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SPECTRAL PROPERTIES OF BACKWARD STIMULATED SCATTERING IN CARBON DISULFIDE LIQUID

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Spectral structure of backward stimulated scattering from a 10 cm-long CS_2-liquid cell is investigated by using Q-switched 10-ns and 532-nm laser pulses with different spectral linewidths. Under a narrow spectral line ($\sim$ 0.1 cm^{-1}) pump condition, very strong sharp lines near the pump wavelength (λ_0) position and the first-order stimulated Raman scattering (λ_{s1}) position can be observed. However, under a wide line ($\approx$ 1 cm^{-1}) pump condition, only a strong and superbroadening spectral band can be observed mainly in the red-shift side of the pump wavelength. The different spectral features under these two conditions can be explained by a competition between stimulated Brillouin, Raman, and Rayleigh–Kerr scattering. Under both pump conditions, the broadening spectral distributions are not consistent with the predictions given by stimulated Rayleigh-wing scattering theories, but can be interpreted well utilizing the theoretical model of stimulated Rayleigh–Kerr scattering.

1. INTRODUCTION

The early observations for spectral broadening (up to $\sim$ 15 cm^{-1}) of the backward stimulated scattering from a Kerr liquid cell were reported in the middle of 1960s and interpreted phenomenologically as stimulated Rayleigh-wing scattering (SRWS) [1–3]. According to SRWS theories [1–4], the normalized exponential gain profile is given by

$$g_{SRWS}(\Delta\nu = \nu_0 - \nu) \propto |E_0|^2(2\pi\Delta\nu\tau)/\left[1 + (2\pi\Delta\nu\tau)^2\right], \quad (1)$$

where ν_0 and ν are the frequency of the pump line and the frequency of SRWS respectively; τ is the molecular reorientational relaxation time of a given Kerr liquid comprising of anisotropic molecules, and E_0 is the amplitude of a monochromatic incident optical electric field. For the same optical intensity level (in units of W/cm^2), a smaller spectral linewidth will yield a greater E_0 value and a higher gain value. According to Eq. (1) one can find that there will be attenuation on the anti-Stokes side of the pump line, and gain on the Stokes side, respectively. In particular the location of the gain maximum on the Stokes side will be determined by

$$\Delta\nu_{max} = 1/2\pi\tau. \quad (2)$$

For CS_2 the measured value of τ is about 1.5–2 ps, so that the value of $\Delta\nu_{max}$ should be $\sim$ 3 cm^{-1}. During the early SRWS studies, it was difficult to accomplish a reliable quantitative comparison between experimental measurements and theoretical predictions due to the influence from strong stimulated Brillouin scattering, the poor spectral resolution, as well as the overexposure effect of the employed photographic films or plates [1, 2].

Since the middle of 1980s, a superbroadening ($> 400\ cm^{-1}$) forward stimulated scattering from a Kerr-liquid-filled hollow fiber system has been reported and systematically investigated [5–10]. The main features of this kind of spectral broadening behavior can not be simply interpreted by either SRWS theories [1, 3, 11], stimulated thermal Rayleigh scattering [12, 13], or self/cross-phase modulation mechanisms [14]. However, they can be explained well based on the theoretical model of stimulated Rayleigh–Kerr scattering (SRKS) [5–9]. According to this model a much broader gain curve on the Stokes side of the pump line should be observed, and can be expressed as [5, 6]

$$g(\Delta\nu \geq 0) = \lambda_0^2 N \sigma(\Delta\nu) I_0 / [4\pi h \nu_0 (\delta\nu_0)]. \qquad (3)$$

Here λ_0 is the pump wavelength, N is the molecular density of the scattering medium, I_0 is the pump intensity, h is the Planck constant, and $\delta\nu_0 = 1/2\pi\tau$ is the spectral linewidth of the elementary Rayleigh–Kerr scattering process. Finally, the scattering cross section $\sigma(\Delta\nu)$ is given by

$$\sigma(\Delta\nu) = (2\pi/c)^4 \nu_0^4 (\alpha_\parallel^2 - \alpha_\perp^2) \cos^2 [f(\Delta\nu)], \qquad (4)$$

where c is the speed of light, $\alpha_\parallel$ and $\alpha_\perp$ are the maximum and minimum molecular polarizabilities of a given Kerr liquid, and $f(\Delta\nu)$ is an increasing function of $\Delta\nu$ which can be experimentally determined for a given scattering medium. A trial function such as

$$f(\Delta\nu) = (a\Delta\nu)^b \qquad (5)$$

can be used to fit the experimental data of the forward stimulated scattering spectra from a CS_2-liquid-filled fiber system. Here, the value of $\Delta\nu$ is in units of cm^{-1}, and the value of $f(\Delta\nu)$ is in units of angular degree. The best fitting parameters for CS_2 liquid were $a = 7.5 \cdot 10^9$ and $b = 0.148$ [5].

