LETTER

Compensation of anisotropy effects in a nonlinear crystal for squeezed vacuum generation

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Abstract
Squeezed vacuum can be obtained by an optical parametric amplifier (OPA) with the quantum vacuum state at the input. We are interested in a degenerate type-I OPA based on parametric down-conversion (PDC) where, due to phase matching requirements, an extraordinary polarized pump must impinge onto a birefringent crystal with a large $\chi^{(2)}$ nonlinearity. As a consequence of the optical anisotropy of the medium, the spatial spectrum of the generated radiation is affected by the transverse walk-off. In this work we describe a method that reduces the spatial distortions, by using two consecutive crystals instead of one. We show that after anisotropy compensation the two-photon amplitude becomes symmetric, allowing for a simple Schmidt expansion, a procedure that in practice requires states that come from experimental systems free of anisotropy effects. Qualitative experimental observations are made for the case of high-gain PDC.

((Some figures may appear in colour only in the online journal)

1. Introduction

Biphoton light (squeezed vacuum, SV) [1] and, more recently, light with even photon numbers (bright squeezed vacuum, BSV) [2, 3], produced in parametric down-conversion (PDC) experiments, have been used by the quantum optics community due to their virtues as a resource for the study of quantum correlations. BSV, in particular, has the potential to be employed as a quantum macroscopic state in a range of applications; for instance, for the enhancement of light–matter interactions through the conditional preparation of large Fock states, for pushing the resolution limits in quantum lithography, for error-free quantum signal transmission, or in high precision measurement of phase and displacement, among many others.

Nevertheless, the usability of BSV is limited by its intrinsic multimodeness. Indeed, a source with a large number of modes and, additionally, a large number of photons per mode is extremely valuable if the possibility to access and control individual modes is experimentally achievable. As a consequence, the selection of a single mode of this radiation, in space and frequency variables, becomes a relevant task although it is experimentally challenging. Such a selection relies on the Schmidt mode decomposition, a procedure that has already been demonstrated on biphoton light [4]. From there it is clear that the usage of the double-Gauss approximation [5–8], a tool that eases greatly the task of finding this expansion analytically, is restricted to the case in which anisotropy is neglected [4, 8]. In other words, the two-photon amplitude (TPA) associated with the SV state of interest must fulfil certain symmetry requirements for the
will refer to it as the extensively for BSV generation \([2, 10, 11]\) and in this work we phase matching regime see figure 3(a).

For an idea of what these distortions look like in the collinear degenerate transverse walk-off by reversing the walk-off angle \(\theta\) approach to this problem consists of reversing the sign of the largely on the correction of anisotropy effects. The empirical small. It is expected that anisotropy will introduce asymmetry the pump diameter becomes small. It is expected that anisotropy will introduce asymmetry in the spectrum of the radiation at the optical parametric amplifier (OPA) output since the PDC generation occurs along a tilted path. In particular, it leads to distortions \([6]\) of the two-photon amplitude (TPA), which is the probability amplitude of creating a photon pair with wavevectors \(k_s\) and \(k_i\) and is used to calculate properties of the biphoton state such as correlation widths, shape of the spectrum, anisotropy effects, etc.\(^3\).

One way to control these effects effectively in the case of faint SV generation is to choose adequate experimental parameters such as large pump diameters and short crystal lengths \([4]\). However, these restrictions may be critical in the case of BSV because the achievement of high gain requires smaller pump sizes and longer crystals \([3]\). This implies that the efficient generation of single-mode BSV depends largely on the correction of anisotropy effects. The empirical approach to this problem consists of reversing the sign of the transverse walk-off by reversing the walk-off angle \(\theta\), a task that in turn is carried out by placing a second birefringent crystal next to the first, with its optic axis \(\zeta\) at an angle \(-\alpha\) to the pump wavevector \(k_p\). We have used this technique extensively for BSV generation \([2, 10, 11]\) and in this work we will refer to it as the compensating two-crystal configuration.

The non-compensating two-crystal configuration in turn corresponds to the case where the first and second crystals are set identically, and they behave effectively as one continuous crystal if the distance between them is negligible. Similar compensation methods are routinely used to correct spatial walk-off between signal and idler photons in type-II PDC \([12]\) and to increase the efficiency of harmonic generation \([13, 14]\).

In this work we go a step further and present an accurate description of this anisotropy compensation method. We model the TPA of an SV state generated in two consecutive crystals with collinear degenerate type-I phase matching. We consider both the compensating and the non-compensating configuration of the crystal pair for two-photon states due to the straightforward accessibility of their TPAs. Our results are shown in terms of two important quantities that characterize the TPA, namely the conditional and unconditional probability distributions. The conditional probability distribution corresponds to the emission of a signal photon at a certain angle to the pump beam given that the idler photon is emitted in a fixed direction, or vice versa. It is obtained from the cross-section of the TPA at a given angle. The unconditional probability distribution corresponds to the emission of a signal or idler photon at a certain angle to the pump. It is obtained from the projection of the TPA on one of its axes. These two characteristic quantities could be recovered in experiment from the coincidence count rates and single count rates observed in two detectors, the subject of our future work. We also present observations of the compensation in the BSV case. Due to the nonlinear amplification of BSV along the crystal, the anisotropy makes the spectrum not only asymmetric but non-collinear. In its turn, anisotropy compensation leads to a dramatic change in the spatial spectrum making the emission collinear and the spectrum nearly Gaussian.

2. Theoretical description of two-crystal anisotropy compensation

From the Hamiltonian of the spontaneous parametric down-conversion (SPDC), considering the evolution of the state in the interaction picture, assuming that the crystal’s transverse dimensions are much larger than the pump’s transverse dimensions and assuming also that the pump has a Gaussian transverse profile, the two-photon state at the output of the OPA is usually written as

\[
|\Psi\rangle \propto \int dk_s \int dk_i F(k_s, k_i) a_s^\dagger(k_s) a_i^\dagger(k_i) |0\rangle. \quad (1)
\]

Here, s and i are the labels for signal and idler fields, \(k_s\) and \(k_i\) are the wavevectors of the signal and idler photons and \(a_s^\dagger\) are the photon creation operators in the signal and idler modes. \(F(k_s, k_i)\) is the TPA for this state and in the absence of anisotropy it can be expressed as \([15]\)

\[
F(k_s, k_i) \propto \exp \left[ -\frac{(d \Delta_\perp)^2}{8 \ln(2)} \sin \left( \frac{L}{2} \Delta_\parallel \right) \right], \quad (2)
\]

\(^3\) For an idea of what these distortions look like in the collinear degenerate phase matching regime see figure 3(a).
Figure 2. Diagram illustrating the TPA calculation. In the first crystal, the frame of reference is rotated from $x, z$ to $x', z'$. In the second crystal, the frame of reference is rotated in the opposite direction, to become $x'', z''$. Here, $\theta$ is the walk-off angle and $L$ is the length of each crystal.

where $d$ is the full width at half maximum (FWHM) of the Gaussian intensity profile of the pump, the sinc function is defined as $\text{sinc}(x) = \sin(x)/x$, $L$ is the length of the crystal and the longitudinal mismatch $\Delta_\parallel$ and transverse mismatch $\Delta_\perp$ are given by

$$
\Delta_\parallel = k_\parallel - k_\parallel \cos \theta_\parallel - k_\parallel \cos \theta_\parallel,
\Delta_\perp = k_\perp \sin \theta_\parallel - k_\perp \sin \theta_\parallel,
$$

where $\theta_\parallel, \theta_\perp$ are the scattering angles of the signal and idler photons. Thus, $F(k_s, k_i)$ is the product of two functions, a Gaussian function (or pump envelope function) and a sinc function (or phase matching function). The pump envelope function is determined by the spatial structure of the pump in near field ($x$-space) which depends on the transverse mismatch. The phase matching function depends on the longitudinal mismatch and it is determined by the properties of the crystal [16, 8].

In [6, 7, 15] the anisotropy effects were modeled by introducing an additional dependence on the phase matching function $\Delta_\perp$ in (2). In our present work we derive an expression that models coherent PDC generation in two consecutive crystals. In particular, we consider the trajectories followed by an orthogonally incident pump on the first and second birefringent crystals, which are given by the walk-off angle $\theta$. PDC radiation at certain angles is generated coherently along these trajectories and it interferes.

