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Generation of high-order rotational lines by four-wave Raman mixing using a high-power picosecond Ti:Sapphire laser

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Abstract. More than thirty rotational lines equally spaced by 587 cm^{-1} are generated simultaneously in the vicinity of the fundamental line by four-wave Raman mixing using a high-power picosecond Ti:Sapphire laser as a pump source and hydrogen as a Raman medium. Since the wavelength of this multifrequency laser emission extends from the near-infrared to the near-ultraviolet, it can be utilized as a tunable light source for picosecond spectroscopy. Because of the wide spectral bandwidth available, this procedure has great potential for the generation of ultrashort laser pulses by mode-locking these emission lines.

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Laser techniques have been used in a variety of scientific fields, both basic and applied. Because the laser has a narrow spectral bandwidth, it is frequently utilized as a light source for spectroscopy. However, most lasers have limited tunability, thus making it difficult to cover a wide spectral region using a single optical system. Recently, several types of solid-state lasers which have wide tunability have been reported. Among these are the Ti:Sapphire laser or an Optical Parametric Oscillator (OPO). In spite of these advances, it still remains difficult to cover the complete visible region, even by employing second harmonic emission of the fundamental beam.

Stimulated Raman Scattering (SRS) has been used for frequency conversion of the tunable solid-state laser [1]. Hydrogen is commonly used as a Raman medium, because of its high conversion efficiency and large frequency shift (4155 cm^{-1}). When the tunable range of the laser is limited, rotational stimulated-Raman emission may be used instead, because of the smaller frequency shift (587 cm^{-1}). However, only a rotational line can be generated in the vicinity of each vibrational line, due to a selection rule in rotational transition, thus making it difficult to cover a wide spectral region.

Recently, in this laboratory, we reported on a multifrequency laser consisting of more than 40 vibrational and rotational Raman lines [2]. The spectral region extends from the near-infrared to the ultraviolet, and, as a result, it allows a continuous coverage of the entire spectral region of interest. This multifrequency laser, which consists of high-order vibrational and rotational lines, is generated by four-wave Raman mixing (FWRM) using a two-color pump beam separated by 587 cm^{-1}, which corresponds to the rotational transition of hydrogen which is used as a Raman medium. Johnson and co-workers reported stimulated Raman emission consisting of vibrational and rotational lines [3], but vibrational lines were more pronounced. Wilkerson and co-workers reported the generation of only high-order rotational lines using a high-power picosecond Nd:YAG laser (1064 nm) [4], but the intensities shown in the spectrum are exactly the same for all rotational lines. These workers did not comment on the reasons for this.

The generation of ultrashort pulses is a big challenge in laser technology. As of this writing, 6-fs pulses have been produced by self-phase modulation of ultrashort dye laser pulses and by compression using pairs of prisms and gratings [5]. It is, however, difficult to compensate the group velocity dispersion further, because of the large frequency-dependence of the refractive indices for a solid laser material and intra-cavity optics. In order to overcome this problem, several approaches have been reported for the generation of ultrashort pulses [6, 7]. In one case, fundamental, frequency-multiplied, or frequency-differentiated beams were coherently superimposed. In another, a multifrequency laser beam generated by harmonic generation was assumed to be mode-locked. However, the intensity distribution obtained is not flat and then the resulting spectral domain available is marginal in terms of the generation of ultrashort pulses. Recently, we proposed a new approach for generation of

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*To whom all correspondence should be addressed ultrashort pulses [8]. In that study, we reported on the possibility of mode-locking all the rotational Raman lines in order to generate ultrashort optical pulses. Generation of 6-fs pulses was estimated by Fourier transformation of the rotational lines, which were observed experimentally, using a computer simulation. The pulse width can be reduced by increasing the number of rotational lines. Thus, shorter pulses are expected to be generated by using a higher-peak-power laser for improvement of efficiency.

In this paper, we report the generation of picosecond multifrequency laser emission covering the spectral region from the near-infrared to the ultraviolet by using only the rotational lines generated. The present results suggest that four-wave Raman mixing can be useful for generation of a widely tunable light source and also for the generation of ultrashort laser pulses.

1 Experimental

Figure 1 shows the experimental apparatus used in this study. An argon-ion laser (Spectra Physics, Model 2060-7S) is used as a pump source for a mode-locked Ti:Sapphire laser (Spectra Physics, Tsunami, 82 MHz). A single picosecond laser pulse is injected by a Q-switch into a regenerative amplifier to generate an 1-mJ pulse. The pulse ejected from the regenerative amplifier by the Q-switch is then passed through two linear amplifiers, which are both pumped by the same harmonic beam of a Nd:YAG laser (Spectra Physics, GCR-6). This system produces 100-ps, 500-MW (50 mJ) pulses at a repetition rate of 10 Hz. For the present study, the emitting wavelength was adjusted to 827 nm. The laser beam is passed through a quarter-wave plate to obtain an elliptically polarized beam and is focussed into a Raman cell (length, 30 cm) with 10-mm thick quartz windows by a quartz lens (focal length, 30 cm). The Raman cell is filled with hydrogen pressurized to 10 atm. The pump beam passes through the Raman cell twice, in order to improve the conversion efficiency. The spectrum of the multifrequency laser emission generated is measured by a monochromator (Jasco, CT-25C) equipped with a photomultiplier (Hamamatsu, R1477). The output signal is measured by a boxcar integrator and is displayed on a strip chart recorder. The pulse width is measured by a combination of a photodiode (Hamamatsu, Model C4258) and an oscilloscope (Iwatsu, Model SAS-6018).

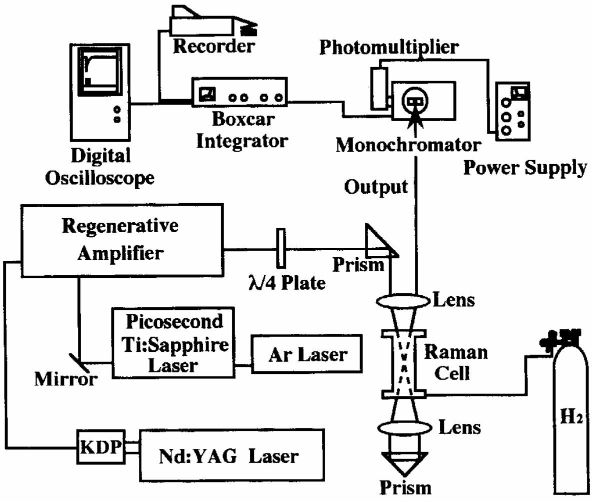

Fig. 1. Experimental apparatus for the generation and detection of multifrequency laser emission

2 Results and discussions

Figure 2 shows the spectrum of multifrequency laser emission generated by FWRM. Numerous high-order rotational lines are observed in the vicinity of the fundamental beam. The rotational lines up to the fifteenth anti-Stokes emission appear in the spectrum. Furthermore, generation of rotational lines up to the twenty-seventh anti-Stokes emission (358 nm) could be confirmed by increasing the sensitivity of the detector. At longer wavelengths, rotational lines at least down to the second Stokes emission appear in the spectrum. Unfortunately, the spectral response of the photomultiplier used in this study decreased rapidly beyond 930 nm. Higher-order Stokes emission might conceivably have been generated in the infrared region, although we cannot confirm this using the present detection system. There are small rotational lines in the vicinity of the first vibrational anti-Stokes line that appears at 615 nm. The seventh rotational anti-Stokes emission is located at 617 nm, so that several split-like lines are observed in the spectrum. In our earlier studies, the vibrationally shifted rotational lines were much stronger than the high-order pure rotational lines around the region where the frequency is displaced by 4155 cm^{-1} from the fundamental line. Thus, pure rotational lines are definitely more pronounced when a high-power picosecond Ti : Sapphire laser is employed as a pump beam.

