Time-dependent adiabatic four-wave mixing in gases

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The self-consistent problem of four-wave mixing in a gas of four-level atoms (or molecules) is solved without the assumption that the interacting fields are weak and without assuming a given pump field. The wave propagation equations have the form of Hamilton's equations (the canonical equations), so the mathematical apparatus of classical mechanics can be employed, and all the integrals of motion can be determined. An exact solution of the problem is derived; energy exchange between waves, a transfer of population, and saturation of the atomic transitions are taken into account rigorously. The problem of determining the conversion efficiency reduces to one of finding the smallest positive root of a polynomial of degree lower than eighth. Possibilities for optimizing the conversion process are pointed out.

1. INTRODUCTION

In the theory of four-wave mixing, the nonlinear polarization of the medium is usually expanded in a power series in the interacting fields, and only the lowest-order terms contributing to the process are retained (Refs. 1–5, for example). The analysis is restricted to short interaction regions, for which the conversion efficiency is so small that one can assume that the pump is fixed and ignore the transfer of population between atomic levels. In that approach, however, one cannot analyze four-wave mixing if the degree of conversion is high, one cannot determine the extent of conversion at which the conversion process reaches saturation, and one cannot find all the integrals of motion. Moreover, that approach becomes incorrect if there are strong resonances, and one of the atomic transitions reaches saturation during the amplification of the weak waves, or if saturation prevails at the very outset, at the entrance to the resonant medium.

In this paper we analyze four-wave mixing for the case of the two-photon excitation of a four-level medium in the conversion $\omega_1 + \omega_2 = \omega_3 + \omega_4$, in each elementary event of which the pumps, of frequencies ω_1 and ω_2, are absorbed, and the photons ω_3 and ω_4 are emitted (and vice versa). The procedure which has been developed makes it possible to solve this problem with an exact account of effects stemming from the coherent saturation of atomic transitions and from energy exchange between waves, to the point at which one wave is totally depleted. Equations derived below [Eqs. (16)] cover not only the reaction mechanism $\omega_1 + \omega_2 \to \omega_3 + \omega_4$, which describes a transfer of intensity from pulses $\omega_{1,2}$ to pulses $\omega_{3,4}$, but also (and rigorously) the inverse mechanism, $\omega_3 + \omega_4 \to \omega_1 + \omega_2$, as well as the interference between these mechanisms. This new computation method is based on finding an integral of motion, (11). This integral of motion cannot be determined by the usual approach (in the case of a collinear interaction, this integral is the quasi-energy of the bound system consisting of the atom and the external field).

2. QUASIENERGY AND POLARIZATION OF AN ATOM

Let us examine the behavior of a four-level system (Fig. 1) in the field of four waves, with carrier frequencies ω_j ($j = 1, 2, 3, 4$) which satisfy the condition $\omega_1 + \omega_2 = \omega_3 + \omega_4$. The lengths of the interacting pulses are assumed to be small in comparison with all the relaxation times of the system. We assume that the pulses are adiabatic, ignoring possible absorption processes. We seek the wave amplitudes in the form

$$E_j(\mathbf{r}, t) = E_j \exp(-i\omega_j t) + \text{c.c.}, \quad E_j = \mathscr{E}_j \exp(i\mathbf{k}_j\mathbf{r}), \tag{1}$$

where $k_j = \omega_j / c$. We assume that the waves propagate at small angles from the vector $\mathbf{k}_1 + \mathbf{k}_2$ (the x axis). In the resonance approximation,[6] we seek a solution of the Schrödinger equation as the following superposition of unperturbed functions ψ_j:

$$\Psi = e^{-i\Omega t}\{a_1\psi_1 + a_2\psi_2 e^{-i\omega_1 t} + a_3\psi_3 e^{-i(\omega_1+\omega_2)t} + a_4\psi_4 e^{-i\omega_4 t}\}. \tag{2}$$

The ground level is adopted as the origin for the energy scale. Substituting (2) into the Schrödinger equation, we find a system of equations for the amplitudes a_j. We write these equations in matrix form:

$$\begin{pmatrix} 0 & -E_1^* d_{12} & 0 & -E_4^* d_{14} \\ -E_1 d_{21} & \hbar\Delta_1 & -E_2^* d_{23} & 0 \\ 0 & -E_2 d_{32} & \hbar\Delta_2 & -E_3 d_{43} \\ -E_4 d_{41} & 0 & -E_3^* d_{34} & \hbar\Delta_3 \end{pmatrix} \begin{pmatrix} a_1 \\ a_2 \\ a_3 \\ a_4 \end{pmatrix} = \hbar\Omega \begin{pmatrix} a_1 \\ a_2 \\ a_3 \\ a_4 \end{pmatrix}, \tag{3}$$

where

$$\Delta_1 = \omega_{21} - \omega_1, \quad \Delta_2 = \omega_{31} - \omega_1 - \omega_2, \quad \Delta_3 = \omega_{41} - \omega_4 \tag{4}$$

is the detuning from resonance, and d_{ij} are the dipole matrix elements, which can be assumed to be real. The quasi-energy Ω is found from the condition under which Eqs. (3) have a nontrivial solution:

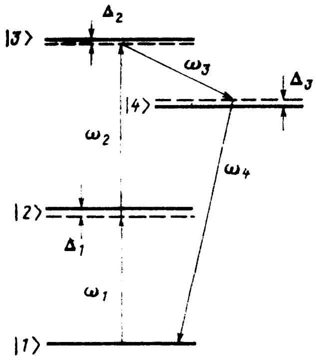

FIG. 1. Level scheme.

$$\hbar^4\Omega(\Omega-\Delta_1)(\Omega-\Delta_2)(\Omega-\Delta_3)-\hbar^2(\Omega-\Delta_2)(\Omega-\Delta_3)d_{12}{}^2|E_1|^2 \\ -\hbar^2\Omega(\Omega-\Delta_3)d_{23}{}^2|E_2|^2-\hbar^2\Omega(\Omega-\Delta_1)d_{43}{}^2|E_3|^2-\hbar^2(\Omega-\Delta_1) \\ \times(\Omega-\Delta_2)d_{41}{}^2|E_4|^2+d_{12}{}^2d_{43}{}^2|E_1E_3|^2+d_{14}{}^2d_{23}{}^2|E_2E_4|^2 \\ -d_{12}d_{23}d_{34}d_{41}(E_1E_2E_3{}^*E_4{}^*+\text{c.c.})=0. \qquad (5)$$

If the interaction is turned on adiabatically, the unperturbed states of the system go into corresponding quasi-energy states, so the roots of Eq. (5) should be numbered in accordance with the conditions $\Omega_1\to 0$, $\Omega_2\to\Delta_1$, $\Omega_3\to\Delta_2$, and $\Omega_4\to\Delta_3$ as $E_j\to 0$. We assume that the system starts out in its ground state. After the interaction is turned on, the state of the system is then described by wave function (2), into which we are to substitute the amplitudes $a_j^{(1)}$, which are the solutions of Eqs. (3) with $\Omega=\Omega_1$. When we write the expression for the polarization of the system in state (2), it becomes obvious that the direct substitution of the amplitudes $a_j^{(1)}$ would put this expression in a totally opaque form. We instead use the method developed in Ref. 7, which does not require finding explicit expressions for the amplitudes $a_j^{(1)}$ and which is convenient for the analysis below. Specifically, the polarization of an atom in state Ψ can be written in the form

$$P=-\hbar\sum_{j=1}^{4}\frac{\partial\Omega_1}{\partial E_j^*}\exp(-i\omega_jt)=-\hbar\sum_{i=1}^{4}\frac{\partial\Omega_1}{\partial\mathscr{E}_j^*}\exp[i(\mathbf{k}_j\mathbf{r}-\omega_jt)]. \qquad (6)$$

We will not reproduce the proof of (6) here; it is similar to a proof in Ref. 8. Note that the matrix equation (3) has the same form as the steady-state Schrödinger equation, with a Hamiltonian which depends on E_j and E_j^* as parameters. Making use of this circumstance and also the known expression for the diagonal matrix element of the derivative of a Hamiltonian with respect to its parameter,[9] we can easily derive expression (6).

3. WAVE PROPAGATION EQUATIONS

Let us examine the self-consistent problem of the propagation of waves $\mathscr{E}_j$ ($j=1, 2, 3, 4$) through a four-level medium which fills the half-space $x>0$. We treat the one-dimensional problem, assuming that the phase-matching conditions hold along the directions transverse with respect to the x axis. Substituting (6) into the wave equation, and carrying out a truncation operation, we obtain

$$\frac{d\mathscr{E}_j}{dx}=-i\frac{2\pi N\hbar\omega_j}{c}\frac{\partial\Omega_1}{\partial\mathscr{E}_j^*}, \qquad (7)$$

where N is the density of atoms. The usual approach to this problem corresponds to an expansion of the quasi-energy in a power series in the quantities $\mathscr{E}_j$. Substitution of the first term of the expansion of Ω_1, found from (5), into (7), leads to the well known equations for the resonant refractive indices at the frequencies ω_j:

$$n_1=1+\frac{2\pi Nd_{12}{}^2}{\hbar\Delta_1}, \quad n_2=n_3=1, \quad n_4=1+\frac{2\pi Nd_{14}{}^2}{\hbar\Delta_3}.$$

If we were to retain the terms of the expansion of the next order, we would obtain the equations which were studied in Ref. 1 and several other places. Those equations ignore coherent saturation and cannot be used to determine all the integrals of motion.

