

FIG. 2. Comparison of the Franck-Condon distribution for the products of the resonance dissociation of hydrogen molecules (process (2)) with the distribution calculated with Eq. (20) using the parameter values $E_0 \approx 2.4 \cdot 10^{-1}$ and $\Omega \approx 1.5 \cdot 10^{-1}$ from^[6] and the experimental value $\Gamma \approx 7.4 \cdot 10^{-3}$ from^[9]. The dashdot curve shows the Franck-Condon spectrum normalized to unity at the peak, and the full curve shows the spectrum calculated with allowance for the finite dwell time of the system in the $(1 s\sigma_{e})(2p\sigma_{w})^{2}\Sigma_{e}^{*}$ state of H₂⁻.

 $\frac{\Delta E(D_2) - \Delta E(H_2)}{\Delta E(HD) - \Delta E(H_2)} = 3$

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¹⁾Transitions following a single rapid change of the interaction are described by the theory of sudden perturbations and have been well investigated for specific systems.^[1]

- ²⁾Earlier published papers on the resonance interaction of electrons with molecules were based on numerical integration of Eq. (4) or on direct summation of expression (3).
- ³⁾As a specific example we give the asymptotic expansion of the amplitude for the elastic process following sudden removal of the Coulomb potential with its subsequent restoration a time τ later $(V_{\alpha} = V_{\beta} = -z/r, R_{\alpha} = R_{\beta} = 2rz^{3/2}e^{-rr})$:

$$a_{ii}(\tau) = 1 - i \frac{z^2 \tau}{2} - \frac{5}{8} (z^2 \tau)^2 + \frac{32}{15} \left(\frac{i}{2\pi}\right)^{\frac{1}{2}} (z^2 \tau)^{\frac{5}{2}} + \cdots$$

- ⁴⁾The known result $a_{\alpha\beta} \sim \tau^{-3/2}$ for free motion in the intermediate state (see, e.g., ^[8]) follows from Eq. (14) with $\lambda = 1/2$ ($\gamma = 0$).
- ¹L. D. Landau and E. M. Lifshits, Kvantovaya mekhanika (Quantum mechanics), Fizmatgiz, 1963.
- ²F. B. Bronfin and A. I. Ermolaev, Vestnik LGU 22, 29 (1971).
- ³J. N. Bardsley, A. Herzenberg, and F. Mandl, Proc. Phys. Soc. Lond. 89, 305 (1966).
- ⁴D. T. Birtwhistle and A. Herzenberg, J Phys. B 4, 53 (1971).
- ⁵J. C. Y. Chen and J. L. Magee, J. Chem. Phys. 36, 1407 (1972).
- ⁶J. C. Y. Chen, J. Chem. Phys. 40, 3513 (1964).
- ⁷G. V. Skrotskii and G. I. Solomakho, Opt. Spektrosk. 39,
- 827 (1975) [Opt. Spectrosc. (USSR) 39, 470 (1975)].
- ⁸Yu. N. Demkov, Zh. Eksp. Teor. Fiz. 46, 1126 (1964) [Sov. Phys. JETP 19, 762 (1964)].

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Stimulated Raman emission and frequency scanning in an optical waveguide

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Equations are derived for stimulated Raman emission in an optical waveguide. The arbitrary number of components of the radiation and also the dependence of the refractive index on the light intensity are taken into account in the equations. Solutions of the equations are obtained for some cases of practical interest. On the basis of the solutions the following phenomena are predicted and investigated: "ladder" scanning of the optical frequency in a fixed cross section of the waveguide; a "multiplication" effect of the initial scanning range due to mutual transformation of the radiation components; the possibility of controlling the scanning process by means of a weak input (Stokes) pulse. The possibilities of "quenching" stimulated Raman emission in a wave guide are also considered.

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INTRODUCTION

It is known that various nonlinear phenomena can be observed in optical waveguides. Ippen *et al.*^[1] have observed broadening of the spectrum of picosecond light pulses passing through a multimode optical waveguide. This broadening was attributed by them to phase modulation due to the dependence of the refractive index of the waveguide material on the light intensity. In an earlier paper⁽²⁾ I called attention to the fact that the passage of an intense light pulse with an initially fixed field-oscillation frequency through a single-mode optical waveguide can be used to obtain¹⁾ broadband scanning of the frequency in this pulse, such that the scanning interval can exceed, for example, 10^{14} rad/sec.

Frequency scanning uncovers great possibilities for selective excitation of a specified level of multilevel

⁹G. V. Dubrovsky, V. D. Ob'edkov, and R. K. Janev, V. ICPEAC, Abstracts of papers, Leningrad, 1967.

quantum-mechanical systems with non-equidistant energy spectrum as a result of a succession of adiabatic transitions (in the optical band) between neighboring states, for example in atoms, impurity centers in solids, etc. A similar method (adiabatic fast passage) for the excitation of quantum states in spin systems is well known for the microwave band (see, e.g., $^{[3,4]}$), and its possible use in the optical region to obtain population inversion of two levels has been discussed in^{15]}. Adiabatic inversion of the populations in a system of two vibrational levels of ammonia was recently investigated by Loy^[6] and Hamadani et al.^[7] Loy^[6] worked with a fixed laser-beam frequency, and varied the vibrational-transition frequency via the Stark effect by applying an electric-field pulse to a gas (the range of variation of the frequency amounted in this case to ~ 5×10^9 rad/sec). Hamadani *et al.*^[7] effected a laserbeam frequency deviation in the range $\sim 10^9$ rad/sec by varying the length of the laser resonator. We note also that Strel'tsov and I^[8] have called attention to the possibility of exciting higher vibrational levels by successive adiabatic transitions (in a gas with a well-resolved rotational level structure), provided that the range of frequency variation "spans" the corresponding nonequidistance of the system of vibrational-rotational levels, which usually does not exceed 10^{12} rad/sec. This excitation can also be produced by scanning the frequency of the field oscillations of the light pulse in the optical waveguide.

