Journal of Modern Optics

Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713191304

Generation of entangled four-photon states by parametric down-conversion

Surya P. Tewari [a]; P. Hariharan [b]
[a] School of Physics, University of Hyderabad, Hyderabad, India
[b] School of Physics, University of Sydney, New South Wales, Australia

Online Publication Date: 01 March 1997
To cite this Article: Tewari, Surya P. and Hariharan, P. (1997) 'Generation of entangled four-photon states by parametric down-conversion', Journal of Modern Optics, 44:3, 543 - 553
To link to this article: DOI: 10.1080/09500349708232920
URL: http://dx.doi.org/10.1080/09500349708232920

JOURNAL OF MODERN OPTICS, 1997, VOL. 44, NO. 3, 543–553

Generation of entangled four-photon states by parametric down-conversion

SURYA P. TEWARI
School of Physics, University of Hyderabad, Hyderabad 500 046, India

and P. HARIHARAN
School of Physics, University of Sydney, New South Wales 2006, Australia

(Received 22 May 1996)

Abstract. Pairs of photons are commonly generated in an entangled state by parametric down-conversion of an ultraviolet pump beam in a nonlinear crystal. We show how this process can be extended to generate entangled four-photon states from two pump photons by using two non-collinear pump beams.

1. Introduction

The generation of correlated photon pairs has made possible a number of tests of quantum mechanics using two-photon interferometry, including tests of Bell's inequality [1–3]. However, a shortcoming of these experiments has been the need to assume that the pairs of photons that are detected are representative of the entire ensemble [4, 5]. One way to close this loophole is through experiments involving entangled states of three or more photons [6]. However, so far no technique has emerged to generate significant signal for such multiphoton states.

Pairs of photons in an entangled state are commonly generated by parametric down-conversion [7–9]. In this process, a single ultraviolet photon spontaneously decays into two photons with frequencies close to half the ultraviolet frequency. The efficiency of the down-conversion process is increased by using a birefringent crystal to achieve phase matching at the two wavelengths. We present, in this paper, an extension of this method by which a non-centrosymmetric crystal can be used with two non-collinear pump beams to generate a variety of entangled four-photon states by parametric down-conversion of two pump photons.

2. Phase matching

Generation of four-photon entangled states from two pump photons is possible by achieving the required phase-matching condition with two non-collinear pump beams. We consider two pump beams of radian frequency ω_p with coplanar wave-vectors $\mathbf{K}_1$ and $\mathbf{K}_2$, inclined to one another at an angle $2\theta_p$ and assume that, as shown in figure 1, four photons are emitted, each with radian frequency ω_a and wave-vector $\mathbf{K}_a$, along the axis of symmetry. These four photons are produced by absorption of two pump photons, one from each pump beam. The longitudinal

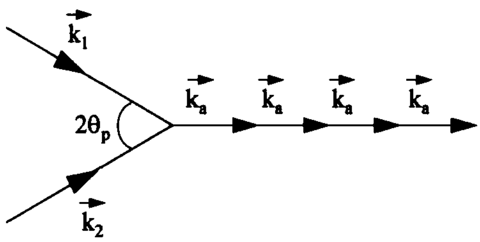

Figure 1. Arrangement using two non-collinear pump beams to achieve phase matching for generating entangled four-photon states by parametric down-conversion.

phase-matching condition, along the direction of the symmetry axis in figure 1, is then satisfied for an angle θ_p if

$$\cos \theta_p = \frac{n(\omega_a)}{n(\omega_p)}, \tag{1}$$

where $n(\omega_a)$ is the refractive index of the crystal at the radian frequency ω_a of the degenerate down-converted photons, and $n(\omega_p)$ is the refractive index of the crystal at the radian frequency ω_p of the pump photons. The requirement for transverse phase matching does not impose any additional condition for such degenerate photons. Since, for normal dispersive media,

$$n(\omega_p) > n(\omega_a), \tag{2}$$

it follows from equation (1) that the angle θ_p is physically realizable.

The use of inclined beams for phase-matching has been studied earlier in coherent anti-Stokes Raman scattering [10], and in second-harmonic generation [11]. An extension is the use of conical beams in second-harmonic generation [12] and in third-harmonic generation [13], for which a detailed theory has recently been given [14, 15]. However, the use of non-collinear pump beams for the generation of a four-photon field has not been discussed so far.

