

Waveguide SRS due to vibrational excitation of molecules

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Bright conical radiation has been experimentally observed in the angular spectrum of the first Stokes component of vibrational SRS in molecular hydrogen. It is shown that the apex angle of the conical radiation exceeds substantially the divergence of the focused laser pump, depends on the laser power, the gas pressure and the experimental geometry, and is determined by the conditions for excitation of the waveguide SRS that is possible in vibrational excitation of the molecules.

INTRODUCTION

Stimulated Raman scattering of light (SRS) is accompanied by a substantial redistribution of the particle in energy states, which occurs in many experiments and is used to solve some spectroscopic problems.¹ The population of the upper level of the Raman transition is so effective that during the scattering time there are observed emission lines corresponding to SRS with already excited vibrational states ($v = 1, 2, \dots$). As a result of all this, the refractive index n_0 of the medium increases by some amount Δn (Ref. 2), since the polarizability of the vibrationally excited molecules exceeds that of the unexcited ones. This can lead to SRS self-focusing and to the onset of an "SRS lens" and an "SRS prism" (Ref. 3), which, depending on the experimental conditions, alter the divergence of the interacting waves as well as the propagation direction of the Stokes scattering component, by a certain angle relative to the initial beam.

On the other hand, an elongated region of the medium containing a sufficient number of excited molecules, can form a lightguide with core and sheath refractive indices $n_0 + \Delta n$ and n_0 , respectively. In this case, Stokes rays for which the waveguide-propagation condition is satisfied⁴ can become amplified in the field of the directed pump in addition to the Stokes wave propagating in the axial direction. As a result, the angle spectrum of the scattered radiation at the Stokes frequency will include conical radiation with an apex angle determined by the mode-excitation conditions in the induced waveguide.

Note that conical radiation at the frequency of the SRS first Stokes component has also been observed⁵ under hard focusing of laser radiation into a cell with compressed hydrogen. The phenomena observed were attributed there to manifestations of effects of propagation at a definite deviation of the Stokes frequency from that corresponding to the resonance condition. In the analysis of this effect in Ref. 5 no account was taken of the motion of the level population of the Raman transition, shown by numerous experiments to influence substantially the SRS process as a whole, especially in the case of hard focusing of the pump radiation.

Waveguide conical radiation at the frequency of the first Stokes component differs fundamentally from the conical radiation at the frequency of the first anti-Stokes component and of the higher Stokes and anti-Stokes scattering components that result from four-photon parametric processes. For these processes, the apex angles of the scattered-light cones at the corresponding wavelengths are determined by the conditions of linear waveguide synchronism,⁶ and

also by the angle spectrum of the pump and by the geometry of the experiment,⁷ but are independent of the power of the laser exciting the SRS. It is therefore of interest to investigate in greater detail the SRS induced in a waveguide by vibrationally excited molecules and reported briefly in Ref. 4. In the present paper we describe a more detailed investigation of this phenomenon.

CONDITIONS FOR OBSERVING WAVEGUIDE SRS

Let us estimate the maximum scattering angle of the conical component of the first SRS Stokes component resulting from waveguide generation. Assuming that the refractive indices of the core and sheath of the induced waveguide are $n_0 + \Delta n$ and n_0 , respectively, the maximum total reflection angle for such a light guide is given by

$$\beta \approx (2\Delta n)^{1/2}. \quad (1)$$

The change Δn of the refractive index of the medium by the vibrational excitation of the medium, if all the particles are only in the ground and first-excited levels, is defined as²

$$\Delta n = \frac{1}{3\epsilon_0} \frac{(n_0^2 + 2)^2}{6n_0} N \eta \Delta \alpha, \quad (2)$$

where ϵ_0 is the dielectric constant; $\Delta n = n(\eta) - n(\eta = 0)$ is the refractive-index change due to the redistribution of the particles among the levels; N , N_0 , and N_1 are the total number of molecules and the number of molecules in the vibrational states $v = 0$ and $v = 1$ per unit volume, respectively; $\eta = N_1 / (N_1 + N_0)$, $\Delta \alpha = \langle \alpha(1) \rangle - \langle \alpha(0) \rangle$, and $\langle \alpha(0) \rangle$ and $\langle \alpha(1) \rangle$ are the polarizabilities of one particle in the vibrational states 0 and 1, respectively. It follows from (1) and (2) that

$$\beta \approx \left(\frac{1}{\epsilon_0} N \eta \Delta \alpha \right)^{1/2}. \quad (3)$$

