially in the laser being probed, can be used to measure the effect of the inelastic process.

5.1. The sech pulse case

In order to further support this ansatz without being limited by a minimum value of the pulse duration, we can consider the simplified case of the collision of two plane waves modulated by a sech envelope. Here we will show qualitatively how, when the duration of the pulses is reduced, the dependence of the number of diffracted photons changes, which we show to be due
Figure 5. The increasing importance of the inelastic process with increasing bandwidth. Plotted is the proportion of the total number of diffracted photons that are due to vacuum four-wave mixing, against $\tau_a$, for $P_b = 10/3$ PW, $P_a + P_b = 10$ PW, $\lambda_a = 0.91$ $\mu$m, $\lambda_b = 0.2275$ $\mu$m, $\tau_b = 2$ fs, $w_{a,0} = 0.91$ $\mu$m, $w_{b,0} = 50$ $\mu$m, $\epsilon_a = (1, 0, 0)$, $\epsilon_b = (0, 0, 1)$, $\psi_a = \psi_b = 0$.

Figure 5. The increasing importance of the inelastic process with increasing bandwidth. Plotted is the proportion of the total number of diffracted photons that are due to vacuum four-wave mixing, against $\tau_a$, for $P_b = 10/3$ PW, $P_a + P_b = 10$ PW, $\lambda_a = 0.91$ $\mu$m, $\lambda_b = 0.2275$ $\mu$m, $\tau_b = 2$ fs, $w_{a,0} = 0.91$ $\mu$m, $w_{b,0} = 50$ $\mu$m, $\epsilon_a = (1, 0, 0)$, $\epsilon_b = (0, 0, 1)$, $\psi_a = \psi_b = 0$.

to an increasing contribution from the inelastic process. For this sub-section, we redefine the quantities used in the Gaussian pulse setup to pertain to the present, sech-pulse scenario:

\[
E_a(y + t) = \hat{\epsilon}_a E_a \cos[\omega_a(y + t)] \operatorname{sech}\left(\frac{y + t}{\tau_a}\right),
\]

\[
E_b(y - t) = \hat{\epsilon}_b E_b \cos[\omega_b(y - t)] \operatorname{sech}\left(\frac{y - t}{\tau_b}\right).
\]

These fields satisfy Maxwell’s vacuum equations exactly, thus removing the limitations on conceivable pulse lengths brought about by using a perturbative solution. The analysis proceeds just as for the Gaussian case but with the difference that now the fields are not bound in the transverse plane. Therefore, in order to avoid a divergence, we only consider the resulting vacuum polarization and magnetization to be non-zero up to a finite transverse radius $\rho_0$. It can be shown that this curtailing of the interaction region then allows us to integrate over the present equation (9) as usual. The diffracted field $\tilde{E}_d(x, \omega)$ then becomes

\[
\tilde{E}_d(x, \omega) = \frac{\omega^2 \alpha^2}{360\pi^2 m^4 r} \left( \epsilon_a^2 \hat{\epsilon}_b v_1 \tilde{Z}_1 + \epsilon_a \epsilon_b v_2 \tilde{Z}_2 \right),
\]

where $v_j$ are geometrical factors as in the Gaussian case equations (A.1) and (A.2), $\tilde{Z}_j$ are integrals of dimension length to the fourth power, given in equation (A.5), and $z = 0$ has already been set for simplicity.

To display the contribution from the elastic and inelastic processes, we plot the spectrum of the number of diffracted photons $d\tilde{N}_d(\omega)$ for decreasing $\tau_a$, with $\tau_b = 6.7$ fs, $\lambda_a = \lambda_b = 0.8$ $\mu$m,
The spectra of $d \tilde{N}_d(\omega)$ against $\omega_\alpha \tau_\alpha$ with $\omega_\alpha = \omega_b$ held constant, with the white dotted lines representing the elastic ($\omega = \omega_b$) and inelastic ($\omega = 3 \omega_b$) contributions. Only for short pulses $\omega_\alpha \tau_\alpha \lesssim 1$, does the inelastic signal become important.

