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Influence of amplified spontaneous emission and fluorescence of β-carotene on stimulated Raman scattering of carbon disulfide

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β-carotene with double fluorescence characteristics and large third-order optical nonlinearities, which is dissolved in the carbon disulfide (CS_2) as the core medium of a liquid core optical fiber (LCOF), is applied in the study of the CS_2 stimulated Raman scattering (SRS). The results of this study show that when the concentrations of solution are more than 3.72×10^{-7} mol/L, the amplified spontaneous emission (ASE) of β-carotene is the main factor influencing the threshold and intensity of Stokes lines; when the concentrations of solution are lower than 3.72×10^{-7} mol/L, the ASE disappears and the fluorescence plays the key role: The high-order Stokes lines may be observed at very low input-laser power, and the Stokes thresholds decrease as the solution concentration increases. The result may be widely used in the study of broadband stimulated radiation laser and seeding laser.

β-carotene, ASE, fluorescence, SRS, LCOF

SRS has important applications in the areas of nonlinear optics, plasma, Raman laser, and Raman amplifier[1−6]. The SRS of a silica fiber has been extensively studied[7−11], and the results of the studies on SRS have enriched the physical mechanisms of the coherent light radiation and widened the range of the wavelength of the stimulated radiation. To lower the threshold of SRS and enhance its intensity, SRS of a weak-gain Raman mode in a micron dimension liquid droplet was enhanced by dye fluorescence[12−15]. However, this technology is impractical to be widely used, because of the difficulties in controlling the size of the liquid droplet, the intensity fluctuation, the direction of Stokes scattering light, and signal reception. Recently, LCOF technology was successfully used to lower the threshold of SRS[4,16]. The technology of the fluorescence seeding enhancement SRS in the liquid droplet and the technology of LCOF were used together in this study to lower the threshold of SRS and improve the collection and control of the scattering light.

β-carotene is a biologic molecule that has the double fluorescence characteristics and a very broad fluorescent band (380—800 nm). Its fluorescence productivity is lower than that of fluorescent dye and it has low absorbability. These characteristics make it appropriate for a core medium used at a low concentration in LCOF. β-carotene has the great linear fluorescence enhancement to SRS, and it has the low absorption loss to the pump light and the Stokes light. The experiment indicated that the number of particles increased with the increase of the concentration, and the interaction between the Stokes lines and the ASE by laser excitation was intensified, which leaded to a great increase in the exponent gain factor of SRS. Therefore, the solution of β-carotene in CS_2 has been used as the core medium of LCOF to study the effect of its concentration on the SRS

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of CS_2, with the concentration of solution ranging from 3.72×10^{-12} to 3.72×10^{-5} mol/L. The followings are the theoretical explanation of this experiment model. When the concentrations of solution are more than 3.72×10^{-7} mol/L, the ASE of β-carotene is the main factor influencing the threshold of CS_2 and the intensify of Stokes lines; when the concentrations of solution are within 3.72×10^{-12} — 3.72×10^{-7} mol/L, the fluorescence of β-carotene plays the key role, and the ASE of β-carotene disappears. Because both the fluorescence of β-carotene and the LCOF have the ability to enhance the SRS, the high-order SRS Stokes lines may be observed at very low input-laser power. For example, the 7th-order Stokes line was detected using β-carotene solution of 10^{-7} mol/L, 1 Hz frequency, and 0.86 mJ pulse energy. The threshold is gradually lowered as the concentration increases within the range from 3.72×10^{-12} mol/L to 3.72×10^{-7} mol/L. The study is of great significance to further understand the mechanism of the fluorescence enhancement SRS, develop new types of the broadband Raman laser and widen the application of β-carotene in the non-biological fields.

1 Experiment

In the experiment, β-carotene was dissolved in CS_2 solution to form the solutions of different concentrations that ranged from 3.72×10^{-12} to 3.72×10^{-5} mol/L. Then the solutions were injected into the hollow core optical fibers with the core diameter of 400 μm and the outer diameter of 600 μm, and 100 cm in length. The facture of liquid core optical fiber was shown by previous papers[17−19]. The experimental setup is shown in Figure 1.

