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ANOMALOUS BROADENING OF SPECTRAL LINES IN NONLINEAR LIQUIDS AND ITS EFFECT ON STIMULATED SCATTERING PROCESSES

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Results of an experimental investigation of the spectral structure of laser radiation passing through a self-focusing liquid are reported. For complete isolation of the generator from the scattering medium, the second harmonic of a neodymium laser is employed as the radiation source. The generator can operate under incoherent-mode or mode-locked conditions. The spectral structure of the radiation was investigated in the direction of the incident light as well as at an angle of 180°. Broadening of a quasicontinuous nature and broadening involving a set of discrete lines, the distance between which was, as a rule, not related to the incident radiation spectrum, were recorded. An analysis of the experimental results shows that the spectral line broadening may be ascribed to interaction between space and time modulation of the beam during its self-focusing. Some data pertaining to stimulated Raman scattering under conditions of anomalous broadening of a laser pulse spectrum are reported. In particular, the strong sensitivity of the scattering indicatrix to modulation of the initial radiation is noted.

1. INTRODUCTION

THE anomalous broadening of spectral lines, occurring when intense light beams propagate in liquids (see, for example,[1-6]), still remains among the least explained problems in nonlinear optics, and calls for further research. In spite of the fact that by now a large number of models have been proposed for the broadening of spectral lines (all of which resort in one form or another to the nonlinear effects connected with the dependence of the refractive index on the intensity; see[1-3,7-14]), it is very difficult to apply them to a clear cut quantitative interpretation of the experimental facts. The latter is connected with two circumstances. First, there is no doubt that the experimentally observed broadenings are connected with a complicated interaction of various mechanisms. On the other hand, in a large number of respects the experimental data are insufficient, and sometimes even simply contradictory (the latter pertains in particular to the question of the role of the mode structure of the scattering radiation and to the observation of a discrete structure in broadened spectra; see[2,4,13]).

The question of the anomalous broadenings of spectral lines in nonlinear media was first distinctly formulated by Bloembergen and Lallemand[1], who observed broadening of SRS lines in a multimode pumping field, and who presented a simple interpretation of the effects on the basis of the quasistatic theory of the conversion of amplitude modulation into phase modulation in a nonlinear medium (AM → PM conversion). Further investigations[2,3] that broadening occurs not only in the SRS lines, but also in the laser line itself. Whereas the broadening obtained using the radiation of a multimode ruby laser[2] fitted well the AM → PM conversion scheme, Brewer[3] observed, with a single-mode laser, broadening of a different type, having the character of a relatively small (not more than 10 cm^{-1}) and variable frequency shift. Although the causes of such shifts (particularly the causes of variations of the frequency shifts) remained unclear, Brewer uniquely established that the broadening of this type is due to spatial self-focusing of the beam.

In succeeding investigations, two types of broadening of the laser lines and of the SRS were observed—broadening in the form of a continuous spectrum[4,5] and in the form of a discretely continuous spectrum[2,4,13]. The absolute values of the broadenings reached in some cases 100—cm^{-1}.

The present investigation was undertaken primarily

to refine the existing data and to accumulate new experimental data on the broadening of spectral lines in nonlinear media. In the formulation of the experiment we started from the following considerations.

1. The overwhelming majority of the experimental investigations cited above were performed with ruby lasers. In many cases, no measures were taken to decouple the generator from the scattering medium. Under such conditions, an appreciable contribution to the broadening of the spectral lines may be made by the mixing in the medium of the spectral components of the fundamental radiation and the components scattered at 180° and returning to the medium after being amplified in the generator (according to[15], effects of optical of optical-photon mixing are capable of producing anomalous broadening by 20—40 cm^{-1}). An important role in such an experimental geometry, as noted by Fabelinskiĭ and co-workers[16], may be played by the "trigger" amplification of weak modes.

It was deemed advisable to perform the experiments in an "absolutely" decoupled system; very convenient sources, from this point of view are optical-harmonic generators[17]. Therefore we used in our experiments single-mode and multimode second-harmonic neodymium-laser generators ($\lambda = 0.53\ \mu$). The choice of such a source is of interest also from another point of view. According to[13], one of the possible causes of the appearance of periodic modulation in the envelope of the broadened spectrum is the excitation of new internal motions in the medium; one could expect the condition for their excitations to be different for different wavelengths.

2. Different broadening mechanisms (AM-PM conversion, stimulated Rayleigh-wing scattering (SRWS), broadening due to cross modulation of the pulses, broadening due to nonstationary character of the self-focusing) should obviously be differently manifest in the spectrum of the radiation scattered in the forward and backward direction. Simultaneous registration of the spectra of the emission scattered in two opposite directions is particularly important obviously, in the case of complete decoupling of the generator from the scattering medium.

3. A promising method for experimentally separating different broadening mechanisms is to vary the duration of the laser pulses. In the investigations cited above, only nanosecond laser pulses were used; it was deemed of interest to carry out similar measurements also with picosecond pulses, of duration $\tau \simeq 10^{-11}$—10^{-12} sec.

4. Finally, the mutual influence of the broadening of the spectral lines and of effects of stimulated scattering is worthy of a more detailed investigation. Besides the already discussed influence of the stimulated scattering on the broadening (SMBS and SRWS can themselves cause anomalous broadening), worthy of attention is also the inverse problem, that of the influence of anomalous broadening on certain characteristics of stimulated scattering. We present below the results of experiments performed in accordance with the foregoing considerations.

2. EXPERIMENTAL SETUP

The experiments were performed using the second harmonic of a neodymium laser ($\lambda = 5300$ Å) as the source of powerful light beams. The active element (10×120 mm, glass LGS-224) was pumped with two IFP-2000 lamps connected in series. The rod and of the lamps were cooled with distilled water. To separate one axial mode, the output mirror was a Fabry-Perot type selector, consisting of two quartz plates 15 mm thick with distance between them 0.01 mm. Separation of one transverse mode was with the aid of a diaphragm with diameter d = 2 mm, placed between the crystal and the selector. The generator radiation was amplified with two amplification stages. The amplifiers were neodymium rods 16×240 mm, pumped with IFP-5000 lamps. Synchronous generation of the second harmonic was effected in a KDP crystal ($10 \times 10 \times 40$ mm); the second-harmonic generation was cut off from the fundamental radiation by a SDS-21 filter. The parameters of the second-harmonic were as follows: energy 0.04 J, $\tau_p \approx 10$—12 nsec, integral power $P \approx 3$ MW at a power density $I \approx$ MW/cm^2. The second-harmonic spectrum was monitored with a Fabry-Perot interferometer (dispersion region $\Delta\nu \approx 0.16$—1.6 cm^{-1}). The scattering medium was carbon disulfide (cell length 10—30 cm). The spectral composition of the radiation scattered in the forward and backward directions was registered with a DFS-8 diffraction spectrograph (linear dispersion 6.2 Å/mm).

3. EXPERIMENTAL RESULTS

Broadening of Second Harmonic Spectrum ($\lambda = 5300$ Å) and of First SRS Stokes Component. Form of Emission Spectra Scattered in the Forward and Backward Directions

The experimental setup is shown in Fig. 1. The radiation scattered in the forward direction illuminated the upper half of the spectrograph slit; the back-scattered radiation illuminated the lower half. Thus, in each flash we registered simultaneously the emission spectrum scattered in the forward and backward directions. Two types of spectra were observed: monotonic broadening and a discrete structure (Fig. 2). An appreciable deformation of the spectrum was observed only in the radiation scattered in the forward direction, and in this case both the pump spectrum and the spectrum of the first Stokes component of SRS were broadened. The widths of the Mandel'shtam-Brillouin components and of the back-scattered SRS Stokes com-

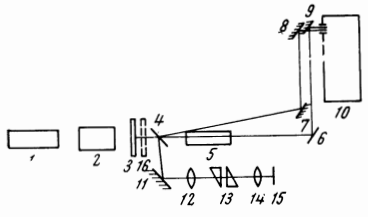

FIG. 1. Experimental setup for the investigation of the spectral characteristic of radiation scattered forward and backward: 1 – laser with two amplification stages, 2 – KDP crystal frequency doubler, 3 – SZS-21 filter transmitting the second-harmonic generation $\lambda = 0.53\mu$, 4, 6 – beam-splitting plates, 5 – cell with liquid, 7, 8, 9, 11 – aluminum-coated mirrors, 10 – DFS-8 spectrograph, linear dispersion 6.2 Å/mm, 12 – lens, 13 – Fabry-Perot etalon, 14 – UF-84 camera, 15 – photographic plate, 16 – removable neutral filter.

ponents did not exceed the apparatus width of the instrument. The observed deformation of the spectra is critical to the pump power: it disappears when the second-harmonic power decreases by a factor 2–2.5, but the SRS components are still reliably registered in this case[1)].

Great interest attaches to the discrete spectra observed in some cases. In our experiments, such spectra (Fig. 2c) were observed with the generator operating both in a single mode and in two or three equidistant or non-equidistant modes with distances $\delta\nu \approx 0.12-0.5$ cm^{-1} between them. In the latter case (multimode regime) the period of the structure registered by the DFS-8 spectrograph was not connected at all with the intermode intervals $\delta\nu$. In the discrete spectra observed by us, the lines were strictly equidistant, and their intensity decreased monotonically towards the edges, the rate of intensity decrease in the low-frequency region being much larger than in the high-frequency region. In those cases when the discrete spectrum was observed at the pump frequency, a similar structure was observed at the first forward-scattered Stokes component. The interval between the discrete lines of the first Stokes component $\delta\nu$ coincides with the interval at the fundamental frequency. This interval changes from flash to flash in a wide range, $\delta\nu \approx (2.70-9.6)$ cm^{-1}.

A very interesting circumstance, which becomes evident immediately upon examination of the spectra of Fig. 2, is the large difference between the widths of the spectral lines for the radiation scattered forward and backward. In the case of the discrete spectra at frequencies close to the laser frequency, the ratio of the widths of the spectra of the radiation scattered forward and backward, $\Delta\nu_f/\Delta\nu_b|_{\omega=\omega_0} \approx 30-50$ (typical values are $\Delta\nu_f|_{\omega\approx\omega_0} \approx 150$ cm^{-1}). For the first Stokes component of SRS we have $\Delta\nu_f/\Delta\nu_b|_{\omega\approx\omega_0-\Omega} \approx 10-15$ (with $\Delta\nu_f|_{\omega\approx\omega-\omega_0-\Omega} \approx 30-40$ cm^{-1}).

It should be noted that the experiments described above were performed with beams having a complicated amplitude profile over the cross section (the diameter of the diaphragm in the generator was d = 4 mm). In the case when the generator operated in the lowest TEM_{00} mode, no noticeable broadening of the spectral lines was observed.

We attribute this fact to the conditions under which the spectra were registered. As is well known, in the case of self-focusing of a Gaussian beam, a small number of thin filaments is produced in the near-axis part of the beam. In our installation, the distance from the cell to the spectrograph was 3 meters, and it was extremely difficult to adjust this system in such a way that the radiation from the filaments was certain to fall into the spectrograph. It was therefore necessary to purposely "spoil" the beam as it entered into the saw, so that during each flash the number of thin filaments in the cross section of the beam was sufficient.

In succeeding experiments, the output window of the cell was projected on the spectrograph slit on a reduced scale. In such a scheme, the broadenings described above were observed also when operating with a beam that was transversely single-mode.

The appearance of discrete spectra had a random character. We were unable to establish any correlation between the registered beam parameters and the appearance of such spectra.