Figure 7. On the left-hand side is the relative difference in $N_d$ due to mixed frequencies for different durations of the polarizing pulse $\tau_\alpha$ ($\omega_\alpha = \omega_b$ is held constant); on the right-hand side, the corresponding total number of diffracted photons $N_d$ and elastic contribution $N^e_d$.

$r = 1$ m, $x/r = 0.1$ and pulse width $\rho_0 = 10 \mu$m, shown in figure 6. We note that for $\omega_\alpha \tau_\alpha \gtrsim 1$, most of the diffracted photons have the same frequency as the probe beam and correspond to elastic scattering, which is especially the case in the right-hand plot of figure 6 for $\omega_\alpha \tau_\alpha \gg 1$. However, for $\omega_\alpha \tau_\alpha \lesssim 1$, the mixed-frequency signal from near $\omega = \omega_b + 2 \omega_a$ becomes dominant. The position of the expected inelastic contribution is indicated in figure 6, but it is clear that the signal is slightly displaced from this. Although only positive frequencies are plotted, the image is very similar for negative frequencies. To quantify the ratio of elastic to inelastic scattering, the relative difference of the number of elastically and total diffracted photons is plotted against $\tau_\alpha$ in figure 7. The total scattered is calculated summing over all frequencies in figure 6 and the elastically scattered by summing just contributions around $\omega_b$. In figure 7, we note that the number of diffracted photons can be substantially altered by the mixed-frequency signal.
(more than 80% in the plot) in very short pulses, with the difference compared to the total number of diffracted photons being more than 10% for pulses as long as $\omega_a \tau_a \approx 4$, similar to the case of the previous Gaussian treatment figure 5. We also note the fluctuations in $N_d$, which correspond to the edges of those islands in the spectrum figure 6, in which the resonance condition for inelastic scattering at $x/r = 0.1$ is fulfilled.

The essence of why the mixed-frequency signal can become larger than the elastic one is included in the interaction term, which contains terms such as $E_a^2 E_b$. The oscillating term then contains $\cos^2(\phi_a)\cos(\phi_b) = f(\phi_b) + g(\phi_a, \phi_b)$ for phases $\phi_c$ and $f(\phi_b) = \cos(\phi_b)/2$, $g(\phi_a, \phi_b) = \cos(2\phi_a)\cos(\phi_b)/2$. When many cycles are integrated over a pulse shape function, the inelastic contribution $g(\phi_a, \phi_b)$ will vanish with respect to the elastic one $f(\phi_b)$. However, for $\phi_c \ll 1$, $g(\phi_a, \phi_b) \approx f(\phi_b)$, so that the relative difference of the mod-squares, $1 - |f|^2/|f + g|^2$, can clearly become as large as $3/4$.

These results for few- and sub-cycle polarizing pulses demonstrate the new behaviour occurring for short pulse durations or equivalently large bandwidths and so further support the ansatz that just one beam split into two counter-propagating sub-cycle pulses is sufficient for accessing the process of vacuum four-wave mixing. Since this is an effect occurring in the frequency domain, it seems logical to suggest that the addition of focusing terms will not greatly influence this conclusion. A suggestion for further work would be to investigate the role of the carrier-envelope phase as well as a chirped frequency.