In an anisotropic crystal, attention has to be paid to the polarization of the pump photons and the down-converted photons. Note that anisotropic crystals behave as isotropic media for the ordinary (O) polarization, as the normal surfaces for different frequencies are concentric spherical surfaces. Consequently, the longitudinal mismatch between the wave-vectors $\mathbf{K}_1$, $\mathbf{K}_2$ and $\mathbf{K}_a$, can be circumvented by choosing an appropriate angle θ_p for any orientation of the crystal, provided that the polarization state of the pump and the down-converted photons corresponds to the O ray; which means taking $n(\omega_a) = n^o(\omega_a)$, and $n(\omega_p) = n^o(\omega_p)$ in equation (1), where the superscript o refers to the O ray.

As an example, we consider a uniaxial crystal (of arbitrary thickness) cut with its optic axis parallel to its faces. The pump beams are incident on the crystal face with their polarization perpendicular to the optic axis and behave as O rays inside the crystal. Two such pump beams, at angles $\pm\theta_p$ as shown in figure 1, produce, in the direction normal to the faces, four degenerate photons from two pump photons, one from each of the two oblique pump beams. For a KH_2PO_4 crystal, and an output wavelength $\lambda_a = 2\lambda_p = 600\,\text{nm}$, $n_p^o = 1{\cdot}545\,570$, $n_a^o = 1{\cdot}509\,274$

Pump beam angle θ_p for KH_2PO_4 for different polarizations.

ω_a	ω_p	Equation determining θ_p	Condition	θ_p for $\lambda_a = 2\lambda_p = 600$ nm (degrees)
O ray	E ray	$\tan^2\theta_p = \frac{n_o^2(w_p)}{n_o^2(w_a)} - \frac{n_o^2(w_p)}{n_e^2(w_p)}$	$n_e(w_p) > n_o(w_a)$	Not possible
E ray	E ray	$\tan^2\theta_p = \frac{n_o^2(w_p)}{n_e^2(w_a)} - \frac{n_o^2(w_p)}{n_e^2(w_p)}$	$n_e(w_p) > n_e(w_a)$	11·816 29
E ray	O ray	$\cos\theta_p = \frac{n_e(w_a)}{n_o(w_p)}$	$n_o(w_p) > n_e(w_a)$	18·197 78

[16], the angle $\theta_p = 12{\cdot}416°$. The polarization of the four down-converted photons is the same as the pump photons and is perpendicular to the optic axis.

Similar examples can be constructed by choosing both pump and down-converted beams with extraordinary (E) polarization, or pump and down-converted beams with O and E polarizations respectively. The resultant values of the angle θ_p for these combinations of polarizations are summarized in the table. Note that negative as well as positive crystals can be used in this fashion.

While we have considered, in the first instance, the degenerate case, the four photons generated by two photons from the two oblique pump beams need not necessarily be degenerate in frequency. The other possibilities are as follows: case (A), three degenerate photons, $3(\omega_a)$ and one with a different frequency, ω_b; case (B), two pairs of degenerate photons, $2(\omega_a), 2(\omega_b)$; case (C), two degenerate and two different, $2(\omega_a), \omega_b, \omega_c$; case (D), all different, $\omega_a, \omega_b, \omega_c, \omega_d$. In contrast with the case of four degenerate photons, the down-converted photons in the other cases can be emitted at angles to the symmetry axis of figure 1. Clearly the angles in each case depend on the crystal properties, as well as the combinations of wavelengths and polarizations of the relevant beams.

In order to get an insight into these cases involving off-axis emission, we consider a representative case, namely case (A), involving three degenerate photons and one with a different frequency, $3(\omega_a), \omega_b$. In such a case, two distinct conditions have to be satisfied: the condition for longitudinal phase matching, and that for transverse phase matching. The longitudinal phase-matching condition relates the components of the wave-vectors on the symmetry axis of figure 1. For case (A) we have

$$2\omega_p n_p \cos\theta_p = 3n_a\omega_a \cos\theta_a + n_b\omega_b \cos\theta_b, \qquad (3)$$