Thanks to representation (6) of the polarization, we are able to solve Eq. (7) exactly. We convert (7) and the equations which are its complex conjugates through the substitution

$$\mathscr{E}_j=|\mathscr{E}_j|\exp(i\varphi_j)=(2\pi\hbar\omega_jI_j/c)^{1/2}\exp(i\varphi_j), \qquad (8)$$

where $I_j=c|\mathscr{E}_j|^2/(2\pi\hbar\omega_j)$ is the photon flux density. Separating the real and imaginary parts, we find from (7)

$$\frac{dI_j}{dx}=N\frac{\partial\Omega_1}{\partial\varphi_j}, \quad \frac{d\varphi_j}{dx}=-N\frac{\partial\Omega_1}{\partial I_j}. \qquad (9)$$

Equation (5) for the quasienergy contains the phases φ_j only in the linear combination $\theta=\varphi_1+\varphi_2-\varphi_3-\varphi_4+qx$, where q is the x projection of the vector $\mathbf{q}=\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_3-\mathbf{k}_4$. It is thus a simple matter to derive the relations

$$\frac{\partial\Omega_1}{\partial\varphi_1}=\frac{\partial\Omega_1}{\partial\varphi_2}=-\frac{\partial\Omega_1}{\partial\varphi_3}=-\frac{\partial\Omega_1}{\partial\varphi_4},$$

with the help of which we find from (9)

$$\begin{gathered} I_1+I_2+I_3+I_4=I_{10}+I_{20}+I_{30}+I_{40}, \\ I_1-I_2=I_{10}-I_{20}, \\ I_3-I_4=I_{30}-I_{40}, \end{gathered} \qquad (10)$$

where I_{j0} are the entrance values of the quantities I_j. The meaning of the integrals of motion in (10) is obvious, since we are dealing with four-wave mixing in a lossless dielectric. Using (10) and the relations $I_1=I_{10}-J$, $I_2=I_{20}-J$, $I_3=I_{30}+J$, and $I_4=I_{40}+J$, we can switch from the four unknowns I_j to the single unknown $J=J(x)$, which is a measure of the energy exchange between waves. The initial condition on J is $J=0$ at $x=0$.

We introduce

$$\Omega_{10}=\Omega_1+qJ/N. \qquad (11)$$

Substituting $\Omega_1=\Omega_{10}-qJ/N$ and either $|\mathscr{E}_j|^2=\gamma_j(I_{j0}-J)$ (for $j=1, 2$) or $|\mathscr{E}_j|^2=\gamma_j(I_{j0}+J)$ (for $j=3, 4$) into (5), we find the equation

$$F=f\cos\theta, \qquad (12)$$

which determines Ω_{10} as a function of the parameters J and θ. Here γ_j, $F=F(J,\Omega_{10})$, and $f=f(J)$ are given by

$$f=[(I_{10}-J)(I_{20}-J)(I_{30}+J)(I_{40}+J)]^{1/2}, \qquad (13)$$

$$F=\frac{1}{2(\gamma_1\gamma_2\gamma_3\gamma_4)^{1/2}}\left\{\left(\Omega_{10}-\frac{qJ}{N}\right)\left(\Omega_{10}-\Delta_1-\frac{qJ}{N}\right)\right.$$

$$\times\left(\Omega_{10}-\Delta_2-\frac{qJ}{N}\right)\cdot\left(\Omega_{10}-\Delta_3-\frac{qJ}{N}\right)-\left(\Omega_{10}-\Delta_3-\frac{qJ}{N}\right)$$

$$\times\left[\gamma_1(I_{10}-J)\left(\Omega_{10}-\Delta_2-\frac{qJ}{N}\right)\gamma_2(I_{20}-J)\left(\Omega_{10}-\frac{qJ}{N}\right)\right]$$

$$-\left(\Omega_{10}-\Delta_1-\frac{qJ}{N}\right)\left[\gamma_3(I_{30}+J)\left(\Omega_{10}-\frac{qJ}{N}\right)\right.$$

$$\left.+\gamma_4(I_{40}+J)\left(\Omega_{10}-\Delta_2-\frac{qJ}{N}\right)\right]+\gamma_1\gamma_3(I_{10}-J)$$

$$\left.\times(I_{30}+J)+\gamma_2\gamma_4(I_{20}-J)(I_{40}+J)\right\}, \tag{14}$$

$$\gamma_j=\frac{2\pi\omega_j d_j^2}{\hbar c},\qquad d_1=d_{12},\qquad d_2=d_{23},\qquad d_3=d_{34},\qquad d_4=d_{41}. \tag{15}$$

Carrying out a canonical transformation, we find the following equations for J and θ from (9):

$$\frac{dJ}{dx}=-N\frac{\partial\Omega_{10}}{\partial\theta},\quad \frac{d\theta}{dx}=N\frac{\partial\Omega_{10}}{\partial J}. \tag{16}$$

Equations (16) obviously have the form of Hamilton's equations (the canonical equations) for classical one-dimensional motion, if we treat J and θ as a generalized coordinate and a generalized momentum. The role of the time is played by the coordinate x, and the role of the Hamiltonian is played by $N\Omega_{10}$. It can be seen from (12)–(14) that Ω_{10} does not depend explicitly on x, so it is an integral of motion $(d\Omega_{10}/dx=0)$. The value of this integral of motion can be found easily by examining the interaction at $x=0$. Since $J=0$ at $x=0$, it follows from (11) that the value of Ω_{10} is determined by the value of the quasi-energy at the entrance to the resonant medium, $\Omega_{10}=\Omega_1\,(x=0)$. This value can be found by substituting the initial values of the field amplitudes, $\mathscr{E}_{j0}=\mathscr{E}_j\,(x=0)$, into (5).

