Investigation of cooperative Raman scattering of light

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A study was made of the cooperative Raman scattering by the $Q_{01}(1)$ line of molecular hydrogen. It was found that in the case of coherent transient excitation there was a delay between a Raman scattering pulse and the maximum of a pump pulse. The delay time was inversely proportional to the H_2 pressure and to the power density of laser radiation. Intensity pulsations were observed in the scattered radiation. An analysis was made of the influence of the four-wave parametric interaction on the cooperative scattering process. The experimental results were found to be in qualitative agreement with the theory.

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1. INTRODUCTION

Several experimental studies have been made recently of the phenomenon of cooperative emission (see, for example, Refs. 1–4) predicted by Dicke[5] in 1954. It was demonstrated in Refs. 6–8 that similar phenomenon should occur also in the scattering. When light is scattered, a correlation should appear between molecules and this should alter drastically the characteristics of the scattered radiation compared with the scattering by isolated molecules. In particular, a light pulse scattered by correlated molecules should exhibit a delay (t_0) relative to the leading edge of a step-like exciting pulse[7,8] and a substructure consisting of a series of short spikes[8] of duration $\tau_0 < t_0$.

The first observation of this cooperative Raman scattering (CRS) was reported in Ref. 9. We shall give the results of an experimental study of this effect.

In addition to general features characteristic of any cooperative process, cooperative scattering has a number of special properties due to the presence of fields of different frequencies participating in the scattering process. For example, in the experiments described below the scattering is simultaneous into two spectral components: Stokes and anti-Stokes. The four-wave interaction between the components of the scattering and the exciting field has a considerable influence on the scattering process. A theoretical analysis of this interaction is given in Sec. 5 below.

2. ESTIMATE OF CONDITIONS FOR THE OBSERVATION OF COOPERATIVE RAMAN SCATTERING

The cooperative scattering can be observed only in the case of fast coherent excitation of a medium: the duration of the leading edge of the exciting pulses t_l should be less than the polarization relaxation times T_2 and T_2^*, where T_2 is the relaxation time due to collisions and T_2^* is due to the thermal motion of the molecules. The coherence time of a pulse should be sufficiently long. Under these conditions the polarization of a medium at the frequency of the scattered radiation rises more slowly than the field of the exciting pulse and it lags behind the leading edge of the latter pulse by the time needed to establish a correlation between the scattering molecules. This gives rise to a delay between the leading edges of the exciting and scattered pulses.[7,8] A "photon avalanche" initiating the correlation between molecules develops if the growth increment of any mode in the avalanche exceeds sufficiently the loss factor at the frequency of the scattered radiation in this mode for a superscattering state to appear in a time shorter than the relaxation time T_1 of the level population. In an extended medium the propagation effects are important and the scattering process is spatially inhomogeneous. In this case the scattered radiation consists of a series of short pulses separated by intervals comparable with their duration.[8] The experimental condition for the observation of the CRS should be such that the characteristic duration of a scattered-radiation pulse τ_0 is less than or comparable with the polarization relaxation time T_2. Thus, for a given density of particles, the CRS has a power threshold governed by the conditions $t_0 \lesssim T_1$ and $\tau_0 \lesssim T_2, T_2^*$.

A convenient object for the investigation of the CRS may be molecular hydrogen with large relaxation times T_1 and T_2. In the CRS case the time T_1 is governed by the vibrational relaxation and its value is[10] 10^{-3} sec·atm (all the relaxation times are given for room temperature). According to Ref. 11, the relaxation time T_2 for the vibrational-rotational transition $Q_{01}(1)$ in the hydrogen molecule represents 7.5×10^{-9} sec·atm. The translational relaxation time of the polarization for the same transition in low-pressure hydrogen is $T_2^* = 2(|\mathbf{k}-\mathbf{k}_s|\bar{v})^{-1} = 5\cdot10^{-10}$ sec for $\mathbf{k}\uparrow\uparrow\mathbf{k}_s (k = 2\pi/\lambda, k_s = 2\pi/\lambda_s$; λ and λ_s are the wavelengths of the exciting and scattered radiation, and $\bar{v}$ is the average thermal velocity). When the gas density is increased to 2 amagat, the mean free path of molecules becomes less than $(|\mathbf{k}-\mathbf{k}_s|)^{-1}$, so that the line width of the spontaneous Raman scattering decreases (Dicke effect). According to Ref. 12, the width of the spontaneous Raman line at $p = 2$ atm corresponds to a relaxation time of 2×10^{-9} sec.

