

EFFECT OF ROTATIONAL SRS ON THE ANGULAR SPECTRUM OF LASER RADIATION IN THE ATMOSPHERE

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Stationary rotational SRS on nitrogen molecules in wide-aperture laser beams was studied. The effect of four-photon parametric processes on the singular spectrum of the radiation was investigated. The results of numerical calculations of the beam profile are presented. It is shown that the parametric processes increase the width of the angular spectrum of wide-aperture laser beams almost by an order of magnitude and distort the beam profile.

When powerful laser radiation propagates in the atmosphere different nonlinear effects can occur. In the visible and UV regions of the spectrum stimulated Raman scattering on rotational sublevels (RSRS) $J = 8 \rightarrow J = 10$ of the nitrogen molecule has the lowest threshold.¹ In spite of the fact that SRS was discovered a long time ago and has been studied extensively, qualitatively new physical results arise in the RSRS in the atmosphere. The characteristic features of RSRS in wide-aperture beams in air are connected with the small rotational frequency shift ($\nu_R = 76 \text{ cm}^{-1}$), the weak dispersion of the medium, and the resulting long interaction lengths. It was found to be impossible to simulate such conditions in laboratory experiments.

The purpose of this work is to investigate the basic characteristics of rotational SRS processes in wide-aperture beams and determine their effect on the angular spectrum of the radiation.

Stimulated Raman scattering changes not only the frequency but also the angular spectrum of radiation. This happens for two reasons. First, parametric four-photon processes convert part of the pumping radiation into Stokes and anti-Stokes radiation, propagating at an angle to the direction of the main wave. The angle φ , under which, for example, the first anti-Stokes component propagates, is proportional to the frequency of the rotational transition of the nitrogen molecule $\omega_R = 2\pi\nu_{RC}$ and depends on the dispersion properties of air²

$$\varphi \approx \omega_R \left[\frac{1}{k_L} \frac{\partial^2 k}{\partial \omega^2} \right]^{1/2}, \tag{1}$$

where k_L is the wave number of the pumping radiation. In the UV region of the spectrum $\varphi \sim 3 \times 10^{-5}$ rad.

The second reason that SRS changes the angular spectrum of the radiation is as follows. Since SRS develops from spontaneous noise the growing Stokes signal has a random spatial structure. The angular

spectrum of such radiation turns out to be much wider than that of the initial radiation.^{3,4}

In this work we studied the stationary stimulated Raman scattering on the rotational transition $J = 8 \rightarrow J = 10$ (RSRS) of the nitrogen molecule. The laser radiation is assumed to be monochromatic and linearly polarized, and in addition $\omega_L \gg \omega_R$ and the frequency of the main wave (the pump wave) and the Raman frequencies are widely separated from electronic resonances. The system of equations for the slowly varying amplitudes of the photon flux density

$(A_i = \sqrt{\frac{c}{4h\omega_i}} \cdot E_i)$, taking diffraction into account has the following form:

$$\left[\frac{\partial}{\partial z} + \frac{1}{2ik} \Delta_{\perp} + \alpha \right] A_n = R \sum_{m, k, p} A_m A_k A_p^* \delta_{nmkp} e^{i\Delta_{nmkp} z} + i\mu \left[2 \sum_m |A_m|^2 - |A_n|^2 \right], \tag{2}$$

where

$$\delta_{nmkp} = \begin{cases} +1, & k - p = n - m = -1, \quad n \neq p \\ -1, & k - p = n - m = +1, \quad n \neq p \\ 0 & \text{in all the other cases;} \end{cases} \tag{3}$$

$$\Delta_{nmkp} = k(\omega_m) + k(\omega_k) - k(\omega_n) - k(\omega_p). \tag{4}$$

A_n is the amplitude of the n -th harmonic of SRS $\omega_n = \omega_L + n\omega_R$ ($n = 0, \pm 1, \pm 2, \dots$), ω_L is the frequency of the laser radiation, ω_R is the transition frequency,

$$R = \hbar \left[\frac{4\pi\omega_L}{c} \right]^2 \gamma, \quad \mu = \hbar \left[\frac{4\pi\omega_L}{c} \right]^2 \beta, \tag{5}$$