The first vibrational and rotational Stokes lines are generated by the stimulated Raman effect. The gain, G, of

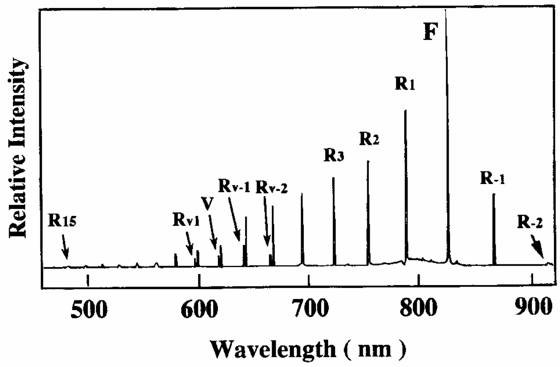

Fig. 2. Spectrum for multifrequency laser emission. F: fundamental line; R: rotational lines; Rv: vibrationally-shifted rotational lines; V: vibrational line. The subscript indicates the order of the rotational line

SRS is described by (1) [9]:

$$G = \exp(g_R I_p l), \tag{1}$$

where g_R is the Raman gain coefficient, I_p is the intensity of a pump beam, and l is the interaction length. The parameter, g_R is described by

$$g_R = \frac{32\pi^3 c^2 \Delta N}{n_i^2 h \omega_i^3 \Delta\omega_R}\left(\frac{\partial\sigma}{\partial\Omega}\right), \tag{2}$$

where c is the velocity of light. ΔN is the difference of the populations in the excited and ground states, n_i is the refractive index and ω_i is the frequency of the first Stokes emission, respectively. $\Delta\omega_R$ is the Raman linewidth, and $(\partial\sigma/\partial\Omega)$ is the cross-section of Raman scattering. On the other hand, all anti-Stokes and high-order Stokes lines are generated by FWRM. The intensity of the first anti-Stokes beam generated by FWRM, I_{FWRM}, is given by (3), when it is sufficiently weak [10],

$$I_{\mathrm{FWRM}} = \frac{9\omega_s^2 N^2}{(2c)^4 \varepsilon_0^2 n_s n_i n_p^2} |\chi^{(3)}_{\mathrm{FWRM}}|^2 l^2 I_i I_p^2 \sin c^2\left[\frac{\Delta k l}{2}\right], \tag{3}$$

where ω_s is the frequency of the first anti-Stokes beam generated, N is the density of a Raman medium, ε_0 is the dielectric constant of vacuum, n_s and n_p are the refractive indices for the signal and the pump beam, respectively, I_i is the intensity of the first Stokes line, and Δk is the degree of phase mismatching. $\chi^{(3)}_{\mathrm{FWRM}}$ is the third-order nonlinear susceptibility, described by (4) [11]:

$$\chi^{(3)}_{\mathrm{FWRM}} = \frac{N^2}{\Omega^2 \Delta\omega_R^2}\left(\frac{\partial\alpha}{\partial Q}\right)_0^4, \tag{4}$$

where α is the polarizability, Q is the coordinate of molecular vibration, and Ω is the Raman shift frequency. Equation (3) may be used to optimize the experimental conditions in obtaining many rotational Raman lines with a flatter intensity distribution, since occurrence of the first anti-Stokes emission is the first step for generation of higher-order emissions through FWRM.

In this work, the peak power of the Ti:Sapphire laser used as a pump beam was several orders of magnitude larger than the lasers used in our previous studies. The intensity of the first Stokes emission is proportional to the exponential of the laser power. On the other hand, the intensity of the first anti-Stokes emission generated by FWRM is proportional to the square of the pump beam power and is proportional to the first Stokes beam power. The linewidth of the pump laser should be sufficiently narrow so as to increase the gain of SRS to generate the first Stokes emission in order to facilitate the generation of many Stokes and anti-Stokes emissions. It should also be noted that the pump laser is nearly Fourier-transform-limited, and there is no appreciable change in the linewidth between the nanosecond laser previously used, and the picosecond laser used in this study. As a result, high-order rotational lines are very efficiently generated in this system. The pump beam employed in this study is elliptically polarized; the ellipticity is, unfortunately, not optimized to generate many rotational lines efficiently. In fact, we could visually observe a very large number of rotational lines generated at higher efficiencies over a period of several seconds, when attempts were made to optimize the ellipticity of the pump beam. However, serious damage occurred in the Ti:Sapphire laser system, probably due to a strong feedback of the back-scattered beam caused by FWRM, and, as a result, it was not possible to measure the spectrum and the pulse energy using this instrumental setup. Further improvement might be possible by the optimization of experimental conditions, e.g., hydrogen pressure, polarization of the pump beam, the focal length of the lens, etc.

