The stimulated Raman scattering competition between solute and solvent in Rhodamine B solution*

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The competition between the stimulated resonance Raman scattering (SRRS) of Rhodamine B (RhB) and the stimulated Raman scattering (SRS) of ethanol (C_2H_5OH) is observed at the RhB in C_2H_5OH solution. For different concentrations of the solution, the peak wavelengths of the SRRS, the amplified spontaneous emission (ASE), the fluorescence and the absorption of RhB are different. The SRRS of RhB and the SRS of C_2H_5OH are simultaneously generated when the concentration of the solution is 10^{-5} mol/L and the energy of the excitation laser is 20.4 mJ. Otherwise, only either the SRRS of RhB or the SRS of C_2H_5OH is generated. The SRRS can be amplified by the ASE gain when the SRRS is near the peak of the ASE, and the peak wavelength of the SRRS coincides with the wavelength of the maximal intensity ASE.

Keywords: stimulated resonance Raman scattering, Rhodamine B, amplified spontaneous emission

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1. Introduction

Raman lasing based on stimulated Raman scattering (SRS) is becoming an attractive research topic in the field of optical informational technology owing to its wide-band tunability.[1,2] Although Raman lasing can principally be obtained at any wavelength due to the Stokes shift, the gain coefficient of the medium is usually not high enough because of its off-resonant scattering process. On the other hand, light amplification based on stimulated resonance Raman scattering (SRRS) can enhance the gain coefficient and lower the threshold of the SRS.[3−6] Resonance Raman scattering refers to the enhancement of Raman scattering when the frequency of the incident laser is nearly resonant or resonant with an electronic transition of the molecule.[7] In the spontaneous light scattering regime, the observation of resonance Raman scattering for fluorescent molecule Rhodamine B (RhB) is difficult because of the overwhelming amount of fluorescence produced by the resonant excitation. So, the SRRS of the fluorescent molecule can obtain a high gain coefficient and a low threshold, and avoid the fluorescence noise.

In this paper, we demonstrate the SRRS of RhB by using a 532 nm excitation laser, and the fluorescent molecule RhB is dissolved in C_2H_5OH. The experimental results show that the SRRS wavelength of RhB has a red shift with an increase in the solution concentration, and the SRRS thresholds of RhB are dramatically lowered because of the amplified spontaneous emission (ASE) of RhB. Simultaneously, a competition between the SRRS of RhB and the SRS of C_2H_5OH is observed at the RhB in C_2H_5OH solution. This research has a potential application in tunable Raman lasers.

2. Experiment

RhB is dissolved into C_2H_5OH, with the solution concentration ranging from 10^{-2} mol/L to

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10^{-6} mol/L. The solution is injected into a quartz cell of 20 cm in length. As the excitation laser, a frequency-doubled Nd:YAG laser is used, which emits a laser pulse with a duration of 12 ns at a wavelength of 532 nm. It has a repetition rate of 1 Hz. The pulse energy is in the range of 0–150 mJ. The laser is coupled to the quartz cell by a 25× object lens, the output light is focused onto a spectrometer, and the signals are processed by a computer. The experimental setup is shown in Fig. 1.

The absorption spectrum measurement is carried out on a Purkinje General TU-1901 spectrophotometer with 1 nm step. All of the experiments are performed at room temperature in a dark room.

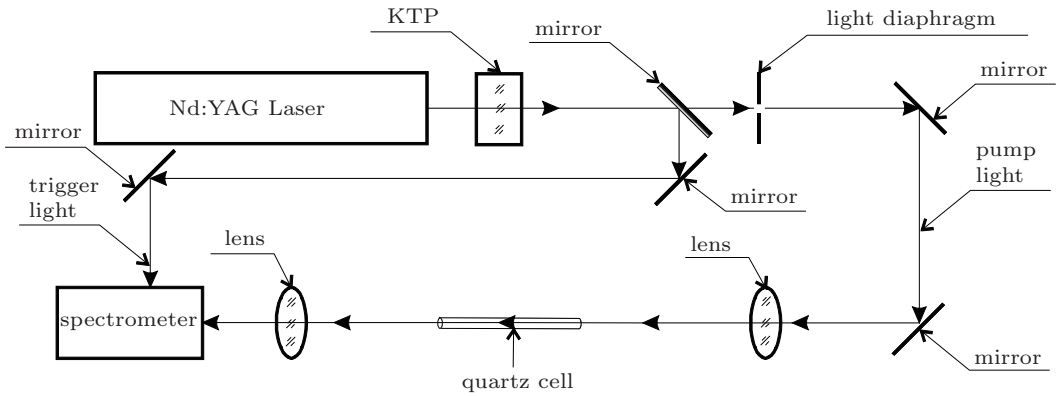


Fig. 1. The experimental setup.

