Resonance stimulated Raman scattering of ultrashort light pulses

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Stimulated Raman scattering of coherent resonance radiation pulses of such short duration that their spectral width considerably exceeds the linewidths of the scattering transitions is investigated theoretically. A set of equations based on the time-dependent Schrödinger equation and Maxwell equations is constructed which describes in a self-consistent manner the time and the spatial variation of a pulse of the first Stokes component in a scattering medium consisting of identical three-level objects (atoms or molecules) in the approximation of a given pumping field. The equations are solved for the case of excitation of scattering of the type indicated by a resonance 2π pulse. The class of solutions obtained describes a Stokes pulse characterized by the absence of phase modulation and predicts the possibility of forming stationary Stokes modes (SSM), i.e., Stokes pulses of constant duration that grow exponentially on penetration into the scattering medium. The gain for the SSM dependent on the intensity of the electric field (G) is in this case directly porportional to the duration (τ_1) of the exciting 2π pulse and, consequently, is inversely proportional to $\sqrt{I_{1\max}}$ where $I_{1\max}$ is its maximum intensity. Numerical estimates carried out for potassium vapor in the case $I_{1\max} \approx 50$ MW/cm^2 (which corresponds to $\tau_1 \approx 10^{-12}$ sec) yield $G \approx 1.4$ cm^{-1}.

1. It is well known that the probability of stimulated Raman scattering (SRS) increases significantly as the frequency of the exciting radiation approaches the frequency of one of the allowed transitions of the scattering system and becomes particularly large in the case of resonance SRS[1,2]. However, in the resonance case a particularly prominent role along with SRS is assumed by the stimulated one-photon processes and processes of nonradiative relaxation. This leads to the situation that in the stationary regime amplification at the Stokes frequency in the case of resonance SRS (RSRS) is possible only if specific and apparently fairly rarely realized relations are satisfied between the relaxation times of the scattering system[1]. Therefore in the present paper we investigate the characteristic features of the Stokes RSRS under conditions when the duration of the pumping radiation and of the Stokes radiation in the medium (respectively τ_1 and τ_2) satisfy the relation

$$\tau_1, \tau_2 \ll T_i \quad (i=1, 2, 3\ldots), \tag{1}$$

where T_i are the times of transverse relaxation of the scattering system.

The validity of (1) corresponds to an essentially nonstationary regime realized when the RSRS is excited by ultrashort laser pulses. An interesting result of this work consists of the fact that when (1) is satisfied a formal analogy exists between expressions describing amplification of Stokes signals in the case of RSRS and in the case of nonresonant SRS[3] which leads to an identity of several qualitative conclusions. It is natural, however, that parameters defining the quantitative picture of development of Stokes pulses are different for the two cases.

2. We consider a system consisting of weakly interacting objects—atoms or molecules playing the role of scattering centers. Taking into account the specific features of RSRS[1] we restrict ourselves in describing each object to three energy levels (cf., diagram), assuming for the sake of simplicity that all the levels are nondegenerate and the states $|1\rangle$ and $|2\rangle$ have the same parity opposite to the parity of the state $|3\rangle$.

We further assume that both the laser and the Stokes waves in the scattering medium are polarized along the z-axis and are propagated along the x-axis so that we have

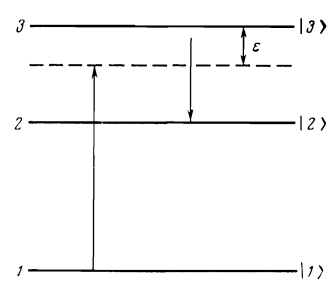
$$E_z(x, t) = E_1(x, t)\cos[k_1 x - \omega_1 t + \varphi_1(x, t)] + E_2(x, t)\cos[k_2 x - \omega_2 t + \varphi_2(x, t)], \tag{2}$$

where the subscripts 1 and 2 refer respectively to the characteristics of the laser and the Stokes waves; $E_i(x, t)$, $\varphi_i(x, t)$ (i = 1, 2) are the slow amplitudes and the additional dispersion terms satisfying the relations

$$\frac{\partial E_i}{\partial x} \ll k_i E_i, \quad \frac{\partial E_i}{\partial t} \ll \omega_i E_i, \qquad \frac{\partial \varphi_i}{\partial x} \ll k_i, \quad \frac{\partial \varphi_i}{\partial t} \ll \omega_i; \tag{3}$$