β is the real part of the cubic susceptibility, the imaginary susceptibility, $\beta = \chi^{(3)'} (\omega = \omega + \omega - \omega)$, and γ is the imaginary part of the cubic susceptibility, $\gamma = \chi^{(3)''} (\omega_0 = \omega_{-1} + \omega_0 - \omega_{-1})$. The coefficient α in the equations accounts for the extinction of the field owing to molecular and aerosol scattering. It should be noted that for $n = k, m = p = n \pm 1$ the phase factor vanishes ($\Delta_{nmkp} = 0$). Such terms in the first sum correspond to the standard SRS, when the spatial matching conditions need not be satisfied. The terms with the exponential factor describe parametric four-photon processes, which require that spatial matching conditions be satisfied. The terms in the last group (in brackets) are responsible for the Kerr effect.

We introduce the characteristic nonlinear length z_{NL} and the wave synchronism length z_D :

$$z_{NL} = \frac{\hbar\omega_L}{RI_L}, \quad z_D = \frac{\pi}{|\Delta_{100-1}|} \quad (6)$$

The weak Stokes wave is amplified by a factor e over a distance z_{NL} . Over a distance z_D the sign of the exponential term, responsible for parametric effects, changes. The characteristic intensity at which the nonlinear length is equal to the wave synchronism length is determined by the equation

$$I_{ND} = \frac{\hbar\omega_L |\Delta_{100-1}|}{\pi R} \quad (7)$$

For the atmosphere (at $p = 1$ atm) in the UV region of the spectrum $I_{ND} \approx 16$ MW/cm² ($\lambda = 350$ nm). For pump intensities $I_L \ll I_{ND}$ the factor in the parametric term can change sign many times before the Stokes component is amplified by a factor of e , i.e., in this case parametric processes do not result in efficient generation of radiation with high spatial frequencies. In the case when $I_L \gg I_{ND}$ ($z_{NL} \ll z_D$) the spatial matching relations are of no importance, since at lengths of $\sim z_D$ all spatial harmonic are amplified by a factor of $\exp(z_D/z_{NL})$. The angular spectrum of the radiation will be very wide in this case. Thus one mechanism of SRS (in which parametric effects play a small role) should be realized for $I_L \ll I_{ND}$ while another mechanism should be realized for $I_L \gg I_{ND}$, where parametric effects result in simultaneous generation of a large number of Stokes and anti-Stokes components.

In this work we used the method of numerical simulation to study the system of equations (2) in the case of the propagation of laser radiation on a vertical path for initial intensities $I_L < I_{ND}$. The beam profile is assumed to be Gaussian with a radius such that the diffraction length of the beam is much greater than the distances studied. The initial intensity $I_L = 0.3 I_{ND}$. A coherent seed signal at the first Stokes frequency with intensity 10^{-8} times the intensity of the pump wave is employed. The seed signal on the remaining Stokes and anti-Stokes frequencies is given in the form of noise. Seven Stokes component and two anti-Stokes components were included in the calculations.

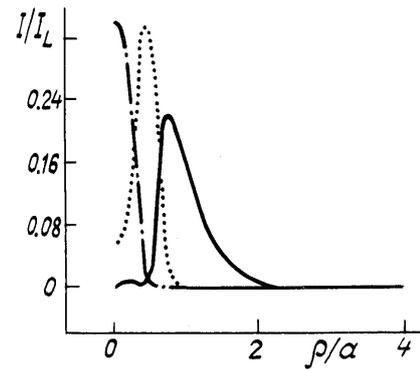


FIG. 1. The transverse distribution of the intensities of the spectral components of SRS at $z = 0.010 z_D$. The intensities are normalized to the input pump intensity at the center of the beam ($I_L = 0.3 I_{ND}$); the transverse coordinate is normalized to the initial width of the pump beam ($a = 30$ cm): solid line – pump (ω_L); dotted line – first Stokes component (ω_{-1}); dot-dashed line – second Stokes component (ω_{-2}); dashed line – third Stokes component (ω_{-3}).

