

NONLINEAR OPTICAL PHENOMENA

PACS numbers: 42.65.Dr; 42.25.Hz; 42.25.Kb
DOI: 10.1070/QE2010v040n12ABEH014398

Interference suppression of SRS

V.P. Kochanov

***Abstract*. The theory of three-wave SRS is developed, which takes into account nonlinear dispersion of a medium for arbitrary phases of the pump waves at the input to the medium. The effect of interference suppression of SRS is predicted for values of the total phase of the three-wave pump $(2n+1)\pi$ ($n = 0, \pm 1, \pm 2\ldots$), the effect being caused by the destructive interference of polarisations of the nonresonant dipole-allowed transitions. The relation between the contributions of the linear and nonlinear dispersions to the SRS is found. It is shown that at a sufficiently large wave detuning, the anti-Stokes wave amplitude experiences spatial oscillations.**

Keywords: *stimulated Raman scattering, interference, coherence, nonlinear dispersion, phase.*

1. Introduction

Stimulated Raman scattering in the general case is a coherent process. Coherence means that the interfering pump and scattering waves interact, and the wave equation for SRS cannot be reduced to equations for the wave intensities. Often, SRS is excited by bichromatic pump waves whose frequency difference is equal to the frequency of a Raman transition. In this case, efficient generation of a broad spectrum of Stokes and anti-Stokes harmonics is observed [1]. Nonlinear dispersion of the medium determines the formation mechanism of the total phase of the waves during their coherent interaction in the active medium [2]. Unlike the linear dispersion when the wavenumbers are directly proportional to real parts of linear polarisabilities, the wavenumbers and phases are found in this case from the solutions of the corresponding differential equations. A qualitative change in the dispersion character, which consists in the mutual adjustment of the phases, leads to a nonlinear trapping and phase jump [2].

It is natural to assume that the above features of coherent SRS will be significantly affected by the phase ratio of the pump waves at the medium input. Amplification of the Stokes and anti-Stokes components of the spontaneous Raman scattering induces the phase difference. The phase difference can also be produced in a controlled manner in excitation of SRS by two- and three-wave pump, i.e., it acts as an independent parameter in the experiments. Earlier, this extra physical factor was not taken into account when considering the SRS; the pump waves in theoretical calculations were assumed in-phase, and the phase relations in the experiments were not controlled.

The aim of this the work is to find out the qualitative features and the degree of influence of the phase ratio of the pump waves at the medium input on the three-wave SRS. This simple version of the SRS is implemented at small optical thickness of the amplifying medium. The medium is a gas of quantum Λ-systems. We restrict our consideration to the intensity-unsaturated SRS and study only the axial scattering in the direction of the pump wave propagation.

2. Medium polarisation

Let us designate the two lowest states of the dipole-forbidden Raman transition by 1 and 2, and the unpopulated upper state by 3. The density matrix equations in the case of homogeneous broadening have the form [3]

$$\dot{\rho}_1 + \gamma\rho_1 = \gamma\rho_1^0 + \frac{2d_1\mathscr{E}}{\hbar}\,\mathrm{Re}\,\mathrm{i}\rho_{31},$$

$$\dot{\rho}_2 + \gamma\rho_2 = \gamma\rho_2^0 + \frac{2d_2\mathscr{E}}{\hbar}\,\mathrm{Re}\,\mathrm{i}\rho_{32}, \quad \rho_3 = 0,$$

$$-\mathrm{i}\dot{\rho}_{31} + \omega_{31}\rho_{31} = \frac{\mathscr{E}}{\hbar}(d_1\rho_1 + d_2\rho_{21}), \qquad (1)$$

$$-\mathrm{i}\dot{\rho}_{32} + \omega_{32}\rho_{32} = \frac{\mathscr{E}}{\hbar}(d_2\rho_2 + d_1\rho_{21}^*),$$

$$\dot{\rho}_{21} + (\Gamma + \mathrm{i}\omega_{21})\rho_{21} = \frac{\mathrm{i}\mathscr{E}}{\hbar}(d_2\rho_{31} + d_1\rho_{32}^*).$$

Here, ρ_{jl} and ω_{jl} are the off-diagonal density matrix elements (polarisations) and the frequency of the transitions $j \leftrightarrow l$, respectively; $\rho_{1,2}$ and $\rho_{1,2}^0$ are the current and equilibrium level populations of the Raman transition; ρ_3 is the population of state 3; $d_{1,2}$ are the matrix elements of dipole moment operators for nonresonant dipole-allowed transitions $1 \leftrightarrow 3$ and $2 \leftrightarrow 3$, respectively; γ and Γ are the longitudinal and transverse relaxation rates of the Raman transition; $\mathscr{E}$ is the electric field amplitude of radiation in the medium; $\hbar$ is Planck's constant.

