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Generation of more than 40 rotational Raman lines by picosecond and femtosecond Ti:sapphire laser for Fourier synthesis

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Abstract. The generation of more than 40 rotational stimulated Raman emission lines in hydrogen gas by use of a picosecond and femtosecond Ti:sapphire laser is reported. The spectral range of the emission lines extends from the near-infrared to the far-ultraviolet region. The resulting broad bandwidth allows the generation of a 0.6- fs pulse by coherent superposition, i.e., Fourier synthesis, of the emission lines.

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An ultrashort pulse laser has been developed, that has great potential for both scientific and industrial applications. In 1987 [1], a report appeared, that described a modelocking technique, followed by selfphase modulation and pulse compression, which allows the generation of 6- fs pulses. No significant improvements, however, have been reported since that time. A major reason for this is the difficulty in the compensation of higher-order group-velocity dispersion, which occurs in the laser material itself, as well as the optics used in the laser system.

More recently, several groups have proposed new techniques for the generation of ultrashort pulses. They are based on coherent superposition of emission lines generated or mixed in the gas medium, which has no or little difficulty related to group-velocity dispersion. One such approach is based on the generation of emission lines consisting of high-order harmonic and mixing frequencies from a single-mode continuous-wave laser and coherent superposition of these emission lines [2]. Another approach is based on the generation of emission lines consisting of an odd-number of high-order harmonic frequencies, which are produced via an intense laser pulse. Since the emission lines extend from the near infrared to the vacuum ultraviolet, the production of attosecond pulses is theoretically possible [3, 4]. Unfortunately, low conversion efficiencies cause low pulse energies for the frequency-multiplied components required for Fourier synthesis to generate ultrashort pulses. In addition, the intensity of the fundamental beam must be reduced by using optics with no group-velocity dispersion. Even for this case, however, it is impossible to avoid a reduction in the pump pulse energy. The use of high-order stimulated Raman emission lines for the generation of a multi-frequency beam represents an alternative approach to this dilemma [5, 6]. The conversion efficiencies are much higher than those from harmonic generation, and, in addition, a high-peak-power ultrashort pulse can readily be obtained without discriminating against the fundamental beam.

It is possible to use vibrational stimulated Raman emission lines as the components for Fourier synthesis, for which the spectral region extends from the infrared to the ultraviolet when hydrogen is used as the Raman medium. The mechanism of phase-locking is described in detail elsewhere [5, 6]. In this case, the emission lines, however, are spaced by a frequency of $4155\,\mathrm{cm}^{-1}$, causing repetitive pulses at time intervals of 8 fs. It is, however, difficult to obtain a single ultrashort pulse instead of highly repetitive pulses, since a short (< 8 fs), high-peak-power and transform-limited laser is required as a pump beam. Unfortunately, no such laser has been developed, even by state-of-the-art laser technology. An alternative approach is the use of rotational stimulated Raman lines. The Raman shift frequency ($587\,\mathrm{cm}^{-1}$) causes a time interval of 57 fs. Such a laser, producing a short (< 57 fs), high-peak-power and transform-limited pulse, is commercially available. For this case, the efficient and simultaneous generation of numerous rotational lines, by using a femtosecond laser pulse, is necessary. In our previous paper, we reported the generation of numerous vibrational and rotational stimulated Raman lines using a 150- fs excimer laser (248 nm) [7]. Similar observations were reported for a frequency-doubled Ti:sapphire laser (400 fs, 390 nm) [8]. In these cases, vibrational and rotational lines with different Raman shift frequencies should be coherently superimposed by Fourier synthesis. It would be desirable to generate many rotational lines and

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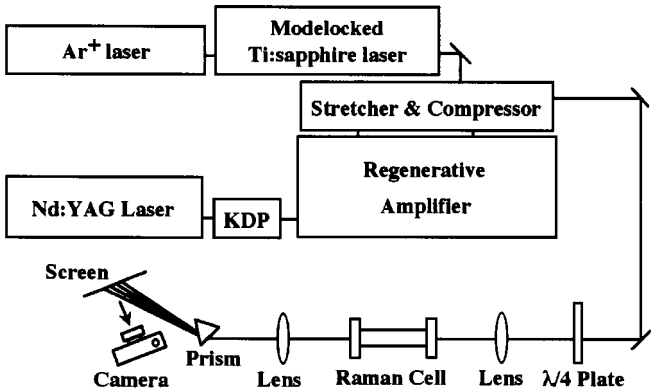

