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Eliminating nonlinear phase mismatch in resonantly enhanced four-wave mixing

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Abstract

Resonantly enhanced four-wave mixing in double-Λ systems is limited by ac-Stark induced nonlinear phase shifts. With counter-propagating pump fields the intensity–phase coupling has minimal impact on the dynamics, but it is of critical importance for co-propagation. The nonlinear phase terms lead to an increase of the conversion length linearly proportional to the inverse seed intensity, while without nonlinear phase-mismatch the scaling is only logarithmic. We here show that the ac-Stark contributions can be eliminated while retaining the four-wave mixing contribution by choosing a suitable five-level system with appropriate detunings.

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Ever since the cancellation of resonant linear absorption and refraction via electromagnetically induced transparency (EIT) [1] was first demonstrated, quantum and nonlinear optics have successfully been exploring the consequences. Many interesting effects have been proposed and investigated [2]. One important application of EIT is optical frequency mixing close to atomic resonances which allows making use of the resonantly enhanced nonlinear interaction without suffering from linear absorption and refraction. It has been predicted that EIT could even lead to a new regime of nonlinear optics on the level of few light quanta [2a,2b,3].

In this paper we consider one particular EIT-based scheme, namely resonantly enhanced four-wave mixing in a double-Λ system as shown in Fig. 1. The two fields with (complex) Rabi frequencies Ω_1 and Ω_2 are initially excited and form the pump fields, while the other fields with (complex) Rabi frequencies E_1 and E_2 are generated during the interaction process. Ω_1 and E_1 are taken to be exactly on resonance while the other two fields are assumed to be detuned by an amount Δ. A finite detuning Δ, large compared to the Rabi frequencies, Doppler broadening and decay rates from the excited states, is necessary to maximize the ratio of nonlinear gain to linear absorption. Decay from

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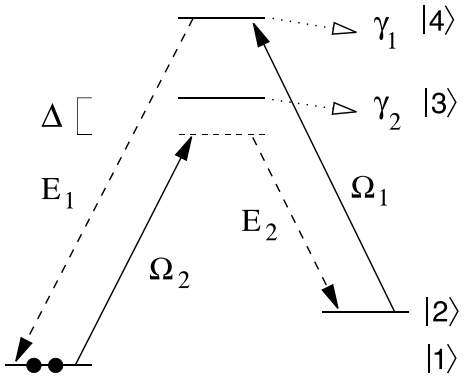

Fig. 1. Parametric amplification in a generic double-Λ system.

the two lower levels is considered to be negligible. Because of energy conservation all fields are in four-photon resonance. It can be shown furthermore that the contributions of the resonant transitions to the *linear* refractive index vanish if the fields are pairwise in two-photon resonance. Phase matching will thus favor two-photon resonance and we assume that this condition is fulfilled. Resonant four-wave mixing has been analyzed both theoretically and experimentally with co-propagating as well as counter-propagating fields [6–13].

Associated with the finite detuning Δ are ac-Stark shifts which lead to intensity dependent dynamical phase shifts of the fields. These phase shifts are of minor consequence in the case where the fields are counter-propagating [13]. They do have a detrimental influence, however, on co-propagating fields. In the following we will concentrate on the latter situation and show how to eliminate these terms, leading to a considerable improvement in nonlinear frequency conversion.

The standard method used to describe the wave mixing process in a resonant medium is to derive density matrix equations for the atomic system, solve them in the steady state, i.e., assuming adiabaticity, and insert the resulting expressions into the Maxwell–Bloch equations. This yields four equations of motion for the fields in the slowly varying amplitude and phase approximation. The field equations can then be further broken down into a set of five coupled equations consisting of four amplitude equations plus the equation of motion governing the relative phase between the fields.

This procedure can be rather cumbersome, particularly when several atomic levels are involved. A much simpler way to derive the field equations is given by the Hamiltonian approach introduced in [14], which we will use in the following. This method makes use of the fact that the polarization P of the medium can be expressed as a partial derivative of the time-averaged interaction energy per atom H with respect to the electric field or, equivalently, the Rabi frequencies E_i

$$P_i = -\frac{Nd_i}{\hbar}\left\langle \frac{\partial H}{\partial E_i^*} \right\rangle e^{-i\nu_i(t-z/c)} + \text{c.c.} \tag{1}$$

A similar expression holds for the drive field polarizations with E replaced by Ω. Here $\langle \ldots \rangle$ denotes quantum-mechanical averaging, d_i the dipole matrix elements of the corresponding transitions, and N is the atomic density. By introducing moving coordinates (ζ, t) with $\zeta = z - ct$ one can directly obtain the field equations of motion in the slowly varying amplitude and phase approximation

$$\frac{dE_i}{d\zeta} = -i\frac{\eta_i}{\hbar}\left\langle \frac{\partial H}{\partial E_i^*} \right\rangle, \tag{2}$$

where $\eta_i = Nd_i^2\omega_i/(2\hbar c\epsilon_0)$. The evaluation of the right-hand side of (2) is particularly simple if an open-system model can be used to incorporate decay in a complex Hamiltonian H and if the atoms adiabatically follow the dynamics of the fields. If the atoms are initially in an eigenstate of H with eigenvalue λ then $\langle H \rangle$ can simply be replaced by λ as the two are equivalent. Thus knowledge of the eigenvalues of the single-atom interaction Hamiltonian is sufficient to directly derive the field equations of motion.

In the basis $(|1\rangle\,|2\rangle\,|3\rangle\,|4\rangle)^{\mathrm{T}}$ the system shown in Fig. 1 can be described by the complex interaction Hamiltonian

$$H = \hbar\begin{bmatrix} 0 & 0 & -\Omega_2^* & -E_1^* \\ 0 & 0 & -E_2^* & -\Omega_1^* \\ -\Omega_2 & -E_2 & \Delta - i\gamma_2 & 0 \\ -E_1 & -\Omega_1 & 0 & -i\gamma_1 \end{bmatrix}. \tag{3}$$

Taking Ω as a characteristic magnitude of the Rabi frequencies involved, to second order in Ω/Δ the relevant eigenvalue of (3) is given by

$$\lambda_0(t) = \frac{1}{\Delta}\left[\frac{\Omega_1\Omega_2E_1^*E_2^* + \Omega_1^*\Omega_2^*E_1E_2}{|\Omega_1|^2 + |E_1|^2} - \frac{|\Omega_1|^2|\Omega_2|^2 + |E_1|^2|E_2|^2}{|\Omega_1|^2 + |E_1|^2}\right]. \quad (4)$$

This eigenvalue corresponds to the instantaneous eigenstate

$$|\lambda_0(t)\rangle = \frac{|E_1|}{\sqrt{|E_1|^2 + |\Omega_1|^2}}\left[-\frac{\Omega_1}{E_1}|1\rangle + |2\rangle\right]. \quad (5)$$

As can be seen, this state is asymptotically connected to $|1\rangle$ at $t \to -\infty$. That is, the eigenstate associated with λ_0 corresponds to the ground state $|1\rangle$ for vanishing E_1 and E_2. If the pump fields change sufficiently slowly, such that the product of the characteristic time scale T and the characteristic energy separation to the next eigenvalue $\lambda_1 - \lambda_0$ is large, we may assume that all atoms stay at all times in $|\lambda_0(t)\rangle$ and $\langle H\rangle$ in (2) can be replaced by $\lambda_0(t)$. This yields the following equations of motion [13]:

$$\frac{\partial}{\partial\zeta}E_1 = -i\eta\frac{\Omega_1^*\Omega_1^2\Omega_2E_2^* - E_1^2E_2\Omega_1^*\Omega_2^*}{\Delta\left(|\Omega_1|^2 + |E_1|^2\right)^2} - i\eta\frac{|\Omega_1|^2\left(|\Omega_2|^2 - |E_2|^2\right)}{\Delta\left(|\Omega_1|^2 + |E_1|^2\right)^2}E_1, \quad (6)$$

$$\frac{\partial}{\partial\zeta}E_2 = -i\eta\frac{\Omega_1\Omega_2E_1^*}{\Delta\left(|\Omega_1|^2 + |E_1|^2\right)} + i\eta\frac{|E_1|^2}{\Delta\left(|\Omega_1|^2 + |E_1|^2\right)}E_2, \quad (7)$$