Differentiating (12) with respect to θ, we find

$$\frac{\partial\Omega_{10}}{\partial\theta}=-\frac{f}{\partial F/\partial\Omega_{10}}\sin\theta=\mp\frac{(f^2-F^2)^{1/2}}{\partial F/\partial\Omega_{10}}. \tag{17}$$

Substituting (17) into the first of Eqs. (16), we find

$$\frac{dJ}{dx}=\pm\frac{N(f^2-F^2)^{1/2}}{\partial F/\partial\Omega_{10}}. \tag{18}$$

The choice of sign in (18) depends on the sign of $\sin\theta$ at $x=0$. Specifically, we take the upper (lower) sign in the case $\sin\theta>0$ ($\sin\theta<0$). The allowed region of motion is from $J=0$ to $J=J_1$, where J_1 is the smallest (or only) positive root of the polynomial $f^2(J)-F^2(J)$. We do not need to solve the second of Eqs. (16), since once we know the dependence $J(x)$ we can work from the relation $\cos\theta=F(J)/f(J)$ to determine the dependence $\theta(x)$.

To get a clearer picture of the structure of Eq. (18), we note that F is a fourth-degree polynomial:

$$F=\sum_{m=0}^{4}A_mJ^m.$$

The derivative $\partial F/\partial\Omega_{10}$ is a polynomial of third degree in J. We will not write out the coefficients A_m, since the general expressions are quite lengthy [they can easily be derived by grouping terms by power of J in (14)]. We will point out that the coefficients $A_{3,4}$ are proportional to q and are thus zero except in the case of a noncollinear ($q\neq0$) interaction. The coefficient A_1 is proportional to δk_0, i.e., to the detuning from phase matching at $x=0$. The free term of the polynomial, A_0, is proportional to the quantity $(I_{10}I_{20}I_{30}I_{40})^{1/2}\cos\theta_0$, where θ_0 is the initial phase. It vanishes if there is no seed at any of the frequencies or if $\cos\theta_0=0$.

Equation (18) describes a process which is periodic in the coordinate x. Let us assume, for example, that one of the following conditions holds: $I_{30}=0$, $I_{40}=0$, $\theta_0=\pm\pi/2$. We can then say that on a certain interval $0\leqslant x\leqslant l_r$ the four-wave mixing develops by the mechanism $\omega_1+\omega_2\rightarrow\omega_3+\omega_4$, i.e., that there is a transfer of intensity from pulses $\omega_{1,2}$ to pulses $\omega_{3,4}$. At $x=l_r$, the quantity J reaches its maximum value $J=J_1$, and the transfer of intensity reverses direction: On the interval $l_r\leqslant x\leqslant 2l_r$ the four-wave mixing occurs by the mechanism $\omega_3+\omega_4\rightarrow\omega_1+\omega_2$ until the value $J=0$ is reached at $x=2l_r$. This picture repeats at a period of $2l_r$. This period is found from (18) to be

$$l_r=\frac{1}{N}\int_0^{J_1}\left|\frac{\partial F}{\partial\Omega_{10}}\right|\frac{dJ}{(f^2-F^2)^{1/2}}. \tag{19}$$

If the root J_1 is degenerate, we conclude from (19) that $l_r=\infty$, i.e., that the transfer of energy does not reverse direction anywhere.

We complete this new formalism with an expression for the refractive index n_j, for the frequency ω_j:

$$n_j=1+\frac{cN(\partial F/\partial I_{j0})}{\omega_j(\partial F/\partial\Omega_{10})}. \tag{20}$$

The analysis below is carried out in terms of some particular cases which are the most typical experimentally. We assume that the intensity of the seed light is considerably lower than that of the pump light and that there is a seed at only one frequency (which we assume to be ω_3). In other words, we assume $I_{10,20}\gg I_{30}$ and $I_{40}=0$. We also assume that the one-photon detuning Δ_1 is large: $|\Delta_1|\gg|\Delta_2|$, $|\mathscr{E}_{j0}d_j|/\hbar$. We will refer to the quantity $J_1/I_{\min}$, where $I_{\min}=\min\{I_{10},I_{20}\}$, as the "conversion efficiency." We wish to stress that we do not need to solve Eq. (18) in order to determine this conversion efficiency; it is sufficient to determine J_1, the smallest positive root of the polynomial $f^2(J)-F^2(J)$. The conversion efficiency can be optimized comparatively easily. To do this, we should choose the parameters of the problem (the entrance angles and detuning) in such a way that the condition $F(J=I_{\min})=0$ holds. In this case, we obtain the conversion efficiency of maximum amplitude, $J_1=I_{\min}$.