Experimental light source was the 2nd harmonic of a Q-switched Nd: YAG laser, the wavelength was 532 nm, which lay within an electronic absorption band of the β-carotene[20], the work pulse duration was 10 ns and the repetition rate was 1 Hz, the work mode was the basic mode, and the output-laser power was within 0—5 mJ. The laser was coupled to the core by 25 times object lens, the output light from the fiber was coupled to a spectrometer, and the signal was displayed and analyzed by a computer. The high-order Stokes lines of the solution of β-carotene in CS_2 (3.72×10^{-12} to 3.72×10^{-5} mol/L) and pure CS_2 were measured. The influence of ASE and fluorescence of β-carotene on the threshold and intensity of Stokes lines was analyzed.

2 Experimental results and discussion

The intensities of spontaneous Raman scattering and resonance Raman scattering were enhanced by 2—3 orders of magnitude; meanwhile, the threshold of SRS was drastically reduced, because the Stokes lines were cumulated by LCOF[17,18].

β-carotene is a biologic molecule with a long-chain structure of 11 π-π^* conjugated double bonds, which gives it the large third-order optical nonlinearities[21]. SRS gain factor of stimulated liquid CS_2 is enhanced because of these characteristics[22].

Consequently, the high-order Stokes lines of CS_2 may be observed at the low input-laser power by the duple effects of the ASE or fluorescence of β-carotene and the LCOF. The experimental result also shows that the concentration of solution and the fluorescence profile are closely related to the threshold and intensity of Stokes lines.

2.1 Influence of ASE of β-carotene on the 1st-order Stokes threshold of CS_2

ASE is that the spontaneous emission is amplified by the stimulated radiation in the propagation process, which belongs to the laser. According to the threshold condition, the ASE may be excited when the gain equals the loss[23]. When the concentrations of solution are more than 3.72×10^{-7} mol/L, the ASE of β-carotene may be generated by the input laser. Because of the absorption loss effect of solution, all light disappears when the concentrations of solution are more than 3.72×10^{-5} mol/L. We detected the ASE of β-carotene in LCOF only, when the concentrations of solution were 3.72×10^{-5}

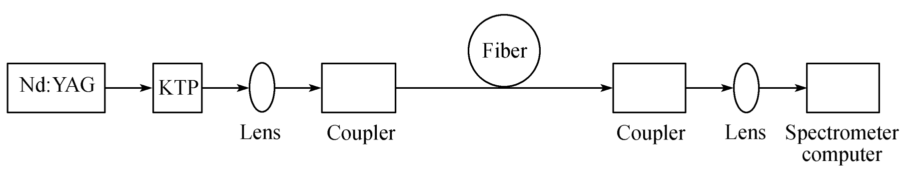

Figure 1 Experimental setup.

mol/L and 3.72×10^{-6} mol/L. Simultaneously, the competition of ASE and SRS was observed. The threshold of the SRS is more than that of ASE. When the input-laser power was 0.12 mJ with the concentration of 3.72×10^{-5} mol/L, the ASE of β-carotene was detected only, without SRS. The 1st-order Stokes line of CS_2 was observed when the power was 0.22 mJ. These thresholds were listed in the Table 1. The 1st-order Stokes line of CS_2 lies in the ASE spectral range from 520 to 560 nm, shown in Figure 2.

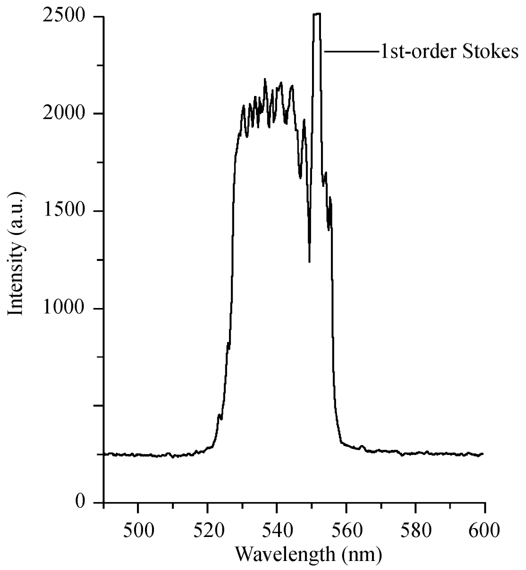

Figure 2 When the input-laser power was 0.22 mJ, the ASE of β-carotene and the 1st-order Stokes line of CS_2 were detected.

Table 1 The threshold of ASE of β-carotene and 1st-order Stokes line of CS_2 at different concentrations

Concentration (M)	Threshold (mJ)	
	ASE	1st order
3.72×10^{-5}	0.12	0.22
3.72×10^{-6}	0.15	0.26

SRS intensity initiating from quantum noise is governed by the equation[13]:

$$I_s(L) = I_{SN}\left\{\exp\left[\frac{gI_0}{\alpha_P}(1-\exp(-\alpha_P L))-\alpha_s L\right]-\exp(-\alpha_s L)\right\}. \quad (1)$$

where I_s is the SRS intensity, I_0 is the pump laser intensity, I_{SN} is the spontaneous noise within the Raman bandwidth, g is the Raman gain factor at the Stokes shift, L is the length of interaction between pump light and Raman scattering medium, and α_s and α_p are the absorption loss coefficient of Stokes lines and the pump laser. When the Stokes line lies in the gain range of ASE, Stokes radiation no longer needs to start from spontaneous Raman noise[24,25]. The intensity of Stokes lines can be exponentially increased and the threshold of Stokes lines can be drastically lowered. G is the ASE gain factor, and the intensify of Stokes lines is shown[14]:

$$I_s(L) = I_{SN}\left\{\exp\left[\frac{(g+G)I_0}{\alpha_p}(1-\exp(-\alpha_p L))-\alpha_s L\right] -\exp(-\alpha_s L)\right\}. \quad (2)$$

2.2 Influence of the concentration of solution on the each-order threshold of CS_2 with the fluorescence enhancement effect

When the concentration of solution is lower than 3.72×10^{-7} mol/L, because the number of particles decreases, the threshold condition of ASE cannot be matched, and only the fluorescence enhances the SRS. With internal seeding by fluorescence, the Stokes radiation no longer needs to start only from spontaneous Raman noise but can also initiate from fluorescence within the Raman bandwidth. Hence,

$$I_s(L) = (I_{SN}+I_f(\omega_s)) \times\left\{\exp\left[\frac{gI_0}{\alpha_p}(1-\exp(-\alpha_p L))-\alpha_s L\right]-\exp(-\alpha_s L)\right\}, \quad (3)$$

where $I_f(\omega_s)$ is the fluorescence intensity at ω_s, which is proportional to the product of the fluorescence cross section and the number of fluorescence molecules, and the spontaneous Raman intensity is proportional to the product of the spontaneous Raman cross section and the number of the CS_2 molecules. The fluorescence cross section is much larger than the Raman cross section[12]. Although the β-carotene concentration is not high, the fluorescence intensity is much larger than that of the spontaneous Raman noise. According to eq. (3), the fluorescence can effectively lower the Stokes threshold.

Table 2 summarizes the threshold of Stokes lines at different concentrations. And the curves of each-order threshold of Stokes lines are shown in Figure 3. Evidently, the thresholds of the solutions are much lower than those of the pure CS_2. In order to match the threshold condition of ASE, we should increase the concentration of β-carotene, which leads to the absorption loss effect of solution being more than the enhancement effect of the ASE and fluorescence to the SRS gain factor[12,20], and results in the threshold of Stokes increasing. Thus, the 1st-order Stokes lines threshold of concentrations of 3.72×10^{-6} and 3.72×10^{-5} mol/L is more than that of other concentrations (3.72×10^{-12}—3.72×10^{-7} mol/L).

Table 2 The thresholds of Stokes lines at different concentrations[a)]

M [b)]	Order 1	2	3	4	5	6	7
	threshold (mJ)						
−7	0.06	0.12	0.15	0.21	0.41	0.5	0.86
−8	0.12	0.18	0.24	0.31	0.45	0.55	
−9	0.12	0.20	0.36	0.54	0.95	1.17	
−10	0.18	0.28	0.38	0.60	1.05	1.25	
−11	0.18	0.31	0.50	0.74	1.25	1.55	
−12	0.20	0.42	0.60	0.80	1.35	1.65	
Pure Cs2	0.71	1.13	1.37	1.55	1.79	2.26	

a) The data in Table 2 are illustrated in Figure 3. The 7th-order Stokes line is observed at the concentration of 10^{-7} mol/L only.

b) M is the exponent (10^{-M} mol/L).