In the case when the CRS spectrum consists of just one Stokes component, the values of the delay time and of a typical duration of a typical duration of a scattered-radiation pulse considered are given in the one-dimensional approximation by the expressions (Ref. 8)[1)]

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$$\tau_0 = 2(\beta z)^{-1} \ln \frac{\pi}{2\theta_0}, \quad t_0 = \frac{\tau_0}{4} \ln \frac{\pi}{2\theta_0},$$
$$\beta = \frac{8\pi^2 n_0 I}{c\hbar k k_s^2} \frac{d\sigma}{d\Omega}, \quad \theta_0 = (\lambda_s n_0 z^2)^{-1/2}. \qquad (1)$$

Here $d\sigma/d\Omega$ is the differential cross section for the spontaneous Raman scattering; I is the power density of the exciting radiation; n_0 is the density of the molecules; z is the length of the scattering region.

We shall estimate the times τ_0 and t_0 and the power density I needed for the observation of the CRS of ruby laser radiation due to the $Q_{01}(1)$ transition in the hydrogen molecule when the scattered radiation is recorded in the direction of propagation of the exciting field. In our case, n_0 denotes the density of the H_2 molecules in the state $v=0$, $J=1$. At room temperature this state is occupied by 66.7% of the molecules. We shall assume that $n_0 = 4.8 \times 10^{19}$ cm^{-3} and $z = 5$ cm. Substituting in Eq. (1) the numerical values for hydrogen $d\sigma/d\Omega = 2.4 \cdot 10^{-31}$ cm$^2 \cdot$ sr$^{-1} \cdot$ mole^{-1} (Ref. 13), $\lambda = 694.3$ nm, and $\lambda_s = 976$ nm we find that $\tau_0 \approx 1.5$ nsec $< T_2, T_2^*$, when $I \gtrsim 7$ GW/cm^2. Under these conditions we have $t_0 \approx 7.5$ nsec $< T_1$. Consequently, we can observe the CRS in hydrogen if the exciting pulses have a leading edge $t_l \approx 10^{-9}$ sec, duration $10^{-8} - 10^{-7}$ sec, and power density $10^9 - 10^{10}$ W/cm^2. It is then possible to observe the effect in question in the part of the spectrum $\lambda_s < 1\mu$ convenient for measurements.

3. APPARATUS

We used the apparatus shown schematically in Fig. 1. A ruby laser 1 emitting a single longitudinal mode generated a pulse of 100 nsec duration at midamplitude with a spectral width $\Delta\nu \leq 10^{-3}$ cm^{-1}. The leading edge of such a pulse was shortened by an electro-optic switch 3. This switch was triggered by a spark gap 2 with laser ignition. After passing through this switch, the duration of the leading edge measured between $0.1 I_{\max}$ and $0.9 I_{\max}$ was varied from 2 to 15 nsec by altering the capacitance shunting the electro-optic cell 3.

The pulse then passed through an amplifier system 4: its duration at the output of the system was $t_p \approx 50$ nsec at midamplitude and its energy was $W \approx 0.5$ J. The divergence of the radiation was reduced by placing a spatial filter between the amplifying stages; this filter consisted of lenses 5 and 7 and a stop 6. The laser radiation was focused by a lens 10 ($F = 50$ cm) into a cell 11 filled with hydrogen (of technical purity) at room temperature. The length of the cell was 30 cm. The influence of the light reflected from the cell windows on the process of scattering in hydrogen was eliminated by inclining the windows at an angle of 5° to the axis. The poor quality of the last amplifying stage made the distribution of the intensity at the focus of the lens 10 inhomogeneous. The nature of the distribution depended on the adjustment of the last amplifier. The results reported below were obtained for the case when the radiation was focused in three spots $d = 0.2$ mm in diameter separated by distances of about 0.5 mm. The results were not affected in the qualitative sense by changes in the structure of these spots in the focal plane.

