mula (9′), the experiment indicates that in the excitation of the $d^3\Pi_u$ states the following approximate selection rules hold fairly well: $\Delta N=0$ for the $d^3\Pi_u^-$ state and $\Delta N=\pm 1$ for the $d^3\Pi_u^+$ state. A critical discussion of this experiment and further development and refinement of the ideas in the present study are contained in Refs. 14.


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# Exact theory of resonant third-harmonic generation in gases


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We consider the self-consistent problem of passage of resonant pump and third-harmonic pulses through a gas consisting of four-level atoms (molecules). An exact solution of the problem is obtained, with account taken of all the terms of the expansion of the nonlinear polarization in powers of the field intensities. It is shown that the wave propagation equation coincides in form with the canonical Hamilton equations, so that the mathematical formalism of classical mechanics can be used. The dependence of the conversion efficiency and of the intensity-transfer period on the pump emission parameters is investigated. The case of small detunings from resonance, in which deviations from the ordinary theory are observed, is considered.



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## 1. INTRODUCTION

In the theoretical analysis of third-harmonic (TH) generation the nonlinear polarization of the medium is usually expanded in powers of the field intensities, and only the terms of lowest order, which contribute to this process, are retained (see e.g., Refs. 1 and 2). This approach, however, is no longer correct in the presence of resonances, when all the terms of the expansion must be retained.[3] The ensuing mathematical difficulties are in part unsurmountable and make it necessary nonetheless either to include a small number of expansion terms[3] or to assume that the resonance condition is satisfied for only one of the transition; otherwise, other approximations must be used. In this paper we develop a procedure for solving this problem exactly, by taking the effects of coherent saturation into account.

## 2. POLARIZATION OF A FOUR-LEVEL SYSTEM

Consider the behavior of a four-level system with nondegenerate levels in the field of two linearly polarized pulses with carrier frequencies $\omega$ and $3\omega$. The pulse durations are assumed to be small compared with all the relaxation times of the system. The pulses will be assumed to be adiabatic, to propagate in the same directions, and to be described by the classical intensity vectors

$$\mathbf{E}_1=E_1(x)\mathbf{e}_1\exp\{i\omega(x/c-t)\}+\text{c.c.},$$

$$\mathbf{E}_3=E_3(x)\mathbf{e}_3\exp\{3i\omega(x/c-t)\}+\text{c.c.}$$

The frequency $\omega$ is assumed close to the system transition frequencies. In the resonance approximation[4] we seek the solution of the Schrödinger equation in the form of the following superposition of unperturbed wave functions $\psi_n$:

$$\Phi=e^{-i\lambda t}\sum_{n=1}^{4}a_n(x)\psi_n\exp\{i(n-1)\omega(x/c-t)\}. \tag{1}$$

The energy is reckoned from the ground state. Substituting (1) in the Schrödinger equation we obtain a system of equations for the amplitudes $a_n(x)$, which we shall write in matrix form:

$$\begin{pmatrix} 0 & -E_1^* d_{12} & 0 & -E_3^* d_{14} \\ -E_1 d_{21} & \hbar\varepsilon_2 & -E_1^* d_{23} & 0 \\ 0 & -E_1 d_{32} & \hbar\varepsilon_3 & -E_1^* d_{34} \\ -E_3 d_{41} & 0 & -E_1 d_{43} & \hbar\varepsilon_4 \end{pmatrix} \begin{pmatrix} a_1 \\ a_2 \\ a_3 \\ a_4 \end{pmatrix} = \hbar\lambda \begin{pmatrix} a_1 \\ a_2 \\ a_3 \\ a_4 \end{pmatrix}, \qquad (2)$$

where $\varepsilon_n = \omega_{ni} - (n-1)\omega$ are the detunings from resonance, and $d_{mn}$ are the dipole-moment matrix elements and can be regarded as real. The quasienergy $\lambda$ is determined from the condition that the solutions of the system (2) not be trivial:

$$\begin{aligned} &\hbar^4\lambda(\lambda-\varepsilon_2)(\lambda-\varepsilon_3)(\lambda-\varepsilon_4) - |E_1|^2\hbar^2[d_{12}^2(\lambda-\varepsilon_3)(\lambda-\varepsilon_4) \\ &+ d_{23}^2\lambda(\lambda-\varepsilon_4) + d_{34}^2\lambda(\lambda-\varepsilon_2)] - |E_3|^2\hbar^2 d_{14}^2(\lambda-\varepsilon_2)(\lambda-\varepsilon_3) \\ &+ |E_1|^4 d_{12}^2 d_{34}^2 + |E_1|^2|E_3|^2 d_{14}^2 d_{23}^2 \\ &- d_{12}d_{23}d_{34}d_{14}(E_1^3E_3^* + E_1^{*3}E_3) = 0. \end{aligned} \qquad (3)$$

If the interaction is turned on adiabatically, the unperturbed states of the system go over into the corresponding quasienergy states, so that the roots of Eq. (3) must be numbered in accord with the condition $\lambda_n \to \varepsilon_n$ as $E_1 \to 0$ and $E_3 \to 0$.

The time-dependent polarization of the system, which corresponds to the transitions $\Phi_n \to \Phi_m$, leads to the appearance of a coherent ($m=n$) TH emission and to an incoherent ($m \neq n$) emission of frequencies close to this harmonic. In this paper we shall take into account only coherent TH emission whose direction coincides with the pump-pulse direction. We assume that initially the system was in the ground state. Then, after turning on the interaction, the state of the system is described by a quasienergy wave function $\Phi_1$. The polarization of the system in the state $\Phi_1$ is of the form

$$\begin{aligned} P_{11} = &(a_1^{(1)*}a_2^{(1)}d_{12} + a_2^{(1)*}a_3^{(1)}d_{23} + a_3^{(1)*}a_4^{(1)}d_{34}) \\ &\times \exp\{i\omega(x/c-t)\} + a_1^{(1)*}a_4^{(1)}\exp\{3i\omega(x/c-t)\} + \text{c.c.}, \end{aligned} \qquad (4)$$

where the amplitudes $a_n^{(1)}$ are solutions of the system (2) with $\lambda = \lambda_1$. Obviously, the direct substitution of the quantities $a_n^{(1)}$ would turn expression (4) into an utterly unmanagable form. The polarization (4) can however be reduced to the form given in Ref. 5, which does not call for finding the explicit form of the amplitudes $a_n^{(1)}$ and which is convenient for further investigation.

We use the fact that the matrix equation (2) is of the same form as the stationary Schrödinger equation. We regard the quantities $E_1$, $E_1^*$, $E_3$, $E_3^*$ as parameters on which the Hamiltonian depends. Then the polarization (4) can be expressed in terms of the derivatives of the Hamiltonian $H$ with respect to the parameters, and in terms of the column matrix $A_1$, with elements $a_n^{(1)}$:

$$P_{11} = -\left(A_1^+ \frac{\partial \hat{H}}{\partial E_1^*} A_1\right) \exp\left\{i\omega\left(\frac{x}{c}-t\right)\right\} - \left(A_1^+ \frac{\partial \hat{H}}{\partial E_3^*} A_1\right) \times \exp\left\{3i\omega\left(\frac{x}{c}-t\right)\right\} + \text{c.c.}$$

Using the known relation for the diagonal matrix element of the derivative of the Hamiltonian with respect to a parameter,[6] we obtain

$$P_{11} = -\hbar\frac{\partial\lambda_1}{\partial E_1^*}\exp\left\{i\omega\left(\frac{x}{c}-t\right)\right\} - \hbar\frac{\partial\lambda_1}{\partial E_3^*}\exp\left\{3i\omega\left(\frac{x}{c}-t\right)\right\} + \text{c.c.} \qquad (5)$$