Fig. 1. Experimental setup. A titanium-doped sapphire oscillator/amplifier laser system producing 100 ps to 70 fs pulses is used as a pump beam

to discriminate against vibrational lines, in order to provide a simpler and more comprehensive scheme for Fourier synthesis. Here, we report the exclusive generation of many rotational lines over an extremely wide spectral region by using a picosecond and femtosecond Ti:sapphire laser (810 nm) as a pump beam.

1 Experimental

The experimental setup is shown in Fig. 1. A modelocked Ti:sapphire laser (Spectra-Physics, Tsunami, 82 MHz, 100 fs, 500 mW, 810 nm) is pumped by an argon ion laser (Spectra-Physics, Beamlok 2060-7S, 7 W). The output pulse is amplified by a regenerative amplifier (Spectra-Physics), which is pumped by second harmonic emission of a Nd:YAG laser (Spectra-Physics, GCR-6, 10 Hz, 6 ns, 600 mJ, 532 nm) equipped with a potassium dihydrogen phosphate (KDP) crystal. The pulse width is stretched to 200 ps by a pulse stretcher (Spectra-Physics). After light amplification, the 200-ps pulse is compressed to 150 fs by a pulse compressor (Spectra-Physics). Although it was possible to further reduce this pulse to 70 fs by a prism pair, this system was not used in this study. The spectral bandwidth of the laser is 12 nm. These results suggest that the laser pulse is nearly completely Fourier transform limited. The laser beam is passed through a quarter wave plate (Sigma, WPQ-7800-4M) to generate an elliptically polarized beam. The typical pulse energy was 5 mJ. The laser beam is focused by a lens (focal length, 1 m) into a Raman cell (length, 1 m) filled with hydrogen gas (10 atm) and again focused, after passing through a quartz prism, on a white screen. The spots dispersed on the screen were then photographed using PROVIA FUJICHROME 400 film. The wavelengths of the emission lines were measured using a monochromator (JASCO, CT-2SCP) equipped with a photomultiplier (Hamamatsu, R636). The pulse width of the pump laser was measured using an autocorrelator (Spectra-Physics, SSA). In this experiment, the pulse width was changed from 150 fs to 100 ps by adjusting the position of a mirror pair in the pulse compressor. This was done to change the beam path length in order to control group velocity dispersion. This caused no change in the pulse energy, the spectrum, or the polarization of the pump beam.

2 Results and discussions

When the pulse width of the Ti:sapphire laser was adjusted to 1.2 ps, more than 40 rotational lines were simultaneously generated from the near infrared (> 945 nm) to the far ultraviolet (< 279 nm). Figure 3 shows a photograph of this result. Figure 4 shows photographs of the output beams, which are recorded at different pulse widths. In all cases, no Stokes line appears, which is probably because of the short exposure time and the low sensitivity of the color film to the longer-wavelength light. A few Stokes components, however, could clearly be observed by the combination of a monochromator and a photomultiplier. When the pump pulse width is adjusted to 100 ps, a few rotational lines are observed in the vicinity of strong 1st–5th anti-Stokes vibrational lines. The 6th–7th vibrational anti-Stokes lines could be observed by using a monochromator equipped with a photomultiplier. When the pulse width is reduced to 1.5 ps, the Raman lines are spread over the entire spectral region from the near infrared to the ultraviolet. When the pulse width is further reduced to 800 fs, a more uniform intensity distribution is observed in the near ultraviolet. In addition, a weak streak appears over the entire spectral region. This is enhanced at shorter pulse widths. Only a strong continuous emission (white-light continuum) is observed at 500 fs (see caption to Fig. 4). This results from self-phase-modulation, which is sometimes observed when a short laser pulse is focused in a solid, a liquid, or a gas. This white-light continuum was further enhanced at shorter pulse widths down to 150 fs. The polarization, the spectral width, and the pulse energy of the pump beam remained essentially unchanged throughout the experiment. As a result, the changes in the spectral patterns, i.e. the basic phenomena, may be attributed to the change in the peak intensity of the pump beam and to the dependence of the laser power on the efficiencies of these phenomena. The tendency can be summarized as follows. (1) The threshold for the generation of rotational lines is higher than that for vibrational ones, but the efficiency under high power pumping is much higher than