Consequently the maximum total-reflection angle β for one and the same population of the upper level of the Raman transition will be larger in the gas whose molecules have the largest polarizability difference $\Delta \alpha$. According to Ref. 2, the largest $\Delta \alpha$ is that of the hydrogen molecule ($\Delta \alpha = 15.5 \cdot 10^{-42} \text{ A} \cdot \text{s} \cdot \text{m}^2/\text{V}$), which furthermore has the largest SRS gain in the vibrational transition $Q_{01}(1)$. This means that molecular hydrogen is the most suitable object for the observation and investigation of waveguide SRS. If complete saturation of the Raman-active transition $Q_{01}(1)$ ($\eta = 0.5$) has been reached in the region of the effec-

tive interaction of laser radiation with hydrogen gas under atmospheric pressure P_0 , we get $\Delta n(\eta = 0.5) = 1.17 \cdot 10^{-5}$. In this case, according to Eq. (3), the maximum total reflection angle in the induced waveguide is $\beta(\eta = 0.5) = 4.84 \cdot 10^{-3}$ rad. At other gas pressures P the scattering angle is given by

$$\beta(\eta=0.5) \approx 4.8 \cdot 10^{-3} \left(\frac{P}{P_0} \right)^{1/2}. \quad (4)$$

Obviously, to observe SRS in a waveguide of excited molecules it is necessary that the angle spectrum of the focused pump be smaller than $2\beta(\eta = 0.5)$, i.e., that the following relation hold:

$$a/F_1 < 2\beta(\eta=0.5),$$

where a is the laser-beam diameter in the plane of the lens and F_1 focal length of focusing lens. This relation can be satisfied in the usual SRS excitation geometry. In fact, for example at $P = 5$ atm and $a = 2$ mm the linear focusing angle a/F_1 is significantly larger than 2β even at $F_1 = 50$ cm. On the other hand, to achieve a noticeable motion of the populations in the SRS process one must ensure a sufficiently hard focusing of the pump radiation into the investigated medium, for in the case of a Gaussian beam with diffractive divergence the effective volume of the nonlinear interaction is proportional to

$$(F_1/a)^4 \lambda^3,$$

where λ is the wavelength of the light. In addition, a fault-free experiment can be performed only at sufficiently low gas pressures, $P \lesssim 10$ atm, when the scattering angles of the higher Stokes and anti-Stokes components of the SRS, which satisfy the linear wave synchronism conditions,⁶ are negligibly small compared with the laser-pump divergence at the exit from the Raman-active medium. In this case, even for single-frequency pumping, the increase of the linewidth of the backward spontaneous Raman scattering⁸ will be suppressed by the back SRS which carries away a significant part of the laser energy and makes the interpretation of the experiment more complicated. It must also be recognized that for a given laser-pulse energy a lower gas density corresponds to a larger fraction of excited molecules over a greater length of the nonlinear interaction of the coherent radiation with the gaseous matter. All this means that the effect of an induced waveguide in hydrogen by SRS will be substantial at higher gas pressures.