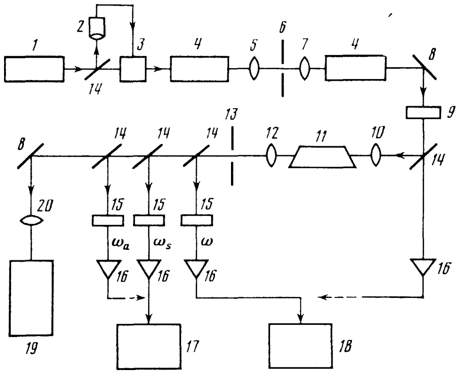

FIG. 1. Schematic diagram of the apparatus: 1) ruby laser; 2) spark gap with laser ignition; 3) electro-optic switch; 4) ruby amplifier system; 5), 7), 10), 12), 20) lenses; 6) stop; 8) mirrors; 9) neutral optical filters; 11) cell with hydrogen; 13) stop; 14) glass plate; 15) optical filters; 16) coaxial photocells; 17), 18) oscilloscopes; 19) DFS-8 spectrograph.

The radiation emerging from the cell was focused by a lens with $F = 27$ cm on a stop 13 with an aperture 0.9 mm in diameter, was made monochromatic by filters 15, and was then recorded by coaxial photocells 16 and two I2-7 oscilloscopes. The relationship between the shapes of the exciting and scattered pulses was determined by simultaneous measurements in two channels. One channel (oscilloscope) received a scattered-radiation pulse at the Stokes or anti-Stokes ($\lambda_a = 538.8$ nm) frequency, and the other received a pulse of the incident or transmitted (by the cell) laser radiation.

4. EXPERIMENTAL RESULTS

When a certain threshold power density was reached, the cell with hydrogen emitted intense Raman scattering pulses. The threshold power density decreased on increase in the hydrogen pressure. When the laser pulse energy was $W = 0.35$ J, intense Raman scattering pulses were observed at pressures $p \geq 2.6$ atm. Our experiments were carried out in the pressure range from 2.6 to 5 atm; at $p \geq 5.5$ atm we observed optical breakdown in hydrogen. The laser radiation was polarized linearly. The spectrum of the scattered radiation consisted of the Stokes I_s and anti-Stokes I_a lines $Q_{01}(1)$ ($\Delta\nu = 4155.2$ cm^{-1} – Ref. 12). The Stokes amd anti-Stokes scattered light had the same linear polarization as the exciting field.

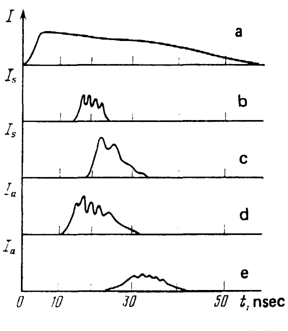

FIG. 2. Oscillograms of the exciting (a), Stokes (b, c), and anti-Stokes (d, e) pulses. The oscillograms b–e were obtained for different shots; $p = 3$ atm, $W = 0.35$ J, $t_l = 3$ nsec.

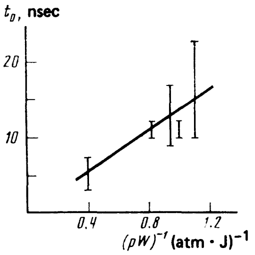

FIG. 3. Dependence of the delay time between the maxima of the exciting and CRS pulses on the H_2 pressure and laser energy; t_l = 3 nsec.

One of the important experimental results was the detection of a considerable delay of the scattered $I_{s,a}$ pulses relative to the maximum of the exciting pulse I. Figure 2 shows oscillograms of the I, I_s, and I_a pulses. A comparison of the oscillograms obtained for different shots demonstrated that the average delay times were the same for the Stokes and anti-Stokes pulses. This observation, and the spatial distribution of the I_a radiation reported below, indicated that the anti-Stokes scattering resulted from a four-photon parameteric process.

Figure 3 shows the dependence of the delay time on the product of the hydrogen pressure and the energy of the laser pulses. It is clear from this figure that the delay time decreased on increase in the gas pressure and also on increase in the energy (i.e., power density) of the exciting pulses, and it was inversely proportional to the product pI.

These results were obtained when hydrogen was excited with pulses with a steep leading edge of duration t_l = 2–3 nsec. When the leading edge of the laser pulses became longer, the delay decreased. For example, for $p=5$ atm, $W=0.2$ J, and $t_l=2$ nsec the delay between the maxima of the exciting and scattered pulses was $t_0 \approx 11$ nsec, but for $t_l=6$ nsec, the delay decreased to 6 nsec. Further increase in the leading edge of the laser pulses to 15 nsec suppressed the delay between the maxima of the exciting and scattered pulses. The experiments described below were carried out (unless otherwise stated) using pulses with t_l =2–3 nsec.

The spatial distribution of the radiation at the anti-Stokes frequency was studied in the focal plane of the lens 12 ($F=27$ cm). The I_a radiation was focused into several spots ~1 mm in diameter. The diameter of the spots corresponded approximately to the emission of a single mode from the active volume: $\lambda_a f/d \approx 0.7$ mm ($d=0.2$ mm). The number of positions of the spots varied from one shot to another. Near the threshold of appearance of the anti-Stokes scattering the spots were observed along a generator of a cone of angle 10–15 mrad relative to the axis. When the energy of the laser pulses or the gas pressure was increased so that the power of I_a increased, the scattered radiation filled the whole cone.

The stop 13 with an aperture 0.9 mm in diameter placed in the focal plane of the lens 12 selected one or two spots from the radiation, i.e., one or two diffraction modes. In this case a scattered-radiation pulse consisted of a series of short spikes separated by an interval which varied from shot to shot ranging from 1.5 to 5 nsec at 2.7 atm. This situation corresponded to the oscillograms shown in Figs. 2b–2e. The apparatus used in these experiments made it possible to record the amplitude modulation of a pulse to a depth at least 10%. Within this accuracy, the shape of the exciting pulses transmitted by the cell was similar to the shape of the incident pulses in the case when the total flux was recorded or when part of it passed through the stop was detected: there were usually no pulsations or dips in the envelope. When hydrogen was excited with a pulse characterized by a long leading edge ($t_l=15$ nsec) a dip was observed in the envelope of the exciting pulse after transmission through the cell and this dip coincided in time with the stimulated Raman scattering pulse (the H_2 pressure was $p=4$ atm).

The depth and period of the modulation of the Raman-scattered pulse, the position of the strongest spike, and the delay of the scattered radiation varied from shot to shot. The scatter of the delay in the case when the total flux was recorded (i.e., when the stop was removed) was considerable (Fig. 3). When the stop 13 was introduced, the scatter increased. In the latter case there were sometimes weak scattered pulses with delays up to 30 nsec. The strongest recorded modulation was about 50%. Some of the scattered pulses had a smooth envelope with a duration 10–15 nsec.

When the whole forward-scattered flux was recorded, a pulse was usually smooth but in some cases there was a substructure with a period of 1.5–5 nsec. At a pressure of $p=2.7$ atm and for a laser energy of $W=0.35$ J the duration of the I_s and I_a pulses was 10–15 nsec when the total flux was recorded. An increase in the gas density and in the laser power increased the duration of the Raman-scattered pulses to 30–40 nsec ($p=5$ atm, $W=0.5$ J.

5. THEORETICAL ANALYSIS OF THE INFLUENCE OF A FOUR-WAVE INTERACTION OF FIELDS

It is known from Ref. 14 that in the case of stimulated Raman scattering under static conditions a four-wave interaction of the Stokes and anti-Stokes components with the exciting field results (in the case of a weak dispersion of the nonlinear polarizabilities) in a strong anisotropy of the angular distribution of the anti-Stokes component; then, for either component there is no gain for the angles of spatial phase matching given by the relationship $2\mathbf{k}=\mathbf{k}_s+\mathbf{k}_a$. In the case of the Stokes component, the gain increases on increase in the parameter $q=2k_z-k_{sz}-k_{az}$, whereas for the anti-Stokes component there is an optimal scattering angle

$$\vartheta_{st}=(2g_s/k)^{1/2}, \qquad g_s=4\pi\omega_s^2\chi_s''|E|^2/c^2k_sz, \tag{2}$$

where ω_s and k_{sz} are the frequency and projection of the wave vector of the Stokes component; χ_s'' is the imaginary part of the nonlinear polarizability at the Stokes frequency.

Reduction in the duration of the interaction between the molecules and the fields, i.e., under transient conditions, does not alter qualitatively the angular distribution of the radiation. A basically new feature is the appearance of time modulation of the scattering phases and amplitudes.

We shall consider the influence of this four-wave interaction in the case of constant populations (linear solution), which corresponds to the initial stage of the scattering process, when the changes in the populations can still be ignored.