V.P. Kochanov V.E. Zuev Institute of Atmospheric Optics, Siberian Branch, Russian Academy of Sciences, pl. akad. Zueva 1, 634021 Tomsk, Russia; e-mail: koch@iao.ru

Received 28 July 2010
Kvantovaya Elektronika **40** (12) 1131 – 1136 (2010)
Translated by I.A. Ulitkin

We will represent the field in the medium in the form

$$\mathscr{E}(t,z) = E\sum_{n=-1}^{1} E_n(z)\cos\Psi_n, \quad \Psi_n = \omega_n t - k_n z + \varphi_n, \tag{2}$$

$$\omega_n = \omega_0 + n\omega_{21}, \; E_0(0) = 1, \; E_{-1}(0) \equiv g_{-1}, \; E_1(0) \equiv g_1,$$

where E_n is the dimensionless field amplitudes; ω_n, k_n, and φ_n are the frequencies, wavenumbers, and initial phases of the fundamental pump wave ($n = 0$), Stokes wave ($n = -1$), and anti-Stokes ($n = 1$) wave; z is the longitudinal coordinate; t is the time.

We will seek the solution of equation (1) in the form

$$\rho_{3j} = \sum_{s=-1,1}\sum_{m=-1}^{1} \exp(\mathrm{i}s\Psi_m)R_{sm}^{(j)}, \tag{3}$$

$$j = 1, 2, \quad \rho_{21} = \exp[\mathrm{i}(\Psi_{-1} - \Psi_0)].$$

Substituting expressions (3) into equation (1) and using the rotating wave approximation [4] lead to the stationary equations, whose solution with allowance for the first terms nonlinear (cubic) in the field amplitudes have the form

$$r = \mathrm{i}n_0\kappa E_0\big(f_{-1}E_{-1} + f_1\mathrm{e}^{-\mathrm{i}\Theta}E_1\big),$$

$$R_{-1-1}^{(1)} = \frac{ad_1E_{-1}}{1-\alpha+\varepsilon}\,\rho_1^0, \quad R_{-10}^{(1)} = \frac{a}{1-\alpha}(d_1E_0\rho_1^0 + d_2E_{-1}r),$$

$$R_{-11}^{(1)} = \frac{a}{1-\alpha-\varepsilon}\big(d_1E_1\rho_1^0 + d_2E_0\mathrm{e}^{\mathrm{i}\Theta}r\big),$$

$$R_{1-1}^{(1)} = \frac{a}{1+\alpha-\varepsilon}\big(d_1E_{-1}\rho_1^0 + d_2E_0 r\big),$$

$$R_{10}^{(1)} = \frac{a}{1+\alpha}\big(d_1E_0\rho_1^0 + d_2E_1\mathrm{e}^{\mathrm{i}\Theta}r\big), \quad R_{11}^{(1)} = \frac{ad_1E_1}{1+\alpha+\varepsilon}\,\rho_1^0, \tag{4}$$

$$R_{-1-1}^{(2)} = \frac{a}{1-\alpha}(d_2E_{-1}\rho_2^0 + d_1E_0r^*),$$

$$R_{-10}^{(2)} = \frac{a}{1-\alpha-\varepsilon}\big(d_2E_0\rho_2^0 + d_1E_1\mathrm{e}^{-\mathrm{i}\Theta}r^*\big),$$

$$R_{-11}^{(2)} = \frac{ad_2E_1}{1-\alpha-2\varepsilon}\,\rho_2^0, \quad R_{1-1}^{(2)} = \frac{ad_2E_{-1}}{1+\alpha-2\varepsilon}\,\rho_2^0,$$

$$R_{10}^{(2)} = \frac{a}{1+\alpha-\varepsilon}(d_2E_0\rho_2^0 + d_1E_{-1}r^*),$$

$$R_{11}^{(2)} = \frac{a}{1+\alpha}\big(d_2E_1\rho_2^0 + d_1E_0\mathrm{e}^{-\mathrm{i}\Theta}r^*\big);$$

$$\alpha \equiv \frac{\omega_0}{\omega_{31}}, \; \varepsilon \equiv \frac{\omega_{21}}{\omega_{31}}, \; \kappa \equiv \frac{d_1d_2E^2}{4\hbar^2\Gamma\Delta}, \; \Delta \equiv \omega_{31}\left(\frac{1}{1-\alpha} + \frac{1}{1+\alpha}\right)^{-1},$$

$$\Theta \equiv (2k_0 - k_{-1} - k_1)z + \Phi, \quad \Phi \equiv \varphi_{-1} + \varphi_1 - 2\varphi_0,$$

$$n_0 \equiv \rho_1^0 - \rho_2^0, \quad a \equiv \frac{E}{2\hbar\omega_{31}}.$$

The medium polarisation is defined as

$$P = 2N\mathrm{Re}(d_1\rho_{31} + d_2\rho_{32}) \equiv \sum_{n=-1}^{1}(P_{sn}\sin\Psi_n + P_{\mathrm{c}n}\cos\Psi_n), \tag{5}$$

where N is the concentration of active molecules; $P_{\mathrm{c}n}$ and $P_{\mathrm{s}n}$ are the real and imaginary parts of the amplitudes of polarisation waves. Calculating the $P_{\mathrm{s}n}$ and $P_{\mathrm{c}n}$ values (5) with the help of (3), (4) yields