6. Summary

In calculating numbers of photons scattered in the collision of two laser beams, we had three aims: (i) to consider a more realistic setup of the colliding beams (including a temporal pulse shape, collision angle, lag and lateral separation), which would produce more accurate qualitative and quantitative predictions for experiment, (ii) to investigate the possibility of using a single laser, split into two beams to measure elastic photon–photon scattering and (iii) to evaluate the ansatz that just two lasers, with sufficiently short pulse durations, can be used to measure the process of inelastic photon–photon scattering (vacuum four-wave mixing). The first of these aims has been met in figure 4 where the dependence on various collision parameters was calculated and found to be consistent with physical reasoning. This led to the second aim, where the inclusion of a pulse form and collision angle led to two orders of magnitude difference over previous elastic photon scattering estimates [33] (the single-slit limit of [17]). In this more complete description, it was shown that when a 10 PW, $\lambda = 0.91$ nm beam is separated into two 30 fs Gaussian pulses, incident at an angle of 0.1, one could expect approximately 0.7, 4 or 13 photons, corresponding to the fundamental, second and fourth harmonic of the probe, respectively (with an assumed loss of 40% per frequency doubling), to be diffracted into detectable regions. As argued in [17], this could be sufficient for measuring elastic photon–photon scattering, here shown using a single 10 PW source. The final aim was partially met, first by considering Gaussian pulses, where it was shown that for $\omega_a \tau_a \lesssim 4$ for the more intense beam $a$, the inelastic scattering process increased and altered the total number of diffracted photons with respect to the purely elastic signal by as much as 20% for $\omega_a \tau_a \approx 2$. However, for these results to be consistent, $\omega_a \tau_a \gg 1$, so the head-on collision of two sech pulses was analysed, for which no such bound applies, where it was shown that again, in this different field background, for $\omega_a \tau_a \lesssim 1$, the majority of photons scattered were due to four-wave mixing.
Appendix A. Integration formulae

Diffraacted field polarization vectors:
\[
\begin{align*}
v_1 &= 4 \left( \hat{e}_a \cdot \hat{e}_b (1 + \cos \theta) - \hat{y} \cdot \hat{e}_b \hat{y} \cdot \hat{e}_a \right) \left[ \hat{e}_a \left( 1 - (\hat{r} \cdot \hat{r}) \right) + (\hat{y} \cdot \hat{e}_a) \wedge \hat{r} \right] \\
&\quad + 7 \left( \hat{e}_a \cdot \hat{y} \wedge \hat{e}_b - \hat{e}_b \cdot \hat{y} \wedge \hat{e}_a \right) \left[ - (\hat{y} \cdot \hat{e}_b) \left( 1 - (\hat{r} \cdot \hat{r}) \right) + \hat{e}_a \wedge \hat{r} \right].
\end{align*}
\]
\[ \text{(A.1)} \]
\[
\begin{align*}
v_2 &= 4 \left( \hat{e}_a \cdot \hat{e}_b (1 + \cos \theta) - \hat{y} \cdot \hat{e}_b \hat{y} \cdot \hat{e}_a \right) \left[ \hat{e}_b \left( 1 - (\hat{r} \cdot \hat{r}) \right) - (\hat{y} \cdot \hat{e}_b) \wedge \hat{r} \right] \\
&\quad + 7 \left( \hat{e}_a \cdot \hat{y} \wedge \hat{e}_b - \hat{e}_b \cdot \hat{y} \wedge \hat{e}_a \right) \left[ (\hat{y} \cdot \hat{e}_b) \left( 1 - (\hat{r} \cdot \hat{r}) \right) + \hat{e}_b \wedge \hat{r} \right].
\end{align*}
\]
\[ \text{(A.2)} \]
where \( \hat{y} \cdot \hat{y} = \hat{y} \cdot \hat{y} = \hat{r} \cdot \hat{r} = 1 \) and \( \hat{r} = (x \cdot \hat{r}) \hat{r} \) for any vector \( x \).

