3D phase-matching conditions for the generation of entangled triplets by $\chi^{(2)}$ interlinked interactions

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Abstract: An analytical calculation of the interaction geometry of two interlinked second-order nonlinear processes fulfilling phase-matching conditions is presented. The method is developed for type-I uniaxial crystals and gives the positions on a screen beyond the crystal of the entangled triplets generated by the interactions. The analytical results are compared to experiments realized in the macroscopic regime. Preliminary tests to identify the triplets are also performed based on intensity correlations.

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1. **Introduction**

The production of multipartite entangled states by means of multiple nonlinear interactions occurring in a single nonlinear crystal has been recently suggested [1, 2, 3] as an alternative to the use of single-mode squeezed states [4, 5, 6] or of two-mode entangled states and linear optical elements [7]. As in the case of bipartite states produced by spontaneous parametric downconversion, such multipartite states display entanglement also in the macroscopic regime in which bright outputs are generated: for this reason they are particularly interesting for all the applications of continuous-variable entanglement [8, 9].

The possibility of realizing the simultaneous phase-matching (PM) of two traveling-wave parametric processes in a single crystal in a seeded configuration has been already demonstrated [10, 11]. In this paper we present the experimental realization of the same interlinked interactions starting from vacuum fluctuations. The output of the nonlinear crystal displays the entire ensemble of inseparable tripartite entangled states satisfying the PM conditions. In order to identify a triplet, we develop a 3D calculation of the characteristics of interlinked PM that reproduce the experimental phenomenology. Finally we perform a preliminary verification of the number of photons conservation law by means of intensity correlations.

2. **Theory**

We consider the Hamiltonian describing the simultaneous PM of a downconversion and an upconversion processes

\[ H_{\text{int}} = \gamma_1 a_4^+ a_4 + \gamma_2 a_5^+ a_3 + h.c. , \]  

where \( \gamma_1 \propto a_4 \) and \( \gamma_2 \propto a_5 \) are coupling coefficients. The two interlinked interactions involve five fields \( a_j \), two of which, say \( a_4 \) and \( a_5 \), enter the crystal and act as non-evolving pumps. When acting on the vacuum as the initial state, \( H_{\text{int}} \) admits the following conservation law

\[ N_1(t) = N_2(t) + N_3(t) , \]  

being \( N_j(t) = \langle a_j^+(t) a_j(t) \rangle \) the mean number of photons in the \( j \)-th mode, and yields a fully inseparable tripartite state [1]. We consider the realization of Eq. (1) in a negative uniaxial crystal in type-I non-collinear PM interaction geometry. The processes must satisfy energy-matching (\( \omega_4 = \omega_1 + \omega_3 \), \( \omega_2 = \omega_3 + \omega_5 \)) and PM conditions (\( k_{\text{eff}}^1 = k_o^1 + k_o^3 \), \( k_{\text{eff}}^2 = k_o^3 + k_o^5 \)), where \( \omega \) are the angular frequencies, \( k \) are the wavevectors and \( o,e \) indicate ordinary and extraordinary field polarizations. In order to investigate the geometrical constraints imposed by the PM conditions, we analytically calculate the output angles of each interacting field.

To simplify calculations, we assume, in the reference frame depicted in Fig. 1, that the two pumps lie in the \( (y,z) \)-plane containing the optical axis (OA) and the normal to the crystal entrance face and that the pump field \( a_4 \) propagates along the normal, \( z \). The solutions corresponding to the effective experimental orientation of the crystal can be obtained by simply calculating the refraction of the beams at the crystal entrance/output faces. Accordingly, the PM conditions for the two interactions simultaneously phase-matched can be written as

\[ k_1 \sin \beta_1 + k_3 \sin \beta_3 = 0 \]  
\[ k_1 \cos \beta_1 \sin \vartheta_1 + k_3 \cos \beta_3 \sin \vartheta_3 = 0 \]  
\[ k_1 \cos \beta_1 \cos \vartheta_1 + k_3 \cos \beta_3 \cos \vartheta_3 = k_4 \]  
\[ k_2 \sin \beta_2 = k_3 \sin \beta_3 \]
the exit face gives: \( \sin \) refraction of the beams at the entrance and exit faces of the crystal. In particular, refraction at being regeneratively amplified at the repetition rate of 500 Hz (High Q Laser Production, Austria). In third-harmonics and the fundamental outputs of a continuous-wave mode-locked Nd:YLF laser, the pump fields were provided by the external angle between the two pumps. Note that not all the calculated output frequencies appear in the experimental part due to the sensitivity of the camera sensor. As the pictures of the experimental outputs were taken on a screen located beyond the crystal normally to the direction of pump \( a_4 \), the calculations had to take into account both the rotation of the crystal and the refraction of the beams at the entrance and exit faces of the crystal. In particular, refraction at the exit face gives: \( \sin \beta_{j,\text{out}} = n_j \sin \beta_j \) and \( \sin \theta_{j,\text{out}} = n_j / (1 - n_j^2 \sin^2 \beta_j)^{1/2} \cos \beta_j \sin \theta_j \).