It is different from the SRWS theory that on the anti-Stokes side of the pump line, an observable spectral broadening is also predicted by the SRKS theory. In this case, the gain curve on the anti-Stokes side of the pump wavelength can be expressed as [5, 6]

$$g(\Delta\nu \leq 0) = g(0) \left[1/(2\pi\tau)^2\right] / \left[(\Delta\nu)^2 + 1/(2\pi\tau)^2\right], \qquad (6)$$

where $g(0)$ is the maximum stimulated scattering gain value at $\Delta\nu = 0$ position, and τ still is the molecular reorientational relaxation time. The experimental results of the forward superbroadening stimulated scattering from a CS_2 liquid-filled hollow-fiber system were basically in agreement with the above theoretical description [5–10]. In these cases, however, someone might not be entirely convinced by thinking that the intensity of the transmitted pump beam is so high, that the possible small-red-shifted SRWS may be covered by the transmitted pump signal, and, also, a cascaded effect may take place for a long hollow-fiber sample. For these reasons, it seems to be necessary to pursue a thorough studies on the spectral property of backward stimulated scattering in a shorter CS_2-liquid cell. In that a case, the intense pump beam background and the spatially cascaded effect can be eliminated.

2. EXPERIMENTAL SETUP

In this work, we report the spectral-broadening measurements of the backward stimulated scattering from a 10 cm-long CS_2-liquid cell pumped with either a narrow (~ 0.1 cm^{-1}) 532-nm laser line or a wide (~ 1 cm^{-1}) 532-nm laser line, respectively. The experimental setup is schematically shown in Fig. 1. The pump source was a frequency-doubled and Q-switched Nd:YAG laser with a $\sim$ 10-ns pulsewidth, $\sim$ 1-mrad beam divergency, $\sim$ 3-mm beam size, and 10-Hz repetition rate. The spectral width of the output laser pulses was ~ 1 cm^{-1} when a Pockels cell was used as a Q-switching element. When a BDN dye-doped acetate sheet was employed as the Q-switching element, the output spectral linewidth was ~ 0.1 cm^{-1} measured by a Fabry–Perot etalon. Exchanging the Q-switching element did not cause any considerable change of the output pulse duration and profile [7]. The incident 532-nm pump laser beam was focused through a $f_1 = 30$ cm lens into a 10 cm-long quartz liquid cell filled with CS_2. The liquid sample was specially purified, i.e., glass distilled twice and then filtered through a 0.2 μm filter. Therefore, the linear absorption due to residual impurities in the liquid and the possible stimulated thermal scattering can be neglected. The special feature of the setup shown in Fig. 1 is that both the forward beam and the backward beam from the CS_2-liquid cell can be measured simultaneously by a spectrographic device. The spectral distributions of the backward and forward stimulated emission from the liquid-cell sample could be measured by three different systems: (i) a low-spectral-resolution (~ 9 cm^{-1}) system consisting of a single grating (1800 grooves/mm), a $f_3 = 60$ cm focusing lens, and an ordinary camera, (ii) a middle-spectral-resolution (-1.8 cm^{-1}) system consisting of a grating spectrograph (Triplemate from SPEX) in conjunction with a vidicon-OMA (optical multichannel analyzer) Ill device (from EG& G Princeton Applied Research), and (iii) a high-spectral-resolution (~ 0.48 cm^{-1}) system consisting of a double-monochromator (Jobin–Yvon) in conjundion with the same vidicon-OMA III device.

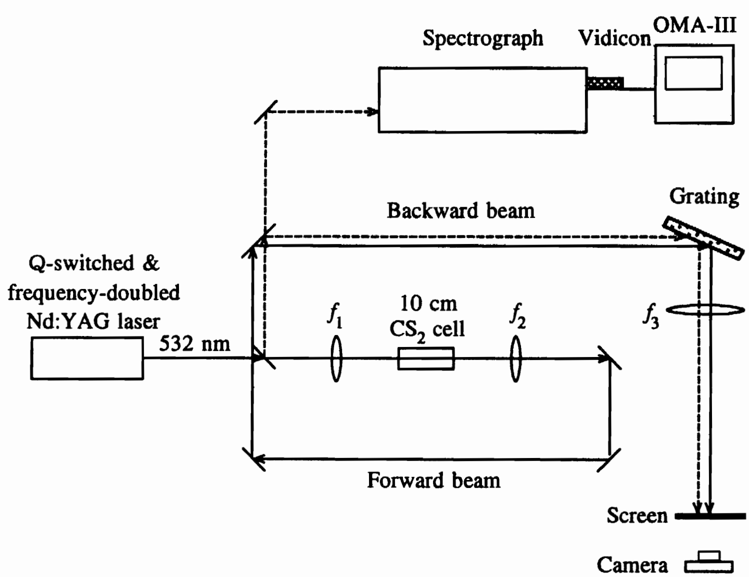

Fig. 1. The experimental setup for spectral measurements of the forward and backward stimulated scattering from a 10 cm-long CS_2-liquid cell

