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Multiplication of Spectral Lines Generated by Two-Color Stimulated Raman Effect

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A dye laser emitting at four different frequencies separated by 293.5 (=587/2) cm^{-1} is made by inserting an etalon in the resonator cavity of the laser. This laser beam is focused into molecular hydrogen (rotational Raman shift frequency, 587 cm^{-1}) to generate a multicolor laser beam consisting of more than 10 rotational lines in the vicinity of the fundamental lines by four-wave Raman mixing. Such rotational lines also occur in the vicinity of the vibrational Raman lines. Thus more than 25 emission lines appear simultaneously. This approach is useful to multiply the line density, i.e., the emission lines within a specified wavelength region.

Key words: multifrequency dye laser, etalon, multicolor laser beam, hydrogen, four-wave Raman mixing

1. Introduction

The two-color stimulated Raman effect, i.e., simultaneous generation of many rotational and vibrational Raman emissions, which is produced by the introduction of a two-color laser beam into a Raman medium, represents a possible method for the generation of multicolor laser emission. When hydrogen is used as a Raman medium, more than 50 emission lines are simultaneously generated; these are separated by 587 cm^{-1} corresponding to the transition energy between the rotational levels of $J=1$ and $J=3$. These lines are also separated by 4155 cm^{-1} corresponding to the transition energy between the vibrational levels $v=0$ and $v=1$.[1,2] Such a multicolor laser could be useful for a variety of applications, e.g., to generate ultimately-short optical pulses,[3] to illuminate artistic presentations such as laser light shows,[4] and as a light source in a laser-radar (lidar) system for remote sensing.[5,6] To increase the line density, i.e., the number of emission lines within a specified wavelength region, it is possible to use a different Raman medium having a smaller Raman shift frequency. Unfortunately, hydrogen is the most efficient Raman medium, and no substitute for it is readily available. An alternate approach might involve the use of para-hydrogen with a smaller rotational Raman shift frequency of 354 cm^{-1}, corresponding to the transition energy between the rotational levels of $J=0$ and $J=2$.[7] However, para-hydrogen is less stable and 3/4 of it is converted into ortho-hydrogen at room temperature. Another method is the use of a multifrequency laser with a frequency separation of $587/n$ cm^{-1}, where n represents the multiplication number. For example, in case of $n=2$, at least four emission lines separated from each other by 293.5 cm^{-1} are needed for the present purpose, in which a pair of lines separated by 587 cm^{-1} acts as pump and seed beams, and another pair separated by 587 cm^{-1} as the other pump and seed beams. The output beams from four independent dye lasers could be combined to construct a laser which emits at four frequencies and which acts as two pump beams and two seed beams in a two-color stimulated Raman effect. However, this system is expensive and adjustment of the laser beams is quite difficult temporally, spatially, and spectrally.

In this study, we report the construction of a dye laser which emits at four different equally spaced frequencies. To achieve this, an etalon is inserted into the dye laser cavity and is slightly tilted as the frequency spacing approaches 293.5(=587/2) cm^{-1}. Coumarin 307, because of its broad gain bandwidth, is employed as a laser dye. This dye laser beam is focused into molecular hydrogen to generate multicolor laser emission by four-wave Raman mixing. This approach allows multiplication of the line density and provides conditions conducive for better reproduction of light and color in illumination work.

2. Experiment

The experimental apparatus constructed in this study is shown in Fig. 1. The dye laser consists of an output coupler (Sigma Koki, beam wedge, quartz), a beam expander (Oriel, magnification 20), a deposited etalon (Technical Optics, ED-200, free spectral range 293.5 cm^{-1}, finesse 100), an end mirror (Sigma Koki, dielectric mirror), and a dye cell (1×1 cm) containing coumarin dye (Exciton, Coumarin 307, 10^{-2} M) which is pumped by focusing a split beam of an excimer laser (Lambda Physik, LPX 205, 380 mJ, 5 Hz) with a cylindrical lens (Sigma Koki, quartz, focal length 10 cm). The etalon is mounted on a rotation stage (Sigma Koki, KSA-120PM) in order to tilt the angle for fine adjustment of the free spectral range. A beam expander is used to enhance the spectral resolution and to reduce the optical damage of the etalon. After amplification of the oscillator beam by passing it through a second dye cell (Exciton, Coumarin 307, 3×10^{-3} M), which is pumped by the remaining portion of the excimer laser by the cylindrical lens (Sigma Koki, quartz, focal length 10 cm), the beam is focused into a hydrogen-filled Raman cell (Iwatani, purity 99.9%) by a spherical lens (Sigma Koki, quartz, focal length 20 cm). The spectrum of the output

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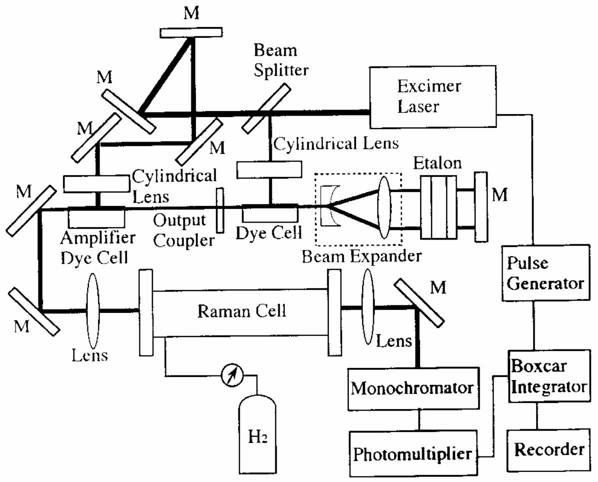

Fig. 1. Block diagram of the experimental apparatus used for the generation of multicolor laser emission.

beam is measured by a monochromator (Jasco, CT-50) equipped with a photomultiplier (Hamamatsu, R1447). The pulse energy of the laser beam can then be measured using either a power meter (Ophir Optronic, DG HH) or a pyroelectric device (Molectron, Joule Meter, J3-05 DW).

3. Results and Discussion

3.1 Dye Laser

The output energies of the dye laser from the oscillator and the amplifier were 0.5 mJ and 25 mJ, respectively. The polarization of the dye laser beam is considered to be perpendicular to the reference plane. The emission spectrum of the amplified beam is shown in Fig. 2. The emission lines were separated from each other by 299.6±7.8 cm^{-1}. The spectral linewidth of each emission line was approximately 0.7 nm (27 cm^{-1}), the finesse being 10. Thus the frequency separation of the emission lines coincides with a value of 293.5(=587/2) cm^{-1} within experimental error. The finesse observed was rather small, a factor which could be due to a defect in the etalon or to poor alignment of the dye laser. The number of emission lines, however, exceeded four and was sufficient for the purposes of the present study. At the beginning of this study, we expected to be able to generate a larger number of emission lines, due to the broad gain bandwidth of Coumarin 307 (479-553 nm,[8] $\Delta\nu$=2794 cm^{-1}, 2794/293.5=9.5). The smaller number of emission lines observed may be ascribable to competition of the emission lines in laser oscillation and amplification, i.e., enhancement of strong lines at the gain center and suppression of weak lines at the wings of the gain curve; the tunable range, however, can be extended to the entire spectral region where the dye laser gain exceeds the loss of the resonator cavity. Other laser dyes such as 4,4'''-bis-(2-butyloctyloxy)-p-quaterphenyl (BBQ) were also investigated, but using these it was difficult to simultaneously generate more than four emission lines so that these dyes were not used in this study. In order to avoid such competition, it is desirable to isolate the pumping region for the laser beams emitting at different wavelengths.[9] Therefore, such a technique might be useful for other laser dyes like BBQ, although the laser cavity would predictably become more complicated.

3.2 Four-Wave Raman Mixing

The multicolor laser emission generated by passing the

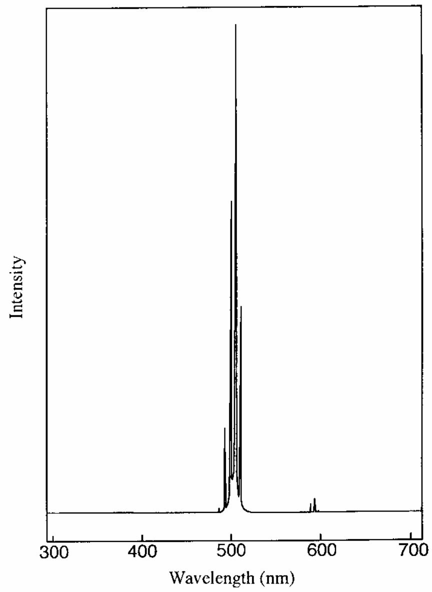

Fig. 2. Emission spectrum of the dye laser. Five emission lines, separated by 587/2 cm^{-1}, are generated. The lines near 600 nm are due to stray light.

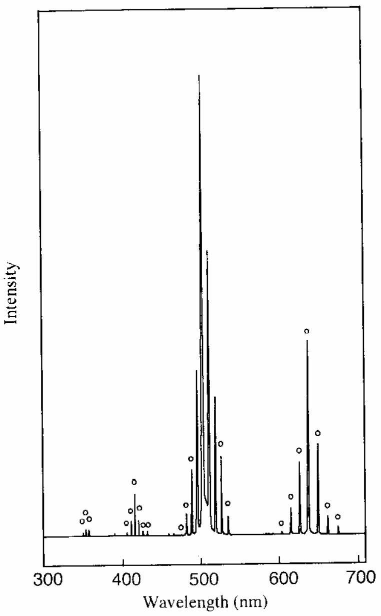

Fig. 3. Emission spectrum of a multicolor laser generated by the two-color stimulated Raman effect. The hydrogen pressure is adjusted to 8 atm. Many emission lines, separated by 587/2 cm^{-1}, are observed in the vicinity of the fundamental and vibrational Raman lines. The peaks marked by open circles are new or strongly enhanced spectral lines through four-wave Raman mixing.

