Stimulated Raman scattering of picosecond optical radiation in an extended dispersive medium

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A study was made of the influence of the dispersion of the group velocities in a medium with inhomogeneous vibrational resonances on the process of stimulated Raman scattering in the field of picosecond radiation. When the wavelength of 30-psec pump pulses was varied near zero dispersion of the group velocities in fused quartz (1.17–1.35 μ), the frequency shift of the Stokes component of stimulated Raman scattering relative to the pump line varied practically from zero to 500 cm^{-1} in a quartz fiber waveguide. The effect was attributed to the difference between the spectral dependences of the length of the region of interaction of the pump and Stokes StRS waves, governed by the dispersion-induced separation of the envelopes of the pulses of different frequencies when the pump wavelength was varied. As a result the spectrum of the increment of the coherent stimulated Raman gain for picosecond pumping depended on the pump wavelength and differed considerably from the usual Raman gain spectrum. Consequently, variation of the pump radiation frequency resulted in successive excitation of coherent molecular vibrations of different energies from the large set of inhomogeneous vibrational resonances of the medium in the case of stimulated Raman scattering.

1. INTRODUCTION

Stimulated Raman scattering (StRS) of light excited by a field of picosecond laser pulses in various liquid, gaseous, and condensed media is of considerable interest from the fundamental point of view and also because of applications (for a review see Auston's paper in Ref. 1). New opportunities are opening up not only for construction of tunable converters of picosecond radiation, but also in studies of transient StRS (when the pumping or excitation time τ_p is comparable with the dephasing time τ_d of molecular vibrations) and, consequently, determination of τ_d. Moreover, the dynamics of StRS excited by picosecond pulses should differ greatly from the steady-state regime and this is related not only to the ratio of the pumping and dephasing times. The effects of dispersion of the group velocities begin to play an important role in the picosecond range and these effects influence the length of the region of interaction of the pump and Stokes StRS waves.[2]

Among the many nonlinear media a special place is occupied by disordered media with inhomogeneously broadened vibrational resonances, usually characterized by low values of the nonlinear coefficients. Nevertheless, the process of StRS in such media (for example, in water) is effective in the picosecond range.[3] One of the most widely used materials which to a greater or lesser extent is subject to laser radiation is the glass in the form of fused quartz. Studies of StRS in glasses are particularly important, especially from the point of view of the influence of StRS on lasing processes, particularly on generation of picosecond pulses in activated glass lasers, and on the processes of propagation of picosecond pulses in glass fiber waveguides.[4]

Intensive studies of StRS in glasses have started since the appearance of quartz fiber waveguides with low optical losses,[5] which not only have made it possible to reduce the threshold power for the process, but also to avoid radiation self-focusing which splits a laser beam into filaments of higher power density and which makes it difficult to interpret the experimental results.[1]

We shall mention several features which deserve close attention in the selection of fiber waveguides for the investigation of StRS as a result of picosecond excitation: 1) the optical losses should be low, the transverse dimensions should be small (the waveguide diameter should be of the order of tens of microns), and it should be possible to retain high spatial coherence of single-mode laser radiation in extremely long single-mode fiber waveguides; 2) the spectrum of spontaneous Raman scattering (inset in Fig. 1) should be

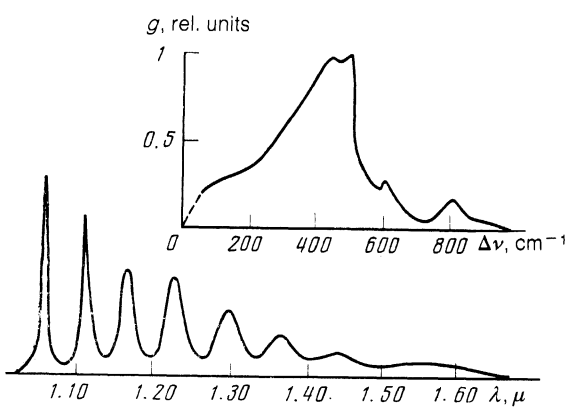

FIG. 1. Overall spectrum of multistage StRS in a fiber waveguide of length 250 m pumped with pulses of $\lambda_p = 1.064\,\mu$ wavelength and $\tau_p \approx 150$ nsec duration. The inset shows the spectrum of the StRS gain g of fused quartz ($\lambda_p = 1.064\,\mu$) plotted using the data of Ref. 6.