The temporal profiles of the pump laser pulse and the backward stimulated scattering pulse can be easily measured by using a 350 MHz oscilloscope (Tektronix 2467 with a C1001 video camera) [7]. Under the pump intensity levels of 150–400 MW/cm^2, the pulse duration of the backward stimulated scattering from the 10-cm long CS_2-liquid-filled cell measured to be 4–6 ns.

3. RESULTS AND DISCUSSION

Typical spectral photographs of the backward stimulated scattering from the 10 cm-long CS_2-liquid-filled cell are shown in Fig. 2 by using the spectral measurement system (i) with a spectral resolution of $\sim$ 9 cm^{-1} at a pump intensity level of $I_0 \approx$ 150 MW/cm^2. The photograph shown in Fig. 2*a* is obtained by using the wide ($\sim$ 1 cm^{-1}) pump line, which shows a smoothly decreasing and superbroadening spectrum mainly on the Stokes side of the

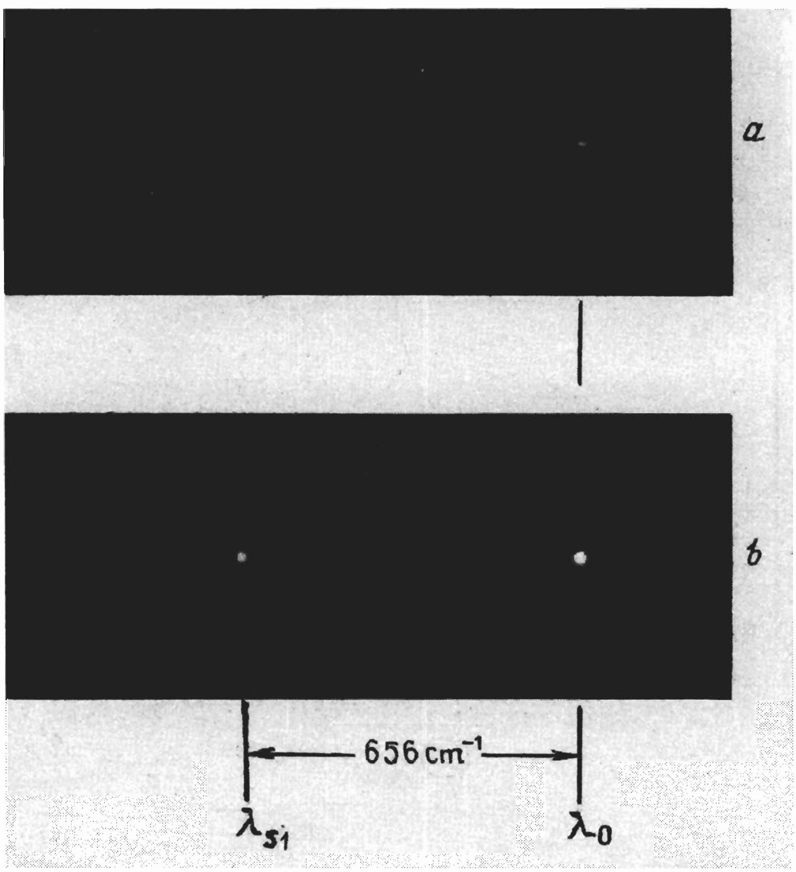

Fig. 2. Spectral photographs of the backward stimulated scattering from the CS_2-liquid sample pumped by $a \sim$ 1 cm^{-1}-wide 532-nm line (*a*) and $\sim$ 0.1 cm^{-1}-wide 532-nm line (*b*), respectively. The pump intensity is $I_0 \approx$ 150 MW/cm^2 and the spectral resolution is $\sim$ 9 cm^{-1}