## 3. WAVE-PROPAGATION EQUATIONS

We consider the self-consistent problem of the passage of pump and TH waves through a gas consisting of identical four-level atoms (molecules) that are uniformly distributed with density $N$. Assume that prior to turning-on the interaction ($t \to \infty$) all the atoms were in the ground state. Then the polarization of each of the atoms in the field of the pump wave and of the TH wave is described by expression (5). Substituting (5) in the one-dimensional wave equation and neglecting the second derivatives, we obtain the following system of equations for the slow field-intensity amplitudes:

$$\frac{dE_1}{dx} = -i\frac{2\pi\hbar\omega N}{c}\frac{\partial\lambda_1}{\partial E_1^*}, \quad \frac{dE_3}{dx} = -i\frac{2\pi\hbar\omega N}{c}\frac{\partial\lambda_1}{\partial E_3^*}. \qquad (6)$$

The usual approach to the considered problem would correspond to expansion of the quasienergy in powers of the quantities $E_1$ and $E_3$. Substituting the first term of the expansion of $\lambda_1$, obtained from (3), into Eqs. (6) we get the known formulas for the resonant refractive indices at the frequencies $\omega$ and $3\omega$:

$$n_1 = 1 + \frac{2\pi N d_{12}^2}{\hbar\varepsilon_2}, \quad n_3 = 1 + \frac{2\pi N d_{14}^2}{\hbar\varepsilon_4}. \qquad (7)$$

Retention of the next-order terms of the expansion would lead to the known equations[1] that do not take into account the effects of coherent saturation.

Since we are able now to represent the polarization in the form (5), we can solve the propagation equations (7) exactly. We transform (7) into their complex conjugates: substituting

$$E_1(x) = (I_1(x))^{1/2}e^{i\varphi_1(x)}, \quad E_3(x) = (I_3(x))^{1/2}e^{i\varphi_3(x)},$$

we have

$$\begin{aligned} \frac{dI_1}{dx} &= \frac{2\pi\hbar\omega N}{c}\frac{\partial\lambda_1}{\partial\varphi_1}, & \frac{d\varphi_1}{dx} &= -\frac{2\pi\hbar\omega N}{c}\frac{\partial\lambda_1}{\partial I_1}, \\ \frac{dI_3}{dx} &= \frac{6\pi\hbar\omega N}{c}\frac{\partial\lambda_1}{\partial\varphi_3}, & \frac{\partial\varphi_3}{dx} &= -\frac{6\pi\hbar\omega N}{c}\frac{\partial\lambda_1}{\partial I_3}. \end{aligned} \qquad (8)$$

It can be readily noted that Eqs. (8) are of the same form as the canonical Hamilton equations for classical motion with two degrees of freedom, if the quantities $I_1$, $I_3$ and $\varphi_1$, $\varphi_3/3$ are regarded as the corresponding generalized coordinates and momenta. The role of the time is satisfied by the coordinate $x$, and the Hamiltonian coincides, apart from a constant factor, with the quasienergy: $H = 2\pi\hbar\omega\lambda_1/c$. We shall investigate Eqs. (8) by using the methods and terminology of classical mechanics.

It is seen from (3) that the quasienergy does not depend on $x$ explicitly. It is therefore an integral of the motion, i.e., $d\lambda_1/dx = 0$. The second integral of motion can be obtained by recognizing that the generalized momenta enter in Eq. (3) only in the form of the linear combination $3(\varphi_1 - \varphi_3/3)$. We carry out a canonical transformation, going over from the two pairs of the canonically conjugate variables $I_1, \varphi_1$ and $I_3, \varphi_3/3$ to new pairs $I, \varphi$ and $I_3, \theta$, where $I = I_1 + I_3$ and $\theta = \varphi_3/3 = \varphi_1$. In terms of the new variable Eq. (3) becomes

$$\begin{aligned} &\hbar^4\lambda(\lambda-\varepsilon_2)(\lambda-\varepsilon_3)(\lambda-\varepsilon_4) - (I-I_3)\hbar^2[d_{12}^2(\lambda-\varepsilon_4)(\lambda-\varepsilon_3) \\ &+ d_{34}^2\lambda(\lambda-\varepsilon_2) + d_{23}^2\lambda(\lambda-\varepsilon_4)] - I_3\hbar^2 d_{14}^2(\lambda-\varepsilon_2)(\lambda-\varepsilon_3) \\ &+ (I-I_3)^2 d_{12}^2 d_{34}^2 + I_3(I-I_3)d_{14}^2 d_{23}^2 \\ &- 2d_{12}d_{23}d_{34}d_{14}(I-I_3)^{3/2}I_3^{1/2}\cos 3\theta = 0. \end{aligned} \qquad (9)$$

