

Adiabatic four-wave mixing in a strongly driven resonant four-level system: Effect of pump depletion

B. Kryzhanovsky

Institute for Physical Research, Ashtarak 378410, Armenia

B. Glushko

Weizmann Institute of Science, Rehovot 76100, Israel

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The exact solution of the self-consistent problem of four-wave mixing in a resonant medium of four-level atoms (molecules) is obtained. The pump depletion, level-population movement, and atomic-transition saturation are explicitly taken into account. It is shown that (i) in the process of transformation the generated pulse is split into a train of subpulses, and their number is strongly related to the length of propagation; in the case of saturation, this splitting does not occur; (ii) the conversion-efficiency definition problem is reduced to solution of a third-order polynomial; (iii) by variation of the initial value of the wave-vector mismatch Δk_0 , the processes of conversion may lead to the complete depletion of one of the pump beams; (iv) a prediction is made of the intersection of quasienergetic terms in the process of energy transfer; this phenomenon is shown to be responsible for the experimentally observed disruption of generation.

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I. INTRODUCTION

The standard approach to treating the four-wave mixing problem includes expansion of the medium polarization as a power series of the field amplitudes of the interacting waves and only terms of lower order are retained [1–12]. Analysis of the propagation effect is usually made for short lengths of interaction, where the rate of conversion is so small that one can use the nondepleted pump beam approach and ignore the atomic level saturation. Such an approach has shortcomings. First, in the framework of this approach we are limited in the rate of conversion. Second, it is impossible to define the maximal value of conversion and optimal length of interaction.

Significant progress has been achieved in a series of recent publications [13–17], where within the framework of the cubic susceptibility model a strong account of pump depletion has been considered. However, the theory developed in these works becomes invalid for the case of intense field and strong resonance, where all orders of polarization should be taken into account. The mathematical difficulties that arise are usually insurmountable and tend to use either a perturbational approach or numerical calculations [18].

In the work presented, the process of four-wave mixing (FWM) in the two-photon resonant four-level system, interacting with ultrashort adiabatic intense pump pulses, is considered. The process evolves according to the scheme $\omega_1 + \omega_2 = \omega_3 + \omega_4$, where $\omega_{1,2}$ and $\omega_{3,4}$ are pumped and generated photons.

The method enables us to solve the problem explicitly, taking into account both the pump depletion and coherent saturation of atomic transitions. The main difficulty in obtaining a mathematical formulation of this problem is the absence of explicit dependence of the

atomic polarization on the interacting field's amplitudes. In order to overcome this obstacle we use the simple and effective relation [19,34] $P = -\hbar \partial \Omega / \partial E$ between field-induced atomic polarization P , the field amplitude E , and quasienergy Ω of the dressed state "atom plus fields" [20]. This relation was initially mentioned by Melikyan [21] and utilized to build up the exact theory of third-harmonic generation in a resonant medium [22]. This mathematical method allows one to solve the problem explicitly, define all integrals of motion, and reduce the solution to one differential equation [19] describing the parametric pendulum oscillations without knowing the real dependence of P on the amplitudes of the interacting fields. The basis of such an approach is a very important integral of motion (12), which is not involved in the traditional approach.

II. QUASIENERGY AND ATOMIC POLARIZATION

Consider the four-level system (Fig. 1) interacting with four waves ω_j ($j=1,2,3,4$), the frequencies of which obey the relation $\omega_1 + \omega_2 = \omega_3 + \omega_4$. The pulse widths must be shorter than any decay time of the system, and the interaction is considered to be adiabatic. The field amplitudes are presented as follows:

$$E_j(\mathbf{r}, t) = E_j \exp(-i\omega_j t) + c.c. \quad (1)$$

The wave function describing the system in the rotating-wave approximation [20] is presented as a superposition of atomic states ψ_i ,

$$\Psi = e^{-i\Omega t} \sum_{n=1}^4 a_n \psi_n e^{-i\Delta_{n-1} t}, \quad (2)$$

where the energy of the ground level is taken to be zero ($\Delta_0=0$); Δ_1 , Δ_2 , and Δ_3 , are, respectively, one-, two-, and three-photon detunings:

$$\begin{aligned}\Delta_1 &= \omega_{21} - \omega_1, & \Delta_2 &= \omega_{31} - \omega_1 - \omega_2, \\ \Delta_3 &= \omega_{41} - \omega_1 - \omega_2 + \omega_3 = \omega_{41} - \omega_4.\end{aligned}\quad (3)$$

Substituting Eq. (2) into the Schrödinger equation, we obtain the system of equations for a_n amplitudes:

$$\begin{pmatrix} 0 & -E_1^* d_1 & 0 & E_4^* d_4 \\ -E_1 d_1 & \hbar \Delta_1 & -E_2^* d_2 & 0 \\ 0 & -E_2 d_2 & \hbar \Delta_2 & -E_3 d_3 \\ -E_4 d_4 & 0 & -E_3^* d_3 & \hbar \Delta_3 \end{pmatrix} \begin{pmatrix} a_1 \\ a_2 \\ a_2 \\ a_4 \end{pmatrix} = \hbar \Omega \begin{pmatrix} a_1 \\ a_2 \\ a_3 \\ a_4 \end{pmatrix}, \quad (4)$$

where $d_1 = d_{12}$, $d_2 = d_{23}$, $d_3 = d_{32}$, and $d_4 = d_{41}$ are the matrix elements of dipole moment that can be treated as real values. The quasienergy Ω is defined by the roots of the determinant of Eq. (4):

$$\begin{aligned}\hbar^4 \Omega (\Omega - \Delta_1) (\Omega - \Delta_2) (\Omega - \Delta_3) - \hbar^2 \Omega (\Omega - \Delta_3) |E_2 d_2|^2 - \hbar^2 \Omega (\Omega - \Delta_1) |E_3 d_3|^2 - \hbar^2 (\Omega - \Delta_2) (\Omega - \Delta_3) |E_1 d_1|^2 \\ - \hbar^2 (\Omega - \Delta_1) (\Omega - \Delta_2) |E_4 d_4|^2 + |E_1 E_3 d_1 d_3|^2 + |E_2 E_4 d_2 d_4|^2 - d_1 d_2 d_3 d_4 (E_1 E_2 E_3^* E_4^* + \text{c.c.}) = 0.\end{aligned}\quad (5)$$