4. CASE OF A WEAK INTERACTION

We will examine the most characteristic features of the evolution of the four-wave mixing in the particular case in which the detuning from resonance is so large that the interaction of an atom with a field remains weak ($|\Delta_1|$, $|\Delta_2|$, $|\Delta_3|\gg|\mathscr{E}_jd_j|/\hbar$) throughout the conversion process. In this limit we have $|\Delta_1|$, $|\Delta_2|$, $|\Delta_3| \gg |\Omega_{10}| \approx |\mathscr{E}_{10} d_1|^2/\hbar^2|\Delta_1|$, and we can use the approximations $\partial F/\partial\Omega_{10} = -\Delta_1\Delta_2\Delta_3$ and $F = A_1 J + A_2 J^2$, in (17), where

$$A_1 = \frac{\Delta_1\Delta_2\Delta_3}{2N(\gamma_1\gamma_2\gamma_3\gamma_4)^{1/2}}\,\delta k_0, \qquad \delta k_0 = q + N\left(\frac{\gamma_1}{\Delta_1} - \frac{\gamma_4}{\Delta_3}\right),$$

$$A_2 = \frac{1}{2(\gamma_1\gamma_2\gamma_3\gamma_4)^{1/2}}\left[\frac{q^2}{N^2}\Delta_1(\Delta_2+\Delta_3)\right.$$

$$\left. - \frac{q}{N}\Delta_1(\gamma_3+\gamma_4) - (\gamma_1\gamma_3+\gamma_2\gamma_4)\right]. \qquad (21)$$

We introduce some notation which we will need:

$$g = (\beta^2 - \delta k_0^2)^{1/2}, \qquad \beta = \frac{2N}{\Delta_1\Delta_2\Delta_3}(\gamma_1\gamma_2\gamma_3\gamma_4 I_{10} I_{20})^{1/2},$$

$$l_r = \frac{1}{g}\ln\frac{16 J_1 J_2}{|J_3|(J_2-J_1)}, \qquad (22)$$

where $J_3 = I_{30}\beta^2/g^2$, and $J_{1,2}$ are the roots of the quadratic equation

$$(I_{10}-J)(I_{20}-J) - (A_1 + A_2 J)^2 = 0. \qquad (23)$$

The quantity J_1 is the smallest (or only) positive root (the condition for the existence of a root $J_1 > 0$ is the same as the condition $\beta > |\delta k_0|$, which we will assume below is satisfied). Using (21)–(23), we can put Eq. (18) in the form

$$\frac{dJ}{dx} = g\left[\frac{J(J-J_1)(J-J_2)(J+J_3)}{J_1 J_2}\right]^{1/2}. \qquad (24)$$

Under the assumptions stated above, we have $|J_{1,2}| \gg |J_3|$, and the solution (24) becomes

$$J = J_3\left(\operatorname{sh}\frac{gx}{2}\right)^2\left\{1 + \exp[2g(x-l_r)]\right.$$

$$\left. + 2\left(\frac{J_1+J_2}{J_2-J_1}\right)\exp[g(x-l_r)]\right\}^{-1}. \qquad (25)$$

Expression (25) was derived by joining two solutions of Eq. (24), one holding in the limit $J \ll I_{10}, I_{20}$ and the other in the limit $J \gg I_{30}$. These two solutions are the same and join over the broad region $I_{30} \ll J \ll I_{10,20}$. Consequently, Eq. (24) has the form of a function which is not periodic in x, and the use of this function is valid only on the interval $0 \leqslant x \leqslant l_r$. Since the periodicity of the function $J(x)$ is known, however, we can make use of (25) to reproduce the value of J for arbitrary x. In particular, at $x = l_r$ we find $J(l_r) = J_1$ from (25). For $x \ll l_r$, the expression in braces (curly brackets) in (25) can be replaced by unity; expression (25) then takes the form of some other well-known expressions.[10-13]

It follows from (22) and (25) that the four-wave mixing develops effectively if the phase mismatch is sufficiently small: $|\delta k_0| < \beta$. The transfer distance l_r is at a minimum in the case $\delta k_0 = 0$. The large number of independent parameters in this problem makes it possible to optimize the four-wave mixing and to achieve a 100% conversion efficiency in many cases, including the case $\delta k_0 \neq 0$. However, the best conditions for conversion are achieved with

$$\delta k_0 = 0, \quad \Delta_2\gamma_4 = \Delta_3\gamma_3. \qquad (26)$$

In this case we have $A_1 = A_2 = 0$, and we find $J_1 = I_{10}$ and $J_2 = I_{20}$ from (23). In other words, the transfer of intensity away from pulses $\omega_{1,2}$ comes to a halt only when one of the waves, ω_1 (in the case $I_{10} < I_{20}$) or ω_2 (in the case $I_{10} > I_{20}$), becomes totally depleted. Conditions (26) are the optimum conditions, since 100% conversion efficiency is achieved at the minimum transfer distance l_r, and under phase-matching conditions.