DESCRIPTION OF EXPERIMENT

The linearly polarized single-mode second harmonic of a YAG:Nd³⁺ laser with passive Q switching by an LiF(F_2^-) crystal was used in the experiment to excite SRS in compressed hydrogen gas on the vibrational transition $Q_{01}(1)$. The laser parameters were: wavelength $\lambda = 532$ nm, pulse duration at half-maximum $\tau = 7$ ns, pulse energy $\mathcal{E}_L \lesssim 30$ mJ, and beam diameter $a = 1.5$ mm. A lens with focal length $F_1 = (14-100)$ cm focused the laser beam into a cell $L = (20-110)$ cm long filled with hydrogen compressed to $P = (1-25)$ atm. To avoid ambiguity, the focusing lens was placed to position the "neck" of the exciting beam in the middle of the molecular-hydrogen cell. The gas pressure was measured with a manometer accurate to 2.5%. The angle

spectrum of the scattered components was analyzed, after preliminary selection by suitable filters, in the focal plane of a second lens ($F_2 = 40, 33$ cm) located at the exit from the hydrogen chamber, and was defined as

$$\theta = \frac{D}{2F_2}, \quad (5)$$

where D denotes the diameters of the rings in the focal plane of an analyzing lens placed directly at the exit from the gas-filled cell. The energies of the pump and of the scattered SRS components were measured with a precisely calibrated multichannel laser-pulse energy recorder based on FD-24K photodiodes. An LFD-2 avalanche photodiode and an S7-10B oscilloscope were used to record the waveforms of the laser-pump light pulses at the entrance and exit of the cell with hydrogen, the first axial and conical Stokes components, and the waveform of the radiation pulses at the first anti-Stokes and the second Stokes components. The use of an optical delay line permitted simultaneous recording of light pulses of two different scattering components in different combinations, so that it was possible to determine the relative times of different light pulses.

EXPERIMENTAL RESULTS AND DISCUSSION

The experiments have shown that at $P \lesssim 7$ atm a first Stokes component is generated both along the laser beam axis and along the generators of a cone with an apex angle 2θ substantially larger than the divergence of the focused pump (see Fig. 1). Given the gas pressure, the scattering angle of the conical part of the first Stokes component increases drastically and to a certain maximum scattering angle θ_{\max} which remains constant at high laser powers. At a lower gas pressure, the character of the $\theta(\mathcal{E}_L)$ dependence remains unchanged, but the corresponding scattering angle θ_{\max} decreases and the pump-energy level at which $\theta(\mathcal{E}_L)$ saturates increases (see Fig. 2).

When the pulse energy is lower than a certain critical value, only axial SRS is observed in the experiment. The increase of the experimentally observed conical radiation angles of the first SRS Stokes component with increase of \mathcal{E}_L is attributed to the increase of the fraction η of the excit-

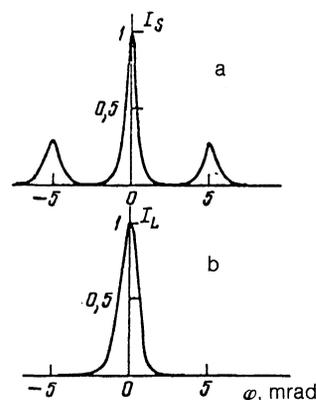


FIG. 1. Angular distributions of the intensities $I_S(\varphi)$ of the first Stokes component (a) and $I_L(\varphi)$ of the focused laser pump at the entrance to the cell (b), obtained at $F_1 = 50$ cm, $L = 64$ cm, $P = 3$ atm, and $\mathcal{E}_L = 15$ mJ. These distributions [$I_S(\varphi)$ and $I_L(\varphi)$] were obtained by scanning a pinhole-input photodiode in the focal plane of an analyzer lens, with multiple averaging of the signal.

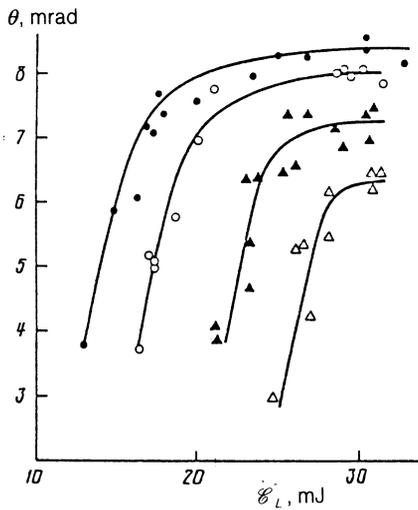


FIG. 2. Experimental dependence of the scattering angle θ of conical radiation at the frequency of the first Stokes component on the pump energy \mathcal{E}_L at $F_1 = 57$ cm, $L = 110$ cm, $P = 5$ (●), 4 (○), 3 (▲), and 2.5 (△) atm.