$$P_{\mathrm{s}-1} = -\frac{b(f_{-1}E_{-1} + f_1\cos\Theta E_1)E_0^2}{(1-\alpha^2)(1-\alpha)(1+\alpha-\varepsilon)},$$

$$P_{\mathrm{s}0} = \frac{b}{(1-\alpha^2)^2}(f_{-1}^2E_{-1}^2 - f_1^2E_1^2)E_0,$$

$$P_{\mathrm{s}1} = \frac{b(f_{-1}\cos\Theta E_{-1} + f_1E_1)E_0^2}{(1-\alpha^2)(1+\alpha)(1-\alpha-\varepsilon)},$$

$$P_{\mathrm{c}-1} = \frac{2EN}{\hbar\omega_{31}(1-\alpha)}\left[\frac{1-\alpha}{1-(\alpha-\varepsilon)^2}\,d_1^2\rho_1^0 + \frac{1-\varepsilon}{1+\alpha-2\varepsilon}\,d_2^2\rho_2^0\right]E_{-1} + \frac{b\sin\Theta E_0^2E_1}{(1-\alpha^2)[(1-\varepsilon)^2-\alpha^2]}, \tag{6}$$

$$P_{\mathrm{c}0} = \frac{2NE}{\hbar\omega_{31}}\left[\frac{1}{1-\alpha^2}\,d_1^2\rho_1^0 + \frac{1-\varepsilon}{(1-\varepsilon)^2-\alpha^2}\,d_2^2\rho_2^0\right]E_0,$$

$$P_{\mathrm{c}1} = \frac{2EN}{\hbar\omega_{31}(1+\alpha)}\left[\frac{1+\alpha}{1-(\alpha+\varepsilon)^2}\,d_1^2\rho_1^0 + \frac{1-\varepsilon}{1-\alpha-2\varepsilon}\,d_2^2\rho_2^0\right]E_1 - \frac{b\sin\Theta E_0^2E_{-1}}{(1-\alpha^2)[(1-\varepsilon)^2-\alpha^2]};$$

$$f_{\mp1} = \frac{1\pm\alpha}{1\pm\alpha-\varepsilon}, \quad b \equiv \frac{Nn_0d_1^2d_2^2E^3}{\hbar^3\Gamma\omega_{31}^2}\left(1-\frac{\varepsilon}{2}\right)^2.$$

Note that in the approximation under study, the real part of polarisation $P_{\mathrm{c}0}$ produced by the pump wave is linear in the field amplitude.

3. Equations for the total phase and slow amplitudes

Differentiation of the equations for the field (2) while retaining only the first derivatives of the amplitudes with respect to the coordinate and substitution of the results into the wave equation

$$\left(\frac{\partial^2}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\mathscr{E} = \frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}P,$$

where c is the speed of light, in the rotating wave approximation yield

$$2\left(k_n + z\frac{\mathrm{d}k_n}{\mathrm{d}z}\right)\frac{\mathrm{d}E_n}{\mathrm{d}z} + \left(2\frac{\mathrm{d}k_n}{\mathrm{d}z} + z\frac{\mathrm{d}^2k_n}{\mathrm{d}z^2}\right)E_n = -\frac{4\pi\omega_n^2}{c^2E}P_{\mathrm{s}n},$$

$$k_n + z\frac{\mathrm{d}k_n}{\mathrm{d}z} = \frac{\omega_n}{c}\sqrt{1 + \frac{4\pi}{EE_n}P_{\mathrm{c}n}} \approx \frac{\omega_n}{c}\left(1 + \frac{2\pi}{EE_n}P_{\mathrm{c}n}\right), \tag{7}$$

$$n = -1, 0, 1.$$

Use of the approximation associated with the expansion of the square root in the last three expressions for the wavenumbers (7) leads (as shown by the corresponding numerical calculations) to underestimating the amplitude of the anti-Stokes wave by approximately 5 % in the region of its output to the stationary value and virtually has no effect on the Stokes wave amplitude and the total phase Θ (4). In what follows, we will restrict ourselves to this approximation.

Expressing the derivatives of the wavenumbers in the first three equations (7) from the last equations with the help of (6) and solving this system for the first derivatives of the amplitudes, in a cubic field approximation we obtain the equations for the amplitudes

$$\frac{\mathrm{d}E_{-1}}{\mathrm{d}\zeta} = \left(1 - \frac{\varepsilon}{\alpha}\right) f_{-1}(f_{-1}E_{-1} + f_1 \cos\Theta E_1)E_0^2,$$

$$\frac{\mathrm{d}E_0}{\mathrm{d}z} = -\left(f_{-1}^2 E_{-1}^2 - f_1^2 E_1^2\right)E_0, \quad (8)$$

$$\frac{\mathrm{d}E_1}{\mathrm{d}\zeta} = -\left(1 + \frac{\varepsilon}{\alpha}\right) f_1(f_{-1}\cos\Theta E_{-1} + f_1 E_1)E_0^2;$$

$$\zeta \equiv Gz, \quad G = \frac{4\pi\alpha(1-\varepsilon/\alpha)^2 N n_0 d_1 d_2 \kappa}{(1-\alpha^2)c\hbar n_{\mathrm{d}}},$$

$$n_{\mathrm{d}} = 1 + \frac{4\pi N\left(d_1^2\rho_1^0 + d_2^2\rho_2^0\right)}{(1-\alpha^2)\hbar\omega_{31}}.$$