pump wavelength and is obviously broader than the previously reported SRWS by more than one order of magnitude [1, 2]. In contrast, the photograph shown in Fig. 2*b* is obtained by using a narrow (~ 0.1 cm^{-1}) pump line, which manifests two strong sharp spectral lines accompanied by a quite weaker broadening component. In Fig. 2*b* the first sharp line was nearly located at the pump line (λ_0) position, and the second line was located at the first-order Stokes stimulated Raman scattering line (λ_{s1}) position with a Raman shift of ~ 656 cm^{-1}. A Fabry–Perot etalon measurement showed that the first sharp line in Fig. 2*b* is the backward stimulated Brillouin scattering line accompanied by a weaker broad wing mainly on the red side. The substantial difference between Fig. 2*a* and Fig. 2*b* can be explained by a competition effect among three major stimulated scattering processes in a CS_2-type transparent liquid: stimulated Brillouin scattering (SBS), stimulated Raman scattering (SRS), and stimulated Rayleigh–Kerr scattering (SRKS) (or possible SRWS scattering?). For different pump conditions, the relative threshold requirements for various stimulated scattering processes may be considerably different even for a given sample. Under the same pump pulse duration and intensity level, the threshold requirements for SBS and SRS quite sensitively depend on the spectral width of the pump line. For instance, the reported experimental results showed that the SBS threshold increased following the pump linewidth increase [15–17]. In fact, the threshold increase (or efficiency decrease) of SBS becomes more severe if the pump linewidth is much greater than the frequency shift of the backward SBS [18, 19]. For CS_2 liquid this shift value is about ~ 0.25 cm^{-1}, therefore, the pump condition in Fig. 2*a* was among the latter case. It is reasonable to assume that in the case of Fig. 2*a*, the backward SBS and SRS were suppressed due to their higher threshold requirements under wide line excitation. Therefore, the backward SRKS process became the dominant mechanism contributed to the observed superbroadband spectral distribution. In contrast, in the case of Fig. 2*b* the SBS and SRS were the dominant processes contributing to the two strong sharp lines due to their relatively lower threshold requirements under the narrow line excitation.

Figure 3 shows the photographs of the spectra for both the forward and backward stimulated emission from the same 10-cm long CS_2-liquid cell sample pumped with the 532-nm line of ~ 1 cm^{-1} width at three different pump intensity levels. In Fig. 3 for each photograph the upper spectrum is corresponding to the forward emission comprising the transmitted pump line and the forward stimulated scattering, meanwhile the lower spectrum is corresponding to the backward stimulated scattering only. It can be seen in Fig. 3 that at a lower pump intensity level (Fig. 3*a*), the forward emission is mainly composed of a transmitted pump line (λ_0) and the first-order Stokes stimulated Raman scattering line (λ_{s1}); whereas at a higher pump intensity level (Fig. 3*c*), there is also an considerable red-shifted broadening component added in the λ_0 line. This is understandable because at a lower pump level the forward broadening stimulated scattering takes only a very small percentage of the total forward beam. As the pump intensity is increased, the ratio between the forward broadening scattering and the transmitted λ_0 emission becomes greater, and the red-spread wing looks broader and stronger. This behavior is essentially the same as that of the forward SRKS observed in a CS_2-liquid cell sample [5, 9]. In addition, it should be noted that the spectral structure of the forward emission shown in Fig. 3 is quite similar to that of the backward stimulated emission shown in Fig. 2*b*. This similarity is understandable because in the latter case instead of the pump line the backward SBS is the predominant component excited by the narrow (~ 0.1 cm^{-1}) pump line; so that the entire feature of the backward SRKS is partially covered by the intense SBS line as shown in Fig. 2*b*. On the other hand, in Fig. 3 one can see that the spectral feature of the backward stimulated scattering remains basically unchanged at the three different pump intensity levels. This fact

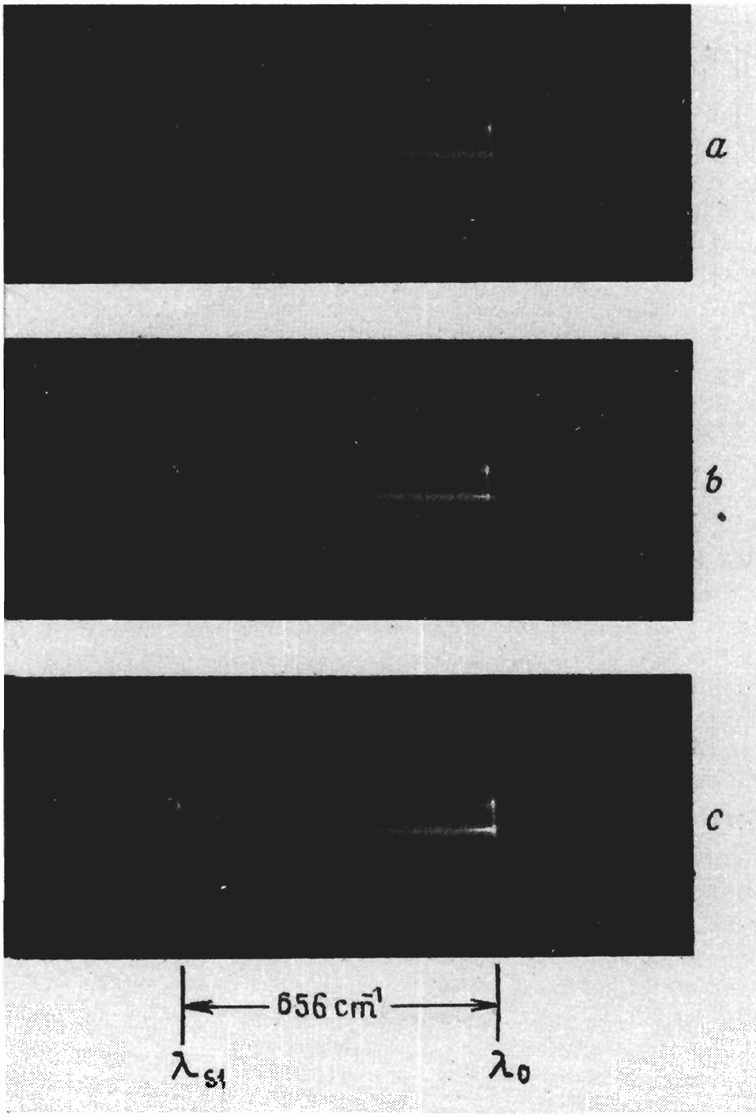

Fig. 3. Spectral photographs of the forward stimulated emission (upper track) and backward stimulated scattering (lower track) pumped by the $\sim$ 1 cm^{-1}-wide 532-nm line at various pump intensity levels: $I_0 \approx 170$ (*a*), 250 (*b*), 400 (*c*) MW/cm^2

can easily be understood because there is no competition with other predominant sharp line.

Now we should further clarify which mechanism, between SRKS and SRWS, is mainly responsible for the observed spectral broadening of the backward emission pumped with either the $\sim$ 0.1 cm^{-1} line or the $\sim$ 1 cm^{-1} line. For this purpose, the quantitative spectral measurements with a higher spectral resolution are needed. In order to observe the detailed spectral distribution pumped with a narrow line ($\sim$ 0.1 cm^{-1}), the spectral measurement system (iii) with a much higher resolution ($\sim$ 0.48 cm^{-1}) was employed to record the incident pump

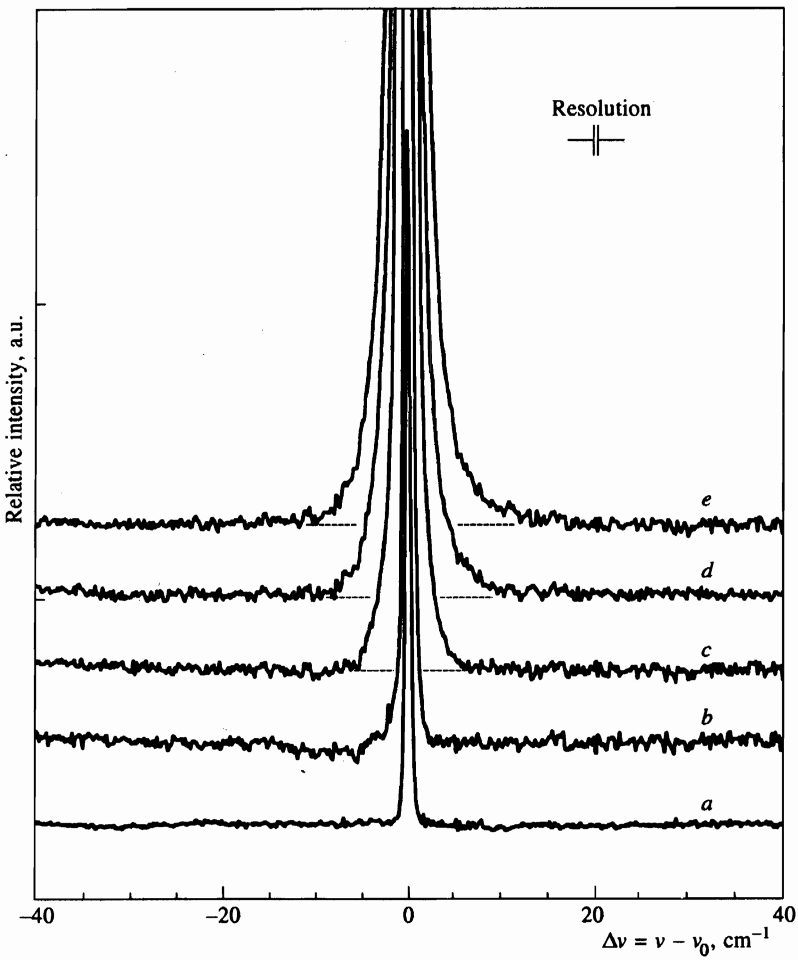

Fig. 4. Detailed spectral distributions of the ~ 0.1 cm^{-1}-wide 532-nm pump line at various attenuation ratios: 1/180 (*a*), 1/90 (*b*), 1/30 (*c*), 1/6 (*d*), 1/1 (*e*). The spectral resolution is ~ 0.48 cm^{-1}

