Enhancement of stimulated Raman scattering of acetone and the generation of three-color laser by using fluorescence dye RB

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The enhancement of stimulated Raman scattering (SRS) of acetone (C_3H_6O) and the generation of three-color lasers in lasing dye rhodamine B (RB) were reported. The first-order Stokes wave (629.9 nm) of SRS of C_3H_6O was amplified by 2.83 times than that of pure C_3H_6O. At the same time, a dye laser of RB at the wavelength from 575 to 598 nm can be generated in a suitable concentration of RB between 3×10^{-5} and 2×10^{-4} mol/L. Thus the green pump laser, yellow dye laser, and red Stokes wave concurred.

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Stimulated Raman scattering (SRS) as an important nonlinear optical effect and method of generating tunable coherent radiation has been extensively studied in the past decades[1−3]. But the lower transition efficient of SRS limited its real applications. Both Rhodamine 6G (R6G) and rhodamine B (RB) dye media, which have wide fluorescence band-width and high fluorescence gain, are commonly used for tunable dye laser[4,5]. Choosing suitable kind of dye fluorescent system, and making the fluorescence gain range match with the Stokes frequency, SRS effect can be enhanced[6,7]. Zhong *et al.*[8] achieved the enhancement of high-order SRS of CS_2 by dissolving RB in CS_2 and proved the selective enhancement of SRS by the dye fluorescence. The key is that the fluorescence dye material and SRS medium should be dissolved each other. A new method for enhancing SRS Stokes waves by using dye fluorescence was reported by Cheng *et al.*[9], in which a lasing dye followed the SRS medium, and with a common-beeline pump configuration. The Stokes wave of SRS medium pumped by a frequency-doubled YAG laser was enhanced in the followed lasing dye solution. A large enhancement factor of 96 was obtained for the second Stokes line of CS_2 in the experiment. Because of the larger nonlinear optical coefficient and the lower self-focusing threshold for CS_2, the output of Stokes wave is usually unstable. In order to obtain the high gain and the high stability of SRS, Raman medium acetone (C_3H_6O) and fluorescent dye RB have been used in this paper. C_3H_6O has a lager Raman shift of 2921 cm^{-1}, less nonlinear optical coefficient, and high self-focusing threshold. The fluorescent dye RB has a broadband fluorescence range of $565-598$ nm. Using C_3H_6O as the SRS medium and RB as the fluorescence enhancement medium, a good stable enhanced Stokes wave with larger Raman shift can be obtained. At the same time, a dye laser of RB could be excited in the dye solution. It means that a novel three-color laser including pump laser, Stokes wave, and dye laser can be gotten in the experimental.

The experimental setup is shown in Fig. 1. A *Q*-switched frequency-doubled Nd:YAG laser was used as SRS and dye laser pump source. The pump laser (532-nm wavelength, 10-ns pulse width, ∼ 15-mJ single pulse energy) was focused by a lens (10-cm focusing length) at the center of the Raman cell (10-cm length) filled with C_3H_6O to produce SRS. A portion of the laser beam was split and used for detection system synchronization. Followed the SRS cell, a dye cell of RB ethanol (C_2H_5OH) solution cell (10-mm length) was used for enhancing the SRS. A grating spectrometer coupled with a CCD optical multi-channel-analyzer (OMA) was used to collect the experimental spectral data both before and after the dye RB solution. Usually, 100 spectra were collected, averaged, and saved in random-access memory (RAM) of the computer.

In the experiment, SRS of C_3H_6O was excited by the frequency-doubled YAG laser, then the Stokes wave of C_3H_6O and the residual pump laser passed through the fluorescent cell containing RB. With a powerful

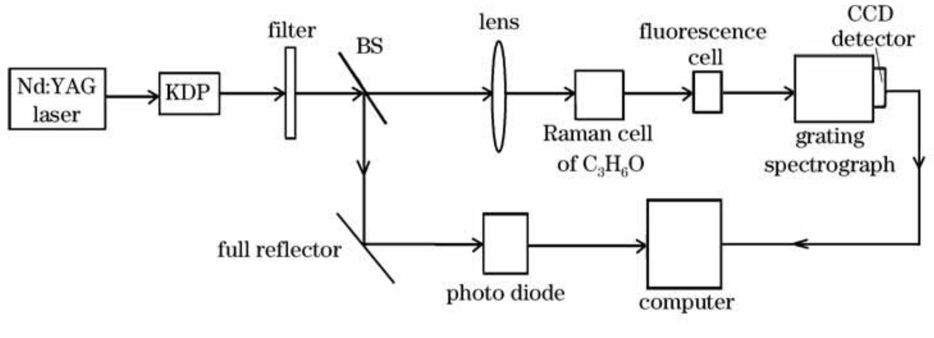

Fig. 1. Schematic diagram of the experimental setup.