Equations (8) have an integral of motion (the Manly–Row relations [3, 5])

$$\frac{E_{-1}^2}{1-\varepsilon/\alpha} + E_0^2 + \frac{E_1^2}{1+\varepsilon/\alpha} = \mathrm{const}$$

$$= 1 + \frac{g_{-1}^2}{1-\varepsilon/\alpha} + \frac{g_1^2}{1+\varepsilon/\alpha} \equiv U. \quad (9)$$

Taking into account the parameters ε and α (4), these relations represent the sum of the ratios of the energy densities of the waves to their frequencies and express the law of conservation of the total number of quanta of all the waves participating in the nonresonance SRS. Because of (9) the intensity of the main pump wave is determined by the intensity of the scattered waves, we will continue to track only the behaviour of the amplitudes of the Stokes and anti-Stokes waves.

The equation for the total phase Θ (4) is derived from equations (7) with the help of series expansion of the linear parts of real polarisations (6) in the small parameter ε up to the second order:

$$\frac{\mathrm{d}\Theta}{\mathrm{d}\zeta} = -\eta + n_{\mathrm{d}} f_{-1} f_1 \sin\Theta\, \frac{(1+\varepsilon/\alpha)E_{-1}^2 - (1-\varepsilon/\alpha)E_1^2}{E_{-1}E_1},$$

$$\eta = \frac{2n_{\mathrm{d}}(3+\alpha^2)\left(d_1^2\rho_1^0 + d_2^2\rho_2^0\right)}{(1-\varepsilon/2)^2(1-\alpha^2)^2 n_0 \kappa d_1 d_2}\,\varepsilon^2, \quad (10)$$

where $\eta = (2k_0 - k_{-1} - k_1)/G$ is a dimensionless wave detuning caused by the real parts of polarisations that are linear in the field amplitudes.

The first and third equations in (8) with allowance for the equality from relation (9)

$$E_0^2 = U - \frac{E_{-1}^2}{1-\varepsilon/\alpha} - \frac{E_1^2}{1+\varepsilon/\alpha} \quad (11)$$

together with equation (10) form a closed system of equations describing a three-wave SRS under conditions of the nonlinear dispersion of the medium. The boundary conditions for this equation have the form:

$$E_{-1}(0) = g_{-1}, \quad E_1(0) = g_1, \quad \Theta(0) = \Phi.$$

In the limit $\varepsilon \to 0$, the resulting system of equations coincides with accuracy to notations with equations (4) from [6]: the only difference is that the latter describe axial and conical scattering.

4. Spatial development of SRS

We will estimate the problem parameters as applied to the SRS on the rotational transition $J = 1 \leftrightarrow J = 3$ of the hydrogen molecules. For the initial parameters $\omega_{21} = 587$ cm^{-1}, $\omega_0 = 1.88 \times 10^4$ cm^{-1} ($\lambda = 532$ nm), $\omega_{31} = 10^5$ cm^{-1}, $N = 2.69 \times 10^{19}$ cm^{-3} (pressure, 1 atm), $\rho_1^0 = 0.493$, $\rho_2^0 = 0.249$, $n_0 = 0.244$, $\Gamma = 0.1$ cm^{-1}, $d_1 = d_2 = 5$D, and the 50-MW cm^{-2} intensity of the main pump component, the calculated parameters are: $\alpha = 0.19$, $\varepsilon = 0.0059$, $\Delta = 4.8 \times 10^4$ cm^{-1}, $f_{-1} = 1.005$, $f_1 = 1.007$, $n_{\mathrm{d}} = 1.00033$, $G = 0.175$ cm^{-1}, $\kappa = 0.0137$, and $\eta = 0.05$.

The numerical solution of equations (8)–(11) shows that in the case of small wave detunings η and relatively large and close-to-the-initial values of the amplitudes of the Stokes and anti-Stokes waves, their amplitude at $\zeta > 0.5$ significantly depend on the initial total phase of the pump waves Φ (Figs 1a, b). When $\Phi = \pm\pi$, the amplitudes of both waves vary with distance much slower than when $\Phi \sim 0$. Thus, the SRS is suppressed. The calculations imply that this effect exists for $g_{-1} \approx g_1 \geqslant 0.1$ and becomes more pronounced with increasing $g_{\pm 1}$.