$k_i = \omega_i \eta_i / c$; η_i is the index of refraction at the frequency ω_i determined by all the nonresonant transitions. In this case we have $\omega_2 = \omega_{32}$, while the carrier frequency of the pumping radiation ω_1 is close to ω_{31}, so that $\omega_{31} - \omega_1 = \epsilon \ll \omega_{31}$.

The significance of the dispersion term φ_2 can be illustrated by the following simple example. If we set $\varphi_2 = \Delta\omega(t - \eta_2 x/c)$, then this corresponds to the description of the usual Stark shift for a stationary SRS. But in the nonstationary case the form of φ_2 is not known be-



forehand. Restricting ourselves to an investigation of isotropic media we assume that the quantity $p_{13} \equiv \langle 1|\hat{p}_z|3\rangle$ and $p_{23} \equiv \langle 2|\hat{p}_z|3\rangle$ ($\mathbf{p}$ is the dipole moment operator) do not change in going over from one scattering center to another. For an isotropic medium consisting of anisotropic molecules one can in this case in the usual manner assume that

$$|p_{13}|^2 = {}^1/_3|\langle 1|\mathbf{p}|3\rangle|^2, \quad |p_{23}|^2 = {}^1/_3|\langle 2|\mathbf{p}|3\rangle|^2.$$

By these simplifications we replace in calculating the polarization response of the medium the procedure, which is complicated in our case, of averaging over the orientations. If (1) is satisfied then in order to describe the change in the state of the three-level scattering center under the action of the field (2) one can utilize the Schrödinger equation an arbitrary solution of which can be represented in the form

$$\Psi = \sum_{K=1} a_K'(x,t)\,\Psi_K(q)\exp\left(-\frac{i}{\hbar}E_K t\right),$$

(K = 1, 2, 3). The x coordinate indicates the position of the center of the scattering object.

Writing down the usual equations[4] for $a_K'(x, t)$, neglecting in them the rapidly oscillating nonresonant terms and carrying out the replacement

$$a_1' = a_1 \exp\left\{-\frac{i}{2}(k_1 x + \varepsilon t + \varphi_1)\right\},$$

$$a_2' = a_2 \exp\left\{\frac{i}{2}(k_1 x + \varepsilon t + \varphi_1) - i(k_2 x + \varphi_2)\right\},$$

$$a_3' = a_3 \exp\left\{\frac{i}{2}(k_1 x + \varepsilon t + \varphi_1)\right\},$$

we arrive at the following system of equations

$$\frac{\partial a_1}{\partial t} - \frac{i}{2}\left(\varepsilon + \frac{\partial \varphi_1}{\partial t}\right)a_1 = \frac{i}{2\hbar}L_1^{1/2}(p_{13}E_1)a_3, \tag{4}$$

$$\frac{\partial a_2}{\partial t} + i\left[\frac{1}{2}\left(\varepsilon + \frac{\partial \varphi_1}{\partial t}\right) - \frac{\partial \varphi_2}{\partial t}\right]a_2 = \frac{i}{2\hbar}L_2^{1/2}(p_{23}E_2)a_3, \tag{5}$$

$$\frac{\partial a_3}{\partial t} + \frac{i}{2}\left(\varepsilon + \frac{\partial \varphi_1}{\partial t}\right)a_3 = \frac{i}{2\hbar}L_1^{1/2}(p_{31}E_1)a_1 + \frac{i}{2\hbar}L_2^{1/2}(p_{32}E_2)a_2, \tag{6}$$

where $L_i = (\eta_i^2 + 2)^2/9$ are the Lorentz correction factors for the local field.