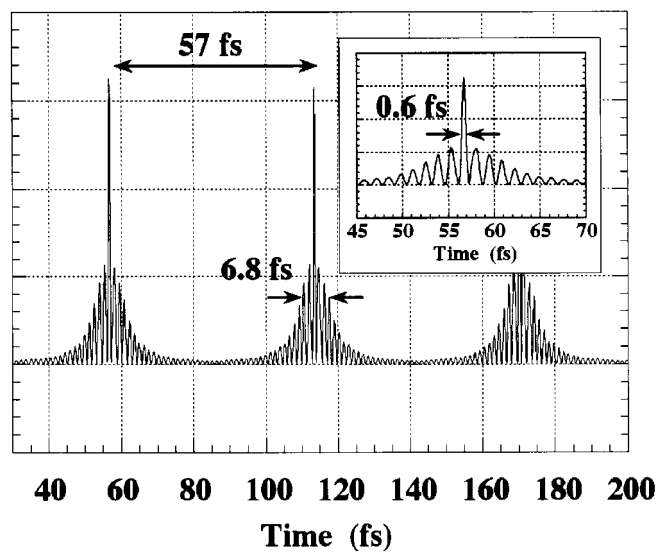

Fig. 2. Temporal profile of ultrashort laser pulses calculated by coherent superposition of the rotational lines observed in Fig. 3. The figure in the window shows an expansion of a single laser pulse

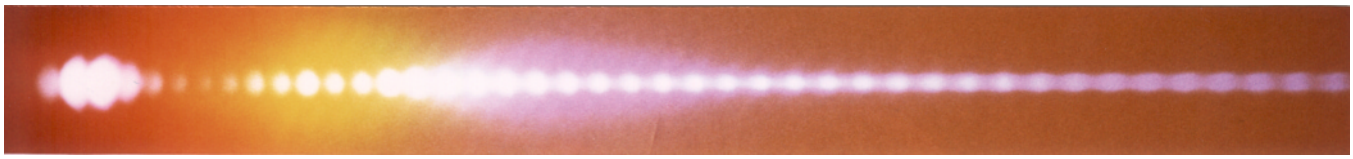

Fig. 3. Photograph of more than 40 rotational emission lines dispersed on a white screen. The pulse width of the pump beam was adjusted to 1.2 ps. The exposure time of the photograph was 60 s