$$\frac{\partial}{\partial\zeta}\Omega_1 = i\eta\frac{\Omega_1^2\Omega_2E_1^*E_2^* - |E_1|^2E_1E_2\Omega_2^*}{\Delta\left(|\Omega_1|^2 + |E_1|^2\right)^2} + i\eta\frac{|E_1|^2(|\Omega_2|^2 + |E_2|^2)}{\Delta\left(|\Omega_1|^2 + |E_1|^2\right)^2}\Omega_1, \quad (8)$$

$$\frac{\partial}{\partial\zeta}\Omega_2 = -i\eta\frac{E_1E_2\Omega_1^*}{\Delta\left(|\Omega_1|^2 + |E_1|^2\right)} + i\eta\frac{|\Omega_1|^2}{\Delta\left(|\Omega_1|^2 + |E_1|^2\right)}\Omega_2, \quad (9)$$

where equal coupling strengths have been assumed. Note that there are no linear absorption terms in (6)–(9), despite the presence of the decay terms γ in Eq. (3). Thus, the process is a parametric one, and the total energy of the electromagnetic fields is conserved. The absence of dissipative loss terms is of course a consequence of the assumed adiabatic following of the system in the instantaneous eigenstate (5) which has no overlap with the decaying bare states $|3\rangle$ and $|4\rangle$. One furthermore finds that the equations have the following three constants of motion:

$$|\Omega_1|^2 + |E_1|^2 = \text{constant}, \quad (10)$$

$$|\Omega_2|^2 + |E_2|^2 = \text{constant}, \quad (11)$$

$$|\Omega_1|^2 - |\Omega_2|^2 = \text{constant}. \quad (12)$$

This allows the problem to be reduced to two degrees of freedom, one corresponding to the exchange energy between the fields and the other to the relative phase $\varphi = \phi_{\Omega_1} + \phi_{\Omega_2} - \phi_{E_1} - \phi_{E_2}$ between the fields.

The terms in the second line of each equation in (6)–(9) are ac-Stark induced, intensity dependent phase terms. They have a considerable impact on the dynamics, particularly in terms of the conversion length, i.e., the distance required for one of the pump modes to attain maximum transfer into one of the generated modes. To see this we solve the field propagation problem analytically, using the methods described in [14,15]. The essence of this formalism is to reduce a set of Maxwell propagation Eqs. (6)–(9) to canonical Hamiltonian equations involving action and angle variables J and φ. The variable $J(\zeta)$ characterizes an amount of energy exchanged between the waves and is determined via the relations:

$$|\Omega_1(\zeta)|^2 = \Omega_{10}^2 - J(\zeta), \quad (13)$$

$$|\Omega_2(\zeta)|^2 = \Omega_{20}^2 - J(\zeta), \quad (14)$$

$$|E_1(\zeta)|^2 = E_{10}^2 + J(\zeta), \quad (15)$$

$$|E_2(\zeta)|^2 = E_{20}^2 + J(\zeta), \quad (16)$$

with the initial condition $J(\zeta = 0) = 0$. Here $\Omega_{i0}^2 = |\Omega_i(\zeta = 0)|^2$ and similarly for E_{i0}^2. The dependence of the eigenvalue λ_0 on the field phases allow us to reduce the Hamiltonian equations

further to a single equation for the exchange energy $J(\zeta)$. A detailed description of this procedure is given in [15]. Integration of this last equation eventually yields an implicit solution for the energy exchange $J(\zeta)$ as function of propagation distance ζ

$$\eta\zeta = \int_0^J S(J')\frac{\mathrm{d}J'}{\sqrt{R(J')}}, \tag{17}$$

where both functions $R(J)$ and $S(J)$ are polynomials in J.

We consider the case where the two pump fields are initially of equal intensity, as are the two seed fields, so that $E_1 = E_2 = E$ and $\Omega_1 = \Omega_2 = \Omega$. Then the functions $R(J)$ and $S(J)$ take the form:

$$R = \Omega_{10}^4 E_{10}^4 \sin^2\varphi_0 + 2\Omega_{10}^2 E_{10}^2\left(\Omega_{10}^2 - E_{10}^2\right) \times \sin^2\left(\frac{\varphi_0}{2}\right)J - 2\Omega_{10}^2 E_{10}^2\cos^2\left(\frac{\varphi_0}{2}\right)J^2, \tag{18}$$

