Observation of stimulated Raman scattering in a waveguide induced with vibrational excitation of hydrogen molecules

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Conical Stokes radiation was observed experimentally. This emission is explained in terms of the excitation of stimulated Raman scattering of light in a waveguide induced with vibrational excitation of hydrogen molecules.

It is known that in the presence of stimulated Raman scattering in hydrogen the upper level is filled efficiently in the $Q_{01}(1)$ vibrational transition.[1] In the process, the index of refraction of the medium n_0 increases by some amount Δn^2.

If it is assumed that the stimulated Raman scattering develops in a volume in the form of a thin and long cylinder, then this cylinder is an optical waveguide whose core and cladding have an index of refraction $n_0 + \Delta n$ and n_0, respectively. In addition to the Stokes wave, which propagates in the axial direction, the Stokes rays making appreciable angles with the axis of the waveguide, for which the condition of waveguide propagation is satisfied, in this case are also excited in the directed pump beam. As a result, apart from the spot corresponding to the axial component, a bright ring with diameter $D = 2\alpha F_2$, where F_2 is the focal length of the analyzer lens of the angular spectrum, and $\alpha \approx \sqrt{2\Delta n}$ is the angle corresponding to the limiting angle of total reflection in the optical waveguide relative to the laser beam, will be observed in the angular spectrum of the Stokes radiation of stimulated Raman scattering.

It is obvious that to observe stimulated Raman scattering in a waveguide consisting of excited molecules, the angular spectrum of the focused pump beam must be less than 2α; i.e., the relation $a/F_1 < 2\alpha$, where a is the diameter of the laser beam at the entrance to the cell with the hydrogen, and F_1 is the focal length of the focusing lens, must be satisfied. In addition, a clean experiment can be performed only at sufficiently low gas pressures ($P < 10$ atm), when the scattering angles of the higher-order Stokes and anti-Stokes components of stimulated Raman scattering, which satisfy the conditions of linear matching,[3] are negligible compared with the divergence of the laser pump beam at the exit from the cell. The inverse stimulated Raman scattering, which carries off a significant part of the energy of the laser, and which makes it more difficult to interpret the experimental results, in this case is suppressed, even in the case of single-frequency pumping. It is also necessary to take into account the fact that for a fixed energy of the laser pulse the larger relative number of excited molecules over a longer nonlinear interaction length corresponds to the lower density of the gas. All this means that the induced-waveguide effect, which accompanies the stimulated Raman scattering, is stronger at lower gas pressures.

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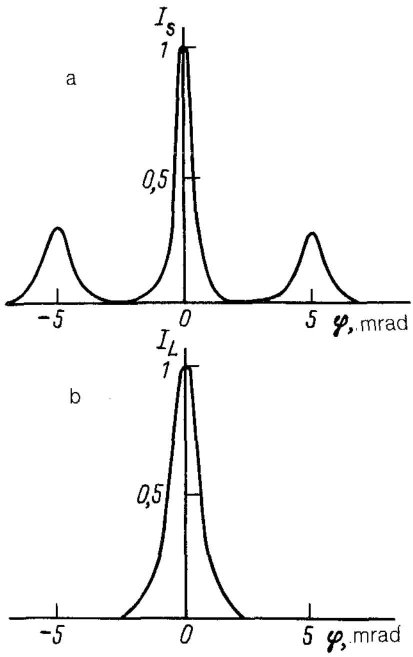

FIG. 1. The angular distribution of the intensity of the first Stokes component $I_s(\varphi)$ (a) and of the laser pump $I_L(\varphi)$ (b), obtained at $P = 2.4$ atm and $E_L = 15$ mJ.

In the experiment, single-mode second-harmonic radiation of a Nd:YAG laser with the following parameters was employed to excite stimulated Raman scattering in hydrogen: the pulse duration was 7 ns, the energy per pulse was $E_L \lesssim 30$ mJ, and the beam diameter was $a = 1.5$ mm. The laser beam was focused by means of a lens ($F_1 = 50$ cm) into a cell ($L = 64$ cm) containing compressed hydrogen ($P = 1$–25 atm). The angular spectrum of the scattered components was analyzed at the focal point of a second lens ($F_2 = 40$ cm).

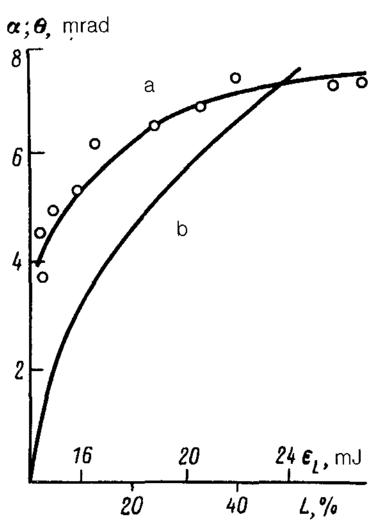

FIG. 2. Plots of the experimental dependence of θ on E_L (curve a) and the computed dependence of the maximum angle of total reflection α on the relative number Z of excited molecules (curve b).

The experiments showed that at pressures $P < 5$ atm the first Stokes component is generated on the axis of the laser beam and on the edges of a cone, whose solid angle 2θ is much greater than the divergence of the focused pump beam (see Fig. 1). As E_L is raised, the scattering angle θ gradually increases (see Fig. 2, curve *a*) and reaches a constant level. If the energy of the laser pulse is less than some critical value, then stimulated Raman scattering in the experiment was observed only on the axis. The increase in the experimentally observed angles of conical emission of the first Stokes component of stimulated Raman scattering with an increase in E_L is attributed to the increase in the relative number Z of excited molecules present in the volume, where the laser pump interacts with the molecular medium. According to Ref. 2, the relative number Z of excited molecules increases with increasing index of refraction of molecular hydrogen. This increases the computed maximum angle of total reflection α (see Fig. 2, curve *b*) in the optical waveguide. This increase is manifested on the experimental curve of $\theta(E_L)$. It follows from Fig. 2 that the angles θ of conical emission of stimulated Raman scattering radiation, which were obtained experimentally at laser pulse energies ($E_L > 24$ mJ), for which the Raman-active transition is saturated, are in good agreement with the value of the maximum angle of total reflection α calculated with $Z \simeq 50\%$.

The generation of the waveguide component of the Stokes component significantly affects the energy characteristics of stimulated Raman scattering as a whole. A decrease of the gas pressure in the range $P = 10$–15 atm for fixed pump energy (see Fig. 3, curve *a*) results in weakening of the associated stimulated Raman scattering; this weakening is caused by four-photon parametric Stokes–anti-Stokes processes, which decrease the rate of growth of the Stokes wave as the wave mismatching decreases.[3] There is no conical stimulated Raman scattering in this pressure range. Further reduction of the hydrogen pressure results in stabilization and some increase in the energy of the Stokes component at the exit from the cell. This behavior is

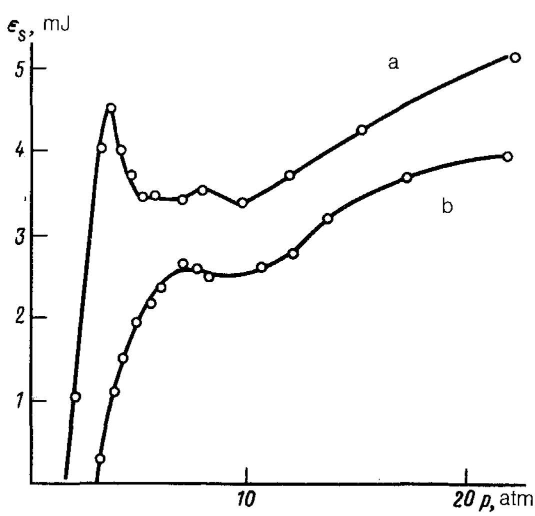

FIG. 3. The energy E_S of the first Stokes component of stimulated Raman scattering as a function of the gas pressure P for $E_L = 18$ mJ (curve *a*) and $E_L = 10$ mJ (curve *b*).

attributed to the appearance of a conical component of stimulated Raman scattering that is not parametrically coupled with the anti-Stokes radiation. At pressures in the range 3.5–5 atm the energy of the axial component of stimulated Raman scattering continues to decrease as the gas pressure decreases, while the energy of the conical radiation increases. At $P = 3.5$ atm the energy of the pulse of conical stimulated Raman scattering is higher than the energy of the axial component.

At low intensities of the laser which excites the stimulated Raman scattering, the relative number of vibrationally excited molecules in the medium decreases, causing the induced waveguide effect to weaken. Thus, for example, at $E_L = 10$ mJ the conical stimulated Raman scattering radiation is not observed in the entire range of pressures studied; this is also manifested in the behavior of the $E_s(P)$ curve (see Fig. 3, curve b).

We have thus observed experimentally the generation of conical Stokes radiation in a waveguide induced in molecular hydrogen as a result of the motion of the populations under conditions of stimulated Raman scattering.

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