line profile as well as the backward stimulated scattering line profile nearby λ_0 position. The measured results are shown in Fig. 4 and Fig. 5, respectively. To ensure a reliable linear display of the spectral intensity distribution nearby the root region of the sharp line, the incident beam on the double monochromator was attenuated by neutral density filters with various attenuation ratios. For the incident pump beam alone (the sample was removed), Fig. 4 shows a nearly symmetric spectral distribution on the root region of the pump line under different attenuation ratios. In contrast, for the backward stimulated scattering beam, Fig. 5 shows a strong sharp line attributed to the backward SBS (its wavelength shift was less than the apparatus resolution) as well as asymmetrically broadened components on the two sides on the root region. One can see in Fig. 5 two features: 1) although the spectral distribution on the Stokes side is stronger and broader, still there is a measurable spectral broadening component on the anti-Stokes side; and 2) no spectral maximum at red-shifted position of ~ 3 cm^{-1} (predicted by the SRWS

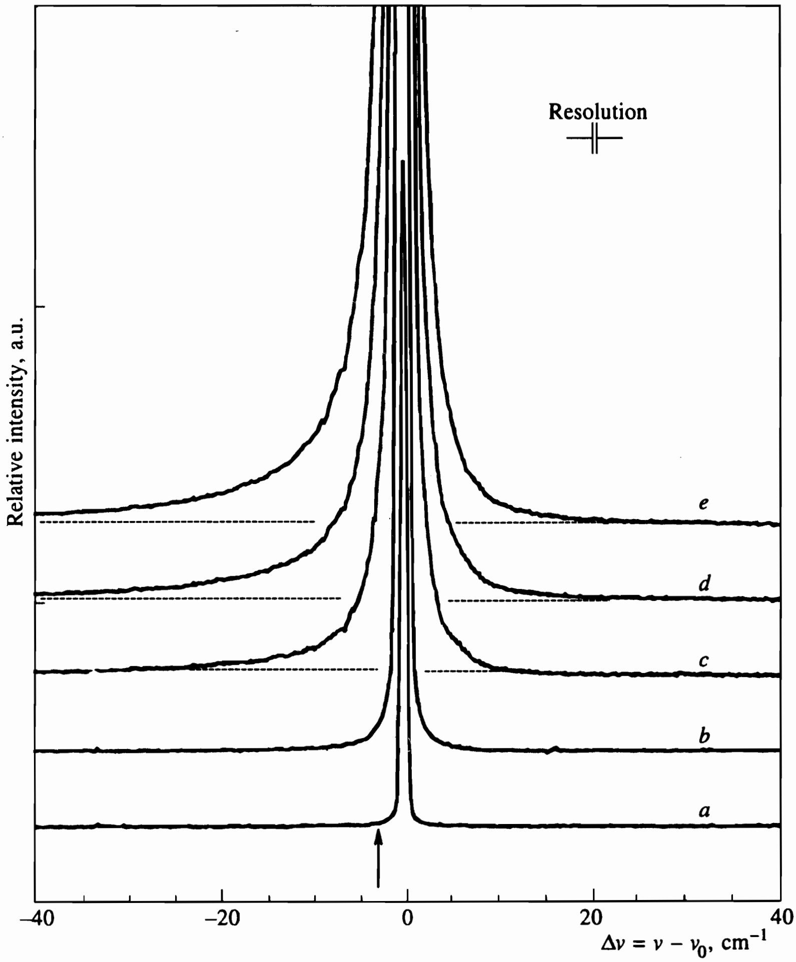

Fig. 5. Detailed spectral distributions of the backward stimulated scattering from the CS_2-sample pumped by the ~ 1 cm^{-1}-wide 532-nm line with various attenuation ratios: 1/180 (*a*), 1/90 (*b*), 1/30 (*c*), 1/6 (*d*), 1/1 (*e*). The pump intensity is $I_0 \approx 150$ MW/cm^2, and the arrow indicates the maximum gain position predicted by the stimulated Rayleigh-wing scattering theory with an assumed value of $\tau = 1.5$ ps

theories [1–4]) was observed. These two features can not be interpreted by the SRWS theories, however, could be well explained based on the superposition of a strong SBS sharp line and a relatively weak SRKS band. This assumption can be further supported by spectral measurements of the backward stimulated scattering pumped with a wide (~ 1 cm^{-1}) laser line by using the spectral measurement system (ii). In this case, no SBS component is expected and the backward SRKS process becomes dominant; the measured spectral distributions at various pump intensity levels are shown in Fig. 6 with a spectral resolution of ~ 1.8 cm^{-1}. It can be found in Fig. 6 that once the pump intensity level is high enough ($\geq$ 200 MW/cm^2) the relative spectral distributions do not change so much, that means a full-scale spectral distribution characteristic of SRKS is established. These results are basically consistent with that shown in Fig. 3 for the backward