Besides frequency scanning of the light pulse, other nonlinear phenomena can be observed in a single-mode optical waveguide. Ippen and Stolen^[9-11] have observed stimulated Raman scattering and stimulated Mandel'shtam-Brillouin scattering in glass waveguides in the cw regime, i.e., at a pump intensity constant in time. They measured the gain at the first Stokes frequency. Owing to the small transverse dimension (usually of the order of $3 \times 10^{-4} - 10^{-3}$ cm) and the large length of propagation without divergence and with practically no linear absorption (this length reaches 10⁵ cm in modern optical waveguides^[12]) the pump power needed to observe the stimulated scattering turned out to be very low, of the order of several watts.^[10-13] At the same time, the use of sufficiently short pump pulses (for example, of duration shorter than 10^{-7} sec) makes it easy to reach peakpower values exceeding $10^3 - 10^5$ W. When a waveguide is excited by such pulses, it is obviously necessary to take into account multiple stimulated scattering (i.e., to take into account an arbitrary number of components of this scattering) and, furthermore, as indicated in^[2], it is necessary to take into account the frequency scanning in the propagating pulses. It should be noted here that even in the cw regime the threshold of stimulated Mandel'shtam-Brillouin scattering usually turns out to be much higher than the calculated value^[13] for strictly monochromatic pumping. The authors of ^[12] attribute this to the fact that the spectral widths of the customarily employed pump sources greatly exceed the width of the spontaneous Mandel'shtam-Brillouin scattering line. Bearing in mind the case of propagation of sufficiently short light pulses in the waveguide, with a frequency that lends itself to scanning, we shall disregard this

type of scattering.

In the present paper we examine jointly stimulated Raman emission and frequency scattering in a singlemode optical waveguide in which light pulses propagate. With respect to the stimulated Raman emission, in contrast to earlier studies, where the gain was estimated at the first Stokes frequency, we have carried out a step-by-step analysis of this phenomenon in the waveguide, taking into account not only the first Stokes component but an arbitrary number of components of higher order. It will be shown below that under these conditions the stimulated Raman emission differs greatly in general from the stimulated Raman scattering (SRS) in homogeneous three-dimensional volumes (see^[14,15]) and from stimulated Raman emission in optical resonators $(see^{[16-19]})$. In typical situations, it has features that can be of practical interest. Our investigation consists in a derivation of equations describing the considered process in the optical waveguide, and the solution of these equations for a number of cases. On the basis of the obtained solutions, in particular, we have predicted and investigated the following: "ladder" scanning of the frequency, effect of "multiplication" of the range of continuous scanning, the possibility of controlling the scanning process with the aid of a weak Stokes input pulse, and also the "quenching" of the stimulated Raman emission in a waveguide in a number of cases.

1. DERIVATION OF EQUATIONS

Let the waveguide parameters be such that only one (fundamental) mode can propagate in it in the considered frequency region. The complex amplitude of the electric field of this wave can be written in the form $(see^{(12,20)})$

$$\mathbf{E} = \mathbf{g}(\mathbf{r}_{\perp}) e^{ikz}, \quad k = \frac{\omega}{c} n_{\text{eff}}^{(0)}(\omega).$$
(1)

Here z and \mathbf{r}_{\perp} are respectively the longitudinal and transverse coordinates, and ω is the frequency of the propagating wave (the function **g** is real). We assume that two or several waves of the type under consideration, with frequencies ω_q determined by the relation

 $\omega_q \approx \omega_0 + q \omega_r, \tag{2}$

where ω_r is the frequency of the vibrational transition with which the stimulated Raman emission is connected, are excited by an external source at the entrance to the waveguide in the plane z=0. Accordingly, the electric field in the waveguide can be written in the form

$$\mathbf{e} = \frac{1}{2} \sum_{q} F_{q}(\mathbf{z}, t) \mathbf{g}_{q}(\mathbf{r}_{\perp}) e^{ik_{q}z - i\omega_{q}t} + \text{c.c.},$$

$$k_{q} = k(\omega_{q}), \qquad (3)$$

with the complex functions $F_q(z,t)$ as can be easily verified with the aid of Maxwell's equations, satisfying the relations

$$\frac{\partial F_q}{\partial t} + v_q \frac{\partial F_q}{\partial z} = \frac{i\omega_q}{2N_q} e^{-ik_q z} \int \mathbf{P}_q^{(\text{extr})} \mathbf{g}_q \, d\mathbf{r}_{\perp} \,, \tag{4}$$

and

$$v_q = \frac{\partial \omega_q}{\partial k_q}, \quad N_q = \frac{1}{4\pi} (n_{\text{eff}}^{(0)})^2 \int \mathbf{g}_q^2 d\mathbf{r}_\perp,$$
 (5)

 $\mathbf{P}_{q}^{(\text{extr})}$ is the complex amplitude of the extraneous polarization, which is taken into account in (4) as a perturbation and is in general a sum of the linear and nonlinear parts of the polarization of the waveguide material at the frequency ω_{q} . The linear part is due to losses caused by linear absorption; the nonlinear part is due in this case to two effects: stimulated Raman emission and the dependence of the refractive index of the material of the waveguide on the light intensity. Accordingly, we write

$$\mathbf{P}_{q}^{(\text{extr})} = \mathbf{P}_{q}^{(L)} + \mathbf{P}_{q1}^{(\text{nl})} + \mathbf{P}_{q2}^{(\text{nl})}, \tag{6}$$

where the linear part $\mathbf{P}_{q}^{(L)}$ satisfies the equality

$$\mathbf{P}_{q}^{(L)} = i \varkappa''(\omega_{q}) F_{q} \mathbf{g}_{q} e^{i k_{q} z}$$
(7)

 (\varkappa'') is the imaginary part of the linear dielectric susceptibility of the material).

The nonlinear part of the polarization $P_{q1}^{(n1)}$, which is connected with the stimulated Raman emission, can be obtained from the material equation of the medium that is active in the Raman-scattering spectrum (see, e.g., ^[14,15]), in analogy with the procedures used in the papers^[16-19] describing stimulated Raman emission in optical resonators:

$$\mathbf{P}_{q1}^{(\mathbf{n}\mathbf{l})} = \frac{1}{2} N \frac{d\alpha}{dx} e^{ik_{q2}} \left[\sum_{l} X_{l}^{*} F_{q+1}(\mathbf{g}_{l} \mathbf{g}_{l+1}) \mathbf{g}_{q+1} \exp\left(i\omega_{ql}^{(2)} t - ik_{ql}^{(2)} z\right) + X_{l} F_{q-1}(\mathbf{g}_{l} \mathbf{g}_{l+1}) \mathbf{g}_{q-1} \exp\left(i\omega_{ql}^{(1)} t - ik_{ql}^{(1)} z\right) \right],$$
(8)

where

N is the density of the molecules active in the Raman center; α is the polarizability of one molecule and is a function of the relative locations of the nuclei (x); the derivative $d\alpha/dx$ is taken at the point corresponding to the equilibrium position, and is a constant coefficient; the quantities X_i are the amplitudes of the molecular os-cillations in the medium and satisfy the equations

$$\dot{X}_{i} = -(h+i\delta_{i})X_{i} + \frac{i}{4m}\frac{d\alpha}{dx}F_{i}F_{i+1}^{\dagger}.$$
(10)

Here

$$\delta_{l} = \omega_{r} + \omega_{l} - \omega_{l+1}, \qquad (11)$$

m is the reduced mass of the molecule, which corresponds to the considered normal vibrations; *h* is the half-width of the spontaneous Raman scattering line $(2h \ll \omega_r)$.

To find that part of the linear polarization which is due to the dependence of the refractive index of the medium on the light intensity, we specify this dependence concretely by assuming, for the sake of argument, that it is connected with the Kerr effect and that the polarizations of all the considered waves are equal. In this case we can start from the relations

$$\mathbf{P}_{2}^{(n)} = \chi \mathbf{e}, \quad \dot{\chi} + h_{1}\chi = h_{1}v_{2}\mathbf{e}^{2}$$
(12)

with a scalar susceptibility χ . For the sake of argument we confine ourselves also to the case when the time required for the Kerr effect to be established greatly exceeds the period of the molecular vibrations $(h_1 < \omega_r)$. Starting from (12) and taking (2) into account, we obtain

$$\mathbf{P}_{q2}^{(nl)} = \frac{1}{2} v_2 F_q g_q e^{ik_q z} \sum_l \chi_l g_l^2,$$
(13)

where the quantities χ_i satisfy the equation

$$\dot{\chi}_{l} + h_{1}\chi_{l} = h_{1}|F_{l}|^{2}$$
. (14)

The relations (4), (6)-(8), (10), (13), and (14) form a closed system of equations for the quantities F_q , X_l , and $\chi_{l'}$.

We simplify this system by considering the case when the scale T of the variation of the quantities $F_i F_{i+1}^*$ as functions of the time is large in comparison with the characteristic time of establishment of the stimulated Raman emission (this condition corresponds to the inequality $hT \gg 1$) and the scale T_1 of the variation of $|F_i|^2$ is large in comparison with the characteristic time of establishment of the Kerr effect ($h_1 T_1 \gg 1$). In this case, putting formally in (10) $\dot{X}_i = 0$ and $\dot{X}_i = 0$ in (14), and expressing next X_i in terms of $F_i F_{i+1}^*$ and χ_i in terms of $|F_i|^2$, we arrive at the following closed system of equations for the quantity F_q :

$$\frac{\partial F_{q}}{\partial t} + v_{q} \frac{\partial F_{q}}{\partial z} = -\mu_{q} F_{q} + \alpha_{q} \left[\sum_{l} a_{ql} \frac{F_{l} F_{l+l} F_{q+1}}{h - i \delta_{l}} \exp\left(i\omega_{ql}^{(2)} t - ik_{ql}^{(2)} z\right) - a_{q-1,l} \frac{F_{l} F_{l+1} F_{q-1}}{h + i \delta_{l}} \exp\left(i\omega_{ql}^{(1)} t - ik_{ql}^{(1)} z\right) \right] + i\beta_{q} F_{q} \sum_{l} b_{ql} |F_{l}|^{2}, \quad (15)$$

where

$$\mu_{q} = \frac{\omega_{q}}{2N_{q}} \int \varkappa''(\omega_{q}) g_{q}^{2} d\mathbf{r}_{\perp}, \qquad \alpha_{q} = \frac{\omega_{q}}{16mN_{q}} \left(\frac{d\alpha}{dx}\right)^{2}, \qquad \beta_{q} = \frac{\omega_{q}}{4N_{q}},$$

$$a_{ql} = \int N(g_{q}g_{q+1}) (g_{l}g_{l+1}) dr_{\perp}, \qquad b_{ql} = \int v_{2}g_{q}^{2}g_{l}^{2} d\mathbf{r}_{\perp}.$$
(16)

Our analysis here includes the case when the quantities \varkappa'' , N, and ν_2 depend on \mathbf{r}_1 ; accordingly, the indicated quantities have been left under the integral sign in (16). We note also that the initial perturbation method (3), (4) is applicable under the condition that all the susceptibilities that determine the extraneous polarization (6) are small in comparison with $\Delta \varepsilon_0/4\pi$, where $\Delta \varepsilon_0$ is the difference between the values of the linear real part of the dielectric constant of the medium inside the wave-guide and on its periphery.

Equations (15) constitute a nonlinear system for the quantities F_q and describe, in accordance with (2) different components of stimulated Raman emission (the subscript q takes on values $0, \pm 1, \pm 2, \ldots$ while the values q = 0, q < 0, and q > 0 correspond to the pump and to

the Stokes and anti-Stokes components, respectively). We shall consider this system below for the case of greatest interest, when the external source excites only two waves—the pump and the first Stokes wave—i.e., we assume

$$F_{q}|_{z=0} = Y_{0}(t), F_{-1}|_{z=0} = Y_{-1}(t),$$

$$F_{q}|_{z=0} = 0, (q \neq 0, -1),$$
(17)

 Y_0 and Y_{-1} are specified functions of the time. In this case the frequencies ω_q (which in general can be chosen arbitrarily within the framework of (2)) can be conveniently defined by the exact equalities $\omega_q = \omega_0 + q(\omega_0 - \omega_{-1})$. We then have $\omega_{ql}^{(1,2)} \equiv 0$ and $\delta_l = \delta_{-1}$. The quantities $k_{ql}^{(1,2)}$, which enter also in (15), are determined by the frequency dispersion of the quantity $n_{eff}^{(0)}(\omega)$ (the only exceptions are the values $k_{qq}^{(2)}$ and $k_{q,q-1}^{(1)}$, which are equal to zero regardless of the dispersion).

2. STIMULATED RAMAN EMISSION IN THE CASE OF STRONG DISPERSION $n_{eff}^0(\omega)$

We consider first the most typical case

$$|\Delta k_q| \gg |s_3 Y_0^2(\tau)|, q=-1, -2, \dots,$$
 (18)

where

$$\Delta k_{q} = k_{q+1} + k_{q-1} - 2k_{q}, \quad s_{3} = \beta_{1}^{(q)} + \beta_{-1}^{(q)} - 2\beta_{0}^{(q)},$$

$$\tau = t - \frac{z}{v}, \quad \beta_{m}^{(q)} = \frac{\beta_{q+m}b_{q+m,q}}{v}, \quad m = 0, \pm 1.$$
(19)

The inequality (18) is usually satisfied, for example, under those conditions when the frequency dependence of $n_{eff}^{(0)}(\omega)$ is determined mainly by the dispersion of the refractive index of the waveguide material, and by the same token, is relatively strong. We confine ourselves here to the following interval in z:

$$0 \leq z < \frac{v_q}{|v_{q-1} - v_{q+1}|} l_p, \frac{v_q}{\mu_q},$$
(20)

where l_p is the length of the train of light pulses propagating in the waveguide. In this interval we can neglect the difference between the group velocities of the pulses of three neighboring components (i.e., we can put formally $v_{q\pm 1} = v_q = v$) and neglect the linear absorption in the waveguide (i.e., we can put $\mu_q = 0$). We assume also that

$$\ln \left| \frac{\Delta k_{q}}{\chi_{12}^{(q)} Y_{0}^{2}(\tau)} \right| \gg 1, \quad q = -1, -2, \dots,$$
 (21a)

$$\ln\left|\frac{Y_{0}(\tau)}{Y_{-1}(\tau)}\right| \gg 1.$$
(21b)

Here and below

$$\chi_{12}^{(q)} = \frac{\alpha_{q-1}a_{q-1,q}}{v(h-i\delta_{-1})}, \quad \chi_{21}^{(q)} = -\frac{\alpha_{q+1}a_{q,q-1}}{v(h-i\delta_{-1})},$$

$$\chi_{11}^{(q)} = \frac{\alpha_{q-1}a_{q-1,q-1}}{v(h-i\delta_{-1})}, \quad \chi_{22}^{(q)} = -\frac{\alpha_{q+1}a_{qq}}{v(h-i\delta_{-1})}.$$
(22)

To find the solution of the initial nonlinear system of differential equations (15) of arbitrary order, we can

employ under our conditions the following device: confining ourselves first to only three neighboring components F_q and $F_{q\pm 1}$, we linearize them with respect to $F_{q\pm 1}$ (a general solution of these linearized equations can be easily obtained) and then "join together" with respect to z the solutions corresponding to different values of q. The corresponding procedure yields

$$F_{q}(\tau, 0 \le z \le z_{-1}) = Y_{0}(\tau) \exp(i\beta_{0}^{(0)} | Y_{0}(\tau) |^{2}z),$$

$$F_{-1}(\tau, 0 \le z \le z_{-1}) = Y_{-1}(\tau) \exp[(\chi_{11}^{(0)} + i\beta_{-1}^{(0)}) | Y_{0}(\tau) |^{2}z],$$

$$F_{q-1}(\tau, z_{q} \le z \le z_{q-1}) = (i\Delta k_{q})^{-1}\chi_{12}^{(q)} F_{q}^{2}(\tau, z_{q} - 0) F_{q+1}^{*}(\tau, z_{q} - 0)$$

$$\times \exp\left[(\chi_{11}^{(q)} + i\beta_{-1}^{(q)}) \frac{\alpha_{q}}{\alpha_{0}} | Y_{0}(\tau) |^{2}(z - z_{q})\right],$$

$$F_{q}(\tau, z_{q} \le z \le z_{q-1}) = F_{q}(\tau, z_{q} - 0) \exp\left[i\beta_{0}^{(q)} \frac{\alpha_{q}}{\alpha_{0}} | Y_{0}(\tau) |^{2}(z - z_{q})\right],$$

$$F_{q+1}(\tau, z_{q} \le z \le z_{q-1}) = F_{q+1}(\tau, z_{q} - 0) \exp\left[(\chi_{22}^{(q)} + i\beta_{1}^{(q)}) \frac{\alpha_{q}}{\alpha_{0}} | Y_{0}(\tau) |^{2}(z - z_{q})\right],$$

where

 $z_{q-1}=z_q+\Delta z_q, z_0=0,$

$$\Delta z_{q} = \frac{1}{\operatorname{Re}\chi_{11}^{(0)}|Y_{0}(\tau)|^{2}} \ln\left\{ \left| \frac{Y_{0}(\tau)}{Y_{-1}(\tau)} \right| \sqrt{\frac{\alpha_{-1}}{\alpha_{0}}} \right\},$$

$$\Delta z_{q} = \frac{\alpha_{0}}{\alpha_{q}} \frac{1}{\operatorname{Re}\chi_{11}^{(q)}|Y_{0}(\tau)|^{2}} \ln\left\{ \left| \frac{\Delta k_{q}}{\chi_{12}^{(q)}Y_{0}^{2}(\tau)} \right| \frac{\alpha_{0}}{\alpha_{q}} \sqrt{\frac{\alpha_{q-1}}{\alpha_{q+1}}} \right\}$$
(24)

(q = -1, -2, ...). In the derivation of these relations we used also the equality

$$|F_q|^2 + \frac{\alpha_q}{\alpha_{q+1}} |F_{q+1}|^2 = \text{const}$$

(q = -1, -2, ...), which follows from (15) with account taken of only two neighboring components and which is valid approximately in the interval $z_{q+1} < z < z_{q-1}$.

It can be verified that expression (23) within the intervals Δz_q are asymptotically exact in terms of the parameter defined by relations (18) and (21). We see that these expressions describe the process of a consecutive practically complete conversion of the pump into the first Stokes component, that of a first component into the second, etc. of the stimulated Raman emission when a light pulse propagates in a waveguide. We note here that we have neglected above the influence of the priming (spontaneous) radiation at the higher Stokes frequencies. This is valid under the conditions

$$\left|\frac{\chi_{12}^{(q)}Y_0^2(\tau)}{\Delta k_{\tau}}\right|^2 \gg \frac{\alpha_0}{\alpha_q} \frac{(k_{q-1}\Lambda_{q-1})^2}{2^8 \pi \operatorname{Re}\chi_{11}^{(q)}|Y_0(\tau)|^2} NQ_0^{(q-1)},$$
(25)

where Λ_q is the scale of localization of the function $\mathbf{g}_q(\mathbf{r}_{\perp})$; $Q_0^{(q)}$ is the total cross section of the spontaneous Raman scattering per molecule at a wavelength $2\pi n^{(0)}/k_q$. We note also that since expressions (23) are valid for the boundary functions $Y_0(t)$ and $Y_{-1}(t)$ of arbitrary form, these expressions yield in essence also the solution of the corresponding statistical problem.²⁾

It follows from (23) that in general the instantaneous frequency of the oscillations of a field in a fixed section z varies with time. We put

$$F_{q}(\tau, z) = |F_{q}(\tau, z)| e^{i\Phi_{q}(\tau, z)}, Y_{m}(t) = |Y_{m}(t)| e^{i\varphi_{m}(t)}$$

(m=0, -1). The instantaneous frequency Ω_q of the oscillations of the field of the q-th component can then be written in the form $\Omega_q = \omega_q + \Delta \Omega_q$, where

 $\Delta\Omega_q = -\partial\Phi_q/\partial t. \tag{26}$

Obtaining from (23) a system of recurrence relations for the quantities $\Phi_q(\tau, z_q - 0)$, solving this system, and carrying out the differentiation (26) (with (24) taken into account), we obtain

$$\begin{split} \Delta\Omega_{0}(\tau, 0 < z < z_{-1}) &= -\frac{d\varphi_{0}(\tau)}{d\tau} - \beta_{0}^{(0)} z \frac{d|Y_{0}(\tau)|^{2}}{d\tau} ,\\ \Delta\Omega_{q}(\tau, z_{q} < z < z_{q-1}) &= -\frac{d\varphi_{0}(\tau)}{d\tau} + q \left(\frac{d\varphi_{-1}(\tau)}{d\tau} - \frac{d\varphi_{0}(\tau)}{d\tau} \right) \\ &+ \frac{1}{\operatorname{Re}\chi_{11}^{(0)}} \left[\beta_{0}^{(0)} \frac{\alpha_{q}}{\alpha_{0}} + q \left(\beta_{-1}^{(0)} + \operatorname{Im}\chi_{11}^{(0)} \right) \right] \frac{d}{d\tau} \ln \left| \frac{Y_{0}(\tau)}{Y_{-1}(\tau)} \right| \\ &- \sum_{m=0}^{q+1} \left(U_{m} + \sum_{l=0}^{m} \eta_{l} \right) \frac{d|Y_{0}(\tau)|^{2}}{d\tau} - \beta_{0}^{(0)} z \frac{\alpha_{q}}{\alpha_{0}} \frac{d|Y_{0}(\tau)|^{2}}{d\tau} , \qquad (27) \end{split}$$

where

$$U_{0}=0, \quad \eta_{0}=0, \quad U_{m}=\frac{\alpha_{0}}{\alpha_{m}} \frac{\beta_{0}^{(0)}}{\operatorname{Re}\chi_{11}^{(m)}|Y_{0}(\tau)|^{2}}, \quad m=-1,-2,\ldots,$$

$$\eta_{l}=\frac{\beta_{0}^{(l+1)}}{\operatorname{Re}\chi_{11}^{(l+1)}|Y_{0}(\tau)|^{2}} - \frac{\beta_{-1}^{(1)}+\operatorname{Im}\chi_{11}^{(l)}}{\operatorname{Re}\chi_{11}^{(l)}|Y_{0}(\tau)|^{2}}, \quad l=-1,-2.\ldots.$$
(28)

Expression (27) for $\Delta\Omega_q$ is given in the interval Δz_q within which a noticable value is possessed only by the *q*-th component (the neighboring components are small inside this interval, and the remaining components, as can be easily verified from (15) are negligible with even higher accuracy). In (27), at a fixed value of *z* and at different τ , the interval $z_q < z < z_{q-1}$ corresponds to the interval $|Y_{0q}|^2 < |Y_0(\tau)|^2 < |Y_{0,q-1}|^2$, where the quantities $|Y_{0q}|^2$, as easily seen from (23) and (24), are determined from the equations

$$\sum_{i=0}^{q+1} \Delta z_i = z.$$
 (29)

Each of these equations can be easily solved by successive approximations with allowance taken of the slow dependence of the logarithm on $|Y_0|$; in the zeroth approximation we have

$$|Y_{0q}|^{2} = \frac{1}{z} \left[\frac{1}{\operatorname{Re} \chi_{11}^{(n)}} \ln \left\{ \frac{|Y_{0}|_{\max}}{|Y_{-1}|_{\max}} \sqrt{\frac{\alpha_{-1}}{\alpha_{0}}} \right\} + \sum_{i=1}^{q+1} \frac{\alpha_{0}}{\alpha_{i}} \frac{1}{\operatorname{Re} \chi_{11}^{(i)}} \ln \left\{ \left| \frac{\Delta k_{i}}{\chi_{12}^{(i)} |Y_{0}|_{\max}^{2}} \left| \frac{\alpha_{0}}{\alpha_{i}} \sqrt{\frac{\alpha_{i-1}}{\alpha_{i+1}}} \right\} \right].$$
(30)

In accordance with (27) and (30), in a fixed section z, with increasing value of $|Y_0|^2$ in the pulse, the frequency of the radiation passing through the waveguide varies continuously initially in a certain interval; then, at $|Y_{0,-1}|^2$, a "jump" of this frequency takes place, followed again by a continuous variation, the next jump of the frequency takes place at $|Y_{0,-2}|^2$ and is followed by a new interval of continuous variation, etc.