Since $\varphi_1$ does not enter in (9) explicitly, we have

$$dI/dx = \partial H/\partial\varphi_1 = 0.$$

Consequently $I(x)$ is a constant, meaning conservation of the summary intensity of waves in a lossless dielectric. The values of the integrals of the motion are determined from the boundary conditions. Assume that the atoms occupy the half-space $x > 0$. The TH intensity at the entrance will be assumed equal to zero, i.e., $I_3(0) = 0$, and we introduce the symbol $I(0) = I_0$. Then one integral of the motion is $I(x) = I_0$, and the other (the quasienergy $\lambda_1$) is determined from Eq. (9) with $x = 0$:

$$\hbar^4\lambda(\lambda-\varepsilon_2)(\lambda-\varepsilon_3)(\lambda-\varepsilon_4)-I_0\hbar^2[d_{12}{}^2(\lambda-\varepsilon_3)(\lambda-\varepsilon_4) +d_{34}{}^2\lambda(\lambda-\varepsilon_2)+d_{23}{}^2\lambda(\lambda-\varepsilon_4)]+I_0{}^2d_{12}{}^2d_{34}{}^2=0. \tag{10}$$

Recall that $\lambda_1$ is that root of Eq. (10) which vanishes together with $I_0$. Thus, since $I_0$ is an integral of the motion and $I_3$ is bounded, the problem reduces to an investigation of a one-dimensional finite motion described by the Hamilton equations

$$dI_3/dx=\partial H/\partial\theta, \quad d\theta/dx=-\partial H/\partial I_3. \tag{11}$$

We consider now the first equation of motion (11). We obtain the quantity $\partial H/\partial\theta$ from Eq. (9). Recognizing that $\lambda_1$ satisfies Eq. (10) we obtain at $s(x) = I_3(x)/I_0$

$$\frac{ds}{dx}=\pm GI_0\frac{(s\{(1-s)^3-(\alpha s+\beta)^2s\})^{1/2}}{\delta+\gamma s}, \tag{12}$$

where

$$G=\frac{12\pi N\omega d_{12}d_{23}d_{34}d_{14}}{c\hbar^3\varepsilon_2\varepsilon_3\varepsilon_4}, \tag{13}$$

$$\alpha=\frac{1}{2}\left(\frac{d_{12}d_{34}}{d_{23}d_{14}}-\frac{d_{23}d_{14}}{d_{12}d_{34}}\right), \tag{14}$$

$$\beta=\frac{1}{2d_{12}d_{23}d_{34}d_{14}}\Big\{\frac{\hbar^2\varepsilon_3}{I_0}(\varepsilon_4d_{12}{}^2-\varepsilon_2d_{14}{}^2)+\frac{\hbar^2\lambda_1}{I_0} \times[(d_{12}{}^2-d_{14}{}^2)(\lambda_1-\varepsilon_3)+d_{23}{}^2(\lambda_1-\varepsilon_4)+d_{34}{}^2(\lambda_1-\varepsilon_2) +\varepsilon_4d_{12}{}^2-\varepsilon_2d_{14}{}^2]-2d_{12}{}^2d_{34}{}^2+d_{14}{}^2d_{23}{}^2\Big\}, \tag{15}$$

$$\delta=\frac{1}{\varepsilon_2\varepsilon_3\varepsilon_4}\Big\{[4\lambda_1{}^3-(\varepsilon_2+\varepsilon_3+\varepsilon_4)\lambda_1{}^2+2(\varepsilon_2\varepsilon_3+\varepsilon_3\varepsilon_4+\varepsilon_2\varepsilon_4)\lambda_1 -\varepsilon_2\varepsilon_3\varepsilon_4]-\frac{I_0}{\hbar^2}[2\lambda_1(d_{12}{}^2+d_{23}{}^2+d_{34}{}^2)-d_{12}{}^2(\varepsilon_3+\varepsilon_4)-d_{23}{}^2\varepsilon_4-d_{34}{}^2\varepsilon_2]\Big\}, \tag{16}$$

$$\gamma=\frac{I_0}{\hbar^2\varepsilon_2\varepsilon_3\varepsilon_4}[2\lambda_1(d_{12}{}^2+d_{23}{}^2+d_{34}{}^2-d_{14}{}^2)-d_{23}{}^2\varepsilon_4-d_{34}{}^2\varepsilon_2 +(\varepsilon_2+\varepsilon_3)d_{14}{}^2]. \tag{17}$$