It follows from (25) that in order to describe the functional dependence $J = J(x)$ we should introduce along with l_r the distance $l_s < l_r$, over which the conversion process essentially reaches saturation. To simplify the discussion we assume that conditions (26) hold. Under these conditions we have $J_1 = I_{10}$ and $J_2 = I_{20}$, with $I_{10} \leqslant I_{20}$. We can then write

$$l_s = g^{-1}\ln(16 I_{10}/I_{30}). \qquad (27)$$

Under the condition $I_{10} \ll I_{20}$ the saturation length is essentially equal to the transfer period ($l_s \approx l_r$). A substantial difference between l_s and l_r arises in another case, $|I_{20} - I_{10}| \ll I_{20}$, in which we have $l_r \gg l_s$. In this case, the four-wave mixing develops in the following way: On the interval $0 \leqslant x \lesssim l_s$ the quantity $J(x)$ increases rapidly, in proportion to $\exp(gx)$. Over a distance $x \gtrsim l_s$ the quantity $J(x)$ essentially reaches its maximum value I_{10}, the four-wave mixing reaches saturation, and the conversion rate on the interval $l_s < x \leqslant l_r$ decreases sharply. On the interval $l_r < x < 2l_r$, the four-wave mixing goes in the opposite direction (Fig. 2). In the case $I_{10} \geqslant I_{20}$, we should interchange I_{10} and I_{20} in (27) and the expressions which follow.

The transfer period l_r depends on the entrance values of the intensities of the interacting pulses, I_{j0}, which are in turn slowly varying functions of the time: $I_{j0} = I_{j0}(\tau)$, where $\tau = t - x/c$. This period, $l_r = l_r(\tau)$, will thus have different values for different parts of the pump pulses. For example, we assume $\delta k_0 = 0$ and that all the pulses have the same shape at the entrance to the medium:

$$I_{j0}(\tau) = I_{j0}(0)\exp(-\tau^2/\tau_0^2)$$

[the curves in Figs. 3 and 4 are plotted for this case, under condition (26)]. From (22) we then find

$$l_{\text{II}}(\tau) = l_{r0}\exp(\tau^2/\tau_0^2),$$

where l_{r0} is the minimum value of the transfer period. This minimum value is reached at the peak of the pump intensity. Because of the τ dependence of l_r, the pulses being amplified,

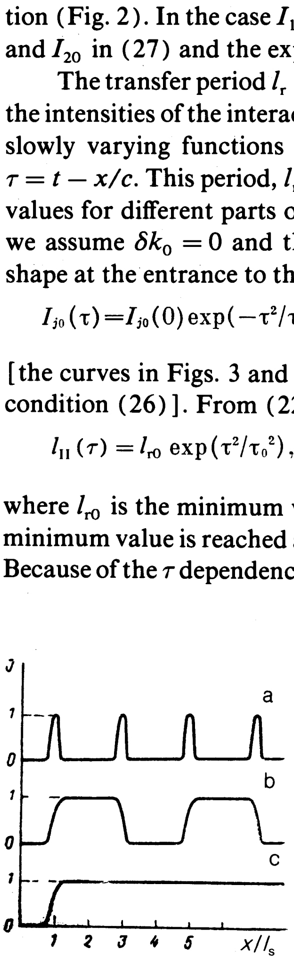

FIG. 2. $J(x)$ for various relations between the pump intensities. a—$I_{10} \ll I_{20}$ ($l_r \approx l_s$); b—$I_{10} = 0.99 I_{20}$ ($l_r = 2l_s$); c—$I_{10} = I_{20}$ ($l_r = \infty$).