where θ_a and θ_b are the angles of the wave-vectors $\mathbf{K}_a$ and $\mathbf{K}_b$ (figure 2 (*a*), diagram (Ia)), with the symmetry axis. In general, the three photons of frequency ω_a can emerge with the wave-vectors $\mathbf{K}_a^{(1)}$, $\mathbf{K}_a^{(2)}$ and $\mathbf{K}_a^{(3)}$, at three different angles θ_a^i, $i = 1, 2, 3$, but, for simplicity, we have set $\theta_a = \theta_a^i$, $i = 1, 2, 3$ in equation (3). From the values of n_a^o and n_b^o and the requirement for energy conservation, we find a range $\theta_a^{min} < \theta < \theta_a^{max}$ in which it is possible, for a value of θ_p determined by equation (1), to obtain

$$|\cos\theta_a| \leqslant 1. \qquad (4)$$

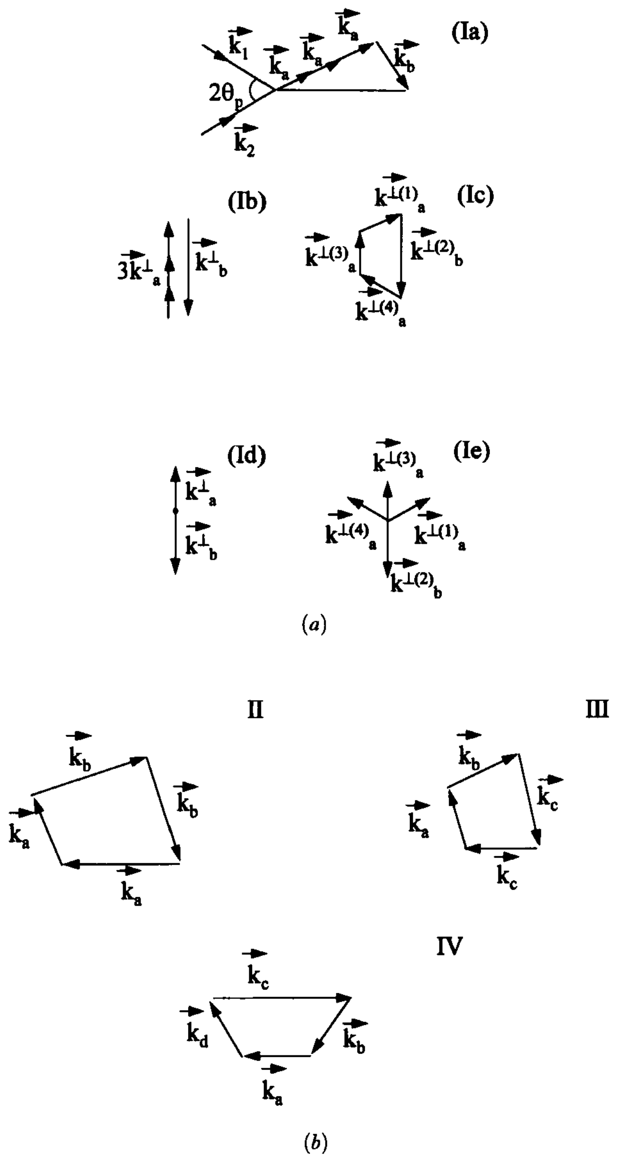

Figure 2. Longitudinal and transverse phase-matching diagrams for (*a*) the degenerate case and (*b*) the cases (B), (C) and (D) discussed in the text.

In this case equation (4) may be satisfied in a crystal when $\omega_a > \omega_p/2$, as well as when $\omega_a < \omega_p/2$, with $\omega_b = 2\omega_p - 3\omega_a$.

The transverse phase-matching condition, which relates the transverse components of the wave-vectors, is satisfied if

$$\mathbf{K}_a^{\perp(1)} + \mathbf{K}_a^{\perp(2)} + \mathbf{K}_a^{\perp(3)} + \mathbf{K}_b^{\perp} = \mathbf{0}. \tag{5}$$