If the interaction is switched on adiabatically, the non-perturbed atomic states are the corresponding quasienergetic ones, therefore the correspondence of the roots Ω_n obey the following relations: $\Omega_n \rightarrow \Delta_{n-1}$ for $E_j \rightarrow 0$. We presume the system to be in the ground state before the interaction is switched on. Thus after switching on the interaction the system will be described by the wave function of Eq. (2) with amplitudes $a_n^{(1)}$ being the solution of Eq. (4) for $\Omega = \Omega_1$. It is obvious that the direct substitution of the $a_n^{(1)}$ into Eq. (2) in order to calculate the polarization $P_{11} = \langle \psi_1 | \hat{d} | \psi_1 \rangle$ will lead us to the useless expression nonexplicitly dependent on field amplitudes E_j . Instead we use the method developed previously [21,22], which does not require knowledge of the explicit a_n dependence on the E_j amplitudes and is very convenient for further investigation. We represent the atomic polarization in the ψ state as

$$P = -\hbar \sum_{j=1}^4 \frac{\partial \Omega_1}{\partial E_j^*} e^{-i\omega_j t} + \text{c.c.} \quad (6)$$

The basis for this relation can be found elsewhere [21–23]. We only mention that the matrix Eq. (4) looks like the stationary Schrödinger equation with the Hamiltonian dependent on E_j and E_j^* as parameters. Taking this into account and using the well-known relation [29] for the diagonal matrix element of the Hamiltonian derivative on the parameter λ , $\langle i | \partial \hat{H} / \partial \lambda | i \rangle = \partial H_{ii} / \partial \lambda$, we can obtain expression Eq. (6).

III. WAVE EQUATIONS: MOTION INTEGRALS

We consider the propagation process of E_j waves in a medium consisting of four-level systems, filling the semispace $z \geq 0$. We solve the one-dimensional problem in the plane-wave approximation, assuming the input beam angles to be small relative to the z axis. This means that

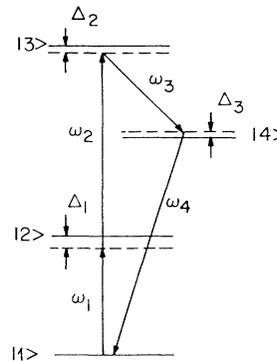


FIG. 1. Four-level system interacting with four laser waves $\omega_1, \omega_2, \omega_3, \omega_4$ ($\omega_1 + \omega_2 = \omega_3 + \omega_4$); $\Delta_1 = \omega_{21} - \omega_1$, $\Delta_2 = \omega_{31} - \omega_1 - \omega_2$, and $\Delta_3 = \omega_{41} - \omega_1 - \omega_2 + \omega_3 = \omega_{41} - \omega_4$ are, respectively, one-, two-, and three-photon detunings. Each wave is supposed to interact with one resonant transition only.

the phase-matching condition in the transverse direction with respect to the z axis is met. For perfect phase matching some amount of buffer gas can be added into the medium, leading to the additional terms in the refractive index δn_{jb} . The wave amplitudes are

$$E_j = A_j e^{i\mathbf{k}_{jb} \cdot \mathbf{r}}, \quad |\mathbf{k}_{jb}| = (1 + \delta n_{jb}) \omega_j / c, \quad (7)$$

where the A_j are slowly varying functions of the variables z and $\tau = t - z/c$. The true value of the wave vector k_j is $k_j = (1 + \delta n_{jb} + \delta n_{ja}) \omega_j / c$, where δn_{ja} results from the interaction with the medium and will be defined in the process of the problem solution.

The substitution of Eqs. (6) and (7) into the wave equation leads to the relation

$$\frac{d A_j}{dz} = -i \frac{2\pi \hbar N \omega_j}{c} \frac{\partial \Omega_1}{\partial A_1^*}, \quad (8)$$

where N is the atomic density. The usual approach involves expansion of Ω_1 as a power series of A_j . Substitution of the first term of the expansion of Eq. (5) into Eq. (8) results in the well-known relation for resonant refractive indices at frequencies ω_j :

$$\delta n_{1a} = \frac{2\pi N d_1^2}{\hbar \Delta_1}, \quad \delta n_2 = \delta n_3 = 0, \quad \delta n_{4a} = \frac{2\pi N d_4^2}{\hbar \Delta_3}. \quad (9)$$

Including the next term of the expansion would lead to the parametric interaction [1–3], where the effect of coherent saturation of atomic transitions has not been considered.

Substituting P polarization in the form of Eq. (6) we are able to solve the problem explicitly. Equation (8) is more convenient to rewrite in the terms of intensity I_j and phase ϕ_j ,

$$A_j = |A_j| e^{i\phi_j}, \quad I_j = \frac{c}{2\pi \hbar \omega_j} |E_j|^2. \quad (10)$$

Then separating the real and imaginary part we obtain from Eq. (8)

$$\frac{dI_j}{dz} = N \frac{\partial \Omega_1}{\partial \phi_j}, \quad \frac{d\phi_j}{dz} = -N \frac{\partial \Omega_1}{\partial I_j}. \quad (11)$$

Equation (5) for the quasienergy includes the phases ϕ_j in the form of one linear combination only: $\phi = \phi_1 + \phi_2 - \phi_3 - \phi_4 + \delta k_b z$, where $\delta k_b = \mathbf{k}_{1b} + \mathbf{k}_{2b} - \mathbf{k}_{3b} - \mathbf{k}_{4b}$ is a projection of the wave-vector mismatch onto the z axis. Therefore the following relations become valid: $\partial \Omega_1 / \partial \phi_1 = \partial \Omega_1 / \partial \phi_2 = -\partial \Omega_1 / \partial \phi_3 = -\partial \Omega_1 / \partial \phi_4$. Then Eq. (11) leads to the set of motion integrals for intensities

$$\begin{aligned} I_1 - I_2 = I_{10} - I_{20}, \quad I_3 - I_4 = I_{30} - I_{40}, \\ I_1 + I_2 + I_3 + I_4 = I_{10} + I_{20} + I_{30} + I_{40}, \end{aligned} \quad (12a)$$

and for quasienergy Ω_1 and wave-vector mismatch δk_b ,

$$N\Omega_1 + \delta k_b I_4 = N\Omega_{10} + \delta k_b I_{40}, \quad (12b)$$

where I_{j0} are the input beam intensities and $\Omega_{10} = \Omega_1(z=0)$. The value Ω_{10} is obtained by substituting into Eq. (5) the values E_{j0} . The expression (12a) is the Manly-Rowe relation, well known in the theory of parametric amplifiers [24]. The integral (12b) determines the quasienergy dependence on propagation distance $\Omega_1 = \Omega_1(z)$; the quasienergy becomes the integral of motion in the case $\delta k_b = 0$ only.