3. Experiment

For the realization of the interaction described by Eq. (1), the pump fields were provided by the third-harmonics and the fundamental outputs of a continuous-wave mode-locked Nd:YLF laser regeneratively amplified at the repetition rate of 500 Hz (High Q Laser Production, Austria). In the refraction indices of the medium, the wavevectors \( k_j \) are defined as \( k_j = n_j(\omega_j, \hat{k}_j) \omega_j / c \), being \( c \) the speed of light in the vacuum and \( n_j(\omega_j, \hat{k}_j) \) the and \( \beta_j \)’s, angles to \((y,z)\)-plane; \( \alpha \), angle to the optical axis (OA).

\[
\begin{align*}
 k_2 \cos \beta_2 \sin \theta_2 &= k_3 \cos \beta_3 \sin \theta_3 + k_5 \sin \theta_5 \quad (7) \\
 k_2 \cos \beta_2 \cos \theta_2 &= k_3 \cos \beta_3 \cos \theta_3 + k_5 \cos \theta_5 \quad (8)
\end{align*}
\]

where the angles are defined as in Fig. 1. The wavevectors \( k_j \) are defined as \( k_j = n_j(\omega_j, \hat{k}_j) \omega_j / c \), being \( c \) the speed of light in the vacuum and \( n_j(\omega_j, \hat{k}_j) \) the refraction indices of the medium. Note that \( \cos \varphi = \cos \beta_2 \cos(\vartheta_2 - \alpha) \) and \( \beta_5 = \beta_4 = \vartheta_4 = 0 \).

For fixed frequencies and propagation directions of the pump fields, we are left with 11 variables \( (\omega_1, \omega_2, \omega_3, \vartheta_1, \vartheta_2, \vartheta_3, \vartheta_5, \beta_1, \beta_2, \beta_3, \alpha) \) and 8 equations only (energy conservation and Eqs. (3) to (8)). The problem can thus be solved by choosing three of the variables (say \( \vartheta_1, \vartheta_5 \) and \( \alpha \)) as parameters. The Appendix contains the algebraic procedure to analytically solve the problem. In order to efficiently handle the dependence of the solutions on the free parameters, we implemented the calculation by using the software Mathematica (Wolfram Research, IL).

In Fig. 2 we show one picture of the movie containing the comparison between measured and calculated results for the output of the crystal as a function of the tuning angle \( \alpha \) for a fixed value of the external angle between the two pumps. Note that not all the calculated output frequencies appear in the experimental part due to the sensitivity of the camera sensor. As the pictures of the experimental outputs were taken on a screen located beyond the crystal normally to the direction of pump \( a_4 \), the calculations had to take into account both the rotation of the crystal and the refraction of the beams at the entrance and exit faces of the crystal. In particular, refraction at the exit face gives: \( \sin \beta_{j,\text{out}} = n_j \sin \beta_j \) and \( \sin \theta_{j,\text{out}} = n_j / (1 - n_j^2 \sin^2 \beta_j)^{1/2} \cos \beta_j \sin \theta_j \).
Fig. 2. (1.271 MB) Movie showing the variation of the measured and calculated outputs of the crystal as a function of the tuning angle $\alpha$ for fixed external angle between the pumps. Left panel: picture of the output of the crystal taken with a commercial digital photocamera (no color correction applied). LHS: a portion of the downconversion cones. RHS: output of the upconversion process. Right panel: calculated output of the upconversion process.

In particular, the third-harmonic field ($\lambda_4 = 349$ nm, $\sim 4.45$ ps pulse duration) was used as the $a_4$ field producing the downconversion cones and the fundamental field ($\lambda_5 = 1047$ nm, $\sim 7.7$ ps pulse duration) as the $a_5$ field pumping the upconversion process.