The TPA for the two-crystal system shown in figure 2 can be calculated by integrating the Hamiltonian over the interaction area, i.e., the area in the nonlinear crystals occupied by the pump. The contribution of the first crystal to the TPA is

$$
F(k_s, k_i) = \int_{-\infty}^{\infty} dx' \int_{0}^{L_s \cos \theta} dz' \exp \left[-\frac{2 \ln 2 (2x')^2}{d^2}\right] \times \exp (i k_s x') \exp \left[-i (k_s x + k_i z')\right] \times \exp \left[-i (k_i x + k_i z)\right],
$$

where $k_{s,x}$ and $k_{s,z}$ are the signal wavevector components in the $x$ and $z$ coordinates and similarly $k_{i,x}$ and $k_{i,z}$ for the idler, and a tilted frame of reference ($x', z'$) has been introduced (figure 2). The second crystal contributes similarly to the TPA, with the replacement $x', z' \rightarrow x'', z''$ and the integration domain in $z''$ being from $L/s\cos \theta$ to $2L/s\cos \theta$. The integration is straightforward after performing the transformations

$$
\begin{align*}
\hat{x'} &= x \cos \theta + x \sin \theta, \\
\hat{y} &= -z \sin \theta + x \cos \theta, \\
\hat{z'} &= (z - L) \cos \theta - (x - \delta) \sin \theta, \\
\hat{z''} &= (z - L) \sin \theta + (x - \delta) \cos \theta,
\end{align*}
$$

where $\delta = L \tan \theta$ (figure 1).

For the compensating case the integration gives

$$
F(k_s, k_i) \propto \exp \left[-\frac{d^2(\Delta_\parallel \cos \theta + \Delta_\perp \sin \theta)^2}{8 \ln 2}\right] \times \exp \left[-\frac{L}{2} \xi \sin \left(\frac{L}{2} \xi\right)\right] + \exp \left[-\frac{d^2(\Delta_\parallel \sin \theta - \Delta_\perp \cos \theta)^2}{8 \ln 2}\right] \times \exp \left(\frac{1}{2} \xi \sin \left(\frac{L}{2} \eta\right)\right),
$$

where $\xi = \Delta_\parallel - \Delta_\perp \tan \theta$ and $\eta = \Delta_\parallel + \Delta_\perp \tan \theta$.

In the non-compensating geometry the second crystal’s optic axis is parallel to the one of the first crystal. Taking this into account is equivalent to replacing $\theta$ by $-\theta$ (see figure 2). The resulting TPA reduces then to the one obtained for a single crystal of length $2L$ instead of two crystals of length $L$ [15].

To summarize, in the presence of anisotropy the TPA is modified in such a way that the pump envelope function and the phase matching function depend on both the transverse mismatch and the longitudinal mismatch. By considering two crystals with the optic axes tilted oppositely we symmetrize this dependence. In what follows we show that this leads to symmetrization of the TPA.

In figure 3 we plot the calculated squared TPAs for one crystal of length $2L$ with the anisotropy taken into account (a) and neglected (b), and then for two crystals, each of length $L$, in the compensating configuration (c). The theoretical predictions are made in terms of scattering angles and use the following parameters: pump wavelength $354.7$ nm, length of each crystal $3$ mm and pump FWHM $70\mu$m, with the latter chosen small enough that it would enhance the anisotropy’s influence on the TPA.

The TPA calculated for one crystal of $6$ mm length is as calculated in [15]. Its corresponding conditional (solid-red) and unconditional (dashed-blue) probability distributions for the signal photons are plotted next to it. Clearly the TPA has a bent structure which is manifested in the asymmetry of its unconditional distribution. This TPA agrees very well with the case of two $3$ mm crystals in the non-compensating configuration, and we omit it for simplicity.

The TPA for the compensating configuration, in which the walk-off angles in the two crystals are opposite, is shown in figure 3(c). From here it is seen that the TPA becomes
Figure 3. Calculated squared TPAs for biphotons emitted via collinear degenerate type-I PDC for a 354.7 nm pump wavelength and 70 µm pump diameter: (a) non-compensated walk-off, one crystal of 6 mm length (or equivalently two consecutive crystals of 3 mm length each), (b) one crystal of 6 mm length, anisotropy neglected, (c) compensated walk-off, two consecutive crystals of 3 mm length each.

symmetric. The similarity between this TPA and the typical symmetric TPA (see figure 3(b)) obtained when the anisotropy has been neglected is promising for the application of this compensation method to the generation of faint SV states free of anisotropy effects. This symmetrization can be qualitatively observed in the angular structure of BSV radiation as will be shown.