As described above, the seventh rotational anti-Stokes emission observed at 617 nm is generated more efficiently than the first vibrational anti-Stokes emission observed at 615 nm. Such a rotational line was less efficient in the previous studies which utilized nanosecond visible dye lasers. The high efficiencies for rotational lines in the present study can probably be ascribed to the longer emitting wavelength of the Ti:Sapphire laser; since the gain coefficient is reciprocally proportional to the cube of the Raman frequency as shown in (2). Therefore, the first vibrational Stokes emission occurs with difficulty in the near-infrared region, which also decreases the efficiency of the vibrational anti-Stokes emission subsequently generated by FWRM.

A continuous-wave (CW) Ti:Sapphire laser has a wide tunable range from 700 to 1000 nm. However, the tunable range for the present high-power picosecond Ti:Sapphire laser is limited to 760–840 nm, because of the limited spectral bandwidth of the dielectric coating used for the mirrors in the regenerative amplifier. This tunable range corresponds to 1253 cm^{-1}, which is much larger than the rotational Raman shift frequency (587 cm^{-1}) but is smaller than the vibrational Raman shift frequency (4155 cm^{-1}). This means that continuous coverage of the visible region cannot be accomplished using vibrational lines but can readily be accomplished using rotational lines. Thus, the present multifrequency laser consisting of high-order rotational lines generated by FWRM is useful as a picosecond laser source tunable in a wide spectral region.

A large number of rotational lines are generated simultaneously using a high-power picosecond laser which emits at long wavelengths. The spectral region covered extends to more than 500 nm. By Fourier transformation of the emitting lines (observed in Fig. 2), the pulse width could be reduced to 5 fs. This result suggests that a high-peak-power laser emitting at long wavelengths is useful for efficient generation of many rotational lines and for pulse shortening by mode locking these emission lines. In our previous report, we found that the efficiency is much less affected by the spectral bandwidth in FWRM than in SRS [11]. In addition, according to (3) and (4), the intensity of the beam generated by FWRM, I_{FWRM}, is proportional to $I_p^2 I_s/\Delta\omega_R^2$; $\Delta\omega_R$ should be replaced with the linewidth of the laser, $\Delta\omega_L$, when $\Delta\omega_L > \Delta\omega_R$. If a pump laser is Fourier-transform-limited, the linewidth is reciprocally proportional to the pulse width which is reciprocally proportional to the peak power when the pulse energy is unchanged. Then, I_{FWRM} is proportional to the peak power of the laser, indicating that a short laser pulse with a high peak power and thus a broad linewidth is preferable for generation of many rotational lines.

When the duration of the pump pulse is much shorter than the dephasing time, T_2, the calculation should be performed using the gain given by Tomov et al. [12]. However, the above discussion is essentially unchanged, even when their equations are applied to the present study. These facts suggest that a femtosecond laser pulse, which has a larger spectral bandwidth, is useful for more efficient generation of a larger number of rotational lines. Thus FWRM has great potential for the generation of ultrashort optical pulses, though other nonlinear optical effects such as self-phase modulation and self-focusing should be carefully minimized.

3 Conclusion

In this study, multifrequency laser emission is generated by FWRM employing a 100-ps Ti:Sapphire laser as a pump beam. This laser system covers the entire visible region and is useful as a widely tunable laser source, because of the larger number of and the small frequency shift of the rotational lines. The results observed in this study suggest that a femtosecond Ti:Sapphire laser has great potential for use as a pump beam for the efficient generation of a larger number of rotational lines, which, in turn, is essential for the generation of ultrashort pulses of less than 1 fs by mode-locking the rotational lines generated.

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