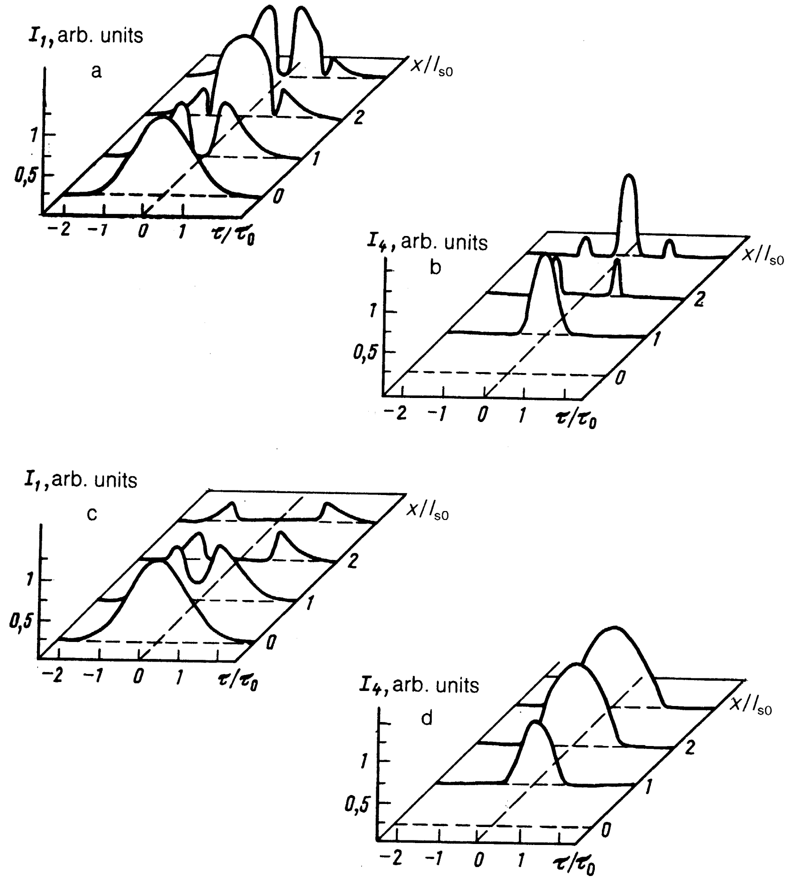

FIG. 3. Shape of pulses ω_1 and ω_4 in various cross sections of the medium. a,b—$I_{10} \ll I_{20}$; c,d—$I_{10} = I_{20}$. All curves are normalized to $I_{10}(\tau = 0)$ $[l_{s0} = l_s(\tau = 0)]$.

$\omega_{3,4}$, break up into a train of subpulses over distances $x > 2l_{r0}$ (Fig. 3). Correspondingly, the envelopes of the pump pulses are modulated. In addition, the dependence $l_r = l_r(\tau)$ rules out a complete transfer of energy from pulse ω_1 (ω_2), even at the maximum conversion efficiency in terms of the amplitude, with $J_1 = I_{10}$ ($J_1 = I_{20}$). The reason is that, if there is a transfer of intensity by the mechanism $\omega_1 + \omega_2 \rightarrow \omega_3 + \omega_4$ in some part of the pump pulse in a given cross section $x > 2l_{r0}$ of the medium, then there are regions in which the transfer goes in the opposite direction: $\omega_3 + \omega_4 \rightarrow \omega_1 + \omega_2$. This circumstance is related to the oscillations in the energies of the pulses being amplified along the coordinate x (Fig. 4). The oscillation period here is $2l_{s0}$. A pattern like that in Fig. 4 has been observed experimentally by Krasnikov *et al.*[14]

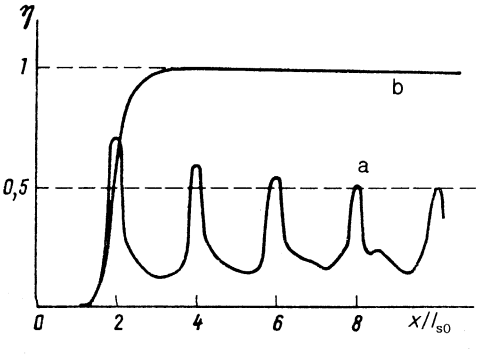

FIG. 4. Quantum conversion efficiency η, defined as the ratio of the number of photons in the pulse being amplified, ω_4, to the initial number of photons in pulse ω_1. a—$I_{10} \ll I_{20}$; b—$I_{10} = I_{20}$.

It can be seen from (22) that as the roots J_1 and J_2 move closer together the value of l_r increases logarithmically. The case $J_1 = J_2$ is of interest. As an example we consider the degenerate pumping process ($I_{10} = I_{20}$), assuming that the optimum conditions for conversion, (26), are satisfied. In the latter case we have $J_1 = J_2 = I_{10} = I_{20}$, and from (24) we find

$$J=I_{30}\left(\operatorname{sh}\frac{gx}{2}\right)^2\left[1+\frac{I_{30}}{4I_{10}}\exp(gx)\right]^{-1}. \tag{28}$$

We see that we have $J \rightarrow I_{10}$ at $x > l_s$. This example is particularly interesting since we find $l_r = \infty$, and the energy transfer does not reverse direction. As a result, it is possible to achieve an essentially complete transfer of energy from the pump wave to the pulses being amplified (Fig. 4).