Integration terms:
\[
\begin{align*}
\mathcal{V}_j &= i A_j (B_j + \Gamma_j (\tau_a / \tau_b)^2) (4 + (\omega_j \tau_j)^2) e^{i \psi_j} \int \frac{1}{[1 + (\hat{y} / y_{1a})^2]^{B_j / 2}} \frac{1}{[1 + (y' / y_{1b})^2]^{B_j / 2}} d^3x' \\
&\quad \times e^{-x^2 \left( \frac{\gamma_j}{\omega_j} \right)^2} \left[ 1 + \frac{1}{e^j} \right] \int \frac{1}{[1 + (\hat{y} / y_{1a})^2]^{B_j / 2}} \frac{1}{[1 + (y' / y_{1b})^2]^{B_j / 2}} d^3x' \\
&\quad \times e^{-x^2 \left( \frac{\gamma_j}{\omega_j} \right)^2} \left[ 1 + \frac{1}{e^j} \right] \int \frac{1}{[1 + (\hat{y} / y_{1a})^2]^{B_j / 2}} \frac{1}{[1 + (y' / y_{1b})^2]^{B_j / 2}} d^3x'.
\end{align*}
\]
\[ \text{(A.3)} \]
\[
\tilde{\psi}_j = \frac{i A_j e^{i \psi_j}}{\sqrt{B_j + \Gamma_j (\tau_a / \tau_b)^2}} \int \frac{1}{[1 + (\hat{y} / y_{1a})^2]^{B_j / 2}} \frac{1}{[1 + (y' / y_{1b})^2]^{B_j / 2}} d^3x' \\
&\quad \times e^{-x^2 \left( \frac{\gamma_j}{\omega_j} \right)^2} \left[ 1 + \frac{1}{e^j} \right] \int \frac{1}{[1 + (\hat{y} / y_{1a})^2]^{B_j / 2}} \frac{1}{[1 + (y' / y_{1b})^2]^{B_j / 2}} d^3x'.
\]
\[ \text{(A.4)} \]
where \( A_j \in \{-2, -1, 1, 2\} \), \( \beta_j, \gamma_j \in \{-2, -1, 0, 1, 2\} \) are given in table A.1, \( \omega_j = \beta_j \omega_a + \gamma_j \omega_b, \psi_j = \beta_j \psi_a + \gamma_j \psi_b, \tau_j = [B_j / \tau_a^2 + \Gamma_j / \tau_b^2]^{-1/2}, B_j + \Gamma_j = 3 \) and \( \Gamma_j = 1 \) for \( j \leq 6 \), otherwise \( \Gamma_j = 2 \).
Table A.1. Sum coefficients that occur in the integrals $\mathcal{V}_j$ and $\tilde{\mathcal{V}}_j$, equations (A.3) and (A.4).

| $j$ | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 |
|-----|---|---|---|---|---|---|---|---|---|----|----|----|
| $A_j$ | 1 | 1 | −2 | −1 | −1 | 2 | 1 | 1 | −2 | −1 | −1 | 2 |
| $\beta_j$ | 2 | −2 | 0 | 2 | −2 | 0 | 1 | 1 | 1 | −1 | −1 | −1 |
| $\gamma_j$ | 1 | 1 | 1 | −1 | −1 | 2 | −2 | 0 | 2 | −2 | 0 | 0 |

A.1. The sech diffracted field formulae

Integration terms:

\[ \tilde{I}_j = \int_0^\infty d\rho \, \rho \int_0^{2\pi} d\varphi \int_{-\infty}^{\infty} d\phi_+ \int_{-\infty}^{\infty} d\phi_+ \, \operatorname{sech}^{2-j}(\phi_+ / \tau_a) \operatorname{sech}^j(\phi_+ / \tau_b) \]

\[ \times \cos^{2-j}(\omega_b \phi_+) \cos^j(\omega_b \phi_+) e^{i \rho \varphi (1 + \xi_2 \cos^2 \varphi)} \exp \left[ \frac{m^+}{2} (\phi_+ - \phi_+ \cos \varphi) \right] \]

\[ \times e^{i \rho \varphi \cos \varphi} \frac{1}{2} \left( (\phi_+ \cos \varphi + (1 - \xi_2)) \right) - \frac{1}{2} (\phi_+ \cos \varphi) \left( (1 - \xi_2) \right) \]  

(A.5)

Appendix B. Comparison of elastically diffracted field numerics with previous calculations

We return to the two orders of magnitude difference in the number of diffracted photons $N_d$ calculated from $\mathbf{E}_d$ compared to the number $N_d^h$ calculated from the literature for the head-on collision of two Gaussian beams of infinite duration, with diffracted electric field $\mathbf{E}_d^h$. As mentioned in the main text, upon comparing the diffracted field intensities in the two cases, $I_d$ and $I_d^h$ shown in figure B.1, the pulse durations in $\mathbf{E}_d$ had to be set much higher than the figures considered in the literature before agreement was reached. To illuminate this discrepancy, the expression for the number of diffracted photons was studied. The integrands for the diffracted fields were reduced to the most significant terms for a head-on, elastic collision and evaluated independently in Mathematica. The simplified expressions for the number of diffracted photons, denoted by a hat $\tilde{N}$, were then given by

\[ \tilde{N}_d(\tau) = \int_{\rho_d^{\min}}^{\rho_d^{\max}} d\rho \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \int_{-\infty}^{\infty} dy' \sqrt{\frac{\pi}{3}} \mathcal{V}(y', \omega, \omega_b, \tau, \rho / r)^2, \]  