The physical mechanism of the SRS suppression is clarified on the basis of equations (8)–(11) in the limit $\eta, \varepsilon \to 0, f_{\pm 1} \to 1$. In the case of $\Phi = \pm\pi$, when the phase in the medium has a constant value $\Theta = \pm\pi$ (Fig. 1c), the factor $\cos\Theta$ in the right-hand sides of equations (8) for the wave amplitudes is equal to -1. The system of equations (8)–(11) then reduces to two equations for the amplitudes:

$$\frac{\mathrm{d}E_{-1}}{\mathrm{d}\zeta} = \frac{\mathrm{d}E_1}{\mathrm{d}\zeta} \approx (E_{-1} - E_1)E_0^2, \quad (12)$$

and equality (11).

Apart from relations (9), equations (12) have the integral of motion

$$E_{-1} - E_1 \approx \mathrm{const} = g_{-1} - g_1. \quad (13)$$

It follows from equations (12), (13) that in the case of $g_{-1} \approx g_1$, the derivatives of the amplitudes of the Stokes and anti-Stokes waves are small and the SRS is suppressed. The difference between the amplitudes (13) at $\Theta = \pm\pi$ is proportional to the imaginary part of the constant component of the polarisation of the Raman transition r (4), which, as seen from the last equation in (1) is proportional, in turn, to the

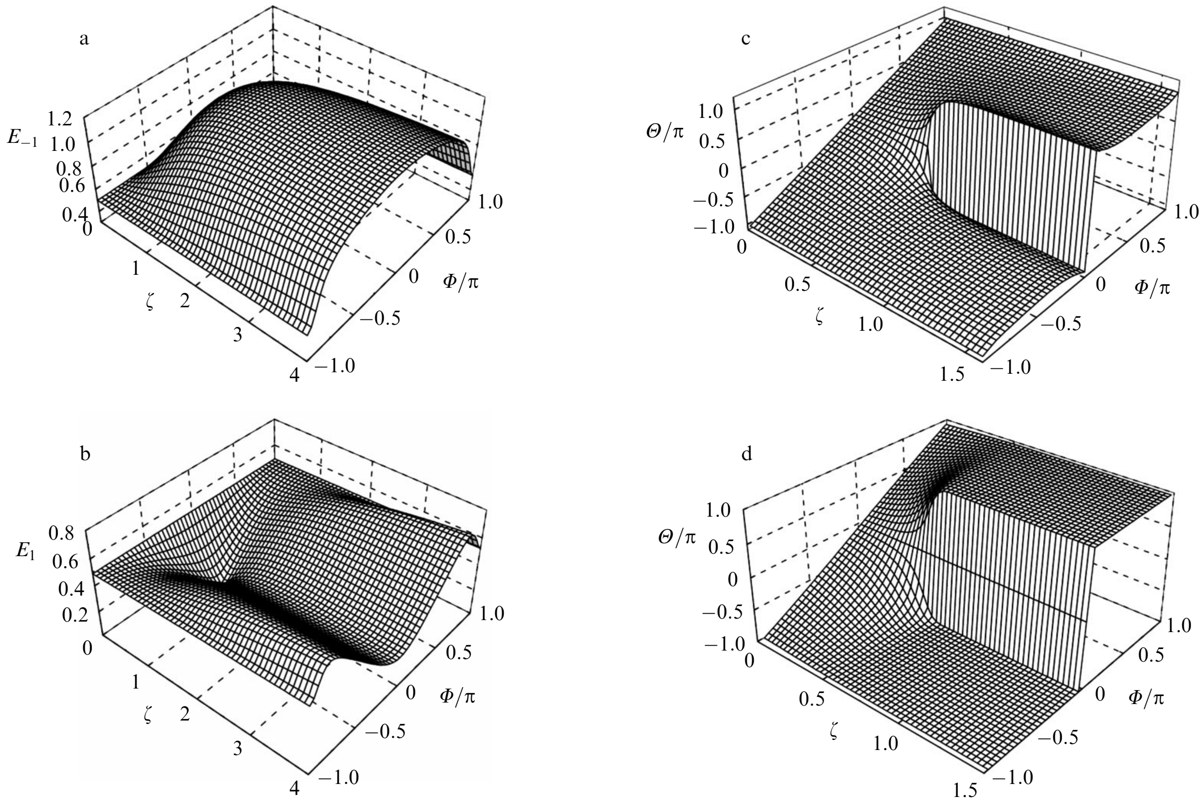

Figure 1. Amplitudes of Stokes (a) and anti-Stokes (b) waves as well as the total phase (c, d) as functions of the dimensionless length ζ and the initial total phase Φ at $\eta = 0.007$, $g_{-1} = 0.5$, $g_1 = 0.49$, $\kappa = 0.1$, $\varepsilon = 0.006$, $\alpha = 0.2$. The phase Θ (Fig. 1d) is calculated by expression (16) for $h_0 = 180$, $q = 0.07$, and $\zeta_0 = 0.7$.

difference between the polarisations of the nonresonant dipole-allowed transitions. Thus, the SRS is suppressed due to destructive interference of these polarisations that is similar to destructive interference, which is the cause of the coherent population trapping [7] in the case of the resonant interaction of two waves with a dipole-allowed transitions of the Λ-system.