5. LIMIT OF A STRONG PUMP

Let us consider a collinear interaction, $q = 0$, in which case the quasienergy Ω_1 is an integral of motion. We assume that the two-photon detuning Δ_2 is small enough ($\gamma_1\gamma_2 I_{10} I_{20} \gg \Delta_1^2\Delta_2^2$) that the pump field saturates the two-photon transition 1–2–3 at the entrance to the medium. In this case we can[6] set $\Omega_{10} = -(\gamma_1 I_{10} + \gamma_2 I_{20})/\Delta_1$ in (14). Phase matching ($\delta k_0 = 0$) is possible in this case only if $\Delta_1\Delta_3 > 0$ and $|\Delta_3| > |\Omega_{10}|$. Assuming that these conditions hold, we find from (14)

$$F=A_1J+A_2J^2, \quad \partial F/\partial\Omega_{10}=-\Delta_1\Delta_3\Omega_{10}(1+bJ), \tag{29}$$

where

$$A_1 = -\frac{\Delta_1\Delta_3\Omega_{10}\delta k_0}{2N(\gamma_1\gamma_2\gamma_3\gamma_4)^{1/2}},$$

$$\delta k_0 = \frac{N(\gamma_1+\gamma_2)}{\Delta_1}\left(1+\frac{\gamma_3\gamma_2 I_{20}+\gamma_4\gamma_1 I_{10}}{\Delta_3\Omega_{10}(\gamma_1+\gamma_2)}\right),$$

$$A_2 = -\frac{\gamma_1\gamma_3+\gamma_2\gamma_4}{2(\gamma_1\gamma_2\gamma_3\gamma_4)^{1/2}}, \qquad b = -\frac{\gamma_3+\gamma_4}{\Delta_3\Omega_{10}}. \tag{30}$$

Using (29), we can put Eq. (18) in the form

$$\frac{dJ}{dx} = \frac{g}{1+bJ}\left[\frac{J(J-J_1)(J-J_2)(J+J_3)}{J_1J_2}\right]^{1/2}, \tag{31}$$

where $J_3 = I_{30}\beta^2/g^2$, and $J_{1,2}$ are the roots of Eq. (23). We should substitute $A_{1,2}$ from (30) into Eq. (23). The quantities g and β are given by expressions (22), into which we should substitute δk_0 from (30), and in which (in the expression for β) we should make the substitution $\Delta_2 \to \Omega_{10}$. Equation (31) differs in form from (24) only in containing a factor $(1 + bJ)^{-1}$. It is thus obvious that in the case $bJ_1 < 1$ the solution of Eq. (31) will differ only slightly from (25). Again in the case $J_1J_2 > 0$, there is no qualitative difference.

Let us consider the case $bJ_1 \gg 1$, assuming $J_2 < 0$. It is sufficient to analyze the four-wave mixing on the interval $0 \leqslant x \leqslant l_r$. At short interaction distances, with $J \ll J_1$, the quantity J increases exponentially, as does the solution of (24).

For large interaction distances $[gx > |\ln(bI_{30})|]$, over which we reach values $bJ > 1$, we find from (31)

$$J = {}^1/_2\{J_1+J_2+(J_1-J_2)\cos[g_e(l_r - x)]\}, \tag{32}$$

where

$$g_e = \frac{g}{b|J_1J_2|^{1/2}}, \qquad l_r = g_e^{-1}\arccos\left(\frac{|J_2|-J_1}{|J_2|+J_1}\right). \tag{33}$$

It follows that if the four-wave mixing develops comparatively rapidly $[J \propto \exp(gx)]$ in the initial stage, the rate of development decreases sharply in a later stage (at $gx \gg 1$) and is determined by an effective gain $g_e \ll g$. In the case $\delta k_0 = 0$ the quantity g_e is independent of the entrance values of the pump intensities:

$$g_e = \frac{4N(\gamma_1\gamma_2\gamma_3\gamma_4)^{3/2}}{|\Delta_1|(\gamma_3+\gamma_4)(\gamma_1\gamma_3+\gamma_2\gamma_4)^2}. \tag{34}$$

The transfer distance is also independent of the intensities of the pump and the seed light, so it is independent of the time. As was mentioned in Sec. 5, this is a favorable circumstance if we wish to increase the conversion efficiency. The maximum efficiency is reached at $\delta k_0 \neq 0$ [this situation is characteristic of a collinear interaction ($q = 0$) and is not a consequence of a saturation of the transitions]. In the case $I_{10} < I_{20}$, for example, the maximum conversion ($J_1 = I_{10}$) is achieved at a detuning

$$\delta k_0 = -\frac{N(\gamma_1\gamma_3+\gamma_2\gamma_4)I_{10}}{\Delta_1\Delta_3\Omega_{10}} \tag{35}$$

from phase matching. In the case $I_{10} > I_{20}$ ($J_1 = I_{20}$), we should replace I_{10} in (35) by I_{20}.

6. CONCLUSION

The exact equations derived above can be used for calculations on four-wave mixing with an arbitrary extent of conversion. Pump depletion can be taken into account, as can coherent-saturation effects, which are important under resonance conditions. Because of the large number of parameters in this problem, we have applied the general results to only two particular cases. We have not discussed the case in which the interaction of the atom with the field is weak at $x = 0$ but becomes strong enough to cause saturation as the waves $\omega_{3,4}$ are amplified. Such a situation generally holds when the detuning Δ_3 is small ($|\Delta_3| \ll |\Delta_1|$, $|\mathscr{E}_{10}d_1|/\hbar$, $|\mathscr{E}_{20}d_2|/\hbar$), and it is characterized by a low conversion efficiency.

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