ed molecules in the volume where the laser pump interacts with the molecular medium. In fact, according to (2) a growth of the fraction η of the excited molecule is accompanied by a growth of the refractive index of the core of the induced waveguide. This increases the calculated maximum total-reflection angle β in the light guide, as is actually observed on the initial section of the experimental $\theta(\mathcal{E}_L)$ plot. The displacement of the $\theta(\mathcal{E}_L)$ curve corresponding to a lower gas density towards higher pump energies is due to the well-known increase of the SRS threshold at low gas densities.

The stabilization of the scattering angle θ at high energies of the SRS-exciting laser can be explained as due to saturation of the combination-active transition at which the refractive index of the induced-lightguide core, and with it the maximum total-reflection angle β , ceases to grow. Upon saturation of the Raman-active transition, when about 50% of the molecules are excited, the maximum total-reflection angle β is determined only by the gas density and decreases with decrease of P in accordance with Eq. (4). The same is in fact manifested in the experimental $\theta(\mathcal{E}_L)$ plots obtained at different gas pressures (see Fig. 2) as a decrease of the maximum scattering angle θ_{\max} with decrease of the molecular-hydrogen density.

The measured scattering angles θ_{\max} of the conical radiation ($L = 86$ cm, $F_1 = 50$ cm) in the pressure range $P = (2-2.5)$ atm is in good agreement with the plot of Eq. (4). This is attested to by Fig. 3a. However, as shown by

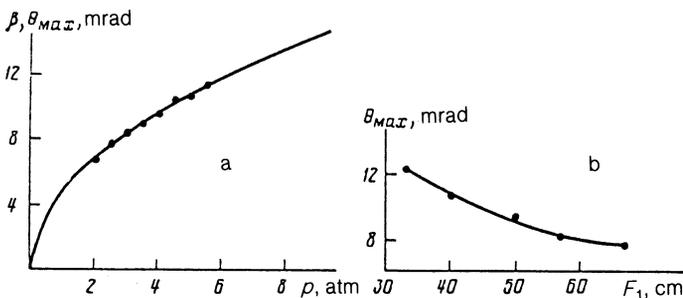


FIG. 3. (a) Experimental points (●) of maximum scattering angle θ_{\max} obtained at $F_1 = 50$ cm, $L = 86$ cm, and various gas pressures P , and also the calculated $\beta(\eta = 0.5, P)$ plot (solid curve) of Eq. (4). (b) Maximum scattering angle θ_{\max} vs the focal length F_1 at $P = 4$ atm and $\mathcal{E}_L = 27$ mJ.

experiments at different F_1 and L , the maximum scattering angle θ_{\max} corresponding to a specified gas pressure depends on the focal length F_1 of the focusing lens, which in turn governs the diameter of the "neck" of the exciting-radiation beam. Indeed, it follows from Fig. 3b that as F_1 decreases (meaning with decrease of the beam diameter at the lens focus) the maximum conical-radiation angle θ_{\max} increases. This shows that it will be more correct to regard conical radiation at the frequency of the first Stokes component as the result of excitation of higher spatial modes of the waveguide induced in the medium by the SRS.