3. Experimental results and discussion

The absorption spectrum, the fluorescent profile, the ASE, and the SRRS of RhB and the SRS of C_2H_5OH are measured with the RhB in C_2H_5OH solutions, respectively. By increasing the input laser energy, the evolvement process of spectra from fluorescence to SRRS can be obtained. The competition between the SRRS of RhB and the SRS of C_2H_5OH is observed by varying the concentration of the solution. The laser wavelength of 532 nm lies within the electronic resonance absorption band of the solution, with a concentration of 10^{-3} mol/L. The absorption band is shown in Fig. 2.

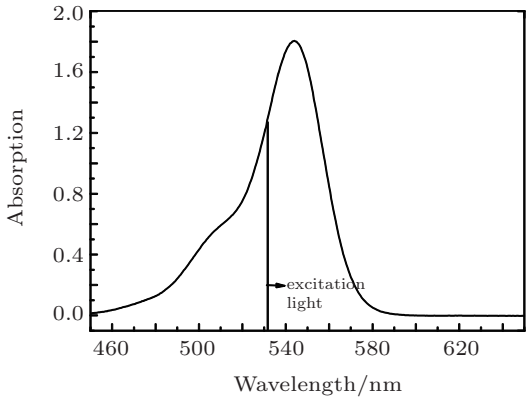


Fig. 2. The absorption spectrum of RhB in C_2H_5OH solution with a concentration of 10^{-3} mol/L.

ASE is the propagation process where the spontaneous emission is amplified by the stimulated radiation from the laser. According to the threshold condition, ASE can be excited when gain equals loss.[8] When the concentration of the solution is larger than 10^{-6} mol/L, the ASE of RhB can be generated by the input laser. Because of the absorption loss effect of the solution, all light will disappear when the concentration of the solution is larger than 10^{-2} mol/L. When the concentrations of the solution are 10^{-3} mol/L and 10^{-4} mol/L, we detect only the ASE of RhB. Simultaneously, the threshold relationship between the ASE and the SRRS is observed. The threshold of SRRS is always higher than that of ASE. When the input laser energy is 9.0 mJ and the concentration of the solution is 10^{-3} mol/L, the ASE of the solution is detected, while no SRRS is observed. By increasing the input laser energy, the ASE intensity decreases, and the SRRS of RhB correspondingly gradually increases. When the input laser energy is 14.0 mJ, the SRRS of the solution is generated, as shown in Fig. 3. The thresholds of the ASE, the SRRS and the SRS are given in Table 1 for different concentrations. The SRRS peak is located at the wavelength of the maximal intensity ASE. This phenomenon is probably attributed to the inhomogeneous broadening of the density of states in RhB.[5]

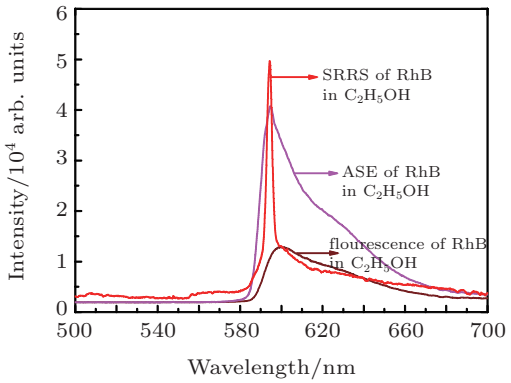


Fig. 3. (colour online) The fluorescent profile, ASE and SRRS of RhB in C_2H_5OH solution with a concentration of 10^{-3} mol/L.

Table 1. The thresholds of ASE, SRRS and SRS at different concentrations, in unit of mJ.