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wide, extending practically from zero to thousands of reciprocal centimeters or even further, and this spectrum should reflect the density of inhomogeneously broadened overlapping vibrational resonances of the medium due to various vibrational modes exhibited by the structure of fused quartz[7]; 3) there should be a wide trasparency "window" from 0.4 to 1.8 μ and it should have regions with positive and negative dispersion of the group velocities, as well as with zero dispersion in the region of the minimum of the optical losses at wavelengths of $\sim 1.3\,\mu$ (Ref. 8). It should be added that the use of fiber waveguides as objects for investigation, particularly of StRS, extends greatly the experimental opportunities for the use of continuously tunable highly stable picosecond pump radiation sources operating at a high repetition frequency and characterized by relatively low powers.[9]

The influence of the group delay on the dynamics of the StRS process, which should be manifested most fully in long nonlinear media, has been practically neglected. The combination of the properties of quartz fiber waveguides listed above together with the use of picosecond tunable light sources makes it possible to study the relationship between the characteristics of the medium such as the dispersion and an inhomogeneous set of vibrational resonances, on the one hand, and StRS growing from the spontaneous noise level, on the other. A study of this relationship was the main task of the present investigation.

2. WIDE-RANGE PICOSECOND SPECTROMETER

A picosecond spectrometer was constructed on the basis of a tunable (in the range 0.74–1.9 μ) parametric oscillator and an optical multichannel analyzer. In the para metric oscillator a nonlinear barium sodium noibate ($Ba_2NaNb_5O_{15}$) crystal was employed; the wavelength was tuned by varying the temperature of the crystal, which was pumped synchronously with the second harmonic ($\lambda = 0.532\,\mu$) of a neodymium garnet laser pumped continuously and operated in the active mode-locking and Q-switching regime.[10] The parametric oscillator generated 200-nsec trains of ultrashort pulses at intervals of 4 nsec and the repetition frequency of such emission was 400 Hz. Ultrashort pulses having no time substructure were of ~ 30 psec duration at midamplitude.

The radiation from the parametric oscillator was coupled into a quartz fiber waveguide of length $L = 250$ m (the diameter of the waveguide core was $\sim 10\,\mu$ and the difference between the refractive indices of the core and cladding was $\sim 4\times 10^{-3}$). The radiation emerging from the waveguide was focused on the entry slit of a grating polychromator and a vidicon was placed in the plane of the exit slit of the polychromator. The optical multichannel analyzer made it possible to determine the overall emission spectrum after several laser shots and to analyze this spectrum by a computer.

The time characteristics of the radiation were determined employing an autocorrelator on the basis of the width of zero-background autocorrelation functions of the second order in the case of noncollinear generation of the second harmonic in a nonlinear ($LiIO_3$) crystal.[1]

3. EXPERIMENTAL RESULTS AND DISCUSSION

The nature of StRS in an extended dispersive medium with a wide spectrum of vibrational resonances depends very strongly on the duration of the pump pulses. Figure 1 shows the spectum of multistage StRS resulting from the pumping of the waveguide with long and smooth (~ 150 nsec) pulses from a neodymium garnet laser ($\lambda = 1.064\,\mu$) operating in the Q-switching regime. In this multistage StRS the radiation of each Stokes component was the pump for the next component and as a result the emission spectrum covered the full infrared transparency range of fused quartz.

The frequency shifts of the Stokes components relative to the pump lines amounted to 440–490 cm^{-1} and were governed by the maxima of the Raman gain curve of Fig. 1 (this curve was obtained from the spectrum of the spontaneous Raman scattering[6] allowing for the important factors of the wavelength and thermal population of the vibrational states[11]). In the case of such long submicrosecond excitation the dispersion of the waveguide played practically no role and the length of the region of interaction between the pump and Stokes waves was equal to the full length of the waveguide and the threshold of appearance of StRS growing exponentially from the spontaneous noise was attained specifically for the Stokes components with the maximum values of the gain $g(\Delta\nu)$ [$g_{max}(\Delta\nu)\approx 10^{-11}$ cm/W at $\lambda = 1.06\,\mu$ (Ref. 5)].