Without taking the inhomogeneous broadening into account the effective polarization at the Stokes frequency can now be represented in the form

$$P_z = P_S \sin(k_2 x - \omega_2 t + \varphi_2) + P_c \cos(k_2 x - \omega_2 t + \varphi_2), \tag{7}$$

$$P_S = 2L_2^{1/2} N \operatorname{Im}(a_2 a_3^* p_{32}), \quad P_c = 2L_2^{1/2} N \operatorname{Re}(a_2 a_3^* p_{32}), \tag{8}$$

where N is the number of particles per unit volume.

3. The required abbreviated equations for the resonance fields of the form $E(x, t)\cos(\omega t - kx + \varphi)$ differ from the usual abbreviated equations of nonlinear optics of transparent media[5] and when (3) is satisfied for the Stokes wave have the form[6,7]

$$\frac{\partial E_2}{\partial t} + \frac{c}{\eta_2}\frac{\partial E_2}{\partial x} = \frac{2\pi\omega_2}{\eta_2^2}P_S, \tag{9}$$

$$\left(\frac{\partial \varphi_2}{\partial t} + \frac{c}{\eta_2}\frac{\partial \varphi_2}{\partial x}\right)E_2 = \frac{2\pi\omega_2}{\eta_2^2}P_c. \tag{10}$$

Analogous equations are valid for E_1 and φ_1, but the system of coupled equations obtained in this case is too complicated. Therefore we subsequently consider only the case of weak scattering ($E_2 \ll E_1$) under the conditions of which the pumping field can be regarded as given. But even excluding the effect of RSRS on the pumping pulse we must take into account the fact that in the resonance case ($\omega_1 \approx \omega_{31}$) the characteristics E_1 and φ_1 of the prescribed pumping are determined not only by the characteristics of the laser radiation at its point of entry into the sample (x = 0) but also by the properties of the resonance transition. Therefore in order to obtain the "given" pumping field it is necessary to solve a system of equations describing the production of the resonance signal by an ensemble of two-level systems (levels 1 and 3 in the diagram).

The solution indicated above, as is well known,[6,7], leads to a description of the effect of self-induced transparency. Specifically this means that for $\tau_1 \ll T_i$ and when the boundary condition

$$\pi < \frac{L_1^{1/2}|p_{13}|}{\hbar}\int_{-\infty}^{\infty} E_1(0,t)\,dt < 3\pi \tag{11}$$

is satisfied the formation of a stationary 2π-pulse occurs near the irradiated surface of the sample, which is then propagated without a change in the profile of its envelope.

Limiting ourselves to the case of weak scattering we shall utilize the field of the stationary 2π-pulse for our given pumping field. The assumption $E_2 \ll E_1$ enables us, moreover, to achieve essential simplification by neglecting on the right hand side of (6) the term $iL^{1/2}(p_{32}E_2)a_2/2\hbar$ compared to $iL_2^{1/2}(p_{31}E_1)a_1/2\hbar$. Then from the complete set of coupled equations (4)—(6), (9), (10) an independent subsystem for a_1 and a_3 is separated out. The solution of this subsystem together with the equations for E_1 and φ_1 which are not reproduced here must in principle give well known formulas describing the 2π-pulse. However, it is sufficient for us to determine a_3 in terms of the known E_1 and φ_1, after which we obtain a closed system of equations (5), (9), (10) for the determination of a_2, E_2, φ_2.

4. The solution of the problem formulated above can be carried out most simply in the case of strictly resonant exciting radiation, i.e., for $\epsilon = 0$. Taking the foregoing into account we then have from (4), (6) the following system of equations for the determination of a_1 and a_3:

$$\frac{\partial a_1}{\partial t} - \frac{i}{2}\frac{\partial \varphi_1}{\partial t}a_1 = \frac{i}{2}\tilde{E}_1 a_3 e^{-i\psi}, \tag{12}$$

$$\frac{\partial a_3}{\partial t} + \frac{i}{2}\frac{\partial \varphi_1}{\partial t}a_3 = \frac{i}{2}\tilde{E}_1 a_1 e^{i\psi}, \tag{13}$$

where

$$\tilde{E}_1 = L_1^{1/2}|p_{13}|E_1/\hbar, \quad \psi = \arg(p_{31}).$$

In the case $\epsilon = 0$ the normalized amplitude $\tilde{E}_1$ and the dispersion term φ_1 of the stationary 2π-pulse have the form[7]

$$\tilde{E}_1 = \frac{2}{\tau_1}\operatorname{sch}\left(\frac{t - x/V}{\tau_1}\right), \quad \varphi_1 = \text{const}, \tag{14}$$

where V is the velocity of the envelope of the 2π-pulse determined in the case when inhomogeneous broadening is absent by the formula

$$V^{-1} = \eta_1 c^{-1} + \alpha_1 \tau_1^2, \quad \alpha_1 = 2\pi L_1 \omega_1 |p_{13}|^2 N/\hbar\eta_1 c. \tag{15}$$

Utilizing (14) we obtain from (12) and (13) under the condition $a_3(x, t = -\infty) = 0$

$$a_3 = \operatorname{sch}\left(\frac{t - x/V}{\tau_1}\right). \tag{16}$$

Equations (5), (9) and (10) with a_3 determined by formula (16) after transition to new independent variables

$$w = t - x/V, \quad x = x$$

and the introduction of real functions Q_S and Q_C in accordance with the formulas

$$Q_s=\mathrm{Im}\left(\frac{p_{32}}{|p_{23}|}a_2\right),\quad Q_c=\mathrm{Re}\left(\frac{p_{32}}{|p_{23}|}a_2\right), \tag{17}$$

take on the form

$$\frac{\partial \tilde{E}_2}{\partial x}-\nu\frac{\partial \tilde{E}_2}{\partial w}=2\alpha_2\,\mathrm{sch}\left(\frac{w}{\tau_1}\right)Q_s, \tag{18}$$

$$\left(\frac{\partial \varphi_2}{\partial x}-\nu\frac{\partial \varphi_2}{\partial w}\right)\tilde{E}_2=2\alpha_2\,\mathrm{sch}\left(\frac{w}{\tau_1}\right)Q_c, \tag{19}$$

$$\frac{\partial Q_c}{\partial w}+\frac{\partial \varphi_2}{\partial w}Q_s=0,$$

$$\frac{\partial Q_s}{\partial w}-\frac{\partial \varphi_2}{\partial w}Q_c=\frac{1}{2}\,\mathrm{sch}\left(\frac{w}{\tau_1}\right)\tilde{E}_2, \tag{20}$$

$$\tilde{E}_2=L_2^{1/2}|p_{23}|E_2/\hbar,\quad \alpha_2=2\pi\omega_2|p_{23}|^2NL_2/\hbar\eta_2 c, \tag{21}$$

where ν is defined by the formula

$$\nu=1/V-\eta_2/c. \tag{22}$$

Below we shall investigate one class of solutions of the system (18)—(21) characterized by the condition $\partial\varphi_2/\partial w = \partial\varphi_2/\partial x = 0$ for arbitrary x and w. This class of solutions is consistent with the natural physical assumption $\partial\varphi_2/\partial x = \partial\varphi_2/\partial w = 0$ at the initial instant $w = -\infty$ for an arbitrary x, and also with the assumption of absence of phase modulation of Stokes radiation at the boundary $x = 0$. Since $Q_C = 0$ for $w = -\infty$, then from (18)—(21) it follows that $Q_C = 0$ for arbitrary w and x, while $\tilde{E}_2$ and Q_S are determined from the system of equations

$$\frac{\partial \tilde{E}_2}{\partial x}-\nu\frac{\partial \tilde{E}_2}{\partial w}=2\alpha_2\,\mathrm{sch}\left(\frac{w}{\tau_1}\right)Q_s, \tag{23}$$

$$\frac{\partial Q_s}{\partial w}+\frac{Q_s}{T_2}=\frac{1}{2}\,\mathrm{sch}\left(\frac{w}{\tau_1}\right)\tilde{E}_2+K(x,w). \tag{24}$$