We shall consider the process of CRS of a pulse traveling along the z axis and characterized by a step-like shape at the boundary of the active medium. We shall describe the exciting field and the scattered fields at the Stokes and anti-Stokes frequencies by

$$E=\frac{1}{2}\mathscr{E}(z,t)\exp i(\omega t-kz)+\frac{1}{2}\mathscr{E}_s(z,t)\exp i(\omega_s t-k_s z)$$
$$+\frac{1}{2}\mathscr{E}_a(z,t)\exp i(\omega_a t-k_a z)+\text{ c.c.},\qquad(3)$$

where $\mathscr{E}$, $\mathscr{E}_a$, and $\mathscr{E}_s$ are the envelopes of the amplitudes of the exciting, anti-Stokes, and Stokes fields, respectively. The scattered fields propagate at certain angles to the z axis and initially their amplitudes are zero.

We shall describe the CRS process (in the approximation of a constant exciting field) by the following system of equations for the elements of the $\hat{\rho}$ matrix and amplitudes of the scattered fields:

$$\frac{\partial n}{\partial t}=4\operatorname{Re} i\left\{\left[\frac{1}{2\hbar}D_{21}{}^{s}\mathscr{E}^{*}\mathscr{E}_s+\frac{1}{2\hbar}D_{21}\mathscr{E}\mathscr{E}_a{}^{*}\exp(2iqz)\right]\rho_{12}\right\},\qquad(4a)$$

$$\frac{\partial}{\partial t}\rho_{12}=-\frac{i}{2\hbar}[D_{12}{}^{s}\mathscr{E}\mathscr{E}_s{}^{*}+D_{12}{}^{a}\mathscr{E}^{*}\mathscr{E}_a\exp(2iqz)]n,\qquad(4b)$$

$$\left(v_s\frac{\partial}{\partial z}+\frac{\partial}{\partial t}\right)\mathscr{E}_s=-4\pi i\omega_s D_{12}{}^{s}\rho_{21}\mathscr{E}.\qquad(4c)$$

$$\left(v_a\frac{\partial}{\partial z}+\frac{\partial}{\partial t}\right)\mathscr{E}_a=-4\pi i\omega_a D_{21}{}^{a}\rho_{12}\mathscr{E};\qquad(4d)$$

$$D_{21}^{s,a}=\frac{1}{2\hbar}\sum_m d_{2m}d_{m1}\left(\frac{1}{\pm\omega_{s,a}+\omega_{m1}}+\frac{1}{\mp\omega_{s,a}+\omega_{m2}}\right),\quad n=\rho_{11}-\rho_{22}.$$

The indices 1 and 2 correspond to the initial and final states.

We shall assume that the projections of the group velocities of the Stokes and anti-Stokes components are equal $v_s=v_a\equiv v$. We shall also assume that the difference between the populations is constant in time and we shall integrate Eq. (4b) with respect to time and then substitute the result in Eqs. (4c) and (4d):

$$\begin{aligned}\frac{\partial^2\theta_s}{\partial z\,\partial\tau}&=\beta_s\theta_s+\beta_s\theta_a e^{2iqz},\\ \frac{\partial^2\theta_a}{\partial z\,\partial\tau}&=-\beta_a\theta_a-\beta_a\theta_s e^{-2iqz},\end{aligned}\qquad(5)$$

where

$$\theta_{s,a}=\theta_{0s,a}+\frac{D_{12}^{s,a}}{\hbar}\int_0^t\mathscr{E}^{*}\mathscr{E}_{s,a}\,dt',\quad\tau=t-z/v,$$

$$\beta_{s,a}=2\pi\omega_{s,a}|D_{12}^{s,a}|^2|\mathscr{E}|^2\,n_0/v\hbar,$$

and $n_0\theta_{0s,a}$ is the initial polarization simulating spontaneous noncoherent Raman scattering.

The first terms on the right-hand sides of the equations in the system (5) represent the Raman scattering, whereas the second terms are due to the four-wave interaction discussed above.

The case of phase matching, $q=0$, is discussed in Ref. 8, where it is shown that if $\beta_s\neq\beta_a$, the problem reduces to the CRS into a single component. Then, the equation should be modified by replacing the combined matrix element D_{12}^{s} with an effective matrix element $D_{12}^{s}(1-|D_{12}^{a}|^2\omega_a/|D_{12}^{s}|^2\omega_s)$. If the frequency dispersion of the terms $|D_{12}^{a,s}|^2\omega_{a,s}$ is negligible, the effective matrix element vanishes, i.e., there is no scattering.