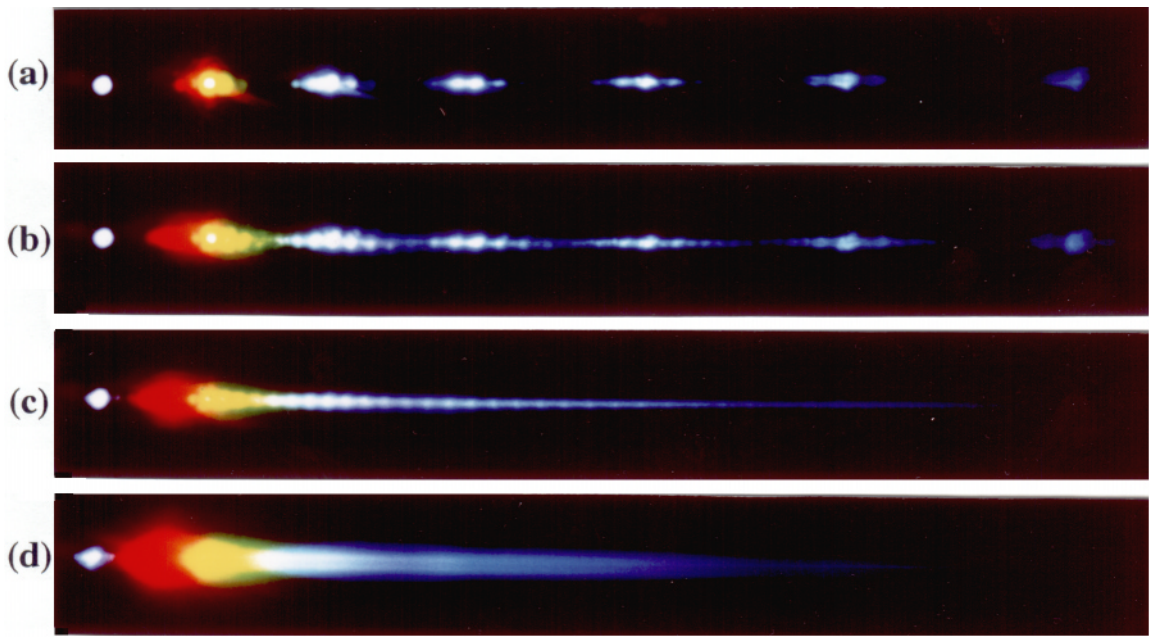

Fig. 4a–d. Photographs of Raman emission lines dispersed on white screen. The pulse width is (**a**) 100 ps, (**b**) 1.5 ps, (**c**) 800 fs, (**d**) 500 fs. The exposure time for the photograph was 30 s. The four groups of colors (red, yellow, green, blue) might originate from four dyes contained in the printing paper for light reproduction and might not reflect the intensity distribution of the white light continuum

that for vibrational ones. (2) A white light continuum, generated by self-phase-modulation, is most efficient under the highest power pumping, and, as a result, the generation of vibrational and rotational Raman lines is suppressed. (3) The optimum pulse width for the generation of many rotational lines is 1.2 ps under the experimental conditions used in this study.

Figure 2 shows the pulse shape calculated with Eq. (1) and a Fourier synthesis of the spectral lines observed in Fig. 3, by assuming that all the rotational lines are coherently superimposed.

$$I(t) = \left(\sum_j E_j \exp\left[\mathrm{i} (\omega_0 + j\Delta\Omega) t \right] \right)^2 , \qquad (1)$$

where $I(t)$ and $\Delta\Omega$ are the temporal profile of the output pulse and the rotational Raman shift frequency of hydrogen, respectively. The group-velocity dispersion that occurred in the hydrogen gas (10 atm, confocal distance) is calculated to be negligible [9], and this term is omitted in (1). The full width at half maximum (FWHM) of the pulse is 0.6 fs. This short pulse width originates as a result of the available spectral region in which the rotational lines are generated. In the study described here, the pulse is generated every 57 fs in an envelope of 1.2 ps. These values are determined by a Raman shift frequency of 587 cm^{-1} and a pump pulse duration of 1.2 ps. It should also be noted that the result shown in Fig. 2 can be obtained by assuming a transform-limited pulse with a 1.2-ps width could generate a multi-frequency laser emission having the same spectral profile shown in Fig. 3.

In the present study, the laser beam is chirped (not transform limited), and, as a result, this is not the case. If the stimulated Raman emissions generated are linearly chirped together with a fundamental beam, they can be compressed to a subfemtosecond pulse by compensating for the group-velocity dispersion. For example, the fundamental beam linearly chirped by a grating pair as well as the chirped rotational Raman beams coherently generated from the same fundamental beam can be compressed to a single pulse with the temporal profile shown in the window of Fig. 2, in theory, by a grating pair, although two weak pulses, in addition to the main pulse, may also appear in an envelope of 70 fs (transform-limited pulse width). If the stimulated Raman emissions are not linearly chirped, it is necessary to employ a transform-limited pulse as a fundamental beam, directly providing a subfemtosecond pulse without any compensation of the group-velocity dispersion.

It is noted that multifrequency Raman emission was generated by using a nearly-transform-limited pulse (200 fs) in our recent work [10]. The linear part of the dispersion, which may occur at a higher hydrogen pressure with a longer path length, can be compensated by a grating pair. It is needless to say that a windowless experiment based on a nozzle-jet

technique is essential, since the group-velocity dispersion of the quartz plate used in the windows for the Raman cell is not negligible, as pointed out previously [5, 7]. Thus, this approach, i.e., chirