EFFECT OF OPTICAL BREAKDOWN ON STIMULATED RAMAN SCATTERING OF XeCl-LASER RADIATION IN GASES AND VAPORS

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The results of experiments on SRS conversion of XeCl-laser radiation in hydrogen and lead vapors with different pump-beam divergence are presented. It is shown that the conversion efficiency is limited primarily by optical breakdown, whose effect increases as the energy and divergence of the converted radiation increase. The total quantum efficiency for conversion of XeCl-laser radiation with a divergence of 0.1 mrad *in hydrogen was* ~ 90%.

The development of laser sources with pulse energy 1–10 J and total efficiency ≳ 1% in the blue-green region of the spectrum remains an important problem in connection with practical applications, such as ranging and sounding of the atmosphere. One of the most promising methods for achieving this goal is stimulated Raman scattering (SRS) of exciplex-laser radiation in gases and vapors. However when plane-parallel cavities are used the output beam of such lasers has a large divergence, which makes it impossible to achieve efficient SRS conversion. The beam divergence can be reduced to practically the diffraction limit either with the help of unstable cavities with a large gain (at the expense of energy) or a generator-amplifier system (at the expense of simplicity of the apparatus). The compromise solution is to use simple types of unstable resonators, which make it possible to obtain an output beam with a divergence 10–100 times higher than the diffraction limit. The efficiency of SRS conversion in this case can be quite high and can reach 7054 (the total quantum efficiency).[1–3] However the problem of increasing the efficiency of such systems as well as the possibility of increasing the energy of the converted radiation has virtually not been studied.

In this work we studied the effect of the divergence and the energy of the pump radiation on the efficiency of SRS. The total quantum efficiency of SRS conversion of XeCl-laser radiation was equal to 90% in hydrogen and 37% in lead vapor. It is shown that the main process that competes with SRS is optical breakdown, whose effect is all the stronger the lower the quality of the beam and the higher the energy of the converted radiation.

The experiments were performed with LIDA-101 (Ref. 4) and LIDA-KT (Ref. 5) lasers, whose active length is equal to 80 and 60 cm, respectively. Three types of unstable cavities were employed: a partially misaligned plane-parallel cavity,[4] a cavity consisting of a concave mirror with $R = 5$ m and a focusing lens with $F = 60$ cm,[6] and a cavity consisting of plane and convex ($R = 1$ m) aluminum-coated mirrors.

In the case when SRS conversion in hydrogen was studied the laser beam was focused with a lens with $F = 60$ cm into a stainless-steel 1 m long and 5 cm in diameter tube equipped with a side window. For SRS conversion in lead vapors a ceramic 50 cm long and 2 cm in diameter tube, on whose outer wall a heater was placed, was employed. In the cold state the cell is filled with helium as a buffer gas up to a pressure of 500 torr. The radiation spectrum was recorded with the help of ISB-30 and ISB-51 spectrographs and the radiation energy was recorded with the help of an IMO-2 N calorimeter using light filters to separate the components. The temporal characteristics of the radiation were recorded with the help of S8-12 and 6 LOR oscillographs. The signal was fed to them from an FEK-22 photodiode placed behind the output slit of an MDR-2 monochromator.

In the case of SRS conversion of XeCl-laser radiation in hydrogen four Stokes and three anti-Stokes components were obtained in the visible and UV regions of the spectrum.[7] The conversion efficiency depended on the divergence of the laser beam and the energy of the laser radiation. Thus the first of the cavities described above made it possible to obtain a beam with a divergence of ~ 2 mrad. The total quantum efficiency of conversion of such a beam did not exceed 30%. The second type of cavity formed radiation with a divergence of ~ 0.8 mrad. The total quantum efficiency of conversion in this case reached 70%. It was found that increasing the energy of the converted radiation without decreasing the divergence of the beam can result in lower conversion efficiency. In addition, the conversion efficiency varies appreciably during the pulse (Fig. 1). When the pump power density at the focal point of the lens exceeds 170 MW/cm^2 the conversion efficiency starts to drop, in spite of the fact that the SRS gain is proportional to the pump power density.

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These data suggest that there exists a nonlinear process that competes with the SRS process.

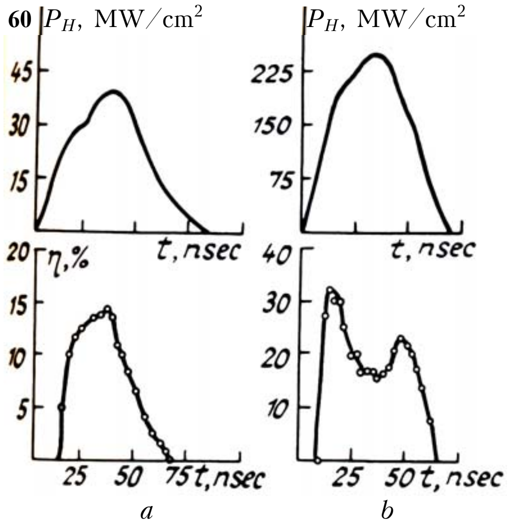

FIG. 1. Oscillograms of pulses of incident radiation and the efficiency of conversion into the first Stokes component of the SRS in hydrogen as a function of time for peak pump power density $P_p = 40$ *(a) and* 250 *(b)* MW/cm^2. *The pump beam divergence was equal to* 0.8 mrad. *The hydrogen pressure was equal to* 16 atm.

Visual observations in the side window of the cell revealed the existence of optical breakdown in the gas, during which strong scattering of the incident and converted radiation in the focal region of the lens was recorded. Figure 2 shows the threshold power density for SRS conversion and optical breakdown as a function of the hydrogen pressure.

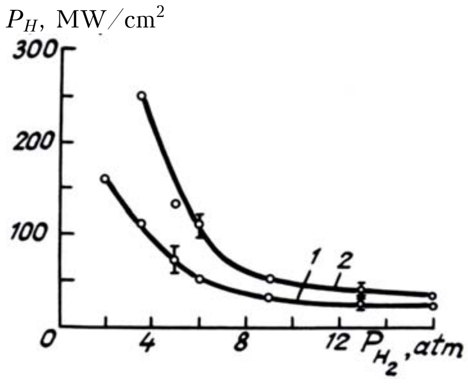

FIG. 2. The thresholds of SRS conversion (1) and optical breakdown (2) as a function of the hydrogen pressure. The divergence of the incident radiation is equal to 0.8 mrad.

For powerful UV radiation with large divergence the threshold of optical breakdown is quite low. This is explained, on the one hand, by the multiphoton ionization of the gas by UV radiation and on the other by the large diffusion length Λ, which determines diffusive losses of electrons and is of the order of the radius of the beam at the waist[8]:

$$\Lambda \sim r = F\Theta/2, \qquad (1)$$

where F is the focal length of the lens and Θ is the divergence of the radiation.

The complete picture of the process can be obtained by analyzing the oscillograms of the incident radiation, the converted radiation, and the pump radiation, scattered from the focal region of the lens into the side window (Fig. 3). After the SRS threshold is reached ($d_1 = 5$ ns) the converted radiation starts to increase (Fig. 3b, curve 1). The power density of the radiation at the focal point continues to increase, reaching at $t = t_2 = 8$ ns the threshold of optical breakdown (Fig. 2). At $t_3 = 15$ ns the density of the plasma formed is high enough that the plasma can appreciably affect the light flux: the power of the converted radiation starts to decrease and at the same time the intensity of the radiation scattered into the side window increases (Fig. 3c).

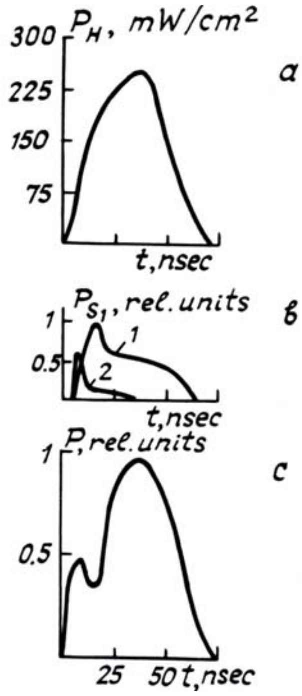

FIG. 3. Oscillograms of the incident radiation pulse (a) and the first Stokes component (b) in hydrogen (1) and in lead vapors (2) and the oscillogram of radiation scattered into the side window of the chamber (c). The divergence of the pump beam was equal to 0.8 mrad, *the hydrogen pressure was equal to* 16 atm.

To suppress optical breakdown its threshold must be increased without changing the SRS threshold. Thus increasing the diffusive losses by decreasing the beam divergence to 0.1 mrad (1) using a cavity of the third type made it possible to increase the conversion efficiency substantially. The total quantum yield for conversion of a 10 mJ beam was equal to ~ 90%. It should be noted that when the beam energy was increased to 100 mJ the threshold of optical breakdown (~ 10 GW/cm^2) was exceeded and correspondingly the efficiency dropped to 70%.

In experiments on SRS conversion of XeCl-laser radiation in lead vapors only a cavity of the second type was employed. Characteristically conversion in the visible range (λ = 458 nm) stopped if the pump power density exceeded a definite magnitude. In addition, radiation in a wide spectral range, characteristic for plasma formed by optical breakdown, was observed at the output of the cell, conversion efficiency was equal to 37%, which is less than the results obtained with beams of higher quality.

Thus optical breakdown is the main process competing with the process of SRS conversion of exciplex-laser radiation in gases and vapors. It limits the efficiency of conversion of radiationwith high per pulse energy. Efficient conversion of radiation of wide-aperture lasers with radiation energy of 1–10 J and higher requires either that the divergence of the output beam be reduced to the diffraction limit or "soft" focusing and collimated beams be employed.[9,10]

In conclusion we present data on the quantum efficiency η of conversion of laser radiation in hydrogen (E_p = 10 mJ) and lead vapors (E_p = 50 mJ) into different SRS components and with different pump beam divergence. The efficiency of conversion of radiation in hydrogen into S_4 (λ = 631 nm), AS_2 (λ = 246 nm), and AS_3 (λ = 223 nm) was insignificant and totalled ≲ 1 (Table I).

TABLE I.

θ mrad	η_{s1}, % (λ = 353 nm)	η_{s2}, % (λ = 414 nm)	η_{s3}, % (λ = 499 nm)	η_{as1}, % (λ = 273 nm)	η_{Σ}, %
Hydrogen					
2	10	12	3	3.5	28.5
0.8	40	20	8	5	73
0.1	50	24	13	3.5	90.5
Lead vapors					
0.8	37	—	—	—	37

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