Let us discuss the behaviour of the phase. At a small initial phase $\Phi \approx 0$, the phase in the medium remains virtually unchanged up to some value $\zeta = \zeta_0 \leqslant 1$, and at the point $\zeta_0 \approx g_1/g_{-1}$ it experiences a jump by π and then quickly takes the stationary value (Fig. 1c). This behaviour has been called nonlinear trapping and phase jump [2, 6]. At positive initial phases, $\Phi > 0$, the phase jump in the medium $\Delta\Theta$ with increasing the length is positive, $\Delta\Theta = \pi$, and at $\Phi \leqslant 0$ – negative, $\Delta\Theta = -\pi$. Consequently, a small change in the initial phases from negative values to positive ones causes a 2π phase jump at the point with the coordinate $\zeta > \zeta_0$. For $|\Phi| > 0.1$, the phase jump quickly disappears. These peculiarities in the behaviour of the phase were not observed earlier in the literature.

The qualitative behaviour of the phase (pulling, jump, and a quick attainment of the stationary value) is due to fundamental changes in the mechanism of dispersion, which in the nonlinear case is determined by differential equations (7), (10). The main feature of equation (10) is the dependence of the phase derivative on $\sin\Theta$, which arises due to nonlinearity (cubic in the field) of the medium polarisation. This mechanism acts as follows. In the limit $\eta \to 0$, equation (10) can be written as

$$\frac{d\Theta}{d\zeta} \approx h \sin\Theta, \quad \Theta(0) = \Phi, \tag{14}$$

where h is the coordinate function determined by the wave amplitudes. To simplify the situation, we set the function h constant. In this case, equation (14) has a solution

$$\Theta = 2\operatorname{arccot}\left(e^{-h\zeta} \cot\frac{\Phi}{2}\right). \tag{15}$$

The behaviour of the function $\Theta(\zeta, \Phi)$ agrees qualitatively with the behaviour of the phase shown in Fig. 1c; however, in contrast to the calculated phase, the function $\Theta(\zeta, \Phi)$ is more smooth and has no jumps. From the calculations of the $h(\zeta)$ dependence under the conditions adopted while plotting Fig. 1, it follows that the function $h(\zeta)$ can be approximated by the expression $h = h_0/[1 + (\zeta - \zeta_0)^2/q^2]$, where h_0, ζ_0, and q are the constants. Using this approximation in equation (14) leads to a solution

$$\Theta = 2\operatorname{arccot}\left\{\exp\left[-h_0 q\left(\arctan\frac{\zeta - \zeta_0}{q} + \arctan\frac{\zeta_0}{q}\right)\right]\cot\frac{\Phi}{2}\right\}. \tag{16}$$

Expression (16) in the case $q \ll 1$ and $h_0 q \gg 1$, with an appropriate choice of parameters, describes quantitatively well the pulling (nonlinear trapping), jump, and a quick attainment of the stationary phase Θ (Fig. 1d). From solutions (16) follows clear explanation of phase jumps when changing the spatial coordinates and changing the sign of the initial phase.

The above analysis of the phase behaviour showed that the phase jump appears when its derivative is large ($h \gg 1$). When $g_{-1} \approx g_1$, this is due to the fact that, as follows from the third equation in (8) with a negative right-hand side, the amplitude of the anti-Stokes wave at $\zeta < \zeta_0$ decreases linearly from an initial value to almost zero*. Then, the phase jump changes the signs of $\cos\Theta$ and of the right-hand side of the third equation in (8) for the amplitude of the anti-Stokes waves; as a result, the amplitude remains positive and increases. However, there is another way of increasing the derivative of the phase – the use of bichromatic pumping. In this case, $g_{-1} \approx 1$, and the value of g_1 is determined by a spontaneous seed that is several orders of magnitude smaller than the initial amplitude of the Stokes pump. As seen from equation (10), $h \approx g_{-1}/g_1 \gg 1$ for $\zeta = 0$. Therefore, the phase jump occurs immediately at the medium input. Figure 2 shows the numerical solution of equations (8) – (11) for this case. The phase behaviour in this case (Fig. 2c) agrees with the above qualitative considerations. Because the nonlinear dispersion begins to affect at $\zeta \ll 1$, amplification of the Stokes and anti-Stokes waves at the medium input is determined only by a small wave detuning η, i.e., occurs in an optimal way in the regime of the coherent SRS. This explains the high efficiency of harmonic generation in the multifrequency SRS using a bichromatic pump [1]. The dependence of the amplitudes of scattered waves on the initial phase Φ completely disappears in this case (Fig. 2a, b), and the amplitude of the anti-Stokes waves coincides up to a factor with the amplitude of the Stokes wave. The latter is due to the fact that in this case, approximate equation (12), (13) are used, where $g_{-1} \gg g_1$.