Concentration/mol·L^{-1}	RhB		C_2H_5OH
	ASE	SRRS	SRS
10^{-3}	9.0	14.0	–
10^{-4}	11.2	15.0	–
10^{-5}	17.0	20.4	20.4
10^{-6}	20.4	–	23.0

When the SRS line lies in the gain range of ASE, Stokes radiation no longer needs to start from the spontaneous Raman noise.[9,10] The intensity of the Stokes line can be exponentially increased, and the threshold of the Stokes line can be drastically lowered. The intensity of the Stokes line is given as[11]

$$I_s(L) = I_{SN}\Big\{\exp\Big[\frac{(g+G)I_0}{\alpha_p}\big(1-\exp(-\alpha_p L)\big) - \alpha_s L\Big] - \exp(-\alpha_s L)\Big\}, \quad (1)$$

where I_s is the SRS intensity, I_0 is the excitation laser intensity, I_{SN} is the spontaneous noise within the Raman bandwidth, g is the Raman gain factor at the Stokes shift, G is the ASE gain factor, L is the length of interaction between the excitation light and the Raman scattering medium, and α_s and α_p are the absorption loss coefficients of the Stokes line and the excitation laser, respectively. So ASE can exponentially enhance the SRRS intensity and lower the SRRS threshold.

The SRRS of RhB and the SRS of C_2H_5OH in solutions of different concentrations are shown in Fig. 4. When the concentration of the solution is 10^{-5} mol/L, the SRRS of RhB and the SRS of C_2H_5OH are simultaneously generated in the solution. However, when the concentration of the solution is lower than 10^{-5} mol/L, only the SRS of C_2H_5OH can be generated in the solution. According to the Raman gain factor[12] $g \approx \dfrac{\lambda^2}{\Delta S} N_a \sigma \dfrac{I(\nu_0)\lambda_0^2}{h\nu_0 \Delta\nu_0}$ (where λ_0 is the laser wavelength, ΔS is the area of the beam spot, N_a is the population of the up energy level, σ is the Raman scattering section, and λ is the scattering optical wavelength), when the concentration of the solution is low, the population inversion that can form the ASE is small, and g_{RhB} is also small, which cannot lead to the generation of SRRS by RhB in the solutions. However, when the RhB concentration is lowered, the C_2H_5OH concentration becomes larger, resulting in an increase in $g_{C_2H_5OH}$. The SRS of C_2H_5OH will suppress the SRRS of RhB in the solution.

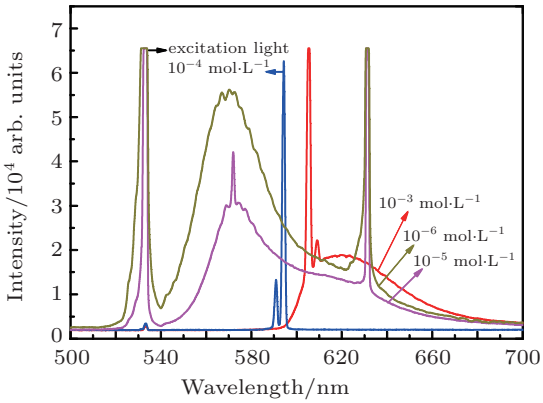


Fig. 4. (colour online) The SRRS of RhB and the SRS of C_2H_5OH in solutions with different concentrations. The SRRS of RhB and the SRS of C_2H_5OH are simultaneously generated in the solution with a concentration of 10^{-5} mol/L.

Another interesting phenomenon is that the SRRS peak wavelength of RhB varies with the concentration of the solution. This phenomenon is attributed to the solvent effect that causes the vibrational level of RhB to change, which leads to a red shift of the SRRS peak as the concentration of the solution increases.

4. Conclusion

The SRRS of RhB and the SRS of C_2H_5OH can be generated at the same time when the Raman gain factors are comparable. However, suppression and competition coexist between the solute SRRS and the solvent SRS, which leads to either the SRRS being generated by the solute or the SRS being generated by the solvent. The solvent effect leads to a red shift of the SRRS peak of RhB. The peak wavelength of the SRRS coincides with the wavelength of the maximal intensity ASE, which is attributed to the inhomogeneous broadening of the density of states in RhB. ASE can exponentially enhance SRRS intensity and lower SRRS threshold.

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