Two terms absent in (21) have been phenomenologically introduced into (24). The term Q_S/T_2 describes the damping of the polarization during the time T_2. Indeed, the introduction of this term leads to the multiplication of Q_S obtained in the case $Q_S/T_2 = 0$, by $\exp(-w/T_2)$, and consequently, to an additional multiplication by the same quantity also of the polarization amplitude P_S, since in accordance with (8) and (16) $P_S \propto \mathrm{sech}(w/\tau_1)Q_S$. But the modulation of the polarization by an exponentially decaying term corresponds to the usual phenomenological method of taking damping into account. However, it should be noted that the damping introduced above does not free us from the limition (1) and must be regarded as a small correction since in obtaining a_3 from equations (13) and (14) the damping processes were ignored. The term K(w, x) takes into account the random external force determined by the fluctuations characteristic of the medium. As in[3,8] it is assumed that K(w, x) is delta-correlated in space and time:

$$\langle K(w,x)K(w',x')\rangle=g\delta(w-w')\delta(x-x'),$$

where g is the intensity of the random force.

The system (23), (24) is formally identical with the system (2) of reference[3], and under the conditions $E_2(x, w = -\infty) = \tilde{E}_2(x = 0, w) = 0$ we have for the averaged intensity of the Stokes radiation under the assumption $\nu > 0$ the following expression[3]:

$$I_2=\alpha\,\mathrm{ch}^{-2}\left(\frac{w}{\tau_1}\right)\int_0^\infty \exp\left(-\frac{2\xi}{T_2}\right)d\xi \times\int_0 \mathrm{ch}^2\left(\frac{w-\xi}{\tau_1}\right)\mathrm{ch}^{-2}\left(\frac{w-\xi+\nu S}{\tau_1}\right)F^2(1+n,1-n,1,y)\,dS, \tag{25}$$

where F is the hypergeometric function, while α, y, n are determined by the following formulas:

$$\alpha=g2\pi\omega_2^2|p_{23}|^2N^2L_2/\eta_2 c,\quad n=(\alpha_2/\nu)^{1/2}\tau_1,$$

$$y=-\mathrm{sh}\left(\frac{\nu S}{\tau_1}\right)\mathrm{sh}\left(\frac{\xi}{\tau_1}\right)\mathrm{ch}^{-1}\left(\frac{w}{\tau_1}\right)\mathrm{ch}^{-1}\left(\frac{w-\xi+\nu S}{\tau_1}\right).$$

An analysis of formula (25) shows[3] that for $n > 1/2$ (here and subsequently in accordance with (1) we neglect the quantity τ_1/T_2 compared to unity) at a distance $x \geq l_2 = \tau_1/\nu$ an exponentially growing stationary Stokes mode (SSM) is formed—i.e., a Stokes pulse of constant duration whose dependence on x is wholly contained in the factor e^{2Gx}:

$$I_2=\beta\exp[2(n-1)w/\tau_1]\mathrm{ch}^{-2n}(w/\tau_1)e^{2Gx}, \tag{26}$$

$$\beta=2^{-2n}g\hbar\omega_2N\Gamma(2n)\Gamma(\tau_1/T_2)\Gamma(2n-1)\Gamma^{-1}(n),\qquad G=(2n-1)\nu/\tau_1, \tag{27}$$

where Γ is the gamma function. The quantity G has the meaning of the amplification coefficient of SSM with respect to intensity. The position of the peak of SSM with respect to the peak of the pumping pulse ($\Delta\tau$) and the duration of the SSM (τ_2) are determined by the formulas

$$\Delta\tau=\frac{\tau_1}{2}\ln(2n-1),\qquad \tau_2=\tau_1\left(\frac{2n}{2n-1}\right)^{1/2}. \tag{28}$$

From (27) it follows that $\beta \to \infty$ as $T_2 \to \infty$, so that taking the finiteness of T_2 into account is necessary for the description of SSM. In accordance with estimates, for the overwhelming majority of real situations the following condition is satisfied

$$|\eta_1-\eta_2|/c\ll\alpha_1\tau_1^2, \tag{29}$$

so that utilizing (15) and (22) we obtain $\nu = \alpha_1\tau_1^2 > 0$, as was supposed in writing down the expression for I_2 in formula (25). When (29) is satisfied the formula for G takes on its simplest form:

$$G=(2n-1)\alpha_1\tau_1,\quad n^2=\frac{L_2\eta_1}{L_2\eta_1}\frac{\omega_2}{\omega_1}\frac{|p_{23}|}{|p_{13}|}. \tag{30}$$