The above analysis was conducted for the case of small dimensionless wave detunings ($\eta \ll 1$), when the nonlinear dispersion of the medium dominates. Let us now find the relation of linear and nonlinear dispersions, which is given by η. As seen from the formula for η (10), the value of η increases with decreasing SRS saturation parameter κ (4) (pump power) and population difference, as well as with increasing Raman transition frequency and angular divergence of radiation [1]. Calculations show different behaviours of the phase and amplitude of the anti-Stokes wave with increasing detuning and varying the initial amplitudes $g_{-1} \approx g_1$. Namely, at sufficiently large detunings the phase jump Θ occurs against the background of its decrease with increasing coordinates by the law $\Theta = -\eta\zeta + \Phi$, which follows from equation (10) (determined by the nonlinear dispersion) with omitted terms in the right-hand side. At some critical value $\eta = \eta_0$, the phase jump completely disappears. The η_0 increases with decreasing initial amplitudes. In particular, $\eta_0 = 3.8$ and 8.2 for $g_{-1} = 0.5$ and 0.2, respectively.

The wave amplitudes, starting with $\eta \simeq 3 - 5$, oscillate with the frequency η. Oscillations are more pronounced for the anti-Stokes wave, and with increasing η, their amplitude decreases, making a few percent for large η ($\eta \approx 80$). The

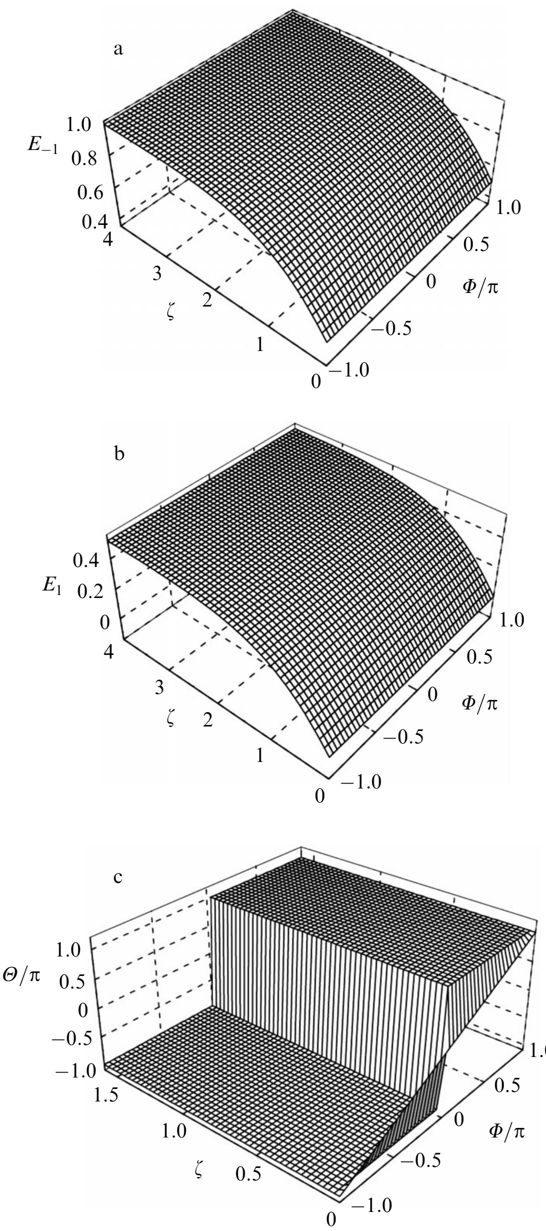

Figure 2. Amplitudes of Stokes (a) and anti-Stokes (b) waves as well as the phase Θ (c) in the case of a bichromatic pump at $g_{-1} = 0.5$, $g_1 = 0.0001$, $\eta = 0.007$, $\kappa = 0.1$, $\varepsilon = 0.006$, $\alpha = 0.2$.

amplitude of the anti-Stokes wave oscillates simultaneously with its general decrease when the length increases up to zero. The example of the behaviour of the wave amplitudes and the phases for the transition values of the wave detuning is shown in Fig. 3.

At $\eta > 100$, the amplitude oscillations in equations (8) are averaged, and these equations are reduced to the equations for dimensionless wave intensities $W_{\pm 1} \equiv E_{\pm 1}^2$:

$$\frac{dW_{-1}}{d\zeta} = 2\left(1 - \frac{\varepsilon}{\alpha}\right) f_{-1}^2 W_{-1}\left(U - \frac{W_{-1}}{1 - \varepsilon/\alpha} - \frac{W_1}{1 + \varepsilon/\alpha}\right),$$
$$\frac{dW_1}{d\zeta} = -2\left(1 + \frac{\varepsilon}{\alpha}\right) f_1^2 W_1\left(U - \frac{W_{-1}}{1 - \varepsilon/\alpha} - \frac{W_1}{1 + \varepsilon/\alpha}\right). \quad (17)$$

* Taking into account only the linear dispersion, as shows the solution of the corresponding equations, the amplitude of the anti-Stokes wave vanishes at point $\zeta = \zeta_0$ and then becomes negative, which means a phase jump by π.

