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Influence of the polarization of radiation on the energy characteristics and threshold of stimulated Raman scattering due to rotational transitions

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It is shown experimentally and theoretically that the threshold intensity for stimulated Raman scattering (STRS) due to rotational transitions depends strongly on the polarization of the pump radiation. The threshold for rotational STRS is lowest for circularly polarized radiation and increases sharply for linearly polarized radiation. The results of investigations of the energy characteristics of rotational STRS on the degree of ellipticity of the pump radiation are presented.

Stimulated Raman scattering (STRS) due to rotational transitions has been studied on many occasions, for example, in Refs. 1–4. However, in these studies the influence of the polarization of the pump radiation on the threshold and energy characteristics of the Stokes and anti-Stokes components was neglected. It was merely shown qualitatively in an experimental study[5] that the threshold for rotational STRS depends strongly on the polarization of the exciting radiation. For circularly polarized pump radiation, rotational STRS had the lowest threshold, whereas for linearly polarized radiation the threshold was not reached even when the pump power was increased fourfold.

The present paper reports an experimental study of the dependence of the rotational STRS threshold and also of the dependences of the energy of the first Stokes, second Stokes, and anti-Stokes components on the degree of ellipticity of the polarization of the pump radiation. A possible theoretical explanation of the dependence of the rotational STRS threshold on the polarization of the exciting radiation is also given.

The gain and thus the STRS threshold depend on the polarization of the pump radiation. For rotational STRS and also for vibrational–rotational STRS when $\Delta J = \pm 2$, the gains of the Stokes components for the various types of polarization of the pump radiation are in the ratio[1]

$$g_{++} : g_{+-} : g_{\parallel} : g_{\perp} = 1 : 6 : 4 : 3, \tag{1}$$

where g_{++} is the gain for circular polarizations of the pump and Stokes component in one direction; g_{+-} is that for circular polarizations in the opposite directions; $g_{\parallel}$ is that for linear polarizations; $g_{\perp}$ is that for linear orthogonal polarizations.

We can find the gain g for elliptically polarized pump radiation from an expression for the polarization vector at the Stokes frequency[6]:

$$\mathbf{P}_s = -i\gamma_s \left[3\left(\mathbf{E}_l^*\mathbf{E}_l\right)\mathbf{E}_s - 2\left(\mathbf{E}_l\mathbf{E}_s\right)\mathbf{E}_l + 3\left(\mathbf{E}_l\mathbf{E}_s\right)\mathbf{E}_l^*\right]. \tag{2}$$

Here $\mathbf{E}_l$ and $\mathbf{E}_s$ are the complex amplitudes of the field at the pump frequency and the Stokes frequency; γ_s is a coefficient that depends on the frequency detuning and on the population difference of the two molecular levels under study, and also on the form of the scattering tensor. Exponential solutions of the type $\mathbf{E}_s = \mathbf{E}_s(0)\exp(gL)$ of a reduced equation for the field at the Stokes frequency

$$d\mathbf{E}_s/dz = 2\pi i \varkappa_s \mathbf{P}_s \tag{3}$$

may be found by using Eq. (2). Here, $\varkappa_s$ is the wave number at the Stokes frequency. Assuming that $g = 2\pi\varkappa_s\gamma_s|\mathbf{E}_l|^2\lambda$ and defining the polarization of the pump radiation by $\mathbf{E}_l = \mathbf{E}_l\binom{\cos\psi}{i\sin\psi}$, we find the dimensionless gain λ:

$$\lambda_{\pm} = {}^1/_2\left(7 \pm \sqrt{49 - 12(3 + \cos 4\psi)}\right). \tag{4}$$

From Eq. (4) for circular polarization ($\psi = \pm\pi/4$), we obtain $\lambda_+ = 6$, $\lambda_- = 1$, and for linear polarization ($\psi = 0, \pi/2$), we obtain $\lambda_+ = 4$, $\lambda_- = 3$, which agrees with the results of Ref. 1. When ψ varies between $\pi/4$ and 0 or $\pi/2$, λ_+ varies monotonically between 6 and 4. In this case, the polarization of the Stokes radiation changes from circular to linear with axes of the polarization ellipse parallel to the axes of the polarization ellipse of the pump radiation.

A stronger dependence of the threshold for rotational STRS on the polarization of the pump radiation than that given by Eq. (4) is observed experimentally. A possible explanation for this discrepancy may be that the influence of the anti-Stokes radiation on the gain of the Stokes component is

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neglected in the theory. It is known that for scalar STRS the magnitude of the polarization at the anti-Stokes frequency agrees (in the Placzek approximation) with that at the Stokes frequency. For a zero wave mismatch the gains are zero and the threshold increases to infinity.[8]

For rotational STRS the wave mismatch is small as a result of the small frequency shift but for circularly polarized pump radiation the polarization vector at the anti-Stokes frequency vanishes. This creates particularly favorable conditions for rotational STRS with circularly polarized pump radiation. In this case, no anti-Stokes radiation is found for any wave mismatch. However, for linearly polarized pump radiation, as can be shown and can be seen from the following analysis as in the scalar case, $g = 0$ is found for a zero wave mismatch.