The situation was radically different in the case of picosecond excitation of StRS when the dynamics of the process began to be affected by the dispersion of the group velocities, transforming greatly the spectrum of the gain increment $g(\Delta\nu)I_pL_{eff}$ because of a change in the effective length (L_{eff}) of the region of interaction between the pump and Stokes waves. In our case when the pump pulses were fairly long (30 psec) the inequality $\tau_p \gg \tau_d$ was obeyed[12] and the process of StRS in fused quartz was of the steady-state nature. The effect of phase self-modulation, associated with the dependence of the refractive index of the medium on the radiation intensity, before the appearance of StRS in the waveguide broadened the pump spectrum to a width not exceeding 20 cm^{-1} (Ref. 9), which should have had practically no influence on StRS because of the very large width of the Raman gain spectrum (Fig. 1). It should be pointed out that before the onset of StRS the process of phase self-modulation did not deform (because the dispersion of the group velocities was too small) the pump pulses in the 250-m long optical waveguide.

Figure 2 shows the StRS spectra obtained on variation of the wavelength of the 30-psec pump pulses of 300 W power: the wavelength was increased from 1.17 to 1.35 μ (the spectral resolution was 6.3 nm). Clearly, in the case of pumping at the shortest wavelength ($\lambda_p \approx 1.18\,\mu$) the StRS spectrum consisted of two wide-band Stokes components, one of which was stronger and overlapped the pump radiation spectrum and the other one was weaker. When λ_p was increased to 1.26 μ, the Stokes component had a clear maxi-

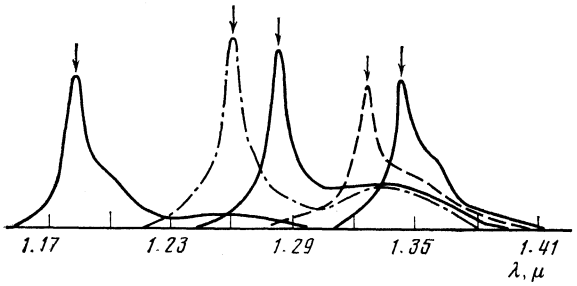

FIG. 2. StRS spectra of a waveguide 250-m long obtained on variation of the wavelength λ_p (identified by arrows) of pump radiation in the form of $\tau_p = 30$ psec pulses. Spectral resolution 6.3 nm.

mum and its center was shifted by about 450 cm^{-1} relative to the pump line. The subsequent increase of λ_p to 1.32–1.35 μ reduced considerably the Stokes shift of the StRS component relative to the pump line (Fig. 2). (It should be noted that at $\lambda_p = 1.17$–$1.35\ \mu$ only one fundamental mode was excited in the waveguide.)

We shall now consider in greater detail the effect of length of the region of interaction between the pump and Stokes waves, and its relationship to the dispersion and radiation spectrum, and we shall also consider its role in the StRS process illustrated in Fig. 2. In the case of steady-state coherent StRS[13] when the phase mismatch between the pump radiation and the Stokes component of the StRS can be ignored, the effective length of the region of interaction between the pump and Stokes waves is governed by the "divergence" length of the envelopes of their pulses:

$$L_d(\lambda_p,\lambda_s) = \tau_p \Big/ \left| \frac{1}{v(\lambda_p)} - \frac{1}{v(\lambda_s)} \right| . \qquad (1)$$

Here, $v(\lambda_p)$ and $v(\lambda_s)$ are the group velocities of the pump and Stokes waves. The effective length of interaction between these waves is then

$$L_{eff}(\lambda_p,\lambda_s) = \begin{cases} L_d(\lambda_p,\lambda_s) & \text{for } L_d \leqslant L \\ L & \text{for } L_d \leqslant L \end{cases} . \qquad (2)$$

It follows from Eqs. (1) and (2) that the effective length of the interaction region between waves of different frequencies is governed by the spectral dependence of the dispersion of the group velocity and by the wavelengths of the pump and Stokes radiation. Our waveguide was characterized by zero dispersion of the group velocities $D = 0$ [$D \equiv d(1/v_g)/d\lambda$] at the wavelength $\lambda_0 = 1.295\ \mu$, by a positive dispersion of the group velocities ($D < 0$) in the range $\lambda < \lambda_0$, and a negative dispersion ($D > 0$) for $\lambda > \lambda_0$. In the spectral range $\lambda = 1.15$–$1.37\ \mu$ the dispersion was $|D| < 10$ psec·nm^{-1}·km^{-1} (Ref. 8) (the linear losses in this range were less than 10 dB/km).