3. Experimental setup

Generation of BSV, by means of a setup such as the one sketched in figure 4, provides for us a qualitative, yet very illustrative, experimental demonstration of how the compensating and non-compensating crystal configurations work. We pump a pair of 3 mm length BBO crystals with the third harmonic obtained from a pulsed Nd:YAG laser source which has 1 kHz repetition rate and 18 ps pulse length. The alignment is performed for a collinear degenerate type-I PDC process. We adjust the crystals into the desired configuration (compensating or non-compensating) by rotating one of them by 180° around the propagation axis. This procedure switches the tilt of the optic axis into the same principal plane. In figure 4, the arrows on the crystals show the compensating configuration as seen from top of the setup. Here we are only concerned with the relative tilt of the optic axes. The direction of the arrow associated with the optic axis is related to the sign of the $\chi^{(2)}$ nonlinearity, which also plays a role in the compensation method [17] but here is kept identical in both crystals for simplicity.

The beam, with a shape close to Gaussian, has a diameter of 150 µm FWHM in between these crystals. Although the waist is larger than in our calculations, the effective width reduces greatly due to the large parametric gain and the resulting nonlinear amplification of BSV. The Rayleigh length is 5 cm, which guarantees that the pump in both crystals is collimated. The PDC radiation is detected in the far field, after a 20 cm focal length lens $L$, on a CCD camera. The average pump power used is 40 mW. The CCD camera allows us to have access to the two-dimensional PDC spatial spectrum which gives immediate information on the unconditional probability distribution of the radiation generated inside the interaction volume. Although retrieval of the conditional probability distribution is also possible, we leave it for our future work [18] and only discuss the structure of the spatial spectrum obtained. We are mostly interested in the profile of the spatial spectrum in both crystal configurations, in the principal plane of the crystals.

4. Experimental results for BSV

Figure 5 shows the obtained spatial spectra. The vertical axis in these pictures (labeled as $\theta$) is in the principal plane of the crystals ($x$–$z$ plane), in which anisotropy plays the main role. Figure 5(a) shows the spectrum obtained in the non-compensating configuration and figure 5(b) shows the one obtained in the compensating configuration.

In the non-compensating configuration two maxima displaced along the principal plane of the crystals are observed. At first sight, this distribution is radically different from what is expected 3(a) and observed [6] for two-photon light. The difference is caused by the fact that for high-gain PDC most of the light is produced in the end of the second crystal while SPDC creates biphotons uniformly along the pump path. For this reason, in the presence of anisotropy the spectrum becomes not just asymmetric but non-collinear (the upper maximum in figure 5(a), corresponding to photons produced along the anisotropic pump path). The lower spot in figure 5(a) is formed by the idler counterparts of these photons. One can see the interference pattern formed due to the ‘induced coherence’ effect [19]: the idler radiation from
the first and second crystals partly interferes because some of the corresponding signal modes are common [17].

The spatial spectrum in the compensated configuration case is shown in figure 5(b). Clearly the spectrum becomes symmetric and Gaussian due to the reduction of the walk-off enabling the application of the Schmidt decomposition. A small asymmetry between the spectrum along the principal plane of the crystals and in the orthogonal plane (θy in the picture) still remains. This occurs because PDC radiation, even after anisotropy compensation, comes from an area stretched in the x–z plane (see figure 2). At the same time this area in the y–z plane is as narrow as the original pump beam.

5. Conclusion

We have considered a method of compensating spatial walk-off for SV generation based on replacing a single nonlinear crystal by two consecutive crystals with their optic axes tilted oppositely. Using the near-field consideration, we have calculated the two-photon amplitude and the corresponding conditional and unconditional probability distributions, giving the expected angular distributions of coincidences and single counts for the case of biphoton light. The model confirms the empirical walk-off compensation method that we have already used extensively: the two-photon amplitude gets straightened and the angular distributions of both single counts and coincidences lose their asymmetry caused by the transverse walk-off. We have experimentally observed the spatial walk-off compensation for the case of BSV. Our experimental results show that while in the non-compensated configuration the BSV is only generated along the walk-off direction and in the conjugate direction, in the compensating configuration the BSV angular spectrum becomes symmetric and nearly Gaussian.

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