Inasmuch as by virtue of (27) and (30) the values of the intervals of continuous variation of the frequency

can greatly exceed the reciprocal values of the corresponding time of this variation (see also the numerical example below), this means that frequency scanning is possible in these intervals. The considered discretecontinuous character of the frequency variation with scanning in the intervals of the continuous variation will as a whole be called a "ladder" scanning of frequency. It follows also from (27) and (30) that in general the position on the frequency scale of each of the individual intervals of continuous scanning (in the vicinity of each of the frequencies ω_{a}) depends on the ratio of the constants of the medium and of the parameters of the input pulses. This feature of the scanning in question can be used in practice in expressions of multilevel quantummechanical systems with quasi-equidistant spectra (owing to the successive adiabatic transitions) to tune the positions of the individual scanning intervals to the frequencies of the corresponding transitions of the multilevel system.

Let us examine expression (27) in greater detail for the case when the frequency deviations with respect to the input pump pulses and the Stokes component are absent $(d\varphi_0(\tau)/d\tau \equiv d\varphi_{-1}(\tau)/d\tau \equiv 0)$, and the duration of the input pulse of the first Stokes component is larger than or of the order of the duration of the input pump pulse. In this case, under the condition $|\operatorname{Im}\chi_{11}^{(q)}| \ll \beta_{-1}^{(q)}$, with allowance for relations (21) and (24), we obtain

$$\Delta\Omega_{q}(\tau, z_{q} < z < z_{q-1}) \approx -\beta_{0}^{(q)} z \frac{\alpha_{q}}{\alpha_{0}} \frac{d|Y_{0}(\tau)|^{2}}{d\tau}.$$
(31)

As seen from (31), under these conditions the frequency scanning $\Delta\Omega_q$ is due to the dependence of the refractive index on the light intensity, and is transferred, as it were, when the components are transformed into one another, from each preceding component to the succeeding one in the sense that with sufficient accuracy $(\Delta \Omega_a \omega_r)$ ω_0) the final frequency of the preceding interval of continuous variation differs from the initial frequency of the preceding integral by an amount ω_0 . By way of illustration, the figure shows the total dependence of the instantaneous frequency on the time in a fixed section z. It is seen that in the typical case the values of the integrals of continuous variation of the frequency increase with increasing |q|, so that the frequency-variation integral of the last component of the stimulated Raman emission can constitute an appreciable fraction of the total interval of the continuous scanning $\Delta \Omega_{tot}$ over all the components (i.e., the sum of all the individual bands of the continuous frequency variation).

We present also an expression for $\Delta\Omega_q$ (in the absence of frequency deviations in the input pulses):

$$\Delta\Omega_{q}(\tau, z_{q} < z < z_{q-1}) \approx \frac{\delta_{-1}}{2\hbar} \left[\frac{q(q+1)}{|Y_{0}(\tau)|^{2}} \frac{d|Y_{0}(\tau)|^{2}}{d\tau} + \dot{q} \frac{d}{d\tau} \ln \left| \frac{Y_{0}(\tau)}{Y_{-1}(\tau)} \right|^{2} \right] ,$$
(32)

which follows from (27) and is valid under the conditions

$$\left|\operatorname{Im}_{\chi_{11}^{(q)}}\right| \gg \beta_{-1}^{(q)}, \quad |q| \ge \ln \left| \frac{\Delta k_q}{\chi_{12}^{(q)} Y_0^2(\tau)} \right|.$$
(33)

We see from (32) that the frequency scanning is possible also in the absence of the nonlinearity connected with the Kerr effect. An individual continuous frequency variation interval, corresponding to the q-th component of the stimulated Raman emission at a fixed value of zand a given waveform of the input pump pulses and of the first Stokes component is usually also enhanced with increasing |q|. We note at the same time that the location of these intervals on the frequency scale, as seen from (30) and (32), differs quite essentially from their location in the preceding case (cf. (30), (31)).

Formula (27) leads to two other consequences of practical interest. If frequency scanning is already present in one of the input pulses $(d\varphi_0(\tau)/d\tau \neq 0, \text{ or } d\varphi_{-1}(\tau)/d\tau)$ \neq 0) and the intensity in these pulses remains constant in time (with $|Y_{0q}|^2 < |Y_0|^2 < |Y_{0,q-1}|^2$), then, as seen from (27), frequency scanning of the q-th component of the stimulated Raman emission (i.e., scanning in the vicinity of the frequency ω_q) appears in a fixed section z ($z_q < z < z_{q-1}$). The scanning interval of the converted radiation exceeds in this case the initial interval by |q|times, i.e., an effect of "multiplication" of the scanning interval sets in, and makes it possible to broaden the initial interval by many times. This effect may be of practical importance also because frequency modulation of a very weak input Stokes pulse, with the pump pulse not modulated $(d\varphi_0(\tau)/d\tau=0)$, suffices for its realization, as can be seen from (27). Finally, if there are practically no frequency deviations in the initial pulses, but the duration of the initial pulse of the Stokes component is much less than the duration of the initial pump pulse, then the decisive term in (27) (see also (32)) may become the term containing $|Y_1(\tau)|$. In the latter case, as is readily seen, it becomes possible to control the scanning process in the waveguide with the aid of the amplitude envelope of the weak (Stokes) input pulse.

Thus, frequency scanning of the stimulated Raman emission components in an optical waveguide has distinguishing features that can be of practical interest. For example, the difference between the ladder scanning considered above and the customary one lies in the discrete-continuous variation of the frequency, and also in the fact that the position, on the frequency scale, of the continuous-scanning intervals does not depend significantly at all on the constants of the waveguide material and on the parameters of the input pulse and Stokes-radiation pulses; in the case of stimulated Raman emission it becomes possible to control in the waveguide the process of frequency scanning of an intense light field with the aid of an additional weak (Stokes) pulse, and also the possibility of the already discussed "multiplication" of the initial-scanning interval.

We note that the expressions given in this section are in general valid under the condition that the individual continuous-scanning intervals, roughly speaking, do not exceed noticably the width of the spontaneous Raman scattering line (a more accurate condition can be easily obtained with the aid of (23), by starting with the requirement that the stimulated Raman emission be quasistationary, $hT \gg 1$). Since these intervals in general increase with increasing number of the component |q|, this condition may be violated, for example, for the higher-order components. In this case it is necessary to start from the nonstationary Eq. (10). The corresponding analysis shows that in general "quenching" of the stimulated Raman emission is possible in this case, in the sense that the effective values of the corresponding quantities $\chi_{ij}^{(q)}$ become smaller.

We present a numerical example. We assume $\Delta k_q \sim 10 \text{ cm}^{-1}$,

$$\chi_{11}^{(q)} \approx |\chi_{12}^{(q)}| \sim 10^{-9} \text{ cgs esu}, \qquad \beta_l^{(q)} \sim 10^{-8} \text{ cgs esu},$$

 $(l=0,\pm 1)$, $|Y_0|^2_{\rm max} = 10^8$ cgs esu (corresponding at a typical value $\Lambda_q \sim 5 \times 10^{-4}$ cm to a pulse peak power $P \sim 3 \times 10^3$ W), $|Y_{-1}|^2 = 10^4$ cgs esu. We have here $\Delta z_q \sim 50$ cm, so that at $z=10^3$ cm the maximum value of |q| is ~ 20 ; the interval $\Delta\Omega_{\rm tot}$ (the sum of all the individual bands of the continuous scanning) is by virtue of (31), $\Delta\Omega_{\rm tot} \sim 10^3/\tau_p$ where τ_p is the duration of the input pump pulse. For example, at $\tau_p = 3 \times 10^{-10}$ sec we have $\Delta\Omega_{\rm tot} \sim 3 \times 10^{12}$ rad/sec. The total interval $\Delta\Omega_{\rm tot}$ of ladder scanning at a typical value $\omega_r \sim 10^{14}$ rad/sec exceeds 10^{15} rad/sec.

3. STIMULATED RAMAN EMISSION AT WEAK DISPERSION $n_{eff}^{0}(\omega)$

Assume that condition (21b) is satisfied. We consider Eqs. (15) with boundary condition (17) in the interval $0 < z < \Delta z_0$, where the value of Δz_0 is determined from the condition that the reactions of the first Stokes component and the first anti-Stokes components on the pump wave can be neglected (see, e.g., (40)). The solution of the corresponding linear system of equations for the amplitudes F_{-1} and F_1 of these components with boundary condition (17) is elementary:

$$F_{-1}(\tau, z) = \frac{Y_{-1}(\tau)}{\xi_1 - \xi_2} \left(\xi_1 e^{\lambda_2 z} - \xi_2 e^{\lambda_1 z} \right) \exp\left\{ i [\zeta + \beta_{-1}^{(0)} |Y_0(\tau)|^2] z \right\},$$

$$F_1(\tau, z) = \frac{\xi_1 \xi_2 Y_{-1}(\tau)}{(\xi_1 - \xi_2) Y_0^2(\tau)} \left(e^{\lambda_2 z} - e^{\lambda_1 z} \right) \exp\left\{ -i [\zeta + \beta_1^{(0)} |Y_0(\tau)|^2] z \right\}.$$
(34)

Here

I

$$\begin{split} \xi_{m} &= (\chi_{12}^{(0)})^{-1} [\lambda_{m} + i\zeta - \chi_{11}^{(0)} | Y_{0}(\tau) |^{2}], \quad m = 1, 2, \\ \lambda_{1,2} &= \frac{1}{2} \{ -s_{1} | Y_{0}(\tau) |^{2} \mp [(s_{1}^{2} + 4s_{2}) | Y_{0}(\tau) |^{4} + 4i (\chi_{22}^{(0)} - \chi_{11}^{(0)}) \zeta | Y_{0}(\tau) |^{2} - 4\zeta^{2}]^{\frac{1}{2}} \}, \end{split}$$



(35)

$$s_{1} = -(\chi_{11}^{(0)} + \chi_{22}^{(0)}), \quad s_{2} = \chi_{12}^{(0)} \chi_{21}^{(0)} - \chi_{11}^{(0)} \chi_{22}^{(0)}, \\ \zeta = -\frac{1}{2} (\Delta k_{0} + s_{3} | Y_{0}(\tau) |^{2}).$$

Expressions (35) for the characteristic exponents $\lambda_{1,2}$ (the real parts of which at fixed τ determine the gain with respect to z of the coupled system of Stokes and anti-Stokes waves in the waveguide) can be of interest for arbitrary ratios of the parameters contained in them, since the dispersion $n_{\rm eff}^{(0)}(\omega)$, depending on the profile of the refractive index of the waveguide, can be strong, weak, or intermediate. In particular, for specially chosen waveguide refractive-index profiles (see^[12]), the dispersion $r_{eff}^{(0)}(\omega)$ can be practically eliminated.

Let us dwell in greater detail on the case of weak dispersion, when

$$|\Delta k_{0}| \ll |s_{3}Y_{0}^{2}(\tau)|.$$
(36)

In this case the characteristic exponents $\lambda_{1,2}$ are proportional to the pump intensity:

$$\lambda_{i,2} = \frac{1}{2} |Y_0(\tau)|^2 \{ -s_i \mp [s_i^2 + 4s_2 + 2is_3(\chi_{i1}^{(0)} - \chi_{22}^{(0)}) - s_3^2]^{\frac{1}{2}} \},$$
(37)

and the corresponding proportionality coefficient is determined both by the susceptibilities $\chi_{11}^{(0)}$ and $\chi_{22}^{(0)}$, contained in s_1 and by the specific waveguide coefficient s_2 and the coefficient s_3 (which determines the degree of mismatch of the frequency scanning of the pump wave and the stimulated Raman emission components).