IV. BASIC RELATIONS

Equations (12) enable the reduction of four variables I_j to one $I = I(z)$, which explicitly characterizes the energy transfer between waves: $I_1 = I_{10} - I$, $I_2 = I_{20} - I$, $I_3 = I_{30} + I$, and $I_4 = I_{40} + I$. The initial value of I is $I(0) = 0$. The canonical transformation of Eq. (11) for variables I and ϕ results in

$$\frac{dI}{dz} = -N \frac{\partial H}{\partial \phi}, \quad \frac{d\phi}{dz} = N \frac{\partial H}{\partial I}, \quad (13)$$

where

$$H = \Omega_1 + \delta k_b I / N. \quad (14)$$

The equation that defines H as a function of parameters I and ϕ is

$$F = f \cos \phi, \quad (15)$$

where

$$f = 2[\gamma_1 \gamma_2 \gamma_3 \gamma_4 (I_{10} - I)(I_{20} - I)(I_{30} + I)(I_{40} + I)]^{1/2} \quad (16)$$

and

$$\gamma_j = \frac{2\pi\omega_j d_j^2}{\hbar c} \quad (j=1, 2, 3, 4). \quad (17)$$

The left part of Eq. (15) is

$$\begin{aligned} F = & \left[H - \frac{\delta k_b I}{N} \right] \left[H - \Delta_1 - \frac{\delta k_b I}{N} \right] \left[H - \Delta_2 - \frac{\delta k_b I}{N} \right] \left[H - \Delta_3 - \frac{\delta k_b I}{N} \right] \\ & - \left[H - \frac{\delta k_b I}{N} \right] \left[\left[H - \Delta_3 - \frac{\delta k_b I}{N} \right] \gamma_2 (I_{20} - I) + \left[H - \Delta_1 - \frac{\delta k_b I}{N} \right] \gamma_3 (I_{30} + I) \right] \\ & - \left[H - \Delta_2 - \frac{\delta k_b I}{N} \right] \left[\left[H - \Delta_3 - \frac{\delta k_b I}{N} \right] \gamma_1 (I_{10} - I) + \gamma_4 (I_{40} + I) \left[H - \Delta_1 - \frac{\delta k_b I}{N} \right] \right] \\ & + \gamma_1 \gamma_3 (I_{10} - I)(I_{30} + I) + \gamma_2 \gamma_4 (I_{20} - I)(I_{40} + I). \end{aligned} \quad (18)$$

Equation (15) is obtained from Eq. (5) by the following substitutions: $|E_j d_j|^2 \rightarrow \gamma_j I_j \hbar^2$ and $\Omega \rightarrow H - \delta k_b I / N$.

It is readily noted that Eq. (13) has the same form as the canonical Hamiltonian equations for one-dimensional classical motion if the variables I and ϕ are regarded as the generalized coordinate and momenta, z replaces the time, and NH is equivalent to the Hamiltonian. Therefore Eq. (13) can be analyzed by the methods and terminology of classical mechanics. It is seen from Eqs. (15)–(18) that H has no explicit z dependence. It is consequently the motion integral, i.e., $dH/dz = 0$. It follows also from Eq. (12b) that $H = \Omega_{10} + \delta k_b I_{40} / N$.

The fact that H is a motion integral results in two conclusions. First, we do not need to find out the explicit polarization P dependence on the parameters E_j and E_j^* , which for arbitrary amplitudes is an unsolvable problem. Second, we do reduce the problem to only one differential equation for I . Now using Eq. (15) in order to exclude ϕ from Eq. (13), we obtain

$$\frac{dI}{dz} = \pm N (f^2 - F^2)^{1/2} \frac{\partial F}{\partial H}. \quad (19)$$

The sign in Eq. (19) is that of $\sin \phi_0$, where $\phi_0 = \phi(z=0)$. Equation (19) describes one-dimensional finite motion, being the analogy of classical parametric pendulum oscillation that has been widely investigated in the theory of elliptic functions [25]. The allowed range of motion is situated between S_4 and S_1 , where S_1 is a least positive root, while $S_4 \leq 0$ is the maximal negative root of the equation

$$f^2(I) - F^2(I) = 0. \quad (20)$$

From here on we consider the case $I_{40} = 0$. In this case $H = \Omega_{10}$, $S_4 = 0$, and I precisely coincides with the wave ω_4 intensity and varies within the limits $0 \leq I \leq S_1$. Then Eq. (19) describes a periodic in z coordinate process $I(z) = I(z + 2l)$. Thus, within the range $0 \leq z \leq l$ the FWM process develops into the channel $\omega_1 + \omega_2 \rightarrow \omega_3 + \omega_4$, i.e., intensity transfer from pump

waves to amplified $\omega_{3,4}$ waves occurs. When $z=l$, the magnitude I reaches its maximal value $I(z)=S_1$ and the direction of energy transfer changes sign: within the range $l \leq z \leq 2l$ the process develops into the channel $\omega_3 + \omega_4 \rightarrow \omega_1 + \omega_2$ up to the point $z=2l$, where $I=0$. The process has an oscillatory character, the spatial period $2l$ being determined from (19) as

$$l = \frac{1}{N} \int_0^{S_1} \left[\frac{\partial F}{\partial H} \right] \frac{dI}{(f^2 - F^2)^{1/2}}. \quad (21)$$

If $I_{40} \neq 0$ the lower limit of the integral should be replaced by S_4 . In the case of degeneration of the root S_1 , $l \rightarrow \infty$, i.e., the energy-transfer direction does not change the sign for any z value. Concluding the formalism developed, we write down the expression for refractive index n_j at the frequency ω_j

$$n_j = 1 + \delta n_{jb} + \frac{cN \left[\frac{\partial F}{\partial I_{j0}} \right]}{\omega_j \left[\frac{\partial F}{\partial H} \right]}. \quad (22)$$

It follows from (22) that both n_j and k_j are complicated functions of the length of interaction. The same holds for $\delta k = \delta k(z)$, which is defined as a projection vector $\delta \mathbf{k} = \mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3 - \mathbf{k}_4$ onto the z axis. Thus in the future we will operate with only one value $\delta k_0 = \delta k(z=0)$, initial wave-vector mismatch.

The function F is in common sense the fourth-order polynomial

$$F = \sum_{m=1}^4 F_m I^m,$$

while its derivative $\partial F / \partial H$ is a third-order polynomial with respect to I . Without writing out the coefficients $F_m = F_m(H_j, I_{j0})$ we note that F_1 is proportional to δk_0 , F_2 and F_3 are proportional to δk_b , therefore for $\delta k_b = 0$ the order of polynomial F is reduced to 2.