FIG. 2. Spectra of the radiation scattered in the forward and backward directions in carbon disulfide. a – Monotonic character of broadening: 1 – pump and first SRS Stokes component radiation scattered in the backward direction, 2 – radiation scattered in the forward direction; b – discrete character of broadening: 1 – pump radiation and radiation of first SRS Stokes component scattered in the forward direction; 2 – radiation scattered in backward direction. In this flash, only one mode was registered in the Fabry-Perot interferometer used to record the spectrum of the initial radiation (the interferometer dispersion region was 0.16 cm^{-1}).

Scattering Indicatrix of the First SRS Stokes Component

In the investigation of the form of the spectral lines of the radiation scattered "forward-backward," we noted an interesting phenomenon: the ratio of the intensities of the first Stokes components scattered forward and backward depend strongly on the pump intensity. In earlier investigations[18,19], such a relation was verified for ruby lasers when the radiation was focused into a cell with a scattering liquid. No special measures were taken in those investigations to decouple the generator and the cell, and, as is well known, the regime of consecutive scattering of the Mandel'shtam-Brillouin components can greatly distort the intensity ratio $\eta = I_{St}(0°)/I_{St}(80°)$. In addition, the dependence of the SRS indicatrix on the pump intensity was not investigated there.

For a correct registration of the ratio η, we first equalized the sensitivities of both arms of the system. To this end, the gas-laser beam was split by a thick (d = 3 cm) plane-parallel plate of high optical quality into two beams of equal (within 4%) intensity. The two beams were incident on the faces of a rectangular prism, which replaced the cell with the liquid, and was diverted by this prism to opposite directions. After reflection from the plates and the mirrors, these beams were incident on the spectrograph slit and were photographed on a film. The adjustment of the plates and of the mirrors in this case was exactly the same as the adjustment used in the investigation of the dependence of η on the pump intensity. The sensitivity of both arms of the system was equalized by neutral filters.

[1)]It must be emphasized that line broadening takes place only in pure and fresh carbon disulfide, after 5–10 flashes the broadening disappears, and the pump lines and the SRS components remain narrow at the same power levels.

Equality was attained when a neutral NS-6 filter was placed in the "forward" arm.

During the performance of the experiment, the intensity of the pump radiation striking the cell was attenuated with neutral filters. It was observed that at low pump levels $\eta \ll 1$; at maximum pump levels, this ratio was reversed (Fig. 3). The effect was so strong that we were unable to measure the ratio η accurately in the two extreme cases, for when working in the region of normal density of the photographic film one of the lines fell either in the region of strong overexposures or very strong underexposures. When the pump intensity was varied from 10 to 100 MW/cm^2 the value of η changed from 0.01 to 1000. The experiments also show that in the case when $\eta \gg 1$ the spectrum of the Stokes component propagating forward is appreciably broadened, i.e., with broadening of the pump spectrum (and of the Stokes component), preferred transformation of the pump energy into the forward Stokes component takes place.

SRS Indicatrix and Spectra of Scattered Radiation in Picosecond Pumping

We have investigated the spectral composition of radiation scattered in a nonlinear medium using as the driving source the second harmonic of a neodymium laser operating in the mode-locking regime. The radiation constituted a train of pulses (20–25) with distances of 10 nsec between pulses and each pulse having a duration 3–4 psec. The pulse duration was determined by the standard procedure of two-photon luminescence. The second-harmonic power in each pulse was 100 MW, at a beam diameter d = 1 mm. Telescoping the beam ensured a forward radiation density of 100 GW/cm^2 in each pulse prior to its entry in the cell. The cell with the liquid was 10 cm long.

We registered in the carbon disulfide an exceedingly strong asymmetrical broadening of the pump line (greatly exceeding the broadening observed with a laser operating in the nanosecond pulse regime), overlapping the frequency interval up to the first SRS Stokes component (656 cm^{-1}). The broadening had a monotonic character, the intensity of the continuous background decreased from the unshifted line towards the edges. At such a strong deformation of the pump line, the SRS Stokes component was excited very weakly

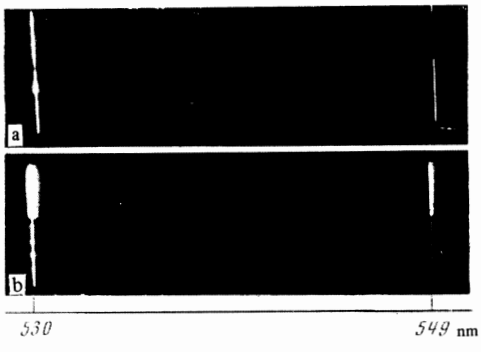

FIG. 3. Intensity ratio of the first Stokes SRS components scattered in the forward and backward directions, at different pump levels: a – second harmonic power 5 MW/cm^2; b – 100 MW/cm^2.

—the intensity of the first SRS Stokes component barely exceeded the intensity of the "wing" of the unshifted line (Fig. 4). It is also interesting to denote that when working with benzene, the pump line was barely broadened, and the conversion into Stokes components of SRS was sufficiently high[20]. The indicated pump power turned out to be below the critical value for the self-focusing in CS_2, and therefore no modulation of the envelope of the broadened spectrum was observed.

In experiments with picosecond pumping, a sharp asymmetry of the SRS indicatrix was observed. The Stokes component scattered at 180° to the pump could not be registered at all[20]. Estimates show that in our case $\eta > 10^5$.

4. DISCUSSION OF RESULTS

1. In our opinion, the results make it possible, first, to narrow down the circle of possible mechanisms that make a noticeable contribution to the broadening of the spectral lines. Investigation of the spectral structure of the radiation scattered forward and backward shows that the effects due to stimulated scattering of the Rayleigh line wing play a small role. Indeed, SRWS should lead to an appreciable broadening of the spectrum of the radiation scattered both forward and backward (see[21]); in the experiments described above, on the other hand, the backward scattering is hardly broadened.

Such a situation is apparently characteristic of the regime of self-focusing, in which the deformation of the spectral line begins without a threshold in the first layers of the medium (unlike SRWS) and accumulates as the distance traversed by the wave in the nonlinear medium increases.

A possible mechanism of the spectrum broadening might be the cross modulation of the pump by a short SRS Stokes pulse. Our investigation of this process leads to the following results (see also[11]). In a medium with a refractive index that depends on the electric field intensity of the light wave $n = n_0 + n_2E^2$, an intense short pulse propagating in opposition to the quasimonochromatic radiation changes the spectrum of the latter. This change reduces to a shift of the central frequency and to a broadening of the spectrum near the shifted frequency. The width of the shifted spectrum and its relative energy are determined by the expressions $\Delta\nu = |\delta|\sigma I_0\tau/\pi\nu_+^2$ and $S/S_0 = \pi\nu_+^3/86I_0\tau\delta^2\tau_p^2$, where $\sigma = kn_2/n_0$, $k = \omega n_0/c$, δ is the growth increment of the SRS Stokes wave ($2|\delta| = gI_p$), I_0 and τ are the intensity and duration of the short pulse of SRS, I_p and τ_p are the intensity and duration of the pump pulse, and ν_+ is the group detuning of the pump and SRS pulses.

For numerical estimates, we shall use the data of[26], where a short SRS pulse was investigated under typical

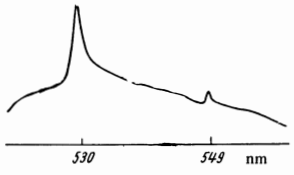

FIG. 4. Spectrum of pump and first SRS Stokes component scattered in the forward direction in carbon bisulfide. The laser operates in the mode-locking regime.

experimental conditions: $I_p = 4 \times 10^2$ MW/cm^2, $I_0 \approx 4 \times 10^3$ MW/cm^2, $\tau_p \approx 3 \times 10^{-11}$ sec, and $n_2 \approx 10^{-11}$ cgs esu (for carbon disulfide). In this case $\Delta\nu \approx 1$ cm^{-1} and $S/S_0 \approx 4 \times 10^{-6}$.

Thus, cross modulation of this type is incapable of explaining the observed broadenings. This conclusion is confirmed by specially organized experiments, in which we observed ultrashort backward Stokes pulses.

In our experimental setup, as already indicated, there is ideal decoupling between the generator and the scattering medium; this means that the observed broadenings are not connected in any way with four-photon processes in successive scattering or with the intensification of the weaker laser modes as a result of the trigger regime[16].

The mechanism of AM → PM conversion, which explains satisfactorily certain characteristics of the broadened spectra observed in[2 13], likewise does not afford a complete interpretation of our experimental data. Indeed, in most of our experiments the envelope of the broadening spectrum decreases monotonically from the fundamental line towards the edges, whereas phase modulation should cause the envelope to increase towards the edges of the broadened spectra. It should also be noted that within the framework of the mechanism of the simple AM → PM conversion it is impossible to explain why the initial amplitude modulation of the beam does not become superimposed on the phase of the wave and becomes necessary to excite some new oscillations of the medium in order to initiate the process of effective phase modulation (in the multimode generation regime, the interval $\delta\nu$ between the lines of the broadened spectrum does not correspond to the distance between these generator modes).

In this connection, we consider it advisable to assess the possibility of periodic auto-modulation and broadening of the spectrum in the process of nonstationary aberration self-focusing of laser beams[2)] [23,24]. As shown[24], owing to the fact that different sections of the amplitude-modulated beam becomes self-focused at different points on the axis, the time profile of the beam becomes distorted, and this is equivalent to the appearance of new bands in the spectra. Thus, if the beam was harmonically modulated on entering the medium, $A(t) = A_0 \cos \Omega t$, then frequencies $\omega_0 \pm n\Omega$ appear in its spectrum as a result of aberration self-focusing. Allowance for the relaxation of the nonlinearity leads to asymmetry of the spectrum. Unlike the AM → PM conversion mechanism, this mechanism leads to a monotonic decrease of the envelope of the broadened spectrum. This is precisely the form of the spectrum observed in our investigation (see Fig. 2c).

Let us turn to the question of what kind of an initial AM of the beam is necessary to actuate the discussed mechanisms. In our experiments, we have unequivocally confirmed that even in the case when the beam entering the medium contains only one longitudinal mode, the broadened spectra have a band form. An investigation of the interval $\delta\nu$ between bands, and also of the temporal and spectral characteristics of the incident radiation, has shown that the observed interval $\delta\nu$ is connected neither with the excitation of the stimulated scattering components (SMBS or SRWS) nor with the temporal characteristics of the initial radiation.

It is interesting that the variations of the structure from flash to flash obtained in our experiments with the second harmonic of a neodymium laser, lie in the same range as when ruby lasers are used[13]. This fact confirms once more that harmonic modulation of the incident radiation occurs in the medium itself. It seems to us that the qualitative explanation proposed by Townes et al. for the effect[13] meets with significant difficulties. Attributing the periodicity of the spectra to the anharmonic motion of the molecules with frequency $\omega_L = |E| \alpha / 2I^{1/2}$, where α is the anisotropic polarizability, E the electric field, and I the moment of inertia of the molecule, does not explain the discrete character, for the field varies continuously in the process of the collapse of the beam into a filament. On the other hand, if it is assumed that ω_L is determined by the field inside the produced thin filament, then the experimentally observed strong scatter of the period of the structure from experiment to experiment cannot be explained, since the field in thin filaments is a constant quantity.