This equation involves four two-dimensional vectors in the plane transverse to the symmetry axis of figure 1, representing the transverse components of the four wave vectors. In general, equation (5) represents a quadrilateral. The vector $\mathbf{K}_b^{\perp}$ may be assumed to be along the x axis ($\phi_b = 0$), and the azimuths for the vectors corresponding to the three degenerate photons have values $\phi_a^{(1)}, \phi_a^{(2)}$ and $\phi_a^{(3)}$. If we take into account the fact that the normal surfaces for O rays are spherical, it is evident that several combinations of these three azimuths are possible for equation (5) to be satisfied as long as

$$|3\mathbf{K}_a^{\perp}| > |\mathbf{K}_b^{\perp}|. \tag{6}$$

It follows that the three degenerate photons in case (A) can be either collinear, as in figure 2 (*a*), diagram (Ia), or non-collinear. In the former case, the **K** vectors of the four photons are coplanar as in figure 2 (*a*), diagram (Ib), while in the latter case they are not coplanar, as shown in figure 2 (*a*), diagram (Ic). To visualize the directions of the wave-vectors of the emerging four photons, one may draw the transverse vectors through the origin, parallel to the sides of the quadrilateral defined by equation (5), as shown in figure 2 (*a*), diagrams (Id) and (Ie). for figure 2 (*a*), diagrams (Ib) and (Ic), respectively. The tips of these vectors in the transverse plane may be taken to represent the directions in which the four photons emerge from the crystal.

Case (A) which has been discussed has implied some restrictions. These restrictions can be relaxed and the consequent changes enumerated. It is of interest to note, here, that a rotation of figures 2 (*a*), diagrams (Id) and (Ie) about the symmetry axis is allowed without changing the polarization of the waves, while satisfying equations (3) and (5). Thus the possible **K** vectors of the emerging photons corresponding to each frequency lie on a cone, even though the incident pump beams are confined to a plane. These characteristics change if one chooses other combinations for phase matching such as E–E, or E–O, instead of the O–O combination which we have considered earlier for simplicity.

The possibilities for phase matching in the cases (B), (C) and (D), can be considered using quadrilaterals of the types II, III and IV, shown in figure 2 (*b*), and an equation similar to equation (3).

3. Entangled four-photon states

It is clear from the five different cases, including the degenerate case, considered above, that a variety of entangled four-photon states are available from the two-photon down-conversion process. The four-photon field is an ensemble of the superpositions of the states:

(i) $|4(\omega_a, \mathbf{K}_a)\rangle$,
(ii) $|3(\omega_a), \mathbf{K}_a^i, i = 1, 2, 3; \omega_b, \mathbf{K}_b\rangle$,
(iii) $|2(\omega_a), \mathbf{K}_a^i, i = 1, 2; 2(\omega_b), \mathbf{K}_b^j, j = 1, 2\rangle$,
(iv) $|2(\omega_a), \mathbf{K}_a^i, i = 1, 2; \omega_b, \mathbf{K}_b; \omega_c, \mathbf{K}_c\rangle$,
(v) $|\omega_a, \mathbf{K}_a; \omega_b, \mathbf{K}_b; \omega_c, \mathbf{K}_c; \omega_d, \mathbf{K}_d\rangle$.

It follows that, depending on the direction of the wave-vectors and the polarizations and frequencies of the emerging photons, one can create or select a variety of correlated pairs of photon-pair states, three-photon states and four-photon states. Such a selection can be made for example, by using downstream from the exit face of the crystal firstly one, two, three or four pinholes, secondly narrow-band frequency filters and thirdly spatial filters to select a narrow range of values of **K** of the emerging photons.

In view of the novelty and possible applications of such a two-photon down-converter, we use the next four sections to establish that such a source can be realized with a reasonable signal strength. For brevity, we shall limit ourselves to the degenerate four-photon case.

4. Semiclassical gain formalism

The intensity of the four-photon field generated by the crystal depends on the magnitude of the nonlinear susceptibility involved in its production, which can be written as [17]

$$\chi^{\mathrm{NL}}(-\omega_{\mathrm{p}}-\omega_{\mathrm{p}};\omega_{\mathrm{a}},\omega_{\mathrm{a}},\omega_{\mathrm{a}})\mathscr{E}_{\mathrm{p}}^{*}\mathscr{E}_{\mathrm{p}}^{*}\mathscr{E}_{\mathrm{a}}^{3}, \tag{7}$$

where $\mathscr{E}$ represents the slowly varying envelope of the electric field E given by

$$E=\mathscr{E}\exp(-\mathrm{i}\omega t+\mathrm{i}\mathbf{K}\cdot\mathbf{r})+\mathscr{E}^{*}\exp(+\mathrm{i}\omega t-\mathrm{i}\mathbf{K}\cdot\mathbf{r}),$$

and the subscripts are self-explanatory. The susceptibility described by equation (7), as a first guess based on perturbation theory, is comparable with the product of $\mathscr{E}_{\mathrm{a}}$ and the square of the susceptibility:

$$\chi^{\mathrm{NL}}(-\omega_{\mathrm{p}};\omega_{\mathrm{a}})\mathscr{E}_{\mathrm{p}}^{*}\mathscr{E}_{\mathrm{a}}, \tag{8}$$

involved in the production of two down-converted photons in the usual parametric process. The growth of the field amplitude due to equation (7), in the approximation of undepleted pump waves and perfect phase matching, is given by the relation

$$\mathscr{E}_{\mathrm{a}}^{2}=\frac{\mathscr{E}_{\mathrm{a}}^{02}}{1-\mathscr{E}_{\mathrm{a}}^{02}(2\pi\omega_{\mathrm{p}}/c)\chi_{\mathrm{I}}^{(4)}\mathscr{E}_{\mathrm{p}}^{2}z}, \tag{9}$$

while the field amplitude due to equation (8) under a similar approximation [18]† is given by

$$\mathscr{E}_{\mathrm{a}}^{2}=\mathscr{E}_{\mathrm{a}}^{02}\exp\left(\frac{2\pi\omega_{\mathrm{p}}}{c}\chi_{\mathrm{I}}^{(2)}\mathscr{E}_{\mathrm{p}}z\right). \tag{10}$$

Note that, while deriving equations (9) and (10), we have assumed, for simplicity, that the field amplitudes are real, with $\chi^{\mathrm{NL}}=\mathrm{i}\chi_{\mathrm{I}}^{(4,2)}$, $\mathscr{E}_{\mathrm{a}}^{0}$ is the initial value of the down-converted field amplitude at a distance $z=0$, in the crystal, along the direction of symmetry, and c is the velocity of light. A short-hand notation has also been adopted for the susceptibilities defined by equations (7) and (8). We find that there is a significant difference in the growth of $\mathscr{E}_{\mathrm{a}}^{2}$ in the two cases. The gain coefficient given by equation (9) is

$$\chi_{\mathrm{I}}^{(4)}\mathscr{E}_{\mathrm{p}}^{2}\mathscr{E}_{\mathrm{a}}^{02}, \tag{11}$$

† The degenerate case with real field amplitudes should be taken.

while that given by equation (10) is

$$\chi_{\mathrm{I}}^{(2)}\mathscr{E}_{\mathrm{p}}. \tag{12}$$

In the non-depleted pump approximation, the amplitude of the down-converted four-photon field is always finite although much weaker than the amplitude of the two-photon field.

5. Initiation by spontaneous emission

The semiclassical result (9) for the generation of the field amplitude $\mathscr{E}_{\mathrm{a}}$, necessitates a non-zero value $\mathscr{E}_{\mathrm{a}}^{0}$ at the entrance face of the crystal. The generation of these initial photons from the vacuum can be explained only by spontaneous emission at the down-converted frequency (see [19] for the usual down-conversion process; see also [20]). The full quantum-mechanical properties of these spontaneously generated photons will be discussed elsewhere. It is sufficient, for the present, to mention that the second derivative of the number operator N_{a} for the four-photon field, in the degenerate case, is given by the relation

$$\begin{aligned}\frac{\mathrm{d}^2}{\mathrm{d}t^2}N_{\mathrm{a}} = \left(\frac{4\chi^{\mathrm{NL}}}{\hbar}\right)^2 &[12N_{\mathrm{v}}^{0}(N_{\mathrm{v}}^{0}-1)+4(7N_{\mathrm{v}}^{02}-7N_{\mathrm{v}}^{0}+3)N_{\mathrm{a}} \\ &+(12N_{\mathrm{v}}^{02}-62N_{\mathrm{v}}^{0}-5)N_{\mathrm{a}}^{2}+2(4N_{\mathrm{v}}^{02}-4N_{\mathrm{v}}^{0}+15)N_{\mathrm{a}}^{3}-10N_{\mathrm{v}}^{0}N_{\mathrm{a}}^{4}+3N_{\mathrm{a}}^{5}].\end{aligned} \tag{13}$$

It follows that even for vanishing values of $N_{\mathrm{a}}(t=0)$ and $(\mathrm{d}/\mathrm{d}t)N_{\mathrm{a}}(t=0)$ there is a parabolic increase with time of the number operator N_{a} for $N_{\mathrm{v}}^{0} \geqslant 2$, where N_{v}^{0} is the number of photons in the state describing the pump field in the region of interaction. Thus the four-photon field can also be initiated by spontaneous emission.