Figure 3 shows the dependences of L_{eff} on the Stokes wavelength λ_s calculated from Eqs. (2) and (1) using measurements of the waveguide dispersion[8] for six values of the pump wavlengths corresponding to the spectra in Fig. 2. It is clear from Fig. 3 that the maximum length of the region of interaction between the pump and Stokes waves, equal to the total length of the waveguide, corresponded either to small

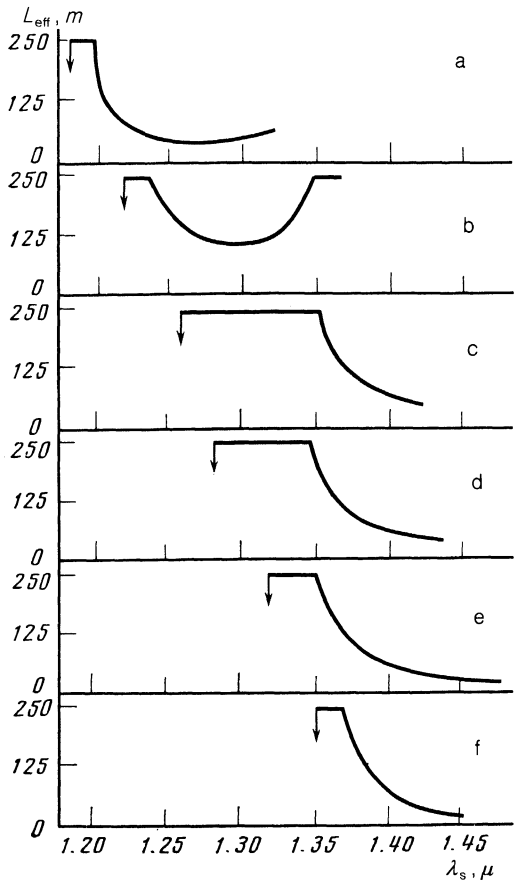

FIG. 3. Dependences of the effective length of the region of interaction between the pump and Stokes waves on the Stokes wavelength λ_s calculated for different pump wavelengths λ_p (identified by arrows) allowing for the dispersion of the group velocities in the case of pulses of $\lambda_p = 30$ psec duration.

frequency shifts between the pump and Stokes lines (Figs. 3a, 3e, and 3f) or occurred in the case of symmetric positions of λ_p and λ_s relative to the zero-dispersion wavelength $\lambda_0 = 1.295\ \mu$, when the group velocities of the StRS and pump waves coincided ($L_d \to \infty$) (Figs 3b–3d).

Thus, in the case of long excitation the StRS shift in fused quartz was governed by the maximum in the gain spectrum of $g(\Delta\nu)$, whereas in the case of picosecond pumping the frequency shift $\Delta\nu$ was generally governed by the product $g(\Delta\nu)L_{eff}(\lambda_p,\lambda_s)$. (Here and later we shall assume that the intensity of the pump radiation was constant in the investigated spectral range.) Figure 4 shows the functions obtained by multiplying the $g(\Delta\nu)$ gain curves (inset in Fig. 1) and the effective length (Fig. 3), yielding $g(\Delta\nu)L_{eff}(\lambda_p,\lambda_s)$ for all the pump wavelengths λ_p shown in Fig. 3. It is clear from Fig. 4 that the functions $g(\Delta\nu)L_{eff}(\lambda_p,\lambda_s)$ representing the StRS gain increment differed considerably for different values of the pump wavelength and had maxima at positions quite different from those in the StRS gain spectrum (Fig. 1), which was the reason why StRS exhibited variable (including small) frequency shifts (Fig. 2).

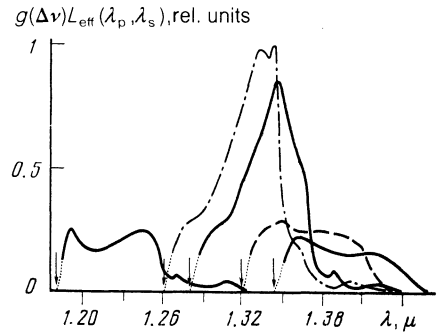

FIG. 4. Spectral dependences of the StRS gain increment (I_p = const) obtained for different pump wavelengths λ_p (identified by arrows) as a result of multiplication of the $g(\Delta\nu)$ curves from Fig. 1 and $L_{eff}(\lambda_p,\lambda_s)$ from Fig. 3.

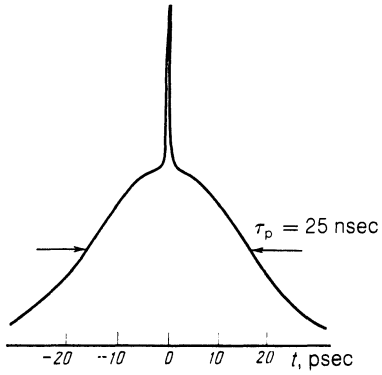

FIG. 6. Autocorrelation function radiation at the Stokes wavelength ($\lambda_s \approx 1.33\,\mu$).