(B.1)

\[ \tilde{N}_d^h(\tau) = 2r^2 \int_{\rho_d^{\min}}^{\rho_d^{\max}} d\rho \int_{-\infty}^{\infty} \frac{d\tau}{4} \int_{-\infty}^{\infty} dy' \lim_{\tau \to \infty} \lim_{\omega \to \infty} \mathcal{V}(y', \omega, \omega_b, \tau, \rho / r)^2, \]  

(B.2)

\[ \mathcal{V}(y', \omega, \omega_b, \tau, \rho / r) = \frac{\alpha I_{a,0} \epsilon_b w_a^2 \omega^{3/2}}{360\pi y I_{c,0} \varepsilon} \left[ 1 + \frac{1}{2} \frac{w_a^2}{w_b^2} \frac{1 + (y' / y_{r,a})^2}{1 + (y' / y_{r,b})^2} \right]^{-1} \]

\[ \times \exp \left[ -\omega^2 \rho^2 (1 + (y' / y_{r,a})^2) + \frac{4i(\omega - \omega_b)y'}{3} - \frac{8y^2}{3\tau^2} - \frac{(\omega - \omega_b)^2\tau^2}{12} \right], \]  

(B.3)
Figure B.1. Numerical comparison of $I_d$ with the results in [33] (denoted by $I_d^h$). Plotted is $\log_{10}$ of the absolute relative difference in $I_d$ for, from top to bottom, $\tau_a = \tau_b = 100, 1000$ and $20\,000\,$fs. Between the dotted lines, the background dominates ($I_b \gg I_d$).

Figure B.2. Left-hand plot (a): $\log_{10} \hat{N}_d$ (upper curve) and $\log_{10} \hat{N}_d$ (lower curve) plotted against $\rho$. Right-hand plot (b): a log–log plot of $\Re \{l(y')\}$ (solid line) with the dashed lines referring to various limits (labelled on the plot).

where $s^2 = (2/w^2_{a,0} + 1/w^2_{b,0})^{-1}$ and $\rho^2 = (x^2 + z^2)/r^2$. The dependence on $\rho/r$ of these two expressions is shown in figure B.2(a), where it can be seen that the monochromatic $\hat{N}_d$ is much larger and more sharply peaked at the centre of the detector. After integrating between the relevant annulus of $\rho_{\min}/r = 0.0032$ and $\rho_{\max}/r = 0.03$, this independent test then gives $\hat{N}_d(\tau) = 0.33$ and $\hat{N}_d^h = 37.0$. The corresponding values for a circular detector of radius 15 cm from the full expression are $\hat{N}_d(\tau) = 0.33$ and $\hat{N}_d^h = 38.0$,
Supporting the two orders of magnitude difference and the claim that equations (B.1)–(B.3) incorporate the main physics. By plotting the exponential dependence on $\rho/r$, which is integrated over to acquire the expected number of photons in the monochromatic case, $N_d^b$:

$$I(y') = \exp \left[ -\frac{\omega^2 \rho^2 (1 + (y'/y_{\text{r,a}})^2)}{4 \left( 2 + \left( \frac{w_{b,0}}{w_{b,0}} \right)^2 \frac{1+(y'/y_{\text{r,b}})^2}{1+(y'/y_{\text{r,b}})^2} \right)^2} \right]. \quad (B.4)$$

we observe the interesting behaviour shown in figure B.2(b). We first note that the decay is not purely exponential, but has two important length scales: $w_{b,0}$ and $y_{\text{r,b}}$, which, in the limit of being infinitely large, correspond to the first and third dashed curves in figure B.2(b). The inclusion of these extra longitudinal length scales in [33], which cannot contribute to photon scattering when the finite pulse length is taken into consideration, then explains the discrepancy in the values of $\hat{N}_d(\tau)$ and $\hat{N}_d^b$. Only the region of the pulses within a distance $\tau$ around their maxima in the longitudinal direction can efficiently contribute to the scattering process, with the rest of the pulse being damped by its Gaussian shape. The finite length of laser pulses probing vacuum photon–photon scattering can then only be neglected when the duration $\tau$ is the largest longitudinal length scale. In the limit $\tau \gg y_{\text{r,b}}$ in the full expression for $E_d$ in equations (11) and (12), the scaling $N_d(\tau) \propto \tau$ of [33] is recovered, supporting this statement (this will also be apparent from figure 4). Furthermore, the results of [33] are expected to remain valid in the case $\pi w_{b,0}^2/\lambda_b \tau_{\text{a,b}} < 1$, meaning for more focused and longer wavelength probe beams as well as for longer pulses. Indeed for the parameters quoted, that the effect would be two orders of magnitude weaker is in no way prohibitive to conducting such experiments. For example in [17, 33], the intensity of the probe beam was taken to be only around $I_p \approx 10^{16}$ W cm$^{-2}$, but as $N_d \propto I_p$, the shortfall could be made up by focusing the probe beam more (if $w_{b,0}$ is set to 60 $\mu$m, $N_d$ increases approximately by a factor 7 in the single-slit and 4.5 in the double-slit case) or increasing the power of the probe (from 10 TW), to which $N_d$ is proportional.

Appendix C. Numerical and analytical calculation of the number of inelastically scattered photons

When the collision of the beams is on-axis ($x_0 = z_0 = \Delta t = \theta = 0$), $y_r = y_{r,a} = y_{r,b}$ and $\rho^2/r^2$ is small, the number density of diffracted photons $dN_d(x, z)$ can be approximated analytically ($N_d = \int dx \, dz \, dN_d(x, z)$). As the number of diffracted photons is proportional to the mod-square of the diffracted terms (which comprises 12 terms), we can write $dN_d(x, z) = \sum_{p,q=1}^{12} dN_{pq}^d(x, z)$ and demonstrate this analysis by concentrating on a single term $dN_{pq}^d(x, z)$ for convenience (the full expression is given in equation (C.4)). One can show that

$$dN_{pq}^d(x, z) \approx \frac{2}{\pi^2} \left[ \frac{\alpha A_d E_{a,0}^0 E_{b,0}^{\tau_q} |w_{(q)}| w_{a,0}^2}{90 \, E_{c\tau}^2} \frac{r_{pq}^2}{16 \, sr \sqrt{1 - t_{pq}^4}} \right]^2 e^{-\frac{\alpha^2 r_{pq}^2}{2(1 - r_{pq}^4)}} \times \int_{-\infty}^{\infty} d\omega |\omega|^3 e^{-\left( \frac{\rho^2 + \omega^2 + \frac{1}{2} r_{pq}^2 (\omega^2 r_{pq}^2)^{1/2}}{2(1 - r_{pq}^4)} \right) \omega^2 + r_{pq}^2 \omega^2 \left[ 1 - \frac{\omega^2 r_{pq}^2 (\omega^2 r_{pq}^2)^{1/2}}{1 - r_{pq}^4} \right] \omega}, \quad (C.1)$$

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where \( s = 1/(B_j + \Gamma_j (w_{a,0}/w_{b,0})^2) \), \( \tau_{qq} = \tau_q/\tilde{\tau}_q \), \( \tilde{\tau}_q^2 = (B_q/\tau_a^2 - \Gamma_q/\tau_b^2)^{-1} \) and \( \tilde{\omega}_q = \tilde{\omega}_a/\omega_q \), \( \tilde{\omega}_{qq} = \tilde{\omega}_q/\omega_q \), under the condition \( T^2/\gamma_q^2 \ll 1 \), for \( T^2 = \gamma_q^2 [1 - \tau_q^4]^{-1} \) and where a condition on \( \omega' : \left| (\omega' (\tau_{qq} - y/r) + \tilde{\omega}_q - \omega_q \tau_{qq}^2) T^2/\gamma_q \right| < 1 \) has been approximated by taking the upper limit of the integration as \( \infty \). To simplify the discussion, let \( \tau_a = \tau_b = \tau \). Then we can see from equation (C.1) that the spectral density for inelastically scattered photons has a different shape compared to the background, namely with a minimum at \( \omega = 0 \) and two maxima, whose positions for the case \( x = z = 0 \) are \( \omega_{\pm} = (\gamma_q \omega_b/2)(1 \pm [1 + 12/(\gamma_q \omega_b \tau)]^{1/2}) \). Setting \( \rho = 0 \) for brevity, the final integral can be approximated by