6. Pulsed pump beams and signal accumulation

The parametric two-photon down-conversion process differs from the normal down-conversion process in two ways: firstly in the manner in which the phase-matching condition is achieved, and secondly in the fact that the gain depends on the second power of the pump amplitude $\mathscr{E}_{\mathrm{p}}$. The latter fact is very important, because it opens up the possibility of signal accumulation by using pulsed pump beams with a very high peak power. The use of such a pulsed pump beam in a process yielding two down-converted photons does not result in any increase in the output, because of the linear dependence of the gain on the amplitude of the pump. The output is therefore proportional to the average pump power, with pulsed as well as with continuous-wave excitation.

However, in a two-photon down-conversion process where the gain is proportional to the second power of the pump amplitude, the output obtained with high-intensity pulses can be several orders of magnitude greater than that obtained with continuous-wave excitation at the same average power.

7. Down-conversion susceptibility and resonance enhancement

It is worthwhile at this stage to make an approximate estimate of the magnitude of the susceptibilities for the generation of four-photon states and two-photon states by parametric down-conversion. From lowest-order perturbation theory, the susceptibility for the generation of four-photon states is given by equation (7) and can be written as [17]

$$\chi_{\mathrm{I}}^{(4)}\mathscr{E}_{\mathrm{p}}^{*2}\mathscr{E}_{\mathrm{a}}^{3}$$
$$\sim \sum_{m_i, i=1,\dots,5}\left(\frac{-\mathrm{i}}{\hbar}\right)^5 \frac{S_1(mm')S_2(mm')d_{gm_5}d_{m_5m_4}\mathscr{E}_{\mathrm{a}}d_{m_4m_3}\mathscr{E}_{\mathrm{a}}d_{m_3m_2}\mathscr{E}_{\mathrm{a}}d_{m_2m_1}\mathscr{E}_{\mathrm{p}}^{*}d_{m_1g}\mathscr{E}_{\mathrm{p}}^{*}}{\Delta_{m_5g}\Delta_{m_4g}\Delta_{m_3g}\Delta_{m_2g}\Delta_{m_1g}}. \quad (14)$$

Similarly the susceptibility for the generation of two-photon states, which is given by equation (8), can be written as

$$\chi_{\mathrm{I}}^{(2)}\mathscr{E}_{\mathrm{p}}^{*}\mathscr{E}_{\mathrm{a}} \sim \sum_{m_1,m_2}\left(\frac{-\mathrm{i}}{\hbar}\right)^2 \frac{S_1(mm')d_{gm_2}d_{m_2m_1}\mathscr{E}_{\mathrm{a}}d_{m_1g}\mathscr{E}_{\mathrm{p}}^{*}}{\Delta_{m_2g}\Delta_{m_1g}}. \quad (15)$$

The summations in equations (14) and (15) are over the complete set of states of the quantum-mechanical system, while d_{mn} and Δ_{mn} represent respectively, the dipole matrix element and the detuning between the states $|m\rangle$ and $|n\rangle$, and $\hbar$ is Planck's constant. The appearance of a larger number of detunings Δ_{mn} in the denominator, as well as a larger number of small dipole matrix elements d_{mn} in the numerator of equation (14), when compared with equation (15), make the susceptibility defined by equation (14) significantly smaller than that defined by equation (15). The term $S(mm')$, which is left undefined mathematically, may be taken operationally as the vertex, in perturbation theory, that mixes the even- and odd-parity states in a non-centrosymmetric medium. Without such an operation the medium is centrosymmetric and $\chi^{(2)}$ is zero. Note that it is necessary to have such an operation twice in evaluating the susceptibility defined by equation (14). This becomes clear by analysing the time-ordered diagram in figure 3 (*a*). The summation over different time orderings of the field amplitude is also implied in the summation processes defined by equations (14) and (15). We represent a