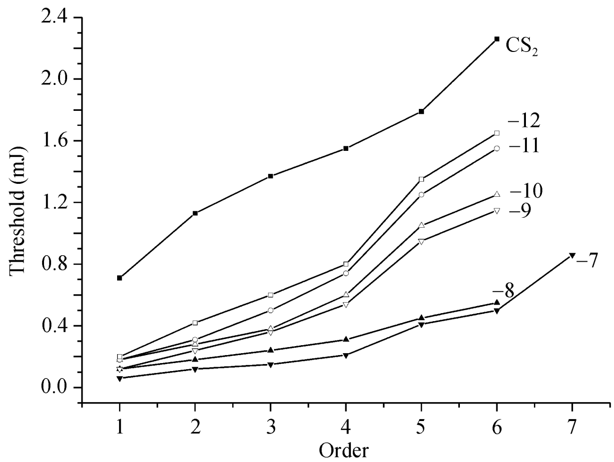

Figure 3 The relationship of each-order threshold of Stokes lines and different concentrations. ■, The pure CS_2; □, the concentration of 3.72×10^{-12} mol/L; ○, the concentration of 3.72×10^{-11} mol/L; △, the concentration of 3.72×10^{-10} mol/L; ▽, the concentration of 3.72×10^{-9} mol/L; ▲, the concentration of 3.72×10^{-8} mol/L; ▼, the concentration of 3.72×10^{-7} mol/L.

The threshold of each-order Stokes lines gradually decreases as the concentration increases: that is, the threshold of Stokes lines increases with the fluorescence intensity decreasing. This change is not linear and it relates to the absorption loss of the solution to pump laser and Stokes lines[20]. According to Raman threshold definition[26]:

$$I_s(L) = I_p(L) = I_0 \exp(-\alpha_p L), \qquad (4)$$

where L is the length of liquid core fiber, and I_p is the pump light intensity from the output end of the fiber. When the length of the liquid core fiber is 100 cm, it may be proved that $g=10^{-9}$ cm/W, $I_{SN}=0.001$ W/cm^2[27]. According to the frequency of light and the concentration of β-carotene, $I_f(\omega_s)$ ranges from 0.008 W/cm^2 to 0.013 W/cm^2. CS_2 has less light absorption. In the experiment, $\alpha_s = \alpha_p = 0.003$ cm^{-1}[28], because the absorption loss of CS_2 solution is so low. Therefore, thresholds at different concentrations can be obtained after simultaneous eqs. (2) and (3) are solved.

For example, at a concentration of 3.72×10^{-9} mol/L, we provided that the value of $I_f(\omega_s)$ is 0.008—0.011 W/cm^2 according to different orders the (the 1st—6th order). The fitting curve is shown in Figure 4, which is well consistent with the experimental data.

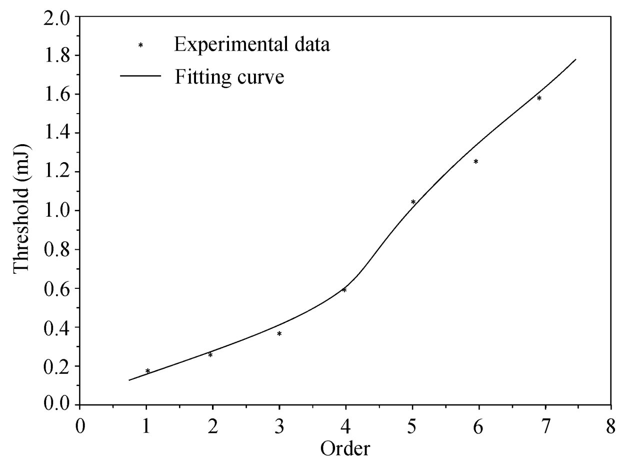

Figure 4 The threshold numerical simulation of the 1st—6th order Stokes lines and the experimental results at a concentration of 10^{-9} mol/L. The full curve is the fitting result, and * is the experimental data.

3 Conclusion

The solution of β-carotene in CS_2, with the double fluorescence characteristic, was applied as the core medium of LCOF. At different concentrations of solution, it had the different enhancement effects of β-carotene on SRS of CS_2. ASE may exponentially enhance the intensity of Stokes lines and lower the threshold of SRS at high concentrations, and the fluorescence may linearly enhance the intensity of Stokes lines and lower the threshold of SRS at low concentrations. Under the duple enhancement effects of ASE or fluorescence and LCOF, SRS may be observed at the very low input-laser power. At the β-carotene concentrations from 3.72×10^{-12} mol/L to 3.72×10^{-7} mol/L, the thresholds of Stokes lines that range from the 1st-order to the 7th-order decrease as the β-carotene concentration increases. This study not only applies the biologic molecule in nonlinear optics fields, but offers a new way to realize the broadband optical fiber Raman laser and widens the theory of the interaction between photon and phonon.

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