The observed phenomena can be better understood after determining the radiation-intensity angular distribution corresponding to the first-order modes of the induced waveguide. One can use for this purpose the known wave equation

$$\Delta E(x, y, z, t) - \frac{\epsilon}{c^2} \frac{\partial^2 E(x, y, z, t)}{\partial t^2} = 0, \quad (6)$$

where $E(x, y, z, t)$ is the electric field, which is a function of the Cartesian coordinates x, y, z and the time t , and ϵ is the dielectric constant. Let the induced waveguide be a cylinder of radius R_0 . Assuming that the refractive index $n(R)$, which depends on the radius R , can be expressed in the form

$$n(R) = n_0 + \Delta n - \Delta n (R/R_0)^2, \quad (7)$$

and also representing the electric field by

$$E(x, y, z, t) = E(x, y) \exp[i(kz - \omega t)]$$

(k is the wave vector, ω is the radiation frequency, and the z axis is aligned with the waveguide axis), we can reduce Eq. (6) to the harmonic-oscillator equation. Using the known solutions of the latter in terms of Hermite polynomials, we obtain the angle spectrum of the radiation intensity of the first-order modes of the induced waveguide:

$$I_{01}(\varphi_x, \varphi_y) = I_{10}(\varphi_x, \varphi_y) = \frac{1}{\pi} k^2 r_0^2 \varphi^2 \exp[-r_0^2 k^2 \varphi^2], \quad (8)$$

where $\varphi_x = k_x/k$, $\varphi_y = k_y/k$, k_x, k_y are the components of the wave vector k along the x and y axes, and

$$r_0 = \left[\frac{R_0}{(\omega/c)(2n_0\Delta n)} \right]^{1/2}, \quad \varphi^2 = \varphi_x^2 + \varphi_y^2.$$

It follows from (8) that $I_{01}(\varphi_x, \varphi_y)$ has a maximum at

$$\varphi_{01} \approx \left(\frac{2\Delta n}{kR_0} \right)^{1/2}. \quad (9)$$

The angle spectrum of the radiation intensity of a first-order mode in the induced waveguide has thus a conical structure. The magnitude and character of the function $\varphi_{01}(\Delta n, R_0)$

defined by Eq. (9) is thus in qualitative agreement with the experimental results given above. It can be shown that the angle spectrum of a higher waveguide mode also has a conical component on top of the axial one. In a real experiment the refractive-index distribution in the cross section of the waveguide can be significantly different from a quadrupole. In addition, the modes of various degree could be excited simultaneously but with different efficiency. All this can cause the actual conical distribution of the resultant SRS radiation to differ from the distribution described by Eq. (8). However, the interpretation of the conical SRS radiation as the result of excitation of an induced-waveguide modes accounts for the preferred scattering of mode emission at a definite angle to the laser-beam axis.

Generation of conical SRS influences substantially the energy characteristics of the scattering process. Thus, for example, there is no stimulated scattering at fixed gas pressure (see Fig. 4) and at a pump energy \mathcal{E}_L below a certain critical \mathcal{E}_{L1} . An increase of the laser power at the input to the chamber with hydrogen produces first an axial first Stokes component of the SRS, whose energy growth gradually gives way to saturation. Conical radiation appears only at $\mathcal{E}_L > \mathcal{E}_{L2}$, when the energy \mathcal{E}_{cen} of the axial Stokes component is high enough and is close to the saturation level. The increasing section of the $\mathcal{E}_{con}(\mathcal{E}_L)$ curve corresponds to almost complete cessation of the growth of the energy of the axial component of the SRS. As a result the total scattering energy at the frequency of the first Stokes component continues to increase with further increase of \mathcal{E}_L , the energy of the conical component becomes comparable with the energy of the axial component and the ratio $r = \mathcal{E}_{con}/\mathcal{E}_{cen}$ approaches unity (see Fig. 4). Experiments have shown that the character of the plots in Fig. 4 remains the same also at other hydrogen pressures. Changes do take place, however, in \mathcal{E}_{L1} , \mathcal{E}_{L2} , in their difference, and also in other characteristic values of the time, in full agreement with the waveguide mechanism of the conical SRS radiation.