In this connection, we consider it more probable that the periodic modulation appears during the process of the nonstationary aberration self-focusing of the light beam. This mechanism was discussed in[24]. As is well known, in a medium with a saturable nonlinearity, when $P/P_{cr} \gg 1$, the width of the beam oscillates beyond the focal point (at $z > R_{nl}$). The spatial scale of the beats is $L = ka_0^2 (P_{cr}/P)^{1/2}$, where a_0 is the beam width at which saturation of the nonlinearity takes place. As a result, at a fixed value of z and at a pulse that varies monotonically in time P(t) the intensity on the axis experiences periodic changes in time (periodic temporal auto-modulation takes place). For a Gaussian beam of duration τ, the average auto-modulation period is $T = \tau L/z$. For our case with $\tau \approx 10^{-8}$ sec, $P/P_{cr} \approx 100$, and $a_0 \approx 10^{-3}$ we obtain $T \approx 10^{-11}$ sec or a modulation frequency $\Omega \approx 3$ cm^{-1}. Under real conditions, this quantity can change from filament to filament as a result of the variation of the ratio P/P_{cr}, of the beam shape, etc.

Thus, both the appearance of the initial amplitude modulation of a single-mode beam and the further enrichment of its spectrum by harmonics can be satisfactorily explained as being due to the interaction between the temporal and spatial modulations of the light beams in self-focusing[3)].

[2)] The mechanism of formation of envelope shock waves in the case of nanosecond pulses, as shown by estimates, should certainly be excluded. Indeed, the distance over which the envelope shock waves are produced, $L_{cr} \approx 0.2\tau v n/\delta n$, amounts to $L_{cr} \approx 5$ even at maximum $\delta n/n \approx 0.1$, for pulses of duration 10^{-8} sec.

[3)] We emphasize that the foregoing pertains even to a Gaussian beam. For such a beam, the total output power $P(t) = \int_0^\infty I(r, t) r dr$, where $I = |A|^2$ is the intensity, changes in time in exactly the same manner as the input intensity $I_0(t)$. This is manifest, however, only in experiments in which the total beam power is investigated with the aid of a photomultiplier. In experiments with spectral instruments, different sections of the self-focused beam are projected on different parts of the film, making it possible to register the deformation of the spectral lines. This pertains to an even greater degree to complicated beams, in which the aforementioned effects are mainly observed.

In the discussion of the results, we would wish to dwell here also on the following question. As shown by Kelley et al.[25], the modulation of the envelope of the spectrum of a thin filament can be due also to the formation in the medium of short pulses with Gaussian form, of duration 5—10 psec (for example, due to stimulated scattering in the backward direction, or else due to self-focusing effects). Our experiments with picosecond pulses show that in the absence of self-focusing the envelope of the broadened spectrum is monotonic (Fig. 4), and in the self-focusing regime, modulation of the envelope, similar to the modulation occurring in the scattering of nanosecond pulses, is indeed observed.

2. The effects of the influence of anomalous broadening of a laser line on the efficiency and SRS indicatrix, observed distinctly in our experiments, can be explained quite clearly.

Under conditions when broadening of the spectral line of the laser takes place, the scattering process becomes essentially nonstationary. In SRS we deal with two types of nonstationary behavior: 1) wave nonstationary processes connected with the effect of group delay of the laser radiation and of the scattered components, and 2) "local" nonstationary effects, appearing under conditions when the characteristic period of the pump wave modulation (predominantly amplitude modulation) becomes comparable with the relaxation time of the molecular oscillations (we note that in a strongly nonlinear medium the character of the pump modulation is determined not so much by the properties of the initial radiation as by the processes that develop in the nonlinear medium). The effects of group delay become manifest in the fact that exponential amplification of the scattered components occurs only over lengths not exceeding the so-called quasistatic lengths L_{qu}. For waves propagating in the forward and backward directions, these characteristic lengths are given by

$$L_{qu}^{\mp} = \tau_m / (u_p^{-1} \mp u_{St}^{-1}),$$

where τ_m is the period of the pump modulation, u_{St} and u_p are the group velocities of the Stokes component of SRS and of the pump; the minus sign pertains to waves propagating in one direction and the plus sign in the opposite direction.