V. STRUCTURE OF GENERATED PULSE

From an applicational point of view, we consider the most interesting case of strong two-photon resonance, assuming the one- and three-photon detunings to be large $|\Delta_{1,3}| \gg |\Delta_2|, E_j d / \hbar$. All the situations different from this one will be pointed out. Thus Eq. (19) takes the form

$$\frac{dI}{dz} = \chi \frac{\sqrt{IU(I)}}{1 + \beta I}, \quad (23)$$

where

$$U(I) = (I_{10} - I)(I_{20} - I)(I_{30} + I) - I(\alpha I + \delta k_0 / \chi)^2, \quad (24)$$

and the wave-vector-mismatch dependence on coordinate is defined by

$$\delta k = \frac{\delta k_0 + 2\chi \alpha I}{1 + \beta I}, \quad (25)$$

where χ is the connection coefficient and δk_0 is the initial

mismatch. In order to clear up the meaning of α and β , we should mention that δk evolution is related to two factors. First, it is the population evolution that occurs in the process of interaction and is taken into account by the term $2\chi\alpha I$ in Eq. (25). Second, it is the exchange of effective two-photon detuning due to the dynamic Stark shift described by the parameter $(1 + \beta I)$. The dependencies of the parameters χ , δk_0 , α , and β on the task parameters are shown in the Appendix. We should mention that the form of Eqs. (23)–(25) will be the same either in the case of weak interaction or in the case of strong one- or three-photon resonance; only the χ , δk_0 , α , and β dependence on the interaction parameters will be modified. Therefore, the conclusion obtained below can be applied to these cases also.

For the sake of simplicity, in the present section we consider the case of $\beta S_1 \ll 1$, supposing the efficient two-photon detuning evolution during the FWM process is negligibly small. Assume that all roots S_j ($j=1,2,3$) of the equation $U(S)=0$ are real and can be set up in the following sequence: $S_1^{-1} \geq S_2^{-1} \geq S_3^{-1}$. (If two roots $S_2 = S_3^*$ are complex the analytical expansion into the complex variable region should be made.) Then Eq. (23) takes the form

$$I = S_1 \frac{\text{sn}^2(gz|\mu)}{1 + \nu \text{cn}^2(gz|\mu)}, \quad (26)$$

where $\text{sn}(x|\mu)$ and $\text{cn}(x|\mu)$ are the elliptic Jacobi functions [25–27] of variable x , and the gain g as well as parameters μ and ν are defined as

$$g = \frac{1}{2} \chi [I_{10} I_{20} I_{30} (S_1^{-1} - S_3^{-1})]^{1/2}, \quad (27)$$

$$\mu = \frac{1 - S_3/S_2}{1 - S_3/S_1}, \quad \nu = -\frac{S_1}{S_3}.$$

Equation (26) describes a periodical function, symmetric with respect to its maxima; its maximal value is equal to S_1 and is reached for $z = (2m + 1)l$, where $m = 0, 1$. The half-period value l is determined by the expression $l = K(\mu)/g$, where $K(\mu)$ is the full first-order elliptic integral. The function $K(\mu)$ has been tabulated [26,27]. However, the relation of Tricomi [31] allows us to obtain an extremely convenient estimation for l :

$$\ln \frac{e^\pi}{1 - \mu} \geq 2gl > \ln \left[\frac{16}{1 - \mu} \right]. \quad (28)$$

Using either the left side of Eq. (28) for $0 \leq \mu \leq 0.8$ or the right side for $0.8 \leq \mu \leq 1$ allows one to estimate l with relative error less than 5%. As is seen from Eqs. (27) and (28), $l \rightarrow \infty$ for strong convergence of two or three roots S_j ($\mu \rightarrow 1$). In the limit $\mu = 1$ the function $I(z)$ becomes nonperiodic, and the energy-transfer direction does not change its sign at all. However, the solution with $l = \infty$ is extremely unstable, i.e., any small deviation of the input variables makes the value l finite and for most practical cases can be estimated by $\pi/2 \leq gl \leq 12$.

Let us consider the temporal structure of the generated pulse ω_4 . In the adiabatic following regime τ is included in the propagation equation as a parameter. Since the transformation length l depends on the $I_{j0}(\tau)$, different

parts of the temporal pulse profile will have different lengths of transformation and reach their maximum at different distances. Therefore, during the evolution the pulses will acquire a complicated multi-peaked structure [Fig. 2(a)]. Consider the most simple example $\delta k_b = 0$, assuming the pulses at the entrance have the same temporal profile $I_{jb} = I_{j0}(0)\exp(-\tau/\tau_0)^2$. In this case $l(\tau) = l_0 \exp\{\tau^2/\tau_0^2\}$, where l_0 is a minimum value of the transfer length, which corresponds to the maximum intensity. The envelope of the ω_4 pulse represents in this case the set of subpulses [Fig. 2(a)]. Generation of the ω_4 wave starts from the central peak appearance at $z = (2m + 1)l$ and its disappearance is at $z = 2ml_0$, where $m = 0.1 \dots$. For $z > 2l_0$ the number of peaks $2n$ is determined by the integer part of the expression $n = [z/2l_0]$. Appearance of the r th peak is related to the time momentum τ_r

$$\tau_r = \pm \tau_0 [\ln(z/2rl_0)]^{1/2}, \quad (29)$$

where $1 \leq r \leq n$. The amplitude of peaks drops as $\exp\{-\tau_r^2/\tau_0^2\}$. The fact that for $z > l_0$ the energy transfer for different parts of the beam occurs in a different way shows that it is in principle impossible to perform the entire energy transfer, even in the ideal case of complete transfer for each separate part of the pulse. The only exclusion is either the case $l_0 = \infty$, or the case of strong medium saturation, where l becomes independent of τ . In both cases the pulse is not broken up into a set of subpulses.