We used the spectra in Fig. 4 to determine for each pump wavelength λ_p the frequency shift at which the function gL_{eff} reached its maximum and this enabled us to plot the dependence of the frequency shifts of the StRS components relative to the pump line as a function of the pump wavelength $\Delta\nu(\lambda_p)$ (continuous curves in Fig. 5). A discontinuity of the function $\Delta\nu(\lambda_p)$ in the region of the wavelength of $\lambda_p \approx 1.2\,\mu$ was due to an approximate equality of the values of the Raman gain increment $g(\Delta\nu)L_{eff}(\lambda_p,\lambda_s)$ for two frequency shifts $\Delta\nu$ (Fig. 4), which in the StRS saturation regime accounted for the simultaneous generation of two Stokes components (Fig. 2) when the pump wavelength was $\lambda_p = 1.18\,\mu$. We included in Fig. 5 the experimental values of the frequency shifts of the Stokes components of StRS obtained in the course of continuous variation of the pump wavelength from 1.15 to 1.35 μ. The scatter indicated in this figure was due to indeterminacy in the position of the central wavelength in the Stokes spectrum and due to the finite spectral resolution of the polychromator. Clearly, the experimental points fitted well the calculated curve, thus confirming the model of the StRS process with an allowance for the group delay effects proposed in the present paper.

It should be noted that the noise origin of the Raman scattering growing from spontaneous fluctuations was manifested also in the StRS saturation regime. Figure 6 shows the autocorrelation function determined by the method of Ref. 9 at the Stokes wavelength ($\lambda_s \approx 1.33\,\mu$). This function has a characteristic narrow spike at the center with the contrast of two and in real time it corresponded to a pulse of 25-psec duration, which was somewhat less than the duration of the pump pulses (τ_p = 30 psec) filled with a noise structure of characteristic fluctuation duration ~0.2 psec. This noise structure of the pulses was mainly responsible for the considerable spectral broadening of the Stokes components (Fig. 2). In contrast to StRS pulses amplified from a regular test Stokes wave because of phase self-modulation,[4] such noise StRS pulses were of limited interest because of their self-compression and formation of picosecond pulses in the region of negative dispersion of group velocities.

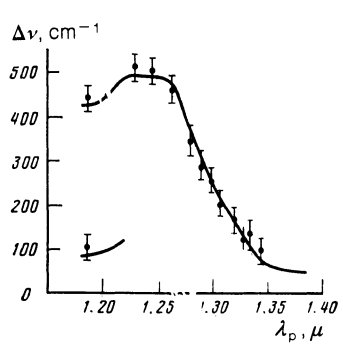

FIG. 5. Dependence of the frequency shifts of the Stokes components of StRS relative to the pump line calculated as a function of the pump wavelength using the data of Fig. 4. (In these calculations an allowance was made for some dependences of gL not shown in Fig. 4.) The experimental points $\Delta\nu(\lambda_p)$ are superimposed on the graph.

4. CONCLUSIONS

It follows from the results of the present investigation that selection of glass fiber waveguides as the objects for investigation together with a frequency-tunable picosecond excitation source made it possible to study the influence of the dispersion of the group velocities on the StRS process. Variation of the wavelength of the picosecond pump pulses in the region of the positive and negative dispersion of the group velocities of the medium resulted in variation of the effective length of the region of interaction between the pump and Stokes component and, consequently, it altered the spectral dependence of the StRS gain increment. Consequently, it was possible to use stimulated scattering of light to excite single coherent molecular vibrational modes from a large set of about ten inhomogeneously broadened vibrational resonances[7] and this was done by varying the excitation energy practically from zero to 500 cm^{-1}. In addition to the known spectroscopic methods involving spontaneous Raman scattering, StRS amplification of a test wave, active Raman scattering spectroscopy,[14] which in the case of biharmonic tunable laser pumping can be used to study selectively excited different vibrations of fused quartz,[12] the above StRS method with picosecond pumping provides an opportunity for obtaining important spectroscopic information on the

intensities of Raman-active inhomogeneous vibrational resonances in strongly disordered glass in media. Tuning of the picosecond pump wavelength near zero dispersion of the group velocities makes it possible to suppress effectively the generation of radiation corresponding to the intensity maximum of the Raman gain band and to control the StRS emission frequency within wide limits.

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