Generation of a waveguide term in the first Stokes component is noticeable also in the relation indicative of the efficiency of the SRS process as a function of the gas pressure. Given the pump energy (see Fig. 5, curve 1), a decrease of the hydrogen pressure in the range $P = (10-25)$ atm weakens the accompanying SRS as a result of four-photon para-

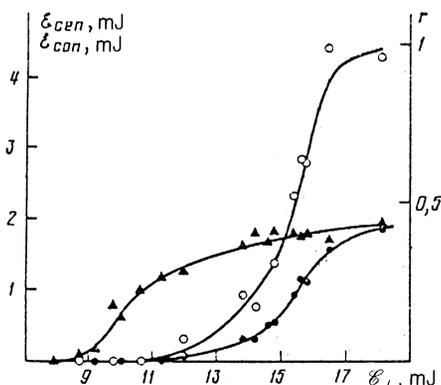


FIG. 4. Energies of the axial \mathcal{E}_{cen} (\blacktriangle) and conical \mathcal{E}_{con} (\bullet) Stokes radiation, and also (O) the ratio $\tau = \mathcal{E}_{con}/\mathcal{E}_{cen}$ vs the energy of the exciting laser pulse at $F_1 = 50$ cm, $L = 64$ cm, and $P = 4$ atm.

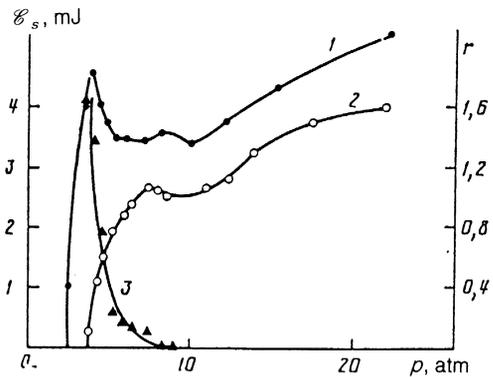


FIG. 5. Total energy \mathcal{E}_s of the first Stokes component of SRS vs the gas pressure P at $\mathcal{E}_L = 18$ (1) and 10 (2) mJ, and plot of the ratio $\tau = \mathcal{E}_{con}/\mathcal{E}_{cen}$ vs P (3) in the case $\mathcal{E}_L = 18$ mJ.

metric Stokes-anti-Stokes processes that decrease the growth rate of the Stokes wave when the initial wave mismatch is decreased. No conical anti-Stokes wave is emitted in this pressure range.

Further decrease of the hydrogen pressure stabilizes and even raises somewhat the total Stokes-component energy level at the exit from the cell. This is due to the onset of a conical SRS component which is decoupled from the anti-Stokes radiation.

At pressures $P \lesssim 8$ atm the energy of the axial component continues to decrease, whereas the energy of the conical radiation increases. This is attested to by curve 3 of Fig. 5, which illustrates the ratio of the conical radiation energy \mathcal{E}_{con} to the energy \mathcal{E}_{cen} of the axial component. The conical SRS radiation pulse energy exceeds the axial SRS energy at pressures close to 4 atm. Further decrease of the gas pressure in the chamber decreases drastically the number of Stokes photons, owing to the decrease of the density of the scattered particles as well as to the increase of the line width of the spontaneous Raman scattering of light on going from homogeneous to Doppler broadening.⁸ At low laser-pump intensity the fraction of the vibrationally excited molecules in the medium decreases, thus weakening the onset of the induced waveguide. Thus, for example, no conical SRS appears at $\mathcal{E}_L = 10$ mJ in the entire investigated pressure range, as manifested by the character of the $\mathcal{E}_s(P)$ dependence (see curve 2 of Fig. 5), which shows no rising section at pressures close to 4 atm.