The group delay affects primarily the front-back asymmetry of the SRS indicatrix[26]. This occurs when the smaller of the L_{qu}, namely L_{qu}^{+}, becomes smaller than the interaction length (the length of the thin filament). In our case, for estimates, it is reasonable to choose for τ_m the average period of the observed discrete structure: $\tau_m = (\delta\nu_{av})^{-1}$. For $\delta\nu_{av} = 5$ cm^{-1} and $(u_{St} - u_p) = 4.5 \times 10^8$ cm/sec[27] we obtain $L_{qu}^{+} \approx 6$ cm. Taking into account the group-delay effect, the ratio η of the intensities of the Stokes components scattered in the forward and backward directions can be estimated as follows:

$$\eta = \exp\{gI_p(L_{qu}^{-} - L_{qu}^{+})\} \approx \exp\{gI_pL_{qu}^{-}\}.$$

In our case at maximum pump intensity $I_p \approx 100$ MW/cm^2 and $g = 0.01$ cm/MW for CS_2 we get $\eta \approx 10^2$, which agrees with the experimental data[4].

In this connection, we consider the reason for the change of the ratio η to be the following: at relatively small laser power levels, prior to the occurrence of noticeable deformations of the spectrum, the intensity of the first backward-scattered Stokes component is larger than that of the forward-scattered component, owing to the intensification of the backward Stokes component of SRS in the pump field[22]. Subsequently, with increasing pump intensity, a noticeable broadening of the pump spectrum takes place, the SRS process becomes nonstationary, and the effect of group delay leads to a predominant scattering of the Stokes component forward.

We note that in order for the group delay to be effective it is necessary to have just amplitude modulation of the beam, for in the case when τ_m is much larger than the relaxation time of the molecular oscillations, the phase relations between the interacting waves are not important (the SRS process is described by the kinetic equations for the intensities). This fact also favors the aforementioned mechanism of spectrum deformation in the process of nonstationary aberration self-focusing.

Thus, the forward-backward scattering indicatrix is very sensitive to the modulation of the pump wave and, primarily, to the time modulation. It is due mainly to wave effects of group delay, and not to effective "local" nonstationary behavior. The foregoing means that in the study of the SRS indicatrix it is necessary to control strictly the temporal and spectral characteristics of the radiation[5].

The "local" nonstationary behavior leads to a decrease of the gain in all directions. In the limit when τ_m of the initial radiation is much smaller than the relaxation time of the molecular oscillations, the gain of the SRS Stokes component is given by $I_{St} = I_{st}(0)\exp(gI_pz)^{1/2}$. This circumstance also explains the sharp decrease of the SRS gain when working with picosecond pulses. A detailed discussion of this question is given in[20].

The authors are grateful to S. A. Akhmanov for interest in the work and for useful discussions of the results.

Note added in proof (29 November 1969). In our latest experiments at 250 GW/cm^2 in a single picosecond pulse, we observed in carbon disulfide self-focusing accompanied by a quasiperiodic modulation of the envelope of the broadened spectrum of the thin filaments.

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[4] It is interesting to note that the case of SRS for two-mode pumping was calculated already in [28]. For carbon disulfide, calculation shows that at a pump modulation depth of 25% and an interaction length L = 10 cm we get $\eta \approx 10^2$.

[5] A weaker dependence of η on the pump power (within one order of magnitude) was observed earlier by Shen and Shaham [14]. They explained qualitatively their results as being due to the variation, with the pump power, of the boundary conditions for the Stokes components scattered in the forward and backward directions. This effect, however, cannot explain the strong variation of η observed in our experiments; such an effect can be due only to a noticeable difference between the increments of the Stokes components scattered in the opposite directions. It seems to us that this effect might have also played an important role in the experiments of [14]; unfortunately there are no data on the spectral investigations in [14].

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