$$S = \Delta\left(\Omega_{10}^2 + E_{10}^2\right), \tag{19}$$

where φ_0 is the initial relative phase difference between the fields. If we define the seed field parameter by $\epsilon = E_{10}^2/\Omega_{10}^2$ then, in the limit $\epsilon \ll 1$, the solution of Eq. (17) is

$$J(\zeta) = \Omega_{10}^2\left\{(1-\epsilon)\sin^2\left[\sqrt{\epsilon}\cos\left(\frac{\varphi_0}{2}\right)\frac{\eta\zeta}{\Delta}\right] - \sqrt{\epsilon}\sin\left(\frac{\varphi_0}{2}\right)\sin\left[2\sqrt{\epsilon}\cos\left(\frac{\varphi_0}{2}\right)\frac{\eta\zeta}{\Delta}\right]\right\}. \tag{20}$$

We see that in this case the conversion length L and conversion efficiency $e = (|E_{\max}|^2 - |E_{\min}|^2)/(|\Omega_{\max}|^2)$ are given by:

$$e = 1 - \epsilon, \tag{21}$$

$$L = \frac{\Delta}{\eta}\frac{1}{\sqrt{\epsilon}}\frac{\pi}{2\cos\frac{\varphi_0}{2}}. \tag{22}$$

One immediately notices that the conversion length scales as $L \sim 1/\sqrt{\epsilon}$, that is, inversely in the seed field amplitude E_0. Consequently for small, let alone vacuum, seed fields, the conversion distance will rapidly become infeasibly long. In addition, as the initial phase angle $\varphi_0 \to \pi$ the conversion length rapidly approaches infinity and no conversion can take place.

On the other hand, if the phase terms in (6)–(9) were to be omitted, the quantity

$$\mathrm{Re}(\Omega_1\Omega_2 E_1^* E_2^*) = |\Omega_1\Omega_2 E_1^* E_2^*|\cos\varphi, \tag{23}$$

is another constant of motion. In this case, if one of the generated fields E_1, E_2 vanishes initially, i.e., if only one of these fields is seeded, then this constant of motion must be zero. This indicates that the relative phase φ can only jump discontinuously between $\pm\pi/2$, which occurs only at the end of each conversion cycle, when at least one of the field amplitudes vanishes. Thus in this case the phase is essentially decoupled from the evolution of the field amplitudes. This makes a considerable difference to the dynamics.

The analytical solution in this case is obtained in the same way as described above. The polynomials $R(J)$ and $S(J)$ are given by:

$$R = \left(E_{10}^2 + J\right)^2\left(\Omega_{10}^2 - J\right)^2 - \Omega_{10}^4 E_{10}^4\cos^2\varphi_0, \tag{24}$$

$$S = \Delta\left(\Omega_{10}^2 + E_{10}^2\right), \tag{25}$$

and the solution for exchange energy can be expressed via Jacobi elliptic sine and cosine functions $\mathrm{sn}[x;p]$ and $\mathrm{cn}[x;p]$:

$$J(\zeta) = E_{10}^2\frac{A}{B}, \tag{26}$$

with

$$A = \left(1+\epsilon-2\epsilon\sqrt{\cos\varphi_0}\right)\left(1+\sqrt{\cos\varphi_0}\right)\mathrm{sn}^2\left[\frac{\eta\zeta}{2\Delta}+\chi_0;p\right] - \left(1+\epsilon\right)\left(1-\sqrt{\cos\varphi_0}\right),$$

$$B = \left(1+\epsilon\right)\mathrm{cn}^2\left[\frac{\eta\zeta}{2\Delta}+\chi_0;p\right] + 2\epsilon\sqrt{\cos\varphi_0}\,\mathrm{sn}^2\left[\frac{\eta\zeta}{2\Delta}+\chi_0;p\right],$$

$$p \approx 1 - 4\epsilon^2\cos\varphi_0, \tag{27}$$

where we have taken $0 < \cos\varphi_0 < 1$ for simplicity, and χ_0 is a complicated function of ϵ and $\cos\varphi_0$, whose precise form is however unimportant for the present discussion. The spatial period of oscillations of $J(\zeta)$ is determined by

$$\frac{\eta}{\Delta}L = 4K(p), \tag{28}$$

with $K(p)$ being a complete elliptic integral of the first kind. In this case, the parameter p is close to unity, so that $K(p)$ can be approximated as $K(p) \approx (1/2)\ln[16/(1-p)]$. Thus the conversion efficiency e and the conversion length L are given by:

$$e = 1 + \epsilon - 2\epsilon\sqrt{\cos\varphi_0}, \tag{29}$$

$$L = \frac{2\Delta}{\eta}\ln\left(\frac{4}{\epsilon^2\cos\varphi_0}\right). \tag{30}$$

It is now evident that the conversion distance scales only as $-\log\epsilon$, and the situation is completely different to the previous case – the conversion length will always remain short. In addition, the phase dependent increase in the conversion length, this time as $\varphi_0 \to \pi/2$, also only takes effect logarithmically.

As an illustration of conversion distance dependence on seed field intensity we have calculated numerical solutions to (6)–(9) with and without the phase terms, and without making the approximation that $\epsilon \ll 1$. We have assumed $E_1 = E_2$, $\Omega_1 = \Omega_2$ and $\varphi_0 = \pi/4$. The results are shown in Fig. 2.

Thus, given the obvious advantages inherent in the omission of these phase terms, the question naturally arises: does there exist a situation in which these terms can be made to vanish?

It has been shown by Harris [16] that in a system of parallel λ transitions with different excited state energies there exists an optimum detuning such that the *nonlinear* index of refraction vanishes. Recently the cancellation of ac-Stark shifts has been discussed in the context of optical magnetometry using dark resonances [17]. A similar idea can be applied here. Noting that both parts of (4) are linear in Δ, but only the first part is linear in each of the fields, suggests a method for canceling the phase terms and at the same time retaining the nonlinear interaction part. To see this, consider the five-level system shown in Fig. 3. Here the fields Ω_2 and E_2 are tuned midway between the two resonances $|1\rangle \to |3\rangle, |4\rangle$ and $|2\rangle \to |3\rangle, |4\rangle$. If the Rabi-frequencies of the transitions involving level $|3\rangle$ are denoted by Ω_2 and E_2, and those involving level $|4\rangle$ by $g\Omega_2$ and fE_2 respectively, $g = \mu_{14}/\mu_{13}$ and $f = \mu_{24}/\mu_{23}$ being the ratios of the dipole moments, the Hamiltonian becomes

$$H = -\hbar\begin{bmatrix} 0 & 0 & \Omega_2^* & g^*\Omega_2^* & E_1^* \\ 0 & 0 & E_2^* & f^*E_2^* & \Omega_1^* \\ \Omega_2 & E_2 & -\Delta + i\gamma_3 & 0 & 0 \\ g\Omega_2 & fE_2 & 0 & \Delta' + i\gamma_2 & 0 \\ E_1 & \Omega_1 & 0 & 0 & +i\gamma_1 \end{bmatrix}. \tag{31}$$

We find that to second order in Ω/Δ or Ω/Δ' the lowest eigenvalue of (31) is given by

$$\lambda_0 = \left[\frac{|E_1|^2|E_2|^2\left(\Delta' - |f|^2\Delta\right) + |\Omega_1|^2|\Omega_2|^2\left(\Delta' - |g|^2\Delta\right)}{\Delta\Delta'(|\Omega_1|^2 + |E_1|^2)} + \frac{E_1E_2\Omega_1^*\Omega_2^*\left(\Delta' - f^*g\Delta\right) + E_1^*E_2^*\Omega_1\Omega_2\left(\Delta' - fg^*\Delta\right)}{\Delta\Delta'(|\Omega_1|^2 + |E_1|^2)}\right]. \tag{32}$$

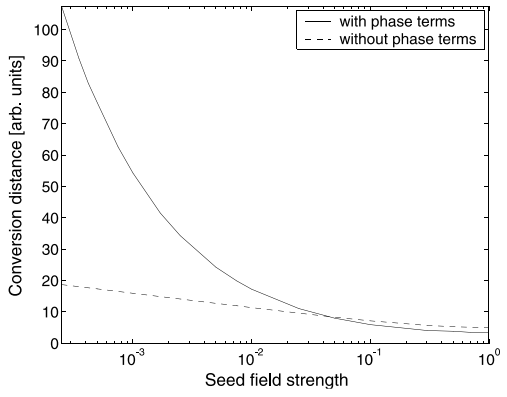

Fig. 2. Distance required for maximum conversion of the pump modes into the generated modes as a function of seed field strength. Initial field intensities taken as $|\Omega_{10}|^2 = |\Omega_{20}|^2$, $|E_{10}|^2 = |E_{20}|^2$, with the seed field intensity defined as $\epsilon = |E_{10}|^2/|\Omega_{10}^2|$.