It is of particular interest to determine the instant when a pulse of conical radiation is produced at the frequency of the first Stokes component relative to the pulse of the axial Stokes and higher Stokes and anti-Stokes scattering components which increase simultaneously with the SRS as a result of four-photon parametric interactions. At the instant when stimulated scattering sets in the number of excited molecules is zero, so that there is no waveguide amplification. As the SRS develops, an increase is observed in the number of inverted molecules in the focal volume of the focusing lens, with lifetimes much longer than the laser-pulse duration.¹ As a result, a waveguide is produced in the medium only after some definite time interval following the onset of the SRS, and conditions for waveguide generation of the Stokes component are realized. Indeed, as shown by experiments performed at $F_1 = 50$ cm, $L = 64$ cm, $P = 4$ atm, and \mathcal{E}_L

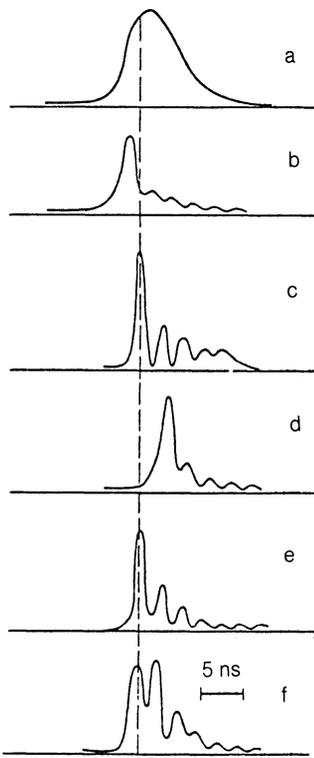


FIG. 6. Relative timing of laser-pump pulses at the entry to the cell (a), at the exit from the cell (b), and of the axial (c) and conical (d) scattered-component pulses of the first Stokes components (e), the first anti-Stokes emission (e), and second Stokes emission (f).

≈ 20 mJ (see Fig. 6), the first Stokes component ($\lambda_s = 683$ nm), the anti-Stokes radiation ($\lambda_a = 436$ nm), and a pulse at the second Stokes frequency ($\lambda_{ss} = 954$ nm) are all produced at the same instant. The time dependences of the anti-Stokes and of the axial-Stokes components are quite similar, owing to the parametric relation between the light waves of these components.

It follows also from Fig. 6 that axial radiation at the frequency of the first Stokes component is produced on the initial section of the pump pulse and has a rather short leading front. It reaches its maximum earlier than the pump radiation. The reason is that the Raman-active transition saturates and as a result the energy transfer from pump to first-Stokes radiation ceases. At that instant, the excitation of SRS in the induced waveguide produces a conical radiation pulse (see Fig. 6) which is not parametrically connected with the anti-Stokes component and for which the higher-mode antinodes lie on the periphery of the waveguide, where the population saturation is weak. This pulse has therefore a larger gain compared with the axial Stokes one and increases quite effectively. The time delay Δt between the peaks of the

axial and conical Stokes components is thus about 3.5 ns. Experiments have shown that at a given gas pressure the delay time Δt increases when the laser pulse energy is decreased and decreases when the energy is increased. All this agrees with the notion of waveguide generation of conical emission of the first SRS Stokes component.

CONCLUSIONS

An understanding of the physical nature of the onset of an initial spatial part of the first SRS Stokes component in the form of conical radiation makes it possible to control the angle spectrum of the scattering. Since the effect described is primarily connected with saturation of the populations of the Raman-active transition by stimulated scattering, its occurrence will be minimal in experiments where no noticeable motion of the populations is reached in the region of effective interaction of the laser beam with the medium. It follows hence that at sufficiently higher pump energies (powers) and at a fixed experimental geometry the manifestation of the induced pump will be smaller the higher the molecular-hydrogen density, and at a fixed gas density the conical SRS will vanish in experiments with longer cells if the radiation is focused into small solid angles, as confirmed by experiment. For example, at $L = 100$ cm, $F_1 = 100$ cm, and $\mathcal{E}_L = 35$ mJ conical emission at the first Stokes frequency was observed only for $P \lesssim 2$ atm.

Our present results show that population motion can be manifested in SRS also in the case of rather soft focusing of the laser radiation. For an adequate theoretical analysis of stimulated Raman scattering of light, especially of the influence of cooperative effects on the scattering, which appear at low gas densities,⁹ account must be taken of the saturation of the upper level of the Raman transition, and of the difference between the molecule-level polarizabilities which produce a waveguide that influences substantially the scattering results.

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