The possibility of the occurrence of SSM in the case of nonresonant SRS in the field of ultrashort pumping pulses under the conditions of normal dispersion was theoretically demonstrated in the paper of Akhmanov et al.[3]. Formulas (30) enable us to bring out the following characteristic features which differentiate between the amplification of SSM in the case of RSRS and the amplification of SSM in the case of SRS. Firstly, the amplification coefficient G is proportional to τ_1 or, in accordance with (14), is inversely proportional to the maximum intensity of the exciting 2π-pulse. Secondly, the necessary condition for the existence of SSM ($G > 0$) depends only on the properties of the medium and is independent of the intensity and the duration of the pumping pulse.

5. We make several remarks concerning some conditions of applicability of the theory developed above which were not discussed previously. It is possible to ignore inhomogeneous broadening if

$$\tau_i\ll T_{i3}^*,\qquad (i=1,2), \tag{31}$$

where $T_{i3}^* = 2\pi g_{i3}(0) \approx 2\pi/\Delta\omega_{i3}$, $g_{i3}(0)$ and $\Delta\omega_{i3}$ are the maximum value and the width of the distribution function which describes the inhomogeneous broadening of the line of absorption or emission in the transition i — 3. The assumption that the pumping pulse is a 2π-pulse requires that the characteristic length l_1 over which the formation of the 2π-pulse occurs should be considerably smaller than the characteristic length for the formation

of the Stokes pulse for which we can take the quantity l_2 introduced above. According to Lamb [7] one can utilize for l_1 the resonance absorption length multiplied by π, i.e., one can set $l_1 = \pi/\alpha_1 T_{13}^*$, and when (29) is satisfied we have $l_2 = (\alpha_1\tau_1)^{-1}$. Then the condition $l_1/l_2 \ll 1$ in essence coincides with the condition (31) for the time τ_1.

6. As an object for carrying out some estimates we consider potassium vapor in which quasistationary RSRS has been observed [9]. The levels 1, 2 and 3 (cf. the diagram) can now be identified with the levels $4p^2P_{3/2}$, $5p^2P_{3/2}$, $6s^2S_{1/2}$. The frequency of the transition $4p^2P_{3/2} - 6s^2S_{1/2}$ coincides with the frequency of the ruby laser maintained at a temperature of $-40°C$ [10]. We choose the axis of quantization along the direction of polarization of the waves, i.e., along the z axis. Then, according to Lamb [7], in order to take into account the degeneracy of the levels in the jm-scheme when (1) is satisfied, we should consider two independent channels consisting respectively of states with $m = \frac{1}{2}$ and $m = -\frac{1}{2}$, the contribution of which to the total polarization is additive. But on the basis of the Wigner-Eckart theorem it can be shown that the dipole moments of the two channels are respectively equal. This enables us to ignore the degeneracy for a field of the form (2) and to consider only one channel assuming that the density of particles in its initial level is equal to the sum of the densities of the particles in the initial levels of both channels.

We set $\tau_1 = 10^{-12}$ sec, which according to Sobel'man [11] guarantees that the condition $\tau_1 \ll T_i^{-1}$, where T_i is the lifetime associated with collision broadening, is satisfied in the vapors of alkali metals under all practically utilized particle densities. Condition (31) is also satisfied at not very high temperatures: for example, for 100–300°C in the case of potassium we have $T_{i3}^* \gtrsim 10^{-10}$ sec^{-1}. Utilizing the values of the oscillator strengths and of the frequencies of transition, reported in [10], we obtain in accordance with formulas (28), (30) for $N = 10^{14}$ cm^{-3}: $G = 1.36$ cm^{-1}, $\tau_2 = 1.4\,\tau_1$ and $\Delta\tau = 0.47\,\tau_1$. The characteristic length l_2 along which the formation of SSM takes place is equal to 1.9 cm. The maximum intensity of the 2π-pulse corresponding to $\tau_1 = 10^{-12}$ sec amounts to approximately 50 MW/cm^2. If the laser pulse on entering the scattering medium is of a duration of the order of 10^{-12} sec, then its peak intensity needed for the formation of a 2π-pulse with $\tau_1 = 10^{-12}$ sec must also be close to 50 MW/cm^2.

7. We note that the equations for the self-induced transparency admit solutions in the form of not strictly resonant 2π-pulses [6,12-14] for which ω_1 differs somewhat from the average frequency ω_{31} of the generally inhomogeneously broadened resonance transition line. But since the present analytic theories do not describe transition processes which occur in the formation of a 2π-pulse, they are not in a position to relate ω_1 to the characteristics of the incident radiation. In carrying out estimates in this case one can assume that ω_1 is equal to the carrier frequency of the incident laser signal, but in essence ω_1 remains an unknown parameter of the theory. The results of a numerical calculation by Diels [15] show that in the process of formation of a 2π-pulse from a not strongly resonant input signal a pull occurs on the central frequency of its spectral decomposition ($\overline{\omega}_1$) until it coincides with $\overline{\omega}_{31}$. But, according to Matulic and Eberly [14], $\overline{\omega}_1 = \omega_1$ for arbitrary 2π-pulses including those which are not exactly resonant. On comparing these results it can be concluded that in some cases for a real 2π-pulse formed from an almost resonant input signal the condition $\omega_1 = \overline{\omega}_{31}$ must be satisfied. The existence of self-tuning of the pumping frequency to exact resonance makes the restriction $\epsilon = 0$ imposed in Sec. 4 to be not so essential when applied under the given experimental conditions.

In the theory developed above it was assumed that $\partial\varphi_1/\partial t = 0$, i.e., that phase modulation (PM) of the pumping signal is absent. For a 2π-pulse this condition is strictly satisfied, if one can neglect the effect of all the nonresonance transitions. The possible PM of the input signal is in this case completely suppressed along the length l_1, from where it follows that the effect of the PM of the input radiation on the RSRS is negligibly small for $l_1/l_2 \ll 1$, in other words, when the condition (31) is satisfied for the time τ_1.

On the other hand the presence of nonresonant transitions can lead in certain cases to PM of ultrashort resonant pulses [7,14,16] even when the inequalities (3) are satisfied. But according to the results of the work of Akhmanov et al. [3,17] PM of the pumping pulse leads also to a reduction in the efficiency of the nonstationary nonresonant SRS and, in accordance with commonly held concepts [3], must play a negative role also in the case of a nonstationary RSRS. However, the existence of a resonance transition can also in this case favor the scattering process leading to a reduction in the PM of the resonance pumping pulse compared with the PM of a similar but a nonresonant signal.

We consider as an example the PM of light pulses produced by the nonlinear dependence of the index of refraction η_1 on the field in accordance with

$$\eta_1 = \eta_0(1+\beta I_1).$$

In this case PM of the stationary 2π-pulse is determined by the formula [14]

$$\frac{\partial\varphi_1}{\partial t} = \frac{3}{4}\frac{\omega_1\eta_0}{c}\beta\frac{I_1}{\alpha_1\tau_1^2}, \tag{32}$$

when the inequality (31) for τ_1 is satisfied, while the PM of a similar nonresonant signal has the form [18]

$$\frac{\partial\varphi_{1nr}}{\partial t} = -\frac{\omega_1\eta_0}{c}\beta\frac{\partial I_1}{\partial t}x. \tag{33}$$

Characteristic values of the quantities $\partial\varphi_1/\partial t$ and $\partial\varphi_{1nr}/\partial t$ can be obtained if in (32) one sets $I_1 = I_{1max}$, and in (33) takes $\partial I_1/\partial t = I_{1max}/\tau_1$, where I_{1max} is the maximum value of the intensity which is chosen to be the same for the nonresonant signal and for the 2π-pulse. Then from (32) and (33) it follows that $|\partial\varphi_1/\partial t| \ll |\partial\varphi_{1nr}/\partial t|$ for $x \gg l_3$, where

$$l_3 = (\alpha_1\tau_1)^{-1}. \tag{34}$$

According to (15) l_3 is inversely proportional to N. For potassium, under the conditions indicated in Sec. 6, we have $l_3 = 1.7$ cm.

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