Further analysis of the system (5) will be made on the assumption that the phase-matching condition is not obeyed, $q\neq 0$, and that the frequency dispersion of the quantities $\beta_{s,a}$ can be ignored. Under these assumptions, the equations in the system (5) have the following integral of motion:

$$\frac{\partial\theta_s}{\partial z}+\frac{\partial\theta_a}{\partial z}e^{2iqz}=C(z).\qquad(6)$$

We shall now consider the spatially inhomogeneous initial conditions

$$\frac{\partial\theta_s}{\partial z}=\frac{\partial\theta_a}{\partial z}=0.\qquad(7)$$

Using the integral of motion (6) and applying the conditions (7), we obtain the following equation:

$$\frac{\partial^3\theta_{s,a}}{\partial z^2\,\partial\tau}\mp 2iq\frac{\partial^2\theta_{s,a}}{\partial z\,\partial\tau}=-2iq\beta\theta_{s,a}.\qquad(8)$$

We shall analyze the case of large values of q, when the terms proportional to q are much greater than the terms containing the third derivative, so that the latter can be ignored. The restrictions on the physical parameters corresponding to these conditions will be specified later. We thus obtain equations which are identical with the linearized equations considered in Ref. 8 and describing the process of CRS into a single component:

$$\frac{\partial^2\theta_{s,a}}{\partial z\,\partial\tau}=\pm\beta\theta_{s,a}.\qquad(9)$$

According to Ref. 8 the asymptotic solutions for large values of η in Eq. (9) is

$$\theta_s=\frac{\theta_{0s}e^{\eta}}{(2\pi\eta)^{1/2}},\quad\theta_a=\theta_{0a}\left(\frac{2}{\pi\eta}\right)^{1/2}\cos\left(\eta-\frac{\pi}{4}\right),\qquad(10)$$

where $\eta=2(\beta z\tau)^{1/2}$. We can see from the solutions given by Eq. (10) that, for a fixed value of z, the Stokes component rises exponentially, whereas the anti-Stokes component oscillates and decays with time.

Using the solutions (10), we obtain the condition for dropping the terms with the third derivative in Eq. (8):

$$\eta<|q|z.\qquad(11)$$

Since the parameter η contains the time, it follows that for a fixed η the inequality (11) is obeyed only for time intervals shorter than

$$\tau_1=q^2z/\beta.\qquad(12)$$

At times shorter than τ_1 we can ignore the influence of the four-wave interaction.

We shall now turn to the case when the terms with a mixed second-order derivative can be stopped from Eq. (8). As above, the restricting condition can be obtained from solutions of the following equation:

$$\frac{\partial^3\theta_{s,a}}{\partial z^2\,\partial\tau}=-2iq\beta\theta_{s,a}. \tag{13}$$

Equation (13) is satisfied by a class of self-similar solutions with a variable $\zeta=3(\beta z^2\tau q)^{1/3}$, for which Eq. (13) is of the form

$$2\theta_{s,a}'''+\frac{3}{\zeta}\theta_{s,a}''-\frac{1}{\zeta^2}\theta_{s,a}'+i\theta_{s,a}=0. \tag{14}$$

Substituting in Eq. (8) the solution dependent on the variable ζ, we can easily show that the condition for neglecting the terms with the second derivative reduces to an inequality which is the reverse of that given by Eq. (11) and which shows that the four-wave interaction is important for time intervals $\tau>\tau_1$.

We shall seek an asymptotic solution of Eq. (14) for large values of ζ in the form

$$\theta_{s,a}=F(\zeta)e^{\lambda\zeta}, \tag{15}$$

where $F(\zeta)$ is a function slow compared with the exponential factor. In the first approximation with respect to the small parameter $1/\zeta$ we have

$$\theta_{s,a}=\sum_{\nu=1}^{3} b_\nu^{s,a}e^{\lambda_\nu\zeta}\zeta^{-1/2}, \tag{16}$$

where $b_\nu^{s,a}$ are the coefficients governed by the condition for the matching of the solutions (10) and (16), and $\lambda_\nu^3=-i/2$.