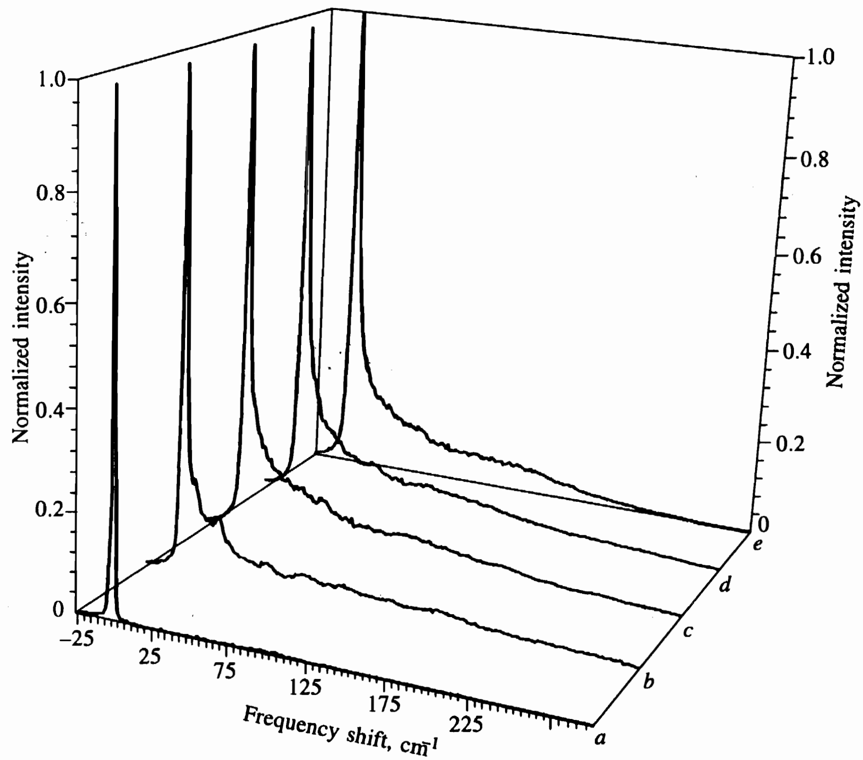

Fig. 6. Normalized spectral distributions of the backward stimulated scattering from the CS_2-sample pumped with the $\sim$ 1 cm^{-1}-wide 532-nm line at various intensity levels: $I_0 \approx 30$ (*b*), 75 (*c*), 150 (*d*), 475 (*e*) MW/cm^2. The spectrum of the incident pump line is shown in (*a*), and the spectral resolution is $\sim$ 1.8 cm^{-1}

stimulated scattering. Another feature shown in Fig. 6 is that still there is a detectable spectral components on the anti-Stokes side of the λ_0 position.

Based on the measured spectral distribution data of the backward stimulated scattering, the corresponding spectral exponential gain curve can be obtained by taking an appropriate logarithm manipulation [5]. As a result, the obtained normalized spectral gain curve of the backward stimulated scattering with a 0.48 cm^{-1} resolution is given in Fig. 7 as well as in Fig. 8 by the solid line, the small random negative spikes were due to a poorer signal/noise ratio near the zero-point. The pump linewidth was $\sim$ 1 cm^{-1} and the pump intensity $\sim$ 500 MW/cm^2. In Fig. 7 the Stokes gain curve predicted by the SRKS theory is given by a dash-dotted line by using Eqs. (3), (4), and (5) with the same fit parameters used in reference [5], meanwhile the anti-Stokes gain curve predicted by Eq. (6) is shown by a dashed line using a fit parameter of $\tau = 1.77$ ps [5]. One can see that the agreement between our experimental results and the theoretical fitting is quite good. In contrast, the normalized gain curve predicted by the SRWS theory is shown in Fig. 8 by a dashed line by using Eq. (1) with a fit parameter of $\tau = 1.5$ ps. In this case, the negative section of the theoretical curve implies an attenuation of the anti-Stokes components, so no anti-Stokes component would be observed. One can see clearly in Fig. 8 that the predictions from the SRWS theory are not consistent with our experimental results.