The choice of the sign in (12) depends on the sign of $\sin 3\theta$ at $x = 0$. The allowed region of motion lies between $s = 0$ and $s = s_1$—the smallest (or only) root of the cubic equation

$$(1-s)^3-(\alpha s+\beta)^2s=0. \tag{18}$$

The quantity $s_1$, which characterizes the ratio of the intensity of TH at the maximum to the intensity of the pump, will be called the conversion efficiency (CE).

It should be noted that nonzero boundary conditions for the TH intensity lead only to appearance in the radicand of (12) of a polynomial of fourth degree with a nonzero free term. The procedure of obtaining and investigating the TH propagation equation, on the other hand, remains unchanged.

## 4. DEPENDENCE OF THE CE ON THE PUMP EMISSION PARAMETERS

The investigation of the dependence of the CE on the parameters of the four-level system itself as well as on the parameters of the pump emission reduces to an analysis of the roots of Eq. (18) at the corresponding values of $\alpha$ and $\beta$. We consider two limiting case.

1. At low pump intensities $I_0 \to 0$ we have from (19) $\lambda_1 \approx -d_{12}^2 I_0/\hbar^2\varepsilon_2$. Substituting $\lambda_1$ in (15) and using formulas (7) and (13), we get

$$\beta\approx\Delta k/GI_0+\mu, \tag{19}$$

where

$$\Delta k=3k_1-k_3=3\omega(n_1-n_3)/c,$$

$$\mu=\frac{d_{12}}{2\varepsilon_2d_{23}d_{34}d_{14}}[(d_{12}{}^2-d_{14}{}^2)\varepsilon_3+d_{23}{}^2\varepsilon_4+d_{34}{}^2\varepsilon_2+\varepsilon_2d_{14}{}^2-\varepsilon_4d_{12}{}^2].$$

As $\beta \to \infty$ we readily find from (18) that $s_1 \approx \beta^{-2}$. Then, recognizing that $s_1 = I_{3m}/I_0$, where $I_{3m}$ is the maximum value of $I_3(x)$, we get

$$I_{3m}\approx G^2I_0{}^3/(\Delta k+\mu GI_0)^2. \tag{20}$$

At $\mu GI_0 \ll \Delta k$ we obtain the known dependence $I_{3m} \sim I_0^3$. However, even in the case of low pump intensities we have $I_{3m} \sim I_0$ if $\Delta k = 0$. The condition $\Delta k = 0$ is an equation linear in the frequency $\omega$; consequently, the indicated situation takes place at only one value of $\omega$.

2. We consider now another limiting case when all the detunings from resonance are small (but remain nonetheless larger than the widths of the atomic levels and the spectral widths of the pulses), so that the conditions $\hbar^2\varepsilon_n^2 \ll I_0D^2$ are satisfied, where $D^2 = d_{12}^2 + d_{23}^2 + d_{34}^2$. In the zeroth approximation in Eq. (10) all the detunings must be set equal to zero. It is then necessary to take into account the fact that although small detunings do not enter in (10) in the presently considered approximation, their signs are important when it comes to numbering the roots of this equation. We make use of the fact that the quasienergies of the considered system can not intersect an any value of $I_0$.[7] Consequently if the relation $\varepsilon_i < \varepsilon_j < \varepsilon_k < \varepsilon_m$ holds for the resonance detunings that are the values of the quasienergies at $I_0 = 0$, then at $I_0 \neq 0$ a similar relation $\lambda_i < \lambda_j < \lambda_k < \lambda_m$ should hold. Thus, the quantity $\lambda_1$ takes the form