above multifrequency laser beam into a Raman cell pressurized with hydrogen at 8 atm is shown in Fig. 3. The pulse energy of the laser beam from the Raman cell was 14 mJ. As in a previous study using a two-frequency beam, numerous rotational lines occur in the vicinity of the fundamental and vibrationally shifted Raman lines. More than 25 emission lines appear simultaneously. Such an experiment was also performed at 6 atm. The rotational lines were more pronounced in the vicinity of the fundamental lines, and more than 10 emission lines could be clearly observed. As expected,[2)] however, the rotational lines in the vicinity of the vibrational lines were more suppressed and the total number of emission lines was reduced to 22. Unfortunately, the threshold for generation of multicolor laser emission (at which the gain is 20–30 and the intensity of the emission line increases to ca. 1% of the fundamental line) was high in the present study, so that the frequency domain covered was narrow in comparison with the case using a two-color beam.[1–4)] This was so even when the pulse energy was increased from 15 mJ to 25 mJ. These results are probably due to the facts that the line density is doubled and two series of emission lines separated by 587 cm^{-1} appear independently. It is apparent that no interaction (e.g., a concerted effect through nonresonant four-wave Raman mixing) occurs, which is probably due to there being similar intensities for these two series of emission lines. Infrared tunable laser emission is strongly enhanced through nonresonant four-wave Raman mixing when a strong fixed-frequency Nd:YAG laser is introduced as a pump beam in addition to a $\mathrm{LiNbO_3}$ optical parametric oscillator.[10,11)] It is also noteworthy that the spectral linewidth of the dye laser is rather broad (0.7 nm), in contrast to 0.007 nm found in our previous studies.[2)] This may provide a higher threshold and lower efficiency, although the efficiency of four-wave Raman mixing responsible for generation of rotational lines is more insensitive to the spectral linewidth than stimulated Raman scattering, which is responsible for the generation of the first Stokes Raman line.[12)]

3.3 Further Developments

For applications to laser illumination it is desirable to generate a multicolor laser beam consisting of many emission lines over the entire visible region. In the present case there are spectral regions where emission lines are absent due to poor efficiency. A transform-limited high-peak-power laser with a narrow spectral linewidth (e.g., in the picometer range) and a short pulse width (e.g., picoseconds) would be especially useful to improve efficiency in the present approach.[12)] To generate multicolor laser emission completely equally-spaced over the entire spectral region, it would require suppressing the vibrational lines and enhancing the rotational lines. This can also be achieved by optimization of conditions, such as hydrogen pressure,[2)] beam focusing conditions,[13)] the linewidth[10)] and the wavelength of the laser.[14)]

4. Conclusions

More than 25 laser emission lines can be generated simultaneously by focusing a laser beam into molecular hydrogen, based on four-wave Raman mixing. The line density is doubled by using a dye laser emitting at four different frequencies separated by 587/2 cm^{-1} as two pump and two seed beams. Such a multicolor laser may have broad applications in areas such as a laser radar (lidar) for atmospheric monitoring, and also as a laser light show for illusional presentation in art.

Acknowledgments

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References

1) T. Imasaka, S. Kawasaki and N. Ishibashi: Appl. Phys. B 49 (1989) 389.
2) S. Kawasaki, T. Imasaka and N. Ishibashi: Appl. Phys. B 52 (1991) 211.
3) S. Yoshikawa and T. Imasaka: Opt. Commun. 96 (1993) 94.
4) S. Ohtake, S. Yoshikawa and T. Imasaka: Appl. Opt. 34 (1995) 4337.
5) Y. Sasano and E.V. Browell: Appl. Opt. 28 (1989) 1670.
6) Z. Chu, U.N. Singh and T.D. Wilkerson: Appl. Opt. 30 (1991) 4350.
7) S. Kawasaki, T. Imasaka and N. Ishibashi: J. Opt. Soc. Am. B 8 (1991) 1461.
8) U. Brackmann: *Laser-grade Dyes* (Lambda Physik GmbH, D-3400 Göttingen, West Germany, 1986) p. 79.
9) M.B. Danailov and I.P. Christov: Appl. Phys. B 51 (1990) 300.
10) Y.-R. Shen: *Nonlinear Infrared Generation*, eds. R.L. Byer, R.L. Herbst (Springer, New York, 1977) Chapt. 3, p. 108.
11) S.J. Brosnan, R.N. Fleming, R.L. Herbst and R.L. Byer: Appl. Phys. Lett. 30 (1977) 330.
12) Y. Irie and T. Imasaka: Opt. Commun. 113 (1994) 105.
13) S. Yoshikawa, S. Kawasaki, T. Imasaka and N. Ishibashi: Jpn. J. Appl. Phys. Lett. 30 (1991) 283.
14) H. Kawano, C.H. Lin and T. Imasaka: Appl. Phys. B 63 (1996) 121.