We shall now write down the values of the roots λ_ν:

$$\lambda_1=2^{-1/3}i,\quad \lambda_{2,3}=2^{-1/3}(-i/2\mp\sqrt{3}/2). \tag{17}$$

All the roots λ_ν have an imaginary component and this corresponds to phase modulation. The imaginary components of the roots λ_1 and $\lambda_{2,3}$ are different so that the intensity of the scattered radiation proportional to $|\partial\theta_{s,a}/\partial\tau|^2$ is time-modulated. It should be noted that for $q=0$ the right-hand side of Eq. (13) vanishes, i.e., there is no scattering into the phase-matched angle. The argument of the growing exponential function in Eq. (16) increases on increase in q. The maximum value of q at which both components increase can be estimated from the equality $q_{\max}z=\eta$. It follows from the solutions (10) and (16) that, for fixed z and τ, there is an optimal value of $|q|$, when the intensity of the anti-Stokes component is maximal. In fact, an increase in $|q|$ from 0 to $q_{\max}$ causes the intensity of the anti-Stokes scattering to increase in accordance with Eq. (16), whereas for $|q|>q_{\max}$ it follows from Eq. (10) that the exponential growth changes to damped oscillations. This dependence on the parameter q corresponds to the existence of an optimal scattering angle for the anti-Stokes component. The value of this angle and the sharpness of the maximum (in the approximation of weak dispersion of the refractive index) are given by

$$\vartheta_{\text{opt}}=2(\beta\tau/k^2z)^{1/4},\qquad \delta\vartheta_{\text{opt}}\approx\vartheta_{\text{opt}}/2; \tag{18}$$

ϑ_{opt} is related to the optimal angle for the steady-state case [Eq. (2)] as follows:

$$\vartheta_{\text{st}}=\vartheta_{\text{opt}}^2(2\pi T_2z/\tau\lambda)^{1/2}. \tag{19}$$

For $\lambda=0.7\mu$, $z=5$ cm, $\tau=5T_2$, and $\vartheta_{\text{opt}}>3\times10^{-3}$ rad, the optimal angle for the steady-state case is always greater than the corresponding angle for the transient scattering.

We shall now discuss qualitatively the essentially nonlinear situation when the difference between the populations changes in the process of scattering. As shown above, the scattering process begins from the growth of the Stokes component and then, because of the four-wave interaction discussed above, the anti-Stokes component also begins to rise. The Stokes component is stronger than the anti-Stokes component and the molecules go over from the initial to the final state. However, when the system is inverted, the Stokes and anti-Stokes component exchange their roles. Consequently, the scattering process becomes periodic and this is followed by a transition from the initial to the final state and back again. This situation is described in Ref. 7 ignoring the propagation effects. The delay time t_0 relative to the leading edge of the exciting pulses is governed primarily by the Stokes scattering. The four-wave interaction reduces the rate of rise to the Stokes component and, therefore, it increases the delay time t_0.

6. DISCUSSION

Characteristic features of the CRS are the delay of the scattered radiation relative to the leading edge of the exciting pulses, and also pulsations of the intensity of the scattered light. These pulsations are accompanied by those of the populations. However, as shown in Sec. 5, the four-wave interaction of the fields may be responsible for pulsations of the intensity when the populations are constant. Then, the anti-Stokes scattering occurs along a certain optimal angle. We shall now use Eq. (18) to estimate the maximum value of this optimal angle. We shall assume that the time is equal to the delay time t_0 of the cooperative avalanche, given by Eq. (1), so that at this moment all the molecules go over to the state 2 and, obviously, the anti-Stokes and Stokes components exchange their roles. At $\tau=t_0$, the optimal angle is given by the expression

$$\vartheta_{\text{opt}}=\left[\frac{2\ln(\pi/2\theta_0)}{zk}\right]^{1/2}.$$

For $n_0=4.8\times10^{19}$ cm^{-3} and $z=5$ cm, the optimal angle is 10 mrad and it is equal to the experimental value.