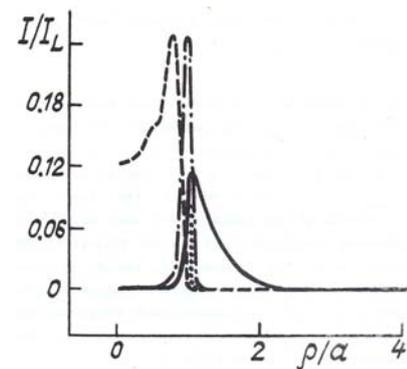


FIG. 2. The transverse distribution of the intensities of the spectral components of SRS at $z = 0.017 z_D$. The intensities are normalized to the input pump intensity at the center of the beam ($I_L = 0.3 I_{ND}$); the transverse coordinate is normalized to the initial width of the pump beam ($a = 30$ cm): solid line – pump (ω_L); dotted line – first Stokes component (ω_{-1}); dot-dashed line – second Stokes component (ω_{-2}); dashed line – third Stokes component (ω_{-3}).

Figures 1 and 2 show the pattern of the change in the spectral composition of the radiation as a function of the distance - for $z = 0.01 L_D$ ($L_D \approx k_L a^2$) and $z \approx 0.017 L_D$. At the starting stage the coherent Stokes signal is amplified. The growth of the first Stokes component is accompanied by parametric generation of a weak signal at the anti-Stokes and second Stokes frequencies. As soon as the intensity of the signal at the first Stokes frequency is equal to that of the pump, the pump is rapidly exhausted on the axis of the beam and energy is transferred from the first Stokes component into the second Stokes component. As one

see from Fig. 1, the second Stokes component has a coherent structure rather than a noise structure. This is because a weak seed signal at the second Stokes frequency is formed owing to parametric processes (of the type $\omega_{-2} = \omega_{-1} + \omega_{-1} - \omega_0$) even before exponential amplification of the second Stokes component in the field of the first Stokes component starts. This signal, though it is weak, is stronger than the spontaneous noise. Since the parametric seed signal at the second Stokes frequency has a different angular spectrum the radiation propagates at an angle to the initial direction. Figures 3 and 4 show the total profile of the radiation intensity for $z = 0.01 L_D$ and $z = 0.017 L_D$, respectively. One can see that the beam profile has changed from Gaussian to hollow. It should be noted that as the altitude increases the parametric effects become more important, since Δ_{100-1} is proportional to the pressure and therefore as the altitude increases I_{ND} diminishes rapidly.

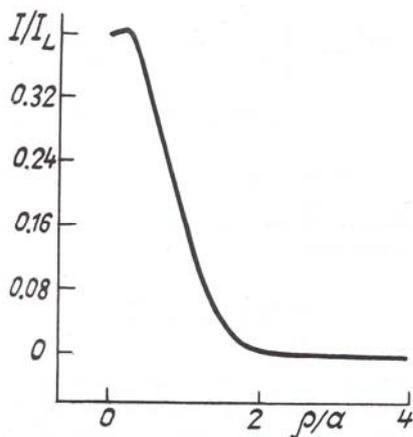


FIG. 3. The transverse distribution of the total intensity of the SRS components normalized to I_L at $z = 0.010 z_D$. The transverse coordinate is normalized to the initial width of the pump beam.

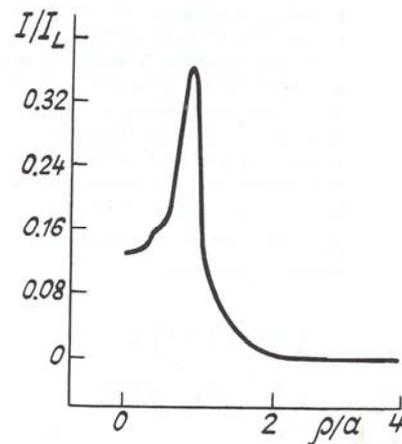


FIG. 4. The transverse distribution of the total intensity of the SRS components normalized to I_L at $z = 0.017 z_D$. The transverse coordinate is normalized to the initial width of the pump beam.

Thus parametric processes can change substantially the angular spectrum of the radiation and the beam profile under conditions of RSRS in the atmosphere. Rotational stimulated Raman scattering processes substantially limit the possibility of efficient directed energy transfer over large distances.

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