We assume also that

$$|s_1^2| \gg 4|s_2|, \quad 2|s_3(\chi_{11}^{(0)} - \chi_{22}^{(0)})|. \tag{38}$$

We then obtain from (37) the following expression for the gains of the considered waves:

$$\operatorname{Re} \lambda_{2} \approx \operatorname{Re} \frac{s_{2}}{s_{1}} |Y_{0}(\tau)|^{2}.$$
(39)

This is much less than the gain $\operatorname{Re}\chi_{11}^{(0)} |Y_0(\tau)|^2$ of the Stokes wave in the case of strong dispersion $n_{eff}^{(0)}(\omega)$ (see (23)), which in turn means a quenching of the stimulated Raman emission (or stimulated Raman scattering) in the waveguide.³⁾ By virtue of (39), the reactions of the considered waves on the pump wave sets in at

$$\widetilde{\Delta} z_0 \approx \left(\operatorname{Re} \frac{s_2}{s_1} |Y_0(\tau)|^2 \right)^{-1} \ln \left| \frac{Y_0(\tau)}{Y_{-1}(\tau)} \right|.$$
(40)

The guenching of the stimulated Raman emission in the waveguide can be used to increase the pump power transmitted through this waveguide without conversion into stimulated Raman emission components, and to increase the interval of continuous scanning of the pump frequency.

In conclusion we call also attention to the possibility of quenching the stimulated Raman emission in the case of strong dispersion $n_{eff}^{(0)}(\omega)$. As seen from (16) and (22), the quantity $\operatorname{Re}\chi_{11}^{(0)}|Y_0|^2$ itself decreases (in the limit, to zero) at large values of the ratio Λ_{-1}/Λ_0 of the scales of the localization of the eigenfunctions $\mathbf{g}_{-1}(\mathbf{r}_{\perp})$ and $\mathbf{g}_0(\mathbf{r}_{\perp})$. It is therefore clear that if the refractiveindex profile at the pump frequency is of the waveguide type $(\Delta n_0 > 0)$, and is not of the waveguide type at the first Stokes frequency ($\Delta n_0 \leq 0$), as is realized at Δn_0 $\sim 10^{-3}$, then the stimulated Raman emission will be completely quenched.

¹⁾It is known that phase modulation of the field of a light beam is observed also in an inhomogeneous nonlinear medium. In a homogeneous medium, however, this modulation differs substantially from modulation in a single-mode waveguide, since the redistribution of the energy over the cross section of a beam propagating in a homogeneous medium makes it practically impossible to produce in the medium scanning, i.e., a monotonic variation of the frequency in the interval $\Delta\Omega \gg 1/\tau$, where τ is the duration of this change. A similar limitation arises also when modes interfere in a multimode waveguide.

²⁾If the assumption $h_1 \ll \omega_r$ made in the preceding section is not satisfied, i.e., if $h_1 \gtrsim \omega_r$, then it can be verified that the form of the solution (23) remains in force also in this case. The differences reduce only to somewhat modified values of the constants that enter in the solution.

- ³⁾In the theory of stimulated Raman scattering in a homogeneous medium (i.e., outside the waveguides) it is known that the gain of the coupled Stokes and anti-Stokes waves can vanish. This occurs when the phase synchronism condition is satisfied. However, a gain of pratically the same value as for the Stokes wave occurs upon deviation from the phasesynchronism condition, which is also always realized in stimulated Raman scattering in three-dimensional homogeneous volumes (see ^[14,15]). There is therefore no quenching of stimulated Raman emission in a homogeneous medium.
- ¹I. E. Ippen, C. V. Shank, and T. K. Gustafson, Appl. Phys. Lett. 24, 190 (1974).
- ²V. N. Lugovoĭ, Pis'ma Zh. Eksp. Teor. Fiz. 22, 416 (1975) [JETP Lett. 22, 200 (1975)].
- ³F. Bloch, Phys. Rev. 70, 460 (1946).
- ⁴A. E. Siegman and R. J. Morris, Phys. Rev. Lett. 2, 302 (1959).
- ⁵E. B. Treacy, Phys. Lett. 27A, 421 (1968); E. B. Treacy
- and A. J. DeMaria, Phys. Lett. 29A, 369 (1969).
- ⁶M. M. T. Loy, Phys. Rev. Lett. 32, 814 (1974).
- ⁷S. M. Hamadani, A. T. Mattick, N. A. Kurnit, and A. Javan, Appl. Phys. Lett. 27, 21 (1975).
- ⁸V. N. Lugovoi and V. N. Strel'tsov, Pis'ma Zh. Eksp. Teor. Fiz. 21, 442 (1975) [JETP Lett. 21, 202 (1975)].
- ⁹E. P. Ippen, Appl. Phys. Lett. 16, 303 (1970).
- ¹⁰R. H. Stolen and E. P. Ippen, Appl. Phys. Lett. 22, 276 (1973).
- ¹¹E. P. Ippen and R. H. Stolen, Appl. Phys. Lett. 21, 539 (1972).
- ¹²S. E. Miller, E. A. D. Markatili, and T. Li, Proc. IEEE 61, 46 (1973) [Transl. into Russian].
- ¹³R. G. Smith, Appl. Opt. 11, 2489 (1972).
- ¹⁴V. N. Lugovoi, Vvedenie v teoriyu vynuzhdennogo kombinatsionnogo rasseyaniya (Introduction to the Theory of Stimulated Raman Scattering), Nauka, 1968.
- ¹⁵N. Bloembergen, Nonlinear Optics, Benjamin, 1965.
- ¹⁶V. N. Lubovoi, Zh. Eksp. Teor. Fiz. 56, 683 (1969) [Sov. Phys. JETP 29, 374 (1969)].
- ¹⁷V. N. Lugovol, Opt. Spektrosk. **27**, 649 (1969). ¹⁸V. N. Lugovol, *ibid.*, 828.
- ¹⁹V. N. Loguvoi, Zh. Eksp. Teor. Fiz. 68, 2055 (1975) [Sov. Phys. JETP 41, 1028 (1975)].
- ²⁰D. Marcuse, Light Transmission Optics, Van Nostrand, 1972 [Russ. Transl., Mir. 1974].
- Translated by J. G. Adashko