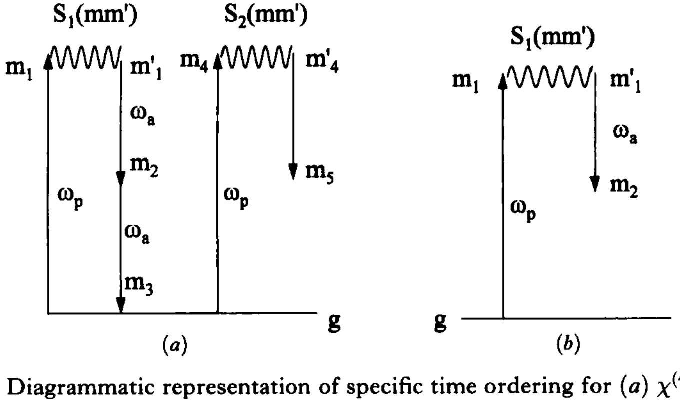

Figure 3. Diagrammatic representation of specific time ordering for (*a*) $\chi^{(4)}$ and (*b*) $\chi^{(2)}$.

particular time-ordered process in figure 3 (*a*) with the convention that time increases as we read from left to right. The vertical arrows imply absorption of a photon, the downward arrows imply emission of a photon, and the horizontal zigzag represents the parity mixing operation $S(mm')$, at the vertex joining $m - m'$. We note that the dominant processes of absorption and emission take place in the dipole approximation, and the phase-matching condition is satisfied only when two photons, one from each beam, are involved in the process leading to the emission of four photons. Now, in the diagram in figure 3 (*a*) for the summation over m_3, the state m_3 can be replaced by the ground state g. Thus one may have a resonant enhancement of the two-photon down-conversion process under the proper phase-matching conditions ($\Delta_{m_3g} \to 0$). Using the above analysis for the time-ordered processes described by figures 3 (*a*) and 3 (*b*), we can approximate equation (14) as

$$\begin{aligned}\chi_{\rm I}^{(4)}\mathscr{E}_{\rm p}^{*2}\mathscr{E}_{\rm a}^{3} &= \left[\sum_{m_1m_2}\left(\frac{-{\rm i}}{\hbar}\right)^2\frac{S_1(mm')d_{gm_2}d_{m_2m_1}\mathscr{E}_{\rm a}d_{m_1g}\mathscr{E}_{\rm p}^*}{\Delta_{m_2g}\Delta_{m_1g}}\right]^2\frac{\mathscr{E}_{\rm a}}{\hbar\Delta_{m_3g}}\\ &= (\chi_{\rm I}^{(2)})^2\frac{\mathscr{E}_{\rm p}^{*2}\mathscr{E}_{\rm a}^{3}}{\hbar\Delta_{m_3g}}. \end{aligned} \tag{16}$$

Equation (16) suggests that a down-conversion process generating four-photon states can have near resonance features, and the susceptibility $\chi_{\rm I}^{(4)}$ is likely to have a much larger value than the small value expected by simply looking at the expression obtained from perturbation theory.

If we use in equation (16) the example of $LiNbO_3$ [21],

$$\chi_{\rm I}^{(2)}(-6\times10^{14}\ {\rm Hz}, 3\times10^{14}\ {\rm Hz})\mathscr{E}_{\rm p} = 2\times10^{-16}\ {\rm MKS}, \tag{17}$$

at an incident pump power of $5\ {\rm MW\ cm^{-2}}$, and a pump frequency $\omega_{\rm p} = 2\omega_{\rm a} = 1{\cdot}2\pi\times10^{15}\ {\rm rad\ s^{-1}}$, one may write

$$\chi_{\rm I}^{(4)}\mathscr{E}_{\rm p}^{*2}\mathscr{E}_{\rm a}^{3} \sim \frac{4\times10^{-32}}{\hbar\Delta_{m_3g}}\mathscr{E}_{\rm a}^{3}. \tag{18}$$

Assuming a modest value of $\Delta_{m_3g} = 10^{12}$ or 10^{13} Hz say (owing to bandwidth effects in solids), we get from equation (16),

$$\chi_{\rm I}^{(4)}\mathscr{E}_{\rm p}^{*2}\mathscr{E}_{\rm a}^{3} \sim 4\times(10^{-9}\ \text{or}\ 10^{-10})\mathscr{E}_{\rm a}^{3}\ {\rm MKS}. \tag{19}$$

This estimate is not large enough to violate the approximation of undepleted pump beams but is not so small as to make experimental trials not worthwhile.