$$\lambda_1=-\frac{\operatorname{sign}\varepsilon_3}{\hbar}\left\{\frac{I_0}{2}[D^2+\operatorname{sign}(\varepsilon_2\varepsilon_3)(D^4-4d_{12}{}^2d_{34}{}^2)^{1/2}]\right\}^{1/2} \quad \text{if} \quad \varepsilon_3\varepsilon_4>0; \tag{21}$$

$$\lambda_1=-\frac{\operatorname{sign}\varepsilon_2}{\hbar}\left\{\frac{I_0}{2}[D^2-(D^4-4d_{12}{}^2d_{34}{}^2)^{1/2}]\right\}^{1/2} \quad \text{if} \quad \varepsilon_3\varepsilon_4<0. \tag{22}$$

Putting $\varepsilon_2 = \varepsilon_3 = \varepsilon_4 = 0$ in (15) and using (21) and (22), we get

$$\beta=\frac{1}{2d_{12}d_{23}d_{34}d_{14}}\{(D^2-d_{14}{}^2)[D^2+\operatorname{sign}(\varepsilon_2\varepsilon_3)(D^4-4d_{12}{}^2d_{34}{}^2)^{1/2}] -4d_{12}{}^2d_{34}{}^2+2d_{14}{}^2d_{23}{}^2\} \quad \text{if} \quad \varepsilon_3\varepsilon_4>0, \tag{23}$$

$$\beta=\frac{1}{2d_{12}d_{23}d_{34}d_{14}}\{(D^2-d_{14}{}^2)[D^2-(D^4-4d_{12}{}^2d_{34}{}^2)^{1/2}] -4d_{12}{}^2d_{34}{}^2+2d_{14}{}^2d_{23}{}^2\} \quad \text{if} \quad \varepsilon_3\varepsilon_4<0. \tag{24}$$

Thus, in the considered limiting case the quantity $\beta$, and with it the root $s_1$ of Eq. (18) are constant, and consequently $I_{3m} \sim I_0$. However, the proportionality coefficient depends essentially on the signs of the detunings. This means, in other words, that asymmetry in the CE must be observed relative to the frequencies of the atomic transitions. A similar picture takes place also in the case of systems for which not all detunings become small simultaneously. Thus, for example, for a system in which the detunings $\varepsilon_2$ and $\varepsilon_4$ are large when the pump frequencies approach the two-photon resonance, the EP will be different on opposite sides of the frequency $\omega_{31}$.

## 5. DEPENDENCE OF THE TH INTENSITY ON THE COORDINATE

The sought dependence can be obtained by integrating Eq. (12). For simplicity we consider here a particular case when $|\beta| \gg 1$ and $|\beta| \gtrsim |\alpha|, |\gamma|, |\delta|$. As shown above, in this case $s(x)$ changes from $s=0$ to $s_1 \sim \beta^{-2}$, so that Eq. (12) can be written in the form

$$\frac{ds}{dx} = \pm \frac{GI_0}{\delta} (s(1-\beta^2 s))^{1/2}. \tag{25}$$

This equation is easily integrated, and when account is taken of the boundary condition $s(0)=0$ we get

$$s = \frac{1}{\beta^2} \sin^2 \frac{G\beta I_0}{2\delta} x. \tag{26}$$

We consider two limiting cases at which $|\beta| \gg 1$, so that formula (26) is valid.

1. At sufficiently low pump intensities or at a large mismatch of the phase velocities, we need retain in (19) only the first term. It follows from (16) that in the approximation in question $\delta = -1$. Then, substituting (19) in (26), we arrive at the well known relation

$$I_3(x) = G^2 I_0^3 \left( \frac{\sin(\Delta k x/2)}{\Delta k} \right)^2. \tag{27}$$

2. We consider the case of small detunings from resonance for systems in which $d_{12} \sim d_{23} \sim d_{34} \gg d_{14}$. We can then neglect in (23) and (24) the terms that contain $d_{14}^2$, and introduce for the parameter $\beta$ a single expression

$$\beta = \frac{(D^4 - 4d_{12}^2 d_{34}^2)^{1/2}}{4d_{12}d_{23}d_{34}d_{14}} [(D^4 - 4d_{12}^2 d_{34}^2)^{1/2} \pm D^2]. \tag{28}$$

The upper sign in (28) must be taken in the case when all the detunings are of the same sign, and the lower sign in all other cases. Putting $\varepsilon_2 = \varepsilon_3 = \varepsilon_4 = 0$ in (13) and (16) and substituting these expressions together with (28) in (26), we get

$$s(x) = \frac{16 d_{12}^2 d_{23}^2 d_{34}^2 d_{14}^2}{(D^4 - 4d_{12}^2 d_{34}^2)[D^2 \pm (D^4 - 4d_{12}^2 d_{34}^2)^{1/2}]^2} \times \sin^2 \left\{ \frac{3\pi\omega N}{2(2I_0)^{1/2} c} [D^2 \pm (D^4 - 4d_{12}^2 d_{34}^2)^{1/2}]^{1/2} x \right\}. \tag{29}$$

It follows from this expression that the CE does not depend on the pump intensity, in accord with the result obtained above. At the same time, in the case of detunings of equal sign the CE is smaller than in all other cases. Thus, for example, for system with "condensing" levels ($\omega_{21} > \omega_{32} > \omega_{43}$) the resonance detunings will have like signs at $\omega < \omega_{43}$ and $\omega > \omega_{21}$. Consequently, for this type of systems the TH generation is most effective when the pump frequency approaches the frequency of one-photon absorption $\omega_{21}$ from below, and decreases sharply at frequencies exceeding $\omega_{21}$, as was in fact observed in experiments[8] on $SF_6$ molecules.

For the coordinate period over which intensity is transferred from the pump pulse into the TH and back, we obtain from (28)

$$X = \frac{2^{1/2} c I_0^{1/2}}{3\omega N} [D^2 \pm (D^4 - 4d_{12}^2 d_{34}^2)^{1/2}]^{-1/2}. \tag{30}$$

We see that in contrast to the case of low intensities, when the period does not depend on $I_0$, in this case the period increases with increasing $I_0$ and also depends on the ratio of the signs of the detunings.

For lengths $x$ small compared with the period, we get from (29)

$$I_3(x) = \frac{18 d_{12}^2 d_{23}^2 d_{34}^2 d_{14}^2 \pi^2 \omega^2 N^2}{c^2 (D^4 - 4d_{12}^2 d_{34}^2)[D^2 \pm (D^4 - 4d_{12}^2 d_{34}^2)^{1/2}]} x^2. \tag{31}$$

It follows from this expression that at $x \ll X$ the TH intensity does not depend on the pump intensity. It was shown earlier[9] that the probability per unit time of TH emission on an isolated atom is likewise independent of the pump intensity in the limit of small detunings. As expected, the results agree at $x \ll X$, when the accumulation effects can be neglected.

## 6. PERIOD OF INTENSITY TRANSFER

It follows from the equation of motion (12) that a periodic (in the coordinate) transfer of the pump pulse intensity to the TH and back takes place, It is easily found from (18) that $s_1 < 1$; consequently only part of the pump intensity is transferred to the TH. For the value of the period we get from (12)

$$X = \pm \frac{2}{GI_0} \int_0^{s_1} \frac{\delta + \gamma s}{[s\{(1-s)^3 - (\alpha s + \beta)^2 s\}]^{1/2}} ds. \tag{32}$$

It was shown above that the period does not depend on the pump intensity at low values of the latter. In the case of small detunings, however, putting $\varepsilon_2 = \varepsilon_3 = \varepsilon_4 = 0$ in (13), (15–(17), and (32), we get

$$X = \frac{c\hbar\lambda_1}{3\pi N\omega d_{12}d_{23}d_{34}d_{14}} \int_0^{s_1} \frac{2\hbar^2\lambda_1^2 - I_0 D^2 + I_0(D^2 - d_{14}^2)s}{[s\{(1-s)^3 - (\alpha s + \beta)^2 s\}]^{1/2}} ds. \tag{33}$$

Since $\beta = \text{const}$ in the considered case, and from (21) and (22) we have $\lambda_1 \sim I_0^{1/2}$, we obtain $X \sim I_0^{1/2}$. Thus, the form of the dependence of the period on the intensity is preserved also at arbitrary ratios of the values of the matrix elements.

We examine now the dependence of the TH intensity at a given point $x_0$ on the gas pressure. It follows from the equation of motion (12) that $I_3(x_0)$ is a periodic function of the atom density $N$. Obviously, the quantity $I_3(x_0)$ will be maximal if the condition $x_0 = (1/2 + k)X \times (k = 0, 1, 2 \ldots)$. is satisfied. From (32) we have $X \sim N^{-1}$. At low densities the pressure is proportional to the density; consequently the maximum of the TH intensity at a given point will be observed at pressures $p = p_0(2k+1)$. At low pump intensities $p_0$ does not depend on $I_0$, and in the case of small resonance detunings, as follows from (33), we have $p_0 \sim I_0^{1/2}$.

## 7. CONCLUSION

The exact equations we obtained allow us to take into account the coherent-saturation effects that are significant under resonance conditions. In view of the large number of parameters in the problem, we have confined ourselves to application of the general results to two particular cases. In the case of low pump intensities we have shown that all the results agree with the known ones. On the other hand, to illustrate the application of the general methods to cases that are not described by the usual theory, we have chosen only that class of systems for which all the resonance detunings become small simultaneously. It must also be emphasized that our procedure is applicable to the analysis of experiments at arbitrary ratios of the resonance detunings, matrix elements, and other parameters.

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# Photon echo from small-area exciting pulses

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A theoretical investigation is reported of photon echo formed in a gas medium, by two linearly polarized exciting pulses of small area. The intensity and polarization are obtained for the echo on an optically allowed transition with arbitrary angular momenta of the levels and with arbitrary ratio of the durations of the exciting pulses and the time of the reversible Doppler relaxation. Analytic expressions are obtained for the form of the echo pulse for both a narrow and a broad spectral line. The results of the theoretical investigations point to the need of performing new experiments on photon echo in gases for the purpose of identifying the resonant transitions.

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With the advent of lasers, various methods of investigating the characteristics of matter, based on nonlinear interaction between the matter and electromagnetic radiation, have found extensive applications. One such method is the photon echo, which uses the nonlinear response of a medium after the passage of two exciting pulses. The photon-echo method yields information on the relaxation characteristics of matter, and permits identification of the corresponding resonant transition. To reduce the experimental results by this method, theoretical relations are necessary between the experimentally recorded quantities, such as the echo intensity and its polarization, on the one hand, and the characteristics of the medium on the other.

The first photon-echo experiment was performed in ruby.[1] Subsequently it came into extensive use also for the investigation of gases. In a gas medium, the resonant levels on which the echo is formed are usually degenerate. This adds to the complexity of the theoretical analysis of the photon echo in gases. We note that up to now there were no calculations for the intensity and polarization of the echo at arbitrary values of the angular momenta of the resonant-transition levels.

An important role in the theoretical investigation of the photon echo phenomenon is played by the relation between the durations $T_1$ and $T_2$ of the exciting light pulses and the time of the reversible Doppler relaxation $T_0$. As a rule[2-4] the calculations are performed within the limits of a narrow spectral line ($1/T_0 \ll 1/T_i$; $i=1, 2$). At the same time, most experiments on photon echo in gases have been performed either for small values of the angular momenta of the levels,[13] or with the degeneracy neglected completely.[14] It follows from the results of Refs. 13 and 14 that on a broad spectral line the echo pulse has a complicated shape, and its polarization on the transitions $j \to j (j>1)$ and $j \rightleftarrows j\pm 1 (j>1/2)$ depends on the intensities of the exciting pulses and on the ratio of $T_0$ and $T_i$. On a narrow spectral line, as follows from Refs. 2–4, the echo polarization on these same transitions also depends on the intensities of the exciting pulses.

It is established in the present paper that in the limit of small areas of the exciting pulses the calculation of

  