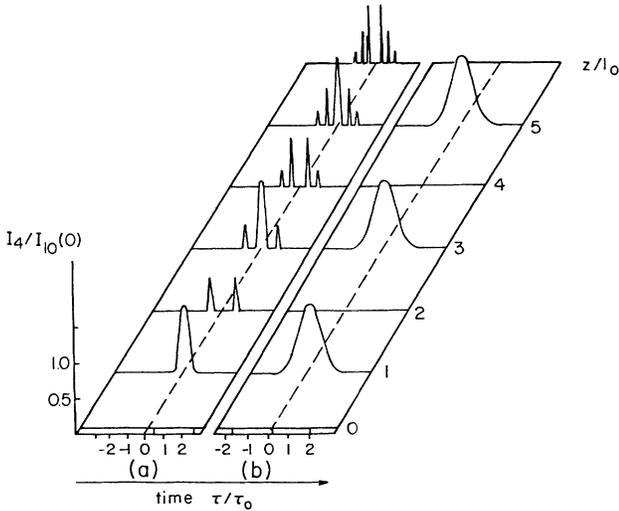


FIG. 2. Evolution of the shape of the ω_4 pulse during the propagation z/l_0 . (a) Weak field case. The generated pulse evolves into the set of subpulses, because of different conversion length for different parts of pulse. (b) Strong field case, with saturation of two-photon transition. The pulse evolves without breaking into the subpulses, since now the conversion length is independent of intensity. (a) and (b) are built up for the optimal condition Eq. (39) and (41a), respectively, supposing that all the pulses at the entrance have the same Gaussian envelope with time duration τ_0 ; $l_0 = l_s(\tau=0)$, $I_{10}(0) = I_{10}(\tau=0)$.

VI. AMPLIFICATION IN THE CASE OF A WEAK PROBE SIGNAL

Consider the evolution of the FWM process for a weak probe signal $I_{30} \ll I_{10}, I_{20}$. In this case the roots S_j are

$$S_3 = -I_{30} \frac{\chi^2 I_{10} I_{20}}{\chi^2 I_{10} I_{20} - \delta k_0^2}, \quad (30)$$

$$S_{1,2} = \frac{I_{10} + I_{20} + 2\alpha\delta k_0/\chi}{2(1-\alpha^2)} (1 \pm \sqrt{1-M}),$$

where

$$M = \frac{4(1-\alpha^2)(I_{10}I_{20} - \delta k_0^2/\chi^2)}{[I_{10} + I_{20} + 2\alpha\delta k_0/\chi]^2}. \quad (31)$$

In the limit considered we have $|S_{1,2}| \gg |S_3|$, thus the parameters μ and ν obey the relations $1 \gg 1-\mu$ and $\nu \gg 1$. Since the function $I(z)$ is periodical and symmetric about its maximum, we can restrict our analysis of this function to the half period $0 \leq z \leq l$. Using the asymmetrical representation of the elliptic functions [25–27], valid for $\mu \rightarrow 1$ and substituting it into Eq. (26), in the case $\beta S_1 < 1$ we obtain

$$I = \frac{S_1 |S_3| \sinh^2(gz)}{S_1 + |S_3| \cosh^2(gz)}, \quad (32)$$

where

$$g = \frac{1}{2} (\chi^2 I_{10} I_{20} - \delta k_0^2)^{1/2}. \quad (33)$$

For small length of interaction $z < l_s$, where $l_s = g^{-1} \ln |S_1/S_3|$, Eq. (32) is reduced to the well-known expressions (see, for example [2–5]) obtained in the nondepleted pump beam approach:

$$I = \frac{\chi I_{10} I_{20} I_{30}}{\chi^2 I_{10} I_{20} - \delta k_0^2} \sinh^2(gz). \quad (34)$$

The value l_s characterizes the length of interaction that leads to saturation of the conversion. In most cases l_s is practically equal to the period l , therefore Eq. (34) is valid for the whole region $0 \leq z \leq l$. A significant difference from the above-mentioned evolution occurs in the case of strong S_1 and S_2 roots convergence, i.e., $|S_1 - S_2| \leq I_{30}$. In this case $l > l_s$, and the nondepleted pump beam approach is valid within the range $0 \leq z \leq l_s$, where the intensity of the generated waves rises exponentially; for $z > l_s$, the variable $I(z)$ approaches its maximum value S_1 .

Now we consider another limit $\beta S_1 \gg 1$, that corresponds to strong two-photon detuning modification during the propagation. In this case the fast exponential intensity growth Eq. (33) may occur for a relatively short length of interaction for which $\beta I \leq 1$. For larger lengths, the rate of growth falls sharply and is determined by

$$I = S_1 \cos^2[g_e(z-l)] + S_2 \sin^2[g_e(z-l)], \quad (35)$$

where l is the half period and g_e is the efficient gain

$$l = \frac{2}{g_e} \arccos \frac{|S_2| - S_1}{|S_2| + S_1}, \quad g_e = \chi \frac{(|1 - \alpha^2|)^{1/2}}{4\beta}. \quad (36)$$

We see that in the case $\beta S_1 \gg 1$, the gain is decreased by factor $\beta |S_1 S_2|^{1/2}$, while a half period of transfer is respectively increased by the same factor in comparison with the case $\beta S_1 < 1$. At the same time the conversion efficiency S_1 does not depend on parameter β completely. For intense pump, which saturates the population of the ground and two-photon excited third levels, the expressions for δk_0 , l , g , and g_e become independent of pump intensity. In this case the pulse ω_4 splitting does not occur [Fig. 2(b)] leading to the ideal conditions for the conversion-efficiency optimization.

VII. CONVERSION EFFICIENCY

It follows from Sec. IV that the rate of conversion can be determined without solving Eq. (19), but simply by finding the value S_1 representing the least positive root of Eq. (20). The optimization of conversion process is thus reduced to the optimization of S_1 as a function of the input parameters. It follows from Eqs. (16) and (17) that $S_1 \leq I_{\min} = \min\{I_{10}, I_{20}\}$, i.e., the energy transfer changes its direction before the complete depletion of any pump waves occurs. However in some cases the conversion efficiency may be optimized up to a limit $S_1 = I_{\min}$. In order to provide it we require that the following condition be met:

$$F(I = I_{\min}) = 0. \quad (37)$$

Since $F(I_{\min}) = 0$, the value I_{\min} in (37) will be one of the roots of Eq. (20). If I_{\min} is also the least positive root, it will automatically mean that FWM process evolution occurs with maximal rate of conversion $S_1 = I_{\min}$, i.e., up to the complete depletion of one of the pump waves. The most simple way to provide it is to vary δk_b by modifying the buffer gas pressure or input wave inclination angles. The function F is the fourth-order polynomial with respect to the parameter δk_b . Therefore the optimal value of δk_b , which provides the complete depletion of one of the pump waves, is defined as one of the roots of Eq. (37), being solved with respect to δk_b . Let us illustrate this for several of the simplest examples.

(i) Analysis of Eqs. (24) and (25) shows that the optimal condition is

$$\delta k_0 = 0, \quad \alpha = 0. \quad (38)$$

In this case, the FWM process evolves in the regime of permanent phase matching $\delta k(z) = 0$ and the energy transfer into the waves $\omega_{3,4}$ will be terminated either for complete depletion of the ω_1 wave if $I_{10} \leq I_{20}$, or for complete depletion of ω_2 if $I_{10} \geq I_{20}$. Such a situation is available in the weak (nonsaturable) pump if $|\Delta_1| \gg |\Delta_{2,3}|$. In this case the conditions $\delta k_0 = 0$ and $\alpha = 0$ correspond to the following relation:

$$\delta k_b = \frac{N\gamma_4}{\Delta_3}, \quad \Delta_3\gamma_3 = \Delta_2\gamma_4. \quad (39)$$

In most cases of interest it is impossible to fulfill condition (38), since the $\alpha = 0$ requires frequency detuning within too wide a range. Thus for example, the analysis of expressions (A1) and (A2) in the Appendix shows that in the case of a strong one-, two-, or three-photon resonance it is impossible to realize the regime of permanent phase matching.

(ii) However, high conversion efficiency can be reached without permanent phase matching in the regime of variable wave-vector mismatch $\delta k(z)$. The optimization procedure can be successfully performed for definite pump frequencies by variation of δk_0 (more precisely δk_b) only. Consider for example the case of a strong two-photon resonance in the limit of weak probe beam $I_{30} \ll I_{10}, I_{20}$ ($I_{20} \geq I_{10}$). The equality $U(S) = 0$ shows that the root S_1 as a function of parameter δk_0 has no maximum for $\delta k_0 = 0$. The value S_1 is increased if δk_0 increases with the sign opposite to the sign of α . The wave-vector mismatch [Eq. (25)] for such a relationship between δk_0 and α is gradually decreased in the process of conversion. The analysis of Eq. (30) shows that the optimization process is determined by the parameter $\alpha^2 I_{10}/I_{20}$.

(a) In the case $\alpha^2 I_{10} < I_{20}$ we get from Eq. (30)

$$S_1 = I_{10} \quad \text{for} \quad \delta k_0 = -\alpha\chi I_{10}. \quad (40a)$$

It is interesting to point out that FWM evolution occurs with periodical sign exchange of the wave-vector mismatch. Thus for $\beta I_{10} \ll 1$ it follows from Eq. (25) that $\delta k = \delta k_0$ for $z = 2ml$ and $\delta k = -\delta k_0$ for $z = (2m + 1)l$, where $m = 0, 1, \dots$.

(b) In the case $\alpha^2 I_{10} > I_{20}$ the optimal condition being defined from Eq. (30) looks as follows:

$$S_1 \rightarrow \frac{2|\alpha|\sqrt{I_{10}I_{20}} - I_{10} - I_{20}}{\alpha^2 - 1} \quad \text{for} \quad \delta k_0 \rightarrow \pm\chi\sqrt{I_{10}I_{20}}. \quad (40b)$$

The sign in (40b) is chosen to be opposite to that of α . We do not use the equality since the gain g in this case tends to be zero. As we see, for large Kerr nonlinearity $|\alpha| \gg 1$ the efficiency of the process sharply falls down.

The dependence of the conversion efficiency on δk_0 is shown in Fig. 3. The curves are bounded on both sides, since for $|\delta k_0| = \chi\sqrt{I_{10}I_{20}}$ breakdown of generation occurs. It is seen that with increasing α we decrease the range of possible δk_0 variation, decreasing in turn the maximally reached conversion efficiency. In the case $\delta k_0 = 0$ the period of transfer l reaches its minimum. In the case of δk_0 variation the efficiency becomes higher, whereas the period l becomes larger (Fig. 4).

(iii) The case $\Delta_2 \rightarrow 0$ is of special interest because of two-photon saturation. In this case, expressions (40) become independent of pump intensity, therefore the optimization condition will be uniform for the whole interacting beam profile, excluding the weak pulse wings. Thus for instance, in the case of degenerated pump ($\omega_1 = \omega_2$), Eqs. (40) are reduced to the following:

$$S_1 = I_{10} \quad \text{for} \quad \delta k_0 = -\frac{2\alpha N\sqrt{\gamma_1\gamma_2\gamma_3\gamma_4}}{|\Delta_1|(\gamma_1 + \gamma_2)} \quad (41a)$$

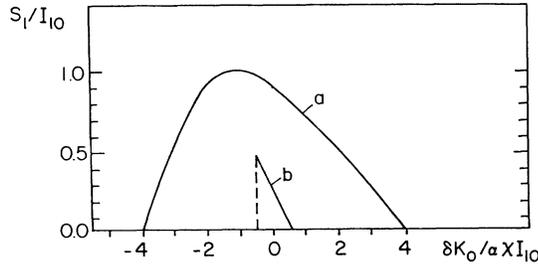


FIG. 3. Conversion-efficiency dependence on the parameter $\delta k_0/\alpha\chi I_{10}$ for different values of $\alpha^2 I_{10}/I_{20}$. (a) $\alpha^2 I_{10}/I_{20} = \frac{1}{4}$. The optimization may reach the maximal point $S_1 = I_{10}$. (b) $\alpha^2 I_{10}/I_{20} = 4$. The maximal value of conversion efficiency is achieved for a value of δk_0 that in turn leads to the disruption of generation. Both curves are built up for the case $I_{20} = 2I_{10} \gg I_{30}$.

if $|\alpha| < 1$, or

$$S_1 \rightarrow \frac{2I_{10}}{|\alpha| + 1} \quad \text{for } \delta k_0 \rightarrow \pm \frac{2N\sqrt{\gamma_1\gamma_2\gamma_3\gamma_4}}{|\Delta_1(\gamma_1 + \gamma_2)} \quad (41b)$$

if $|\alpha| > 1$. The sign in Eq. (41b) is chosen to be opposite to that of α , the value of δk_0 in the limit considered ($\Delta_2 \rightarrow 0$) is determined by the expression

$$\delta k_0 = \delta k_b + \frac{2N(\gamma_1 d_1^2 + \gamma_2 d_2^2)}{\Delta_1(d_1^2 + d_2^2)} - \frac{N(\gamma_4 d_1^2 + \gamma_3 d_2^2)}{\Delta_3(d_1^2 + d_2^2)}. \quad (42)$$

Equation (42) is valid for $d_1 > d_2$; when $d_2 > d_1$ their values should be interchanged $d_1 \leftrightarrow d_2$.

(iv) The ideal for conversion process is expected to be the case of strong probe wave I_{30} ; when $I_{30} \gg I_{10}, I_{20}$, $|\alpha\delta k_0/\chi|$. In this limit we have $S_1 = I_{10}$, $S_2 = I_{20}$, and $S_3 = -I_{30}/(1 - \alpha^2)$. Then, for $\beta I_{10} < 1$ we obtain from (26)

$$I = I_{10} \text{sn}^2(gz|\mu), \quad (43)$$

where the gain g and half period l of $I(z)$ are defined as

$$g = \frac{1}{2}\chi\sqrt{I_{20}I_{30}}, \quad l = K(\mu)/g, \quad \mu = I_{10}/I_{20}. \quad (44)$$

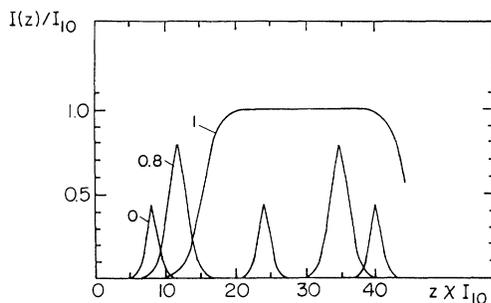


FIG. 4. Conversion-efficiency dependence on normalized length $z\chi I_{10}$ for different values of initial wave-vector mismatch $\delta k_0 = 0, -0.8\alpha\chi I_{10}$, and $-\alpha\chi I_{10}$. An increase of conversion efficiency is achieved together with enlarging of the conversion length. For small lengths the conversion is mainly effective in the case $\delta k_0 = 0$. The curves are built up for the case $\alpha < 1$, $I_{20} = 2I_{10} \gg I_{30}$.

Using the asymptotic representation for the elliptic sine, we may replace Eq. (43) by the following approximation (suppose $I_{10} \leq 0.5I_{20}$):

$$I = I_{10} \sin^2(\pi z/2l). \quad (45)$$

In the case $I_{10} \rightarrow I_{20}$ ($\mu \rightarrow 1$), we have $l \rightarrow \infty$, so Eq. (43) becomes

$$I = I_{10} \tanh^2(gz). \quad (46)$$

The limit $I_{30} \rightarrow \infty$ is characterized by the maximal conversion $S_1 = I_{10}$ and its feature is that the gain g is independent of I_{10} and δk_0 , allowing one to vary δk_0 within wide enough range. The case $\beta I_{10} \gg 1$ differs from Eqs. (43)–(46) by the increasing of transfer period l (approximately by $\beta\sqrt{I_{10}I_{20}}$ times) only. Therefore the optimization procedure in this case should be reduced to the minimization of parameter β in order to decrease the period of transfer. Using Eq. (A1) we can reach $\beta = 0$, if the following condition is fulfilled:

$$\delta k_b = \frac{N(\gamma_3 + \gamma_4)}{2\Delta_3} - \frac{N(\gamma_1 + \gamma_2)}{2\Delta_1}. \quad (47)$$

All the expressions from (39)–(47) are valid in the case $I_{10} > I_{20}$ by simply exchanging $I_{10} \leftrightarrow I_{20}$.

VIII. QUASIENERGY INTERSECTION: DISRUPTION OF GENERATION

In this section we consider the phenomenon of quasienergy crossover related to the well-known self-induced resonance in a three-level system [28–30].

In some cases the polynomial $\partial F/\partial H$ in the denominator of Eq. (19) may have a positive root η less than S_1 . In this case Eq. (19) contains a resonance that becomes important for the lengths close to the critical length l_{cr} determined by the relation $I(l_{cr}) = \eta$. It can be easily shown that the resonance in Eq. (19) for $z \rightarrow l_{cr}$ means physically the intersection of quasienergetic terms of the coupled system atom in the fields, e.g., the self-induced resonance phenomena [28–30]. In other words, for $I \rightarrow \eta$ the Stark shift of atomic levels leads the system to exact one- or two-photon resonance with the incident field. This process is followed by strong absorption of the interacting waves. Thus, for $z \sim l_{cr}$ the conversion process is practically over, therefore its efficiency is limited by η . It is evident that the theory becomes invalid for $z \geq l_{cr}$.

We return to the case of two-photon resonance, where ground and third atomic levels intersect. The effective two-photon detuning with Stark shift included is $\Omega_1 - \Omega_3 = \Delta_{20}(1 + \beta I)$, where Δ_{20} is the value of effective two-photon detuning at $z = 0$ (see Appendix). We see that $\beta = 0$ leads to a constant value of two-photon detuning during the conversion (Fig. 5). For $\beta > 0$ the quasienergies diverge, leading to the increase of two-photon detuning. This leads to an increase of the conversion length. For $\beta < 0$, the quasienergies approach each other, leading to resonant enhancement of the interaction, and a decrease of the conversion length. It is seen from Eq. (23) that the results of Secs. V–VII are still valid for

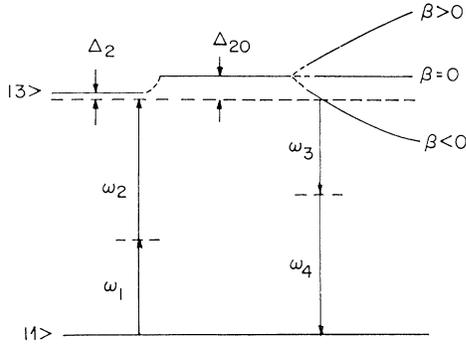


FIG. 5. Evolution of quasienergetic levels in FWM process during the propagation. At the entrance the two-photon detuning is determined by Δ_{20} , which includes the Stark shift of atomic levels. For $\beta=0$ this value is held constant during evolution of the process. For $\beta>0$, the levels diverge, increasing Δ_{20} . For $\beta<0$ the levels approach each other, leading the system at the point $z=l_{cr}$ to exact two-photon resonance.

$\beta S_1 > -1$. The intersection of quasienergies and related saturation of the FWM process is possible for large negative β (e.g., for small two-photon detuning), when $\beta S_1 < -1$. For the last case the generation efficiency is defined as

$$\eta = \frac{1}{|\beta|}. \quad (48)$$

The critical length value can be estimated either by Eq. (33) or Eq. (43). Thus, in the case $I_{10,20} \gg I_{30}$,

$$l_{cr} \sim \frac{1}{(\chi^2 I_{10} I_{20} - \delta k_0^2)^{1/2}} \ln \frac{1}{|\beta I_{30}|}, \quad (49)$$

and in the case $I_{30} \gg I_{20} \gg I_{10}$,

$$l_{cr} \sim \frac{1}{\chi \sqrt{I_{30} I_{20}}} \ln \frac{1}{|\beta I_{10}|}. \quad (50)$$

We see from Eqs. (48)–(50) and Eq. (A1) that for $\beta < 0$ and strong two-photon resonance $\beta \sim 1/\Delta_2$, we have the decreasing of both the conversion efficiency and the length l_{cr} .

The sign of β coincides [see Eqs. (A1)–(A3)] with the sign of $\Delta_2 Q$, where Q is

$$Q = \delta k_b + \frac{N(\gamma_1 + \gamma_2)}{2\Delta_1} - \frac{N(\gamma_3 + \gamma_4)}{2\Delta_3}. \quad (51)$$

We see that for $\Delta_2 Q > 0$ the conversion efficiency is determined by the parameter S_1 , whose magnitude is independent of β , whereas for $\Delta_2 Q < 0$ the conversion drops to $\eta = 1/\beta$, which for $\Delta_2 \rightarrow 0$ is much less than S_1 . In other words, the sign exchange of β (e.g., sign of $\Delta_2 Q$) leads to the disruption of generation. We assume that such phe-

nomena have been observed in the case of frequency sweeping through the resonance [32] as well as in the case of buffer gas or alkali vapor pressure variation [33] (in this case, parameter Q also changes sign).

The maximum resonant enhancement of interaction and shortening of the conversion length may be achieved if $\beta \rightarrow -(1/S_1) + 0$. [However, the efficient two-photon detuning $\Delta_2(1 + \beta S_1)$ should be held larger than the width of absorption line.]

In summary, we should mention that the point $\beta = -1/S_1$ is the separatrix of the one-dimensional phase space, where chaotic modifications of the period of motion as well as the conversion efficiency occur. Therefore, in the vicinity of such a point the motion acquires stochastic character [34]. However, this question needs special consideration.

IX. CONCLUSION

The results enable us to consider the FWM process with arbitrary rate of conversion, taking into account the pump beam depletion as well as the effects of coherent medium saturation, essential for resonant interaction. Because of the large number of parameters involved, we have restricted our consideration by applying the general results to several particular cases. Thus, we did not consider the case where an initially ($z=0$) weak pump field $\omega_{1,2}$ interaction becomes strong during the evolution and leads to conversion into the waves $\omega_{3,4}$. Such a situation is typical for small third-photon detuning Δ_3 ($|\Delta_3| \ll |\Delta_{1,2}|, |E_{j0} d_j / \hbar|$) and is characterized by a low conversion efficiency. The theory developed will be applied to the case of sum frequency generation $\omega_1 + \omega_2 + \omega_3 = \omega_4$ (particularly, for the most interesting case of third-harmonic generation $3\omega_1 = \omega_4$) by the new phase ϕ and wave-vector-mismatch definition $\phi = \phi_1 + \phi_2 + \phi_3 - \phi_4$ and $\delta \mathbf{k} = \mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3 - \mathbf{k}_4$. A corresponding exchange of the Manly-Rowe relation should also be made. Thus for instance, Eqs. (19) and (23) can describe the process $\omega_1 + \omega_2 + \omega_3 = \omega_4$ if the following substitution $I_{30} + I \rightarrow I_{30} - I$ into Eqs. (16), (17), and (24) is made, or the process $3\omega_1 = \omega_4$ if the substitution $I_{10} - I, I_{20} - I, I_{30} + I \rightarrow I_{10} - 3I$ is made. After such substitutions, all the results and conclusions of Secs. V–VIII will be completely valid for these processes.

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APPENDIX

In the case of strong two-photon resonance the dependence of values α , β , δk_0 , and χ on the input parameters is as follows:

$$\chi = \frac{32\pi^2 N d_1 d_2 d_3 d_4 (\omega_2 \omega_3 \omega_4)^{1/2}}{\hbar^2 c^2 |\Delta_1 \Delta_{20} \Delta_3|},$$

$$\delta k_0 = \delta k_b + \frac{N(\gamma_1 |a_{10}|^2 + \gamma_2 |a_{30}|^2)}{\Delta_1} - \frac{N(\gamma_4 |a_{10}|^2 + \gamma_3 |a_{30}|^2)}{\Delta_3},$$

$$\alpha = \pm \frac{\Delta_1 \Delta_3}{2N^2 \sqrt{\gamma_1 \gamma_2 \gamma_3 \gamma_4}} \left[\delta k_b^2 + N \delta k_b \left(\frac{\gamma_1 + \gamma_2}{\Delta_1} - \frac{\gamma_3 + \gamma_4}{\Delta_2} \right) - \frac{N^2 (\gamma_1 \gamma_3 + \gamma_2 \gamma_4)}{\Delta_1 \Delta_3} \right], \quad (\text{A1})$$

$$\beta = \frac{2}{N \Delta_{20}} \left[\delta k_b + \frac{N(\gamma_1 + \gamma_2)}{2\Delta_1} - \frac{N(\gamma_3 + \gamma_4)}{2\Delta_3} \right],$$

where the distance between quasienergetic terms, e.g., the value of efficient two-photon detuning (including the Stark shifts) is

$$\Omega_3 - \Omega_1 = \Delta_{20}(1 + \beta I), \quad \Delta_{20} = \pm (\delta_0^2 + W_0^2)^{1/2}. \quad (\text{A2})$$

The sign of α and Δ_{20} must equal to the sign of Δ_2 ; Δ_{20} is the input ($z=0$) value of efficient two-photon detuning. The values $|a_{10}|^2 = \frac{1}{2}[1 + 1/\sqrt{1+\xi}]$ and $|a_{30}|^2 = \frac{1}{2}[1 - 1/\sqrt{1+\xi}]$ are the coherent populations of atomic $\psi_{1,3}$ states at the entrance ($z=0$):

$$\xi = \frac{W_0^2}{\delta_0^2}, \quad W_0 = 2 \left| \frac{E_{10} E_{20} d_1 d_2}{\hbar^2 \Delta_1} \right| \quad (\text{A3})$$

$$\delta_0 = \Delta_2 + \frac{|E_{10} d_1|^2 - |E_{20} d_2|^2}{\hbar^2 \Delta_1} - \frac{|E_{30} d_3|^2}{\hbar^2 \Delta_3}.$$

For weak pump field ($\xi \ll 1$), $\Delta_{20} = \Delta_2$, $a_{10} = 1$, $a_{30} = 0$, and consideration reduces to the perturbation-theory approach. In the opposite case, when $\xi \gg 1$ the population of $\psi_{1,3}$ levels become equal: $|a_{10}|^2 = |a_{30}|^2 = \frac{1}{2}$.

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