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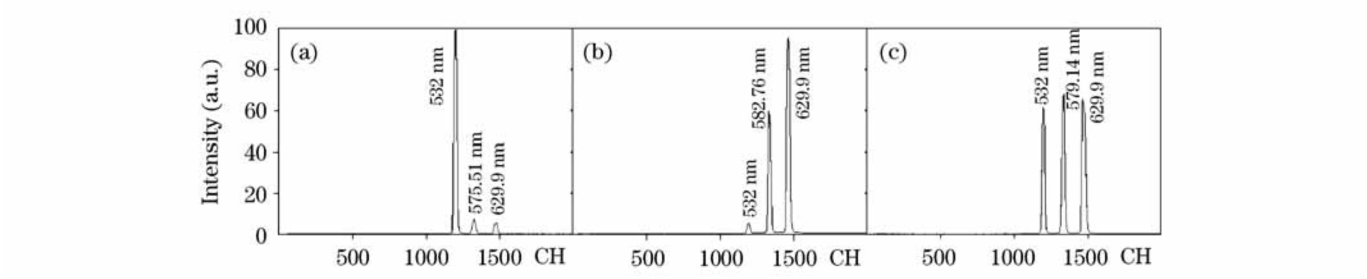

Fig. 2. Experimental spectra of three-color laser at different RB concentrations of (a) 3×10^{-5} mol/L, (b) 8×10^{-5} mol/L, and (c) 4×10^{-5} mol/L.

fluorescence in the dye medium excited by the residual pump laser (532-nm wavelength), the first-order Stokes line of C_3H_6O was enhanced by the RB gain medium. Through controlling the pump laser power and the concentration of RB, both the enhancement of Stokes wave and a dye laser of RB could be generated. Thus a green pump laser with wavelength of 532 nm, an enhanced Stokes line with wavelength of 630 nm, and a dye laser with tunable wavelength from 575 – 598 nm could be emitted simultaneously. In addition, the relationship of intensities of the three-color laser could be changed along with the variety of the dye concentration. The concentration of RB in ethanol can be changed from 2×10^{-5} to 1×10^{-3} mol/L. The three-color laser spectra were shown in Fig. 2. The three lines are the pump laser (532 nm), the dye laser (575-587 nm), and the first Stokes line (630 nm) of C_3H_6O, respectively. And the concentrations of RB are 3×10^{-5}, 8×10^{-5}, and 4×10^{-5} mol/L for Figs. 2(a), (b), and (c), respectively. When the concentration of RB is 4×10^{-5} mol/L, the ratio of the intensities between three-color lasers is closed to 1:1:1 as shown in Fig. 2(c).

Comparing with Figs. 2(a) and (b), the first Stokes line of C_3H_6O had been powerfully enhanced. The enhancement coefficient of Stokes line can be defined as $a_c = I_c/I_0$, where I_0 is the intensity of Stokes line of pure C_3H_6O and I_c is the intensity of the Stokes line after enhanced by the lasing dye of RB. Figure 3 showed the different enhancement coefficients of the first Stokes line under the various concentrations of RB. As observed in Fig. 3, when the dye concentration was increased from 0 to 8×10^{-5} mol/L, the intensity of the first Stokes line was enhanced obviously. When the concentration was increased on and on, the enhancement of Stokes line tended to gentle. At a RB concentration of 4×10^{-4} mol/L, the intensity of Stokes line is increased optimally by about 2.83 times than that of pure C_3H_6O. At that time, when the intensity of pump laser (532 nm) was 13.5 mJ and the pulse width was 10 ns, the output intensity of Stokes wave was 84.4 μJ, then the energy conversion coefficient reached to 0.6%. When the concentration of RB exceeded 4×10^{-4} mol/L, the intensity of Stokes line began to weaken due to the self-absorption of RB for the pump laser of 532 nm. When the concentration of RB reached to 1×10^{-3} mol/L, SRS of C_3H_6O could not be enhanced and the dye laser of RB disappeared.