As depicted in Fig. 3 (a), both pumps were focused and injected into a $\beta$-BaB$_2$O$_4$ crystal (BBO, Fujian Castech Crystals, China, 10 mm $\times$ 10 mm cross section, 4 mm thickness) cut for type-I interaction ($\theta_{\text{cut}} = 34$ deg) at the angle $\theta_{5,\text{ext}} = -34.8$ deg with respect to each other. The required superposition of the two pumps in time was obtained with a variable delay line. For alignment purposes, we used the light emitted by a He-Ne continuous-wave laser (Melles-Griot, etc.).

![Diagram of experimental setup and visible output](image_url)
CA, 5 mW max output power) to seed the process at $\lambda_1 = 632.8$ nm. The beam was collimated and sent to the BBO in the plane containing the pumps at the external angle $\vartheta_{1,\text{ext}} = -2.54$ deg with respect to $a_4$. The seeded interactions produced two new fields: $a_3$ ($\lambda_3 = 778.2$ nm, $\vartheta_{3,\text{ext}} = 3.35$ deg) generated as the difference-frequency of $a_4$ and $a_1$, and $a_2$ ($\lambda_2 = 446.4$ nm, $\vartheta_{2,\text{ext}} = -12.78$ deg) generated as the sum-frequency of $a_3$ and $a_5$. The pump fields were sufficiently intense so as to observe the process starting from vacuum fluctuations: as shown in the picture in Fig. 3b), on a white screen located beyond the crystal it was possible to see both the tunable bright downconversion cones and the two polychromatic half-moons-shaped states generated by the upconversion process together with the spots of the fields generated by the seeded interactions.

The conservation law in Eq. (2) implies strong intensity correlations among the generated fields which are necessary but not sufficient to demonstrate the entangled nature of the triplet. We realized intensity-correlation measurements by filtering each portion of light in frequency and aligning three pin-holes of suitable diameters on the spots of the seeded process. Note that the seeding He-Ne light was switched-off once completed the alignment. The light selected by the pin-holes was focused on three p-i-n photodiodes (two, $D_{1,2}$ in Fig. 3a), S5973-02 and one, $D_3$, S3883, Hamamatsu, Japan). The overall detection efficiencies in the three detection arms were $\eta_1 = 0.44$, $\eta_2 = 0.72$ and $\eta_3 = 0.43$. Each current output was integrated by a synchronous gated-integrator (SGI in Fig. 3a)) in external trigger modality, digitized by a 13-bit converter (SR250, Stanford Research Systems, CA) and recorded in a PC-based multichannel analyzer. Data acquisition and analysis were performed with software LabView (National Instruments, TX). We evaluated the following correlation function on subsequent laser shots ($k$)

$$\Gamma(k) = \frac{\langle m_1(i) - \langle m_1 \rangle \rangle \langle m_2(i+k) + m_3(i+k) - \langle m_2 + m_3 \rangle \rangle}{\sigma(m_1)\sigma(m_2+m_3)}. \quad (11)$$

in which $m_j$ is the number of detected photons and $\sigma(x) = (\langle x^2 \rangle - \langle x \rangle^2)^{1/2}$ is the standard deviation. It can be demonstrated [12] that in the case of bright triplets the correlation coefficient $\varepsilon \equiv \Gamma(0)$ must approach unity. The mean values of the detected photons in the three parties of the triplet were $M_1 = 1.082 \times 10^8$, $M_2 = 2.3 \times 10^6$ and $M_3 = 1.115 \times 10^8$ and the measured correlation coefficient was $\varepsilon = 0.916$.

4. Conclusions

The perfect agreement between calculations and experimental results demonstrates the correctness of the developed model. This enables us to forecast different interaction schemes that in the future will allow to overcome the experimental difficulties experienced in the present setup. In fact, the reason why the measured correlation coefficient is less than unity could be the imperfect selection of the triplet and/or the presence of spurious light in the same location.