We shall find an expression for the dimensionless gain of the Stokes component in the presence of an anti-Stokes one. We shall use expressions for the polarization vectors at the Stokes (ω_s) and anti-Stokes (ω_a) frequencies:[6,7]

$$\mathbf{P}_s = -\gamma_s\left[3\left(\mathbf{E}_l\mathbf{E}_l^*\right)\mathbf{E}_s - 2\left(\mathbf{E}_l^*\mathbf{E}_s\right)\mathbf{E}_l + 3\left(\mathbf{E}_l\mathbf{E}_s\right)\mathbf{E}_l^* + \left(\mathbf{E}_l\mathbf{E}_a^*\right)\mathbf{E}_l + 3\left(\mathbf{E}_l\mathbf{E}_l\right)\mathbf{E}_a^*\right], \tag{5a}$$

$$\mathbf{P}_a = -\gamma_a^*\left[3\left(\mathbf{E}_l\mathbf{E}_l^*\right)\mathbf{E}_a - 2\left(\mathbf{E}_l^*\mathbf{E}_a\right)\mathbf{E}_l + 3\left(\mathbf{E}_l\mathbf{E}_a\right)\mathbf{E}_l^* + \left(\mathbf{E}_l\mathbf{E}_s^*\right)\mathbf{E}_l + 3\left(\mathbf{E}_l\mathbf{E}_l\right)\mathbf{E}_s^*\right]. \tag{5b}$$

Neglecting the wave mismatch and also the difference between $\varkappa_s$ and $\varkappa_a$, γ_s and γ_a in the expressions for $\mathbf{P}_s$ and $\mathbf{P}_a$ and the reduced equations for $\mathbf{E}_s$ and $\mathbf{E}_a$ of the type (3), equations can be found for coupled waves at the frequencies ω_s and ω_a. From these we find $\lambda_\pm$ and the polarization vectors of the coupled waves.

These equations take the form

$$\begin{pmatrix} 3+\cos^2\psi-\lambda & -5\cos\psi\sin\psi & 4\cos^2\psi-3\sin^2\psi & \cos\psi\sin\psi \\ -5\cos\psi\sin\psi & 3+\sin^2\psi-\lambda & \cos\psi\sin\psi & 4\sin^2\psi-3\cos^2\psi \\ 4\cos^2\psi-3\sin^2\psi & \cos\psi\sin\psi & 3+\cos^2\psi+\lambda & -5\cos\psi\sin\psi \\ \cos\psi\sin\psi & 4\sin^2\psi-3\cos^2\psi & -5\cos\psi\sin\psi & 3+\sin^2\psi+\lambda \end{pmatrix}\begin{pmatrix} E_{sx} \\ -iE_{sy} \\ E_{ax}^* \\ iE_{ay}^* \end{pmatrix} = 0. \tag{6}$$

Here, the X and Y axes are directed along the axes of the polarization ellipse of the pump radiation. From these equations we find an equation for λ: $\lambda^4 - 36\sin^2\psi\lambda^2 = 0$. Hence, we obtain

$$\lambda_{1,2} = \pm 6\sin 2\psi, \quad \lambda_{3,4} = 0. \tag{7}$$

It can be shown that $\lambda > 0$ corresponds to coupled field waves at the Stokes and anti-Stokes frequencies having circular polarizations and the intensity ratio

$$|\mathbf{E}_s|^2/|\mathbf{E}_a|^2 = \mathrm{tg}^2(\psi - \pi/4). \tag{8}$$

The following experimental setup was used to study the energy characteristics of the scattered components and dependences of the rotational STRS threshold on the polarization of the pump radiation. Linearly polarized radiation from a neodymium glass laser in the TEM_{00} mode was converted to the second harmonic using a KDP crystal and was then passed through a quarter-wave perpendicular to the incident radiation and mounted on a limb so that it could be rotated to within 1′. In this case, the angle α corresponded to the angle between the optic axis of the plate and the direction of the vector $\mathbf{E}_l$. Thus, the degree of ellipticity of the pump radiation could be varied smoothly between circular and linear. After being passed through the quarter-wave plate the second-harmonic neodymium laser radiation was focused by a lens ($F = 25$ cm) into a cell 50-cm long containing gaseous hydrogen at a pressure of 8 atm. The cell windows were misaligned relative to one another to avoid feedback. The pump parameters were as follows: $W \approx 200$ mJ, $\tau \approx 40$ nsec, $\Delta\nu \lesssim 0.01$ cm^{-1}.

After being collimated, the scattered radiation was fed to the slit of a spectrograph and recorded using a multichannel recording system based on an Élektronika 841 video camera.[9] Either the energy of the transmitted pump radiation and that of the first Stokes and anti-Stokes components or the energy of the transmitted pump radiation and that of the first and second Stokes components was recorded per laser shot.

Figure 1 shows dependences of the threshold pump power for rotational STRS due to the $S_{00}(1)$ transition in the hydrogen molecule on the angle α ($\alpha = \psi - \pi/4$) (curve 1). These dependences do not agree even qualitatively with the theoretical equation (4) (curve 3) neglecting the influence of the anti-Stokes component. Curve 2 plotted for the rotational STRS threshold in accordance with Eq. (7) agrees qualitatively with the experimental curve 1. The faster increase in the experimental curve is attributed to an additional increase in the threshold power since rotational STRS is influenced by STRS due to the $Q_{01}(1)$ vibrational transition. For $\alpha = 21$; the STRS threshold due to the $Q_{01}(1)$ transition is reached.

Figure 2 shows experimental and theoretical dependences of I_a/I_s on the angle α. It is deduced from Eq. (8) that

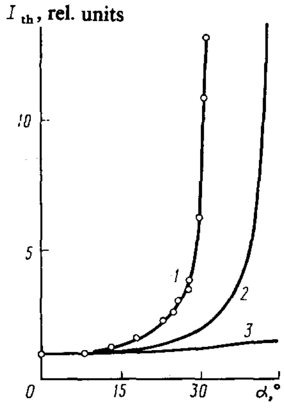

FIG. 1. Dependences of the threhsold intensity for rotational STRS on the polarization of the radiation.

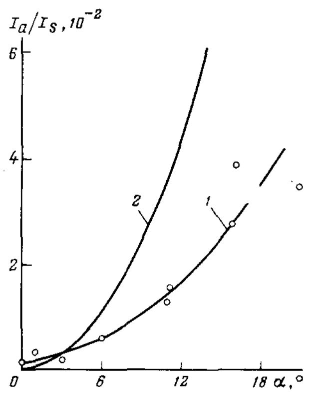

FIG. 2. Experimental (1) and theoretical [Eq. (8)] (2) dependences of the ratio of the intensity of the anti-Stokes component of rotational STRS to the Stokes component on the polarization of the pump radiation.

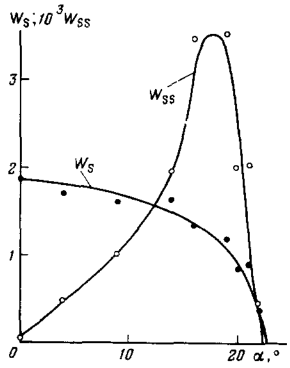

FIG. 3. Dependences of the energy of the first W_s and second W_{ss} Stokes components on the polarization of the radiation from the laser used to excite rotational STRS.

for purely circularly polarized pump radiation ($\alpha = 0$) no anti-Stokes radiation should be found, whilst when the degree of ellipticity of the pump radiation is increased, the ratio I_a/I_s should increase, tending to unity for linearly polarized pump radiation. The experimental curve has the following singularities: $I_a/I_s \neq 0$ for $\alpha = 0$, one of the reasons for this being partial depolarization of the laser beam at the entrance window of the hydrogen cell; as α increases, the ratio I_a/I_s increases more slowly. This may be due to a decrease in $\mathbf{E}_a$ because of a decrease in $\mathbf{E}_l$ caused by the formation of higher Stokes and anti-Stokes components for $\alpha \neq 0$. The second Stokes component, for example may occur as a result of parametric interaction similar to the formation of the anti-Stokes component. In this case, as α increases, the coupling between the second and first Stokes components and the pump wave increases, resulting in an increase in the intensity of the second Stokes component. For $\alpha \neq 0$, no coupling is found between these components and the second Stokes component does not appear.

Figure 3 shows the experimental dependences of the energy of the first and second Stokes components for the $S_{00}(1)$ rotational transition in the hydrogen molecule as a function of the angle α. The pump intensity is 2.2 times higher than the threshold intensity for rotational STRS when $\alpha = 0$. The energy of the first Stokes component is highest for $\alpha = 0$ and decreases with increasing α, mainly due to a reduction in the gain. The energy of the second Stokes component is nonzero for $\alpha = 0$ (this may be due to partial depolarization of the pump radiation at the windows and to the possibility of a cascade process) and increases with increasing α, indicating that the formation of the second Stokes component is a parametric process. For a certain optimum value of α the energy of the second Stokes component is highest and, as α increases further, it begins to decrease rapidly, which may be attributed to a decrease in the energy of the first Stokes component.

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