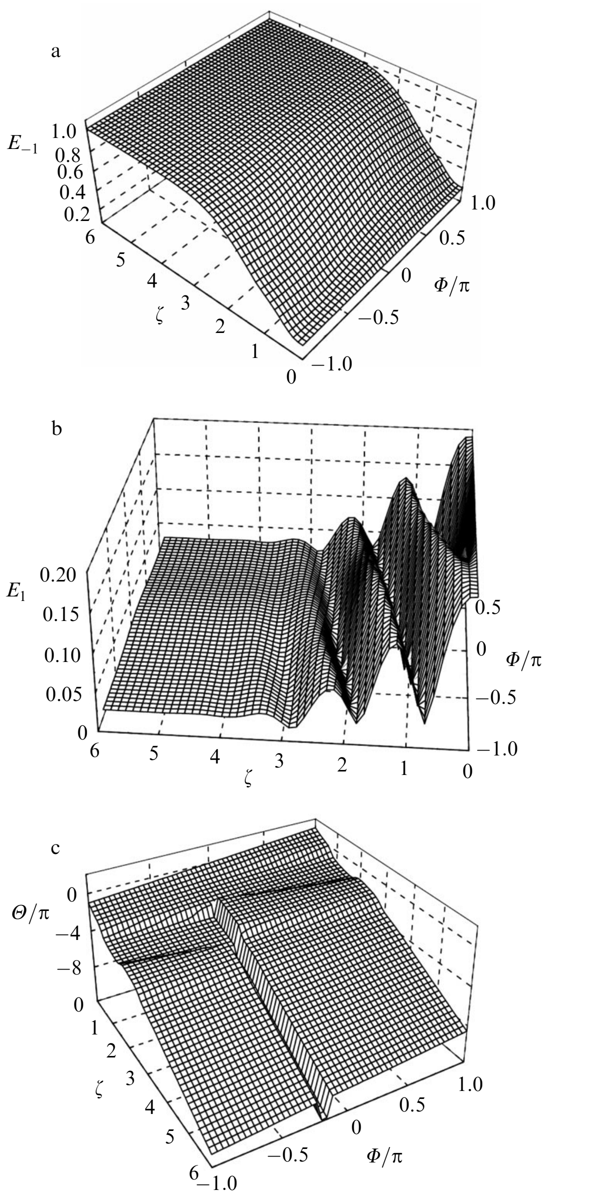

Figure 3. Amplitudes of Stokes (a) and anti-Stokes (b) waves as well as the phase Θ (c) at $\eta = 6$, $g_{-1} = 0.2$, $g_1 = 0.19$, $\varepsilon = 0.006$, $\alpha = 0.2$.

As a result, the SRS becomes noncoherent and phase independent.

When $\varepsilon = 0$, $f_{\pm 1} = 1$, the transcendental solution of equation (17) has the form

$$\zeta = \frac{1}{2Q(g_1^2 - W_1)} \ln\left(\frac{U - 2g_1^2 - Q}{U - 2g_1^2 + Q}\,\frac{U + Q - 2W_1}{U - Q - 2W_1}\right), (18)$$

$$U = 1 + g_{-1}^2 + g_1^2,$$

$$Q \equiv \sqrt{1 + 2g_{-1}^2 + 2g_1^2 + \left(g_{-1}^2 - g_1^2\right)^2},$$

$$W_{-1} = g_{-1}^2 g_1^2 W_1^{-1}.$$

The graphical solution of (18) for different values of g_{-1}^2 and g_1^2 shows that in all cases, the anti-Stokes wave intensity decreases monotonically from its initial value to zero, and the Stokes wave intensity increases monotonically to a limiting value determined by (9).

Note that the effect of the nonlinear dispersion of the medium on the anti-Stokes SRS component is determining at $\eta \leqslant 1$ and has a significant impact in a wide range of values η $(1 < \eta < 50)$, including in the case of small saturation parameters. Therefore, the effects induced by the nonlinear dispersion should be observed upon the SRS not only on rotational but also on the vibrational transitions of molecules.

Acknowledgements. The author thanks M.M. Makogon for useful discussions of this paper.

References

1. Losev L.L., Lutsenko A.P. *Kvantovaya Elektron.*, **23**, 919 (1993) [*Quantum Electron.*, **20**, 1054 (1993)].
2. Butylkin V.S., Kaplan A.E., Khronopulo Yu.G., Yakubovich E.I. *Resonant Nonlinear Interactions of Light with Matter* (Berlin: Springer, 1989; Moscow: Nauka, 1977).
3. Kochanov V.P., Bogdanova Yu.V. *Zh. Eksp. Teor. Fiz.*, **123**, 233 (2003).
4. Allen L., Eberly J. *Optical Resonance and Two-Level Atoms* (New York, Wiley, 1975).
5. Landau L.D., Lifshitz E.M. *Electrodynamics of Continuous Media* (Oxford: Pergamon Press, 1984).
6. Butylkin V.S., Venkin G.V., Protasov V.P,et al. *Zh. Eksp. Teor. Fiz.*, **70**, 829 (1976).
7. Agap'ev B.D., Gornyi M.B., Matisov B.G., Rozhdestvensky Yu.V. *Usp. Fiz. Nauk*, **163**, 1 (1993).