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Appendix

We describe the analytical procedure to solve the equation system (3)-(8).
From the first of Eqs. (10) we write
\[ k_2 = \left[G_2 \cos^2(\vartheta_2 - \alpha) \cos^2 \beta_2 + L_2 \right]^{-1/2}, \]  
where \( G_2 = c^2/\omega_2^2 \left[1/n_2^2(\omega_2) - 1/n_2^2(\omega_2)\right] = 1/k_{2,2}^2 - 1/k_{2,2}', \) and \( L_2 = c^2/\omega_2^2 1/n_2^2(\omega_2) = 1/k_{2,2}'. \) By squaring and summing Eqs. (3), (4) and (5) and defining \( A = (k_3^2 + k_3 - k_1^2)/(2k_3k_4), \) we easily get
\[ \cos \beta_3 = \frac{A}{\cos \vartheta_3}, \]  
whereas by squaring and summing Eqs. (6), (7) and (8) and Eqs. (7) and (8) we get
\[ k_2^2 = k_1^2 + k_3^2 + 2k_3k_5 \cos \beta_3 \left(\sin \vartheta_3 \sin \vartheta_5 + \cos \beta_3 \cos \vartheta_5\right) \]  
and
\[ k_2^2 \cos^2 \beta_2 = k_2^3 \cos^2 \beta_3 + k_2^3 + 2k_3k_5 \cos \beta_3 \left(\sin \vartheta_3 \sin \vartheta_5 + \cos \beta_3 \cos \vartheta_5\right). \]  
By eliminating \( k_2 \) from Eqs. (14) and (15) and using Eq. (13) we find
\[ \cos^2 \beta_2 = \frac{k_2^2 A^2 \tan^2 \vartheta_3 + k_2^2 + 2k_3k_5 A \left(\tan \vartheta_3 \sin \vartheta_5 + \cos \vartheta_5\right)}{k_2^3 + k_3^2 + 2k_3k_5 \left(\tan \vartheta_3 \sin \vartheta_5 + \cos \vartheta_5\right)}. \]  
From Eq. (3) we obtain
\[ \sin \beta_1 = -\frac{k_3}{k_1} \sin \beta_3. \]  
that, once substituted into Eq. (4) together with Eq. (13), gives
\[ \sin \vartheta_1 = -\frac{k_3 A}{\sqrt{k_1^2 - k_3^2 \sin^2 \beta_3}}. \]  
We now divide Eq. (7) by Eq. (8) and use Eq. (13)
\[ \tan \vartheta_2 = \frac{k_3 A \tan \vartheta_3 + k_5 \sin \vartheta_5}{k_3 A + k_3 \cos \vartheta_5}. \]  
By squaring Eq. (12) and inserting Eq. (14) and Eq. (16) into it we get
\[ \cos^2(\vartheta_2 - \alpha) = \frac{1 - L_2 \left[k_3^2 + k_3^2 + 2k_3k_5 \cos \beta_3 \left(\sin \vartheta_3 \sin \vartheta_5 + \cos \beta_3 \cos \vartheta_5\right)\right]}{G_2 \left[k_3^2 \cos^2 \beta_3 + k_3^2 + 2k_3k_5 \cos \beta_3 \left(\sin \vartheta_3 \sin \vartheta_5 + \cos \beta_3 \cos \vartheta_5\right)\right]} \]  
By substituting Eq. (13) into Eq. (20), expanding \( \cos^2(\vartheta_2 - \alpha) \) in Eq. (20) and using Eq. (19) to eliminate \( \vartheta_2, \) after some simplifications we get a quadratic algebraic equation for the variable \( \tan \vartheta_3, \)
\[ a \tan^2 \vartheta_3 + b \tan \vartheta_3 + c = 0, \]  
whose coefficients are
\[ a = G_2 A^2 k_3^2 \sin^2 \alpha \]  
\[ b = 2G_2 A k_3 (A k_3 + k_5 \cos \vartheta_5) \sin \alpha \cos \alpha + 2G_2 A k_3 k_5 \sin \vartheta_5 \sin^2 \alpha + 2L_2 A k_3 k_5 \sin \vartheta_5 \]  
\[ c = G_2 (A k_3 + k_5 \cos \vartheta_5)^2 \left(\cos^2 \alpha - \sin^2 \alpha\right) + 2G_2 k_5 \sin \vartheta_5 (A k_3 + k_5 \cos \vartheta_5) \sin \alpha \cos \alpha \]  
\[ + G_2 (A^2 k_3^2 + k_3^2 + 2A k_3 k_5 \cos \vartheta_5) \sin^2 \alpha + L_2 (k_3^2 + k_3^2 + 2A k_3 k_5 \cos \vartheta_5) - 1. \]  
Once solved Eq. (21), we find \( \vartheta_3 \) and then all the other variables as a function of the free parameters \( \omega_1, \alpha \) and \( \vartheta_5. \)