8. Enhancement by a resonant cavity

In the degenerate case discussed in section 2, where all the four down-converted photons have the same frequency, a further improvement in the down-conversion efficiency is possible by placing the nonlinear crystal in a resonant cavity, as shown in figure 4. If the resonant frequency of the cavity corresponds to the down-conversion frequency, the amplitude of the vacuum held

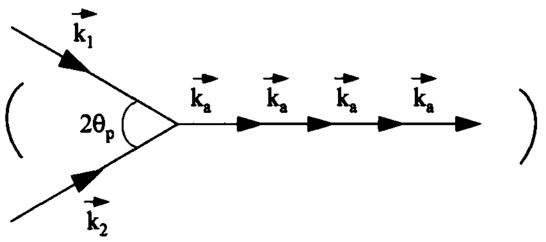

Figure 4. Enhancement of the down-conversion efficiency by means of a resonant cavity.

in the cavity at this frequency can be much higher, resulting in a corresponding increase in the probability of creation of four-photon states.

9. Discussion and conclusions

We have shown how the process of parametric down-conversion in a non-centrosymmetric crystal can be extended to produce a variety of entangled four-photon states by using two non-collinear pump beams. While the susceptibility for this down-conversion process is low, the gain depends on the second power of the pump amplitude, so that a useful output can be obtained by using pulsed pump beams with high peak power. There are also possibilities of obtaining a resonant enhancement of the four-photon emission process.

The configuration described using two pump beams can be generalized by symmetry to allow the use of Bessel beams to produce other novel configurations of the down-converted four-photon field. The down-conversion technique proposed could also form the basis for other higher-order down-conversion processes, to suit specific phase-matching conditions.

Entangled four-photon and multiple-photon states generated by these techniques could open the way to new experiments in hitherto unexplored domains of quantum mechanics.

Acknowledgment

Partial support from the Special Assistance Program at the School of Physics, University of Hyderabad, is gratefully acknowledged.

References

[1] Horne, M. A., Shimony, A., and Zeillinger, A., 1989, *Phys. Rev. Lett.*, **62,** 2209.
[2] Franson, J. D., 1989, *Phys. Rev. Lett.*, **62,** 2205.
[3] Rarity, J. G., and Tapster, P. R., 1989, *J. opt. Soc. Am.* B, **6,** 1221.
[4] Clauser, J. F., Horne, M. A., Shimony, A., and Holt, R. A., 1969, *Phys. Rev. Lett.*, **23,** 880.
[5] Santos, E., 1992, *Phys. Rev.* A, **46,** 3646.
[6] Greenberger, D. M., Horne, M. A., Shimony, A., and Zeillinger, A., 1990, *Am. J. Phys.*, **58,** 1131.
[7] Harris, S. E., Oshman, M. K., and Byer, R. L., 1967, *Phys. Rev. Lett.*, **18,** 732.
[8] Klyshko, D. N., 1967, *JETP Lett.*, **6,** 23.
[9] Burnham, D. C., and Weinberg, D. L., 1970, *Phys. Rev. Lett.*, **25,** 84.
[10] Eckbreth, A. C., 1978, *Appl. Phys. Lett.*, **32,** 421.

[11] Reichert, A., and Betzeber, K., 1992, *Ferroelectrics*, **126**, 9.
[12] Wulle, T., and Herminghaus, S., 1993, *Phys. Rev. Lett.*, **70**, 1401.
[13] Glushko, B., Kryzhanovsky, B., and Sarkisyan, D., 1993, *Phys. Rev. Lett.*, **71**, 243.
[14] Tewari, S. P., Huang, H., and Boyd, R. W., 1995, *Phys. Rev.* A, **51**, RC2707.
[15] Tewari, S. P., Huang, H., and Boyd, R. W., 1996, *Phys. Rev.* A (to be published).
[16] Yariv, A., 1989, *Quantum Electronics*, third edition (New York: Wiley), p. 395.
[17] Butcher, P. N., and Cotter, D., 1990, *The Elements of Nonlinear Optics* (Cambridge University Press), chapters IV and VI.
[18] Yariv, A., 1989, *Quantum Electronics*, third edition (New York: Wiley), p. 409.
[19] Yariv, A., 1989, *Quantum Electronics*, third edition (New York: Wiley), pp. 421–424.
[20] Perina, J., 1984, *Quantum Statistics of Linear and Nonlinear Optical Phenomena* (Deventer: Kluwer), p. 219.
[21] Yariv, A., 1989, *Quantum Electronics*, third edition (New York: Wiley), p. 411.