Finally, someone may consider the possibility of the spectral broadening due to the

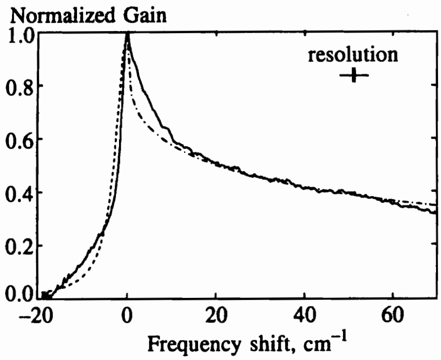

Рис. 7

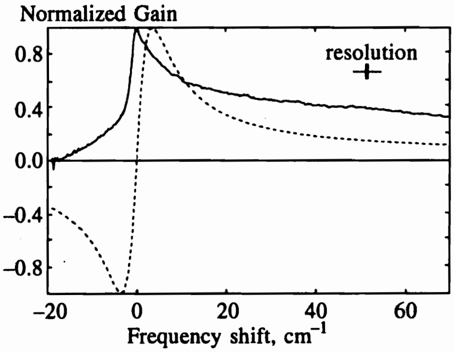

Рис. 8

Fig. 7. Normalized gain curves of the backward stimulated scattering based on the measured data (solid line) and the fitting data (dashed line and dash-dotted line) given by stimulated Rayleigh–Kerr scattering theory. The pump intensity is $I_0 \approx 500$ MW/cm^2, the pump linewidth ~ 1 cm^{-1}, and the spectral resolution ~ 0.48 cm^{-1}

Fig. 8. Normalized gain curves of the backward stimulated scattering based on the measured data (solid line) and the fitting data (dashed line) predicted by the stimulated Rayleigh-wing scattering theory with an assumed value of $\tau = 1.5$ ps. The pump intensity is $I_0 \approx 500$ MW/cm^2, the pump linewidth ~ 1 cm^{-1}, and the spectral resolution ~ 0.48 cm^{-1}

self-phase modulation of the possible sub-pulse structure within the ~ 10 ns pulse envelope, which would not be resolved by our 350-Mz oscilloscope system with a $\sim$ 1-ns resolution. However, this possibility is not likely to be true based on the following considerations. First, according to the uncertainty principle ($\Delta\nu\Delta t \approx 1$), the duration time of the possible sub-pulses would not be less than 300 ps and 30 ps, limited by the spectral linewidth of 0.1 cm^{-1} and 1 cm^{-1}, respectively. If the self-phase modulation plays an essential role, the spectral broadening behavior should strongly depend on the pump pulse duration. In the similar experimental conditions we did use a ~ 100 ps laser pulse and a ~ 0.5 ps laser pulse to pump a CS_2 liquid sample separately, no evidence of self-phase modulation observed from the forward emission excepts the similar spectral broadening behavior as shown in Fig. 2*a* or Fig. 6 [7–10]. Thus the spectral broadening behavior of CS_2 is not dependent sensitively on the pump pulse duration under our experimental conditions. Second, if the self-phase modulation is the major mechanism causing the observed spectral broadening, there should be a periodically modulated spectral structure as demonstrated by the early self-focusing experiments [20–23]. Our spectral measurements with various spectral resolutions (~ 9 cm^{-1}, ~ 1.8 cm^{-1}, and ~ 0.48 cm^{-1}) show that there is no discrete or modulated spectral structure observed. Lastly, if the self-phase modulation is the predominant mechanism, the same spectral broadening should occur with the transmitted pump line which takes the most percentage of the forward emission. However, as shown in Fig. 3, the spectral broadening in the backward direction is much broader than that in the forward direction. All these considerations described above are unfavorable for the self-phase modulation assumption.

4. CONCLUSION

We have accomplished a thorough spectral measurement for the spectral structure of the backward stimulated scattering from a 10 cm-long CS_2-liquid cell pumped by the $\sim$ 10-ns and 532-nm laser pulses with a linewidth of ~ 1 cm^{-1} and ~ 0.1 cm^{-1}, respectively. Under the ~ 1 cm^{-1} line pump condition, only a strong superbroadening spectral band is observed in the backward stimulated scattering. However, under the ~ 0.1 cm^{-1} line pump condition, a strong SBS sharp line and a relatively weak broadening component can be observed together. This difference of the spectral structure of the backward stimulated scattering under different pump linewidths can be explained considering the threshold dependence of SBS on the pump linewidth. In both pump line conditions, the spectral broadening behavior can not be interpreted by either the SRWS theory or the self-modulation assumption, but can be well explained by the SRKS theory.

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