We shall now estimate the period of the oscillations resulting from the four-wave interaction. Using Eqs. (16) and (17), we find that the oscillation period T is given by

$$\operatorname{Im}(\lambda_1-\lambda_{2,3})\delta\zeta=2\pi, \tag{20}$$

$$\delta\zeta=3(\beta z^2q)^{1/3}[(\tau+T)^{1/3}-\tau^{1/3}]\approx(\beta z^2q)^{1/3}\frac{T}{\tau^{2/3}}. \tag{21}$$

Equations (20) and (21) yield the following expression for the oscillation period:

$$T=\frac{2^{7/3}\pi\tau^{2/3}}{(\beta z^2q)^{1/3}}. \tag{22}$$

Substituting in Eq. (22) the maximum value of q and the time equal to the delay time t_0, we find that the period is

$$T\approx\tau_0, \tag{23}$$

where τ_0 is given by Eq. (1) and determines the oscillation period of the cooperative scattering intensity. It is clear from Eq. (23) that the period of the oscillations resulting from the four-wave interaction is of the order of the duration of the characteristic oscillations of the CRS intensity.

The relative depth of oscillations in each separate diffraction mode decreases exponentially with time. However, in view of the phase modulation, the carrier frequencies of different modes may differ and when the scattered radiation is stopped down, the stop may transmit radiation of different modes, giving rise to intensity beats whose relative contrast does not decrease with time. The beat period may then even be greater than the time T_2.

We shall now consider the effects due to the transfer of a considerable proportion of the energy of the exciting field to the scattering components, when the approximation of a constant exciting field is no longer valid. In Ref. 15, we investigated theoretically and experimentally these regimes in the case when the number of molecules transferred from the initial to the final state is negligible. In this case the intensities of the exciting and scattered fields exhibit pulsations and the minimum of the exciting field intensity corresponds to a maximum of the scattered field.

The opposite case was realized in our experiments. There were no pulsations in the intensity of the exciting field and then, judging by the scattering intensity, the redistribution of the populations of the initial and final states was considerable. According to Ref. 8, the condition of a constant exciting field imposes restrictions on the length of the medium. Using the solution given in Ref. 8 [Eqs. (4a) and (4b)], we can obtain a criterion of validity of the approximation of a constant exciting field which is more rigorous than that given in Ref. 8. In the case of scattering into a single component, this criterion is of the form

$$z\leqslant\alpha^{-1}\ln(\pi/2\theta_0), \tag{24}$$

where

$$\alpha=4\pi k_s n_0 D_s;\quad D_s=\left(\frac{d\sigma}{d\Omega}\frac{1}{4kk_s^3}\right)^{1/2}.$$

Substituting in Eq. (24) the parameters $n_0=4.8\times10^{19}$ cm^{-3} and $d\sigma/d\Omega=2.4\cdot10^{-31}$ cm$^2\cdot$sr$^{-1}\cdot$mole^{-1}, we find that $z\leqslant10$ cm. In our experiments the length of the region where the CRS appeared effectively (corresponding to a construction in the laser beam) was less than 10 cm.

Our experiments thus showed that when hydrogen was excited by pulses with a steep leading edge, the Raman scattering appeared after a delay and was of pulsating nature. The delay time agreed, to within a factor of 2, with the predictions of the theory of Ref. 8. The dependence of the delay time on the gas pressure and power density of laser radiation was linear both in the theory and in the experiments. The pulsation period estimated from Eq. (1) was 1.5 nsec and that found experimentally ranged from 1.5 to 5 nsec. A theoretical analysis showed that a transient coherent four-wave interaction, which occurred under experimental conditions, altered the time and spatial characteristics of the scattering. Pulsations with a period exceeding the time T_2 observed for some of the pulses were clearly associated with this interaction.

We shall conclude by considering the relationship between the CRS and other Raman scattering regimes in which the intermolecular interaction also plays the dominant role. They include the photon echo in the Raman scattering[16] and solitons in the stimulated Raman scattering.[16,17,18] The most important difference between the CRS and the above regimes is as follows. In the observation of the photon echo and solitons in the stimulated Raman scattering it is necessary to illuminate a medium with pulses of duration shorter than T_2 at the exciting field frequency and at the Raman frequency. However, in the CRS case it is sufficient that the leading edge of the exciting pulses should be shorter than T_2, although the duration of these pulses can be as long as we please; then a coherent pulse forms spontaneously at the Raman frequency.

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1) The formula for the parameter θ_0 given in Ref. 8 is valid in the case of scattering into a solid angle $\Omega\sim1$. If the diameter of the active volume is much less than its length ($\Omega\ll1$), the formula for θ_0 is identical with that given by Eq. (1) above.

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