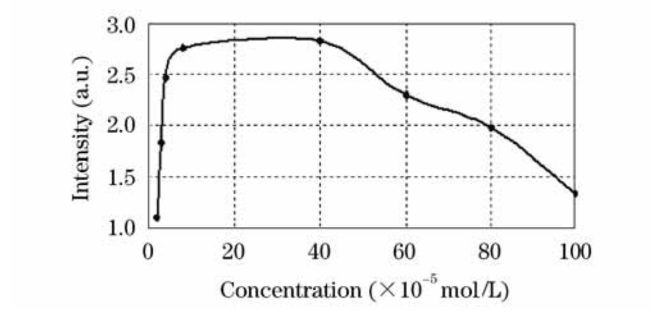

Fig. 3. The relationship between the intensity of the first Stokes line and the concentration of RB.

The mechanism of enhancement of SRS of C_3H_6O by the lasing dye is similar as that of the amplification of a laser beam in a laser gain medium. When the dye medium RB was excited by the residual pump laser at 532 nm, the pump energy was stored in the molecules of RB, thus the RB medium was excited to the state of population inversion. The Stokes wave as a signal light passed through the active medium; the light amplification of stimulated emission process occurred with the reversed particles falling to the ground state. In this case, the Stokes line coming out from the SRS medium of C_3H_6O can be enhanced exponentially in RB dye cell. The relationship among the intensity of Stokes line I_s, the incident intensity $I_s(0)$ and the interaction factor $\exp[I_p(g-\alpha)L]$ is

$$I_s = I_s(0)\exp[I_p(g-\alpha)L], \tag{1}$$

where I_p is the intensity of the residual pump laser at 532-nm in the dye cell, g is the gain coefficient of the dye fluorescence at Stokes wavelength, α is the dye absorption at Stokes wavelength, and L is the dye cell length.

The residual pump laser can also pump the dye laser just in a suitable concentration of RB. In our experiment, the wavelength of the dye laser was from 575 to 598 nm when the concentration of RB increased from 3×10^{-5} to 8×10^{-4} mol/L. With increasing the RB concentration, the peak wavelength of the dye fluorescence appeared red shift. The reason is that the transition of the reversed particles from the excited state to ground state is always to the lower vibration level in a dye laser, if more reversed particles distribute to the higher vibration energy level, so the red shift was emitted when the particles returned to the ground state.

The lasing dye RB with absorption band of 450 – 600 nm absorbs powerfully the pump laser (532 nm). The absorption coefficient of RB for the pump laser increased with the increase of the concentration of RB, when the concentration reached to 4×10^{-4} mol/L, the power of pump laser in RB decreased rapidly. The self-absorption at the high concentration could weaken even vanish the intensity of fluorescence and the enhancement of SRS. For the concentration of RB from 3×10^{-5} to 2×10^{-4} mol/L, the pump laser excited C_3H_6O to generate SRS in Raman cell, enhanced the Stokes wave in fluorescence cell, and pumped the RB in ethanol to produce the dye laser. The three-color lasers were concurrence. Because the intensities of pump laser, dye laser, and Stokes wave were related to the concentration of RB, the concentration could influence the intensity relation of three-color lasers. Choosing a suitable RB concentration, the correctly intensity relationship between the three-color lasers could be gotten. The direction of the dye laser can be changed along with the feedback direction of the sample cell, and the output direction of Stokes wave is the same as that of pump laser. Thus the direction relationship of the three-color lasers can be adjusted.

Similar experimental results had been also carried out in other SRS enhanced by dye fluorescence system. Stokes wave of C_3H_6O was amplified resonantly by using R6G dye fluorescence. When the concentration of R6G in ethanol ranges from 3×10^{-5} to 8×10^{-5} mol/L, the three-color lasers were exported stably. For a concentration of 6×10^{-5} mol/L, the first-order Stokes line was enhanced by 1.97 times than that of pure C_3H_6O. In another system of C_2H_5OH+RB, when the Raman cell length was 20 cm and the dye cell length was 1 cm, the Stokes wave of C_2H_5OH was amplified approximately 4.0 times by RB dye.

The results of enhancement of SRS by dye fluorescence and generation of the three-color lasers give a possibility for increasing SRS efficiency, and enhancing the range of real applications of SRS in tunable laser.

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