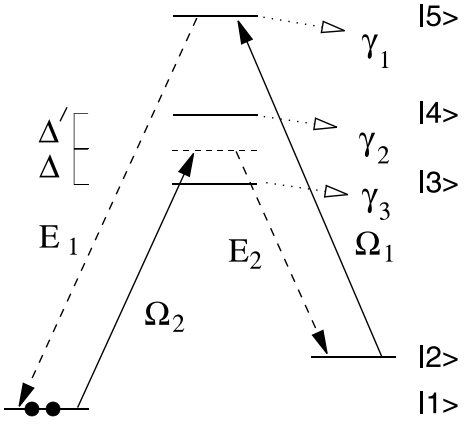

Fig. 3. Modified double-Λ system. Ω_2 and E_2 denote the Rabi-frequencies for the transitions $|1\rangle \to |3\rangle$ and $|2\rangle \to |3\rangle$ respectively. The corresponding Rabi-frequencies for the transitions $|1\rangle \to |4\rangle$ and $|2\rangle \to |4\rangle$ are $g\Omega_2$ and fE_2, where $g = \mu_{14}/\mu_{13}$ and $f = \mu_{24}/\mu_{23}$ are the ratios of the dipole moments μ_{ij}.

The first term represents intensity-dependent contributions to the index of refraction, whose presence results in an increase of the conversion length. These terms can be made to vanish, however, if

$$\frac{\Delta'}{\Delta} = |f|^2 = |g|^2, \tag{33}$$

i.e., if the absolute value of the ratio of the dipole matrix elements from level $|1\rangle$ to levels $|3\rangle, |4\rangle$ is equal to that from level $|2\rangle$ to levels $|3\rangle, |4\rangle$. It should be noted that no condition follows for the absolute value of the dipole matrix elements themselve. The detunings Δ and Δ' have then to be chosen according to (33). In order not to cancel the four-wave mixing terms in (32) at the same time, it is furthermore necessary that $fg^* \neq |fg|$. Together with (33) this implies for real values of g and f

$$g = -f. \tag{34}$$

In this case the eigenvalue reads

$$\lambda_0 = \frac{2}{\Delta}\left[\frac{E_1E_2\Omega_1^*\Omega_2^* + \Omega_1\Omega_2E_1^*E_2^*}{|\Omega_1|^2 + |E_1|^2} + \frac{\delta}{2\Delta}\,\frac{\Delta+\Delta'}{\Delta\Delta'}\,\frac{|E_1|^2|E_2|^2 + |\Omega_1|^2|\Omega_2|^2}{|\Omega_1|^2 + |E_1|^2}\right], \tag{35}$$

where we have also introduced a deviation δ of the laser detuning from the optimum value determined by (33), $\Delta \to \Delta - \delta$. One sees that precise tuning of the laser fields is not crucial since usually $|\delta/\Delta| \ll 1$.

Conditions (33) and (34) are fulfilled for example in alkali atoms with hyperfine splitting. Levels $|1\rangle$ and $|2\rangle$ can be for example the $m_F = \pm 1$ Zeeman sublevels of the $S_{1/2}(F = 1)$ ground state of sodium or rubidium ($I = 3/2$). Levels $|3\rangle$ and $|4\rangle$ are the $m_F = 0$ Zeeman sub-levels of $P_{1/2}(F = 1)$ and $P_{1/2}(F = 2)$. Here we have $|f| = |g| = 1$ and $f = -g$ and the optimum detuning is exactly midway between the hyperfine states.

In summary we have shown that the nonlinear phase contributions arising in resonant forward four-wave mixing due to the ac-Stark effect can be exactly eliminated if a five-state system with appropriate couplings and detunings is used. We have derived a simple effective Hamiltonian for this system and shown that the conversion length scales only logarithmically with the inverse seed intensity whereas with the phase terms present conversion length scales linearly.

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