\[
dN_{qq}^d(x, z) = \sqrt{\frac{2}{\pi^3}} \left[ \frac{\alpha A_q E_{a,0} E_{b,0} \Gamma_q}{180 \ E_{c1}^2} \right]^2 (\gamma_q \omega_b \tau) \left[ 3 + (\gamma_q \omega_b \tau)^2 \right] \times \text{Erf} \left[ \frac{\gamma_q \omega_b \tau}{\sqrt{2}} \right] e^{-\frac{1}{4}(\beta_q \omega_b \tau)^2 - \frac{1}{4}(\gamma_q \omega_b \tau)^2}. \tag{C.2}
\]

These analytical approximations were then used to corroborate the numerical calculation. The numerical integration of the full highly oscillating integrands was performed using the Filon method, which is an approximation to the integral \( \int dt f(t) \cos(\omega t) \) for asymptotically large \( \omega \) (see, e.g., [34]), used with the GNU arbitrary-precision C++ library [35]. Agreement between numerics and analytically for \( w_{a,0} = w_{b,0} = 10 \, \mu\text{m}, y = 1 \, \text{m}, \beta_q = 2, \gamma_q = 1 \) is then shown in figure C.1, in part corroborating our numerical approach. It should be noted that \( \nu_{i(j)} \) is identically zero for \( j > 6 \) at \( r = y \), and so the frequencies \( \omega_a, 2\omega_b \pm \omega_a \) are suppressed, as already argued in the main text.

**C.1. Analytical approximation to \( N_d(x, z) \)**

\[
dN_d(x, z) = \sum_{p,q=1}^{12} dN_{qq}^d(x, z), \tag{C.3}
\]
\[ dN_{d}^{pq}(x, z) = \frac{1}{\pi^{3/2}} \left[ \frac{\alpha}{180} \frac{w_{a,0}^2}{r} \right]^2 E_{R,0}^{R_p + R_q} \frac{E_{b,0}^{R_p + R_q}}{E_{cr}^4} \frac{\nu_{(p)} \cdot \nu_{(q)}}{16^2} A_p A_q \tau_p^2 \tau_q^2 \]

\[ \times \left[ B_p + \Gamma_p \left( \frac{w_{a,0}}{w_{b,0}} \right)^2 \right] \left[ B_q + \Gamma_q \left( \frac{w_{a,0}}{w_{b,0}} \right)^2 \right] \sqrt{1 - \tau_{pp}^4} \sqrt{1 - \tau_{qq}^4} \]

\[ \times \frac{b(6a + b^2)}{a^{7/2}} \operatorname{Erf} \left( \frac{b}{2 \sqrt{a}} \right) e^{\frac{c^2}{b^2}} \]

\[ a = \frac{\rho^2 w_{a,0}^2}{2 \nu^2} + \frac{\tau_p^2}{4} \left[ 1 + \frac{(y/r + \tau_{qq}^2)^2}{(1 - \tau_{pp}^4)} \right] + (p \leftrightarrow q), \]

\[ b = \frac{\tau_p^2}{2} \left[ \nu_p - \frac{(\tilde{\omega}_p - \nu_p \tau_{qq}^2)(y/r + \tau_{qq}^2)}{1 - \tau_{pp}^4} \right] + (p \leftrightarrow q) \]

\[ c = -\frac{\tau_p^2}{4} \left[ \nu_p^2 + \frac{(\tilde{\omega}_p - \nu_p \tau_{qq}^2)^2}{1 - \tau_{pp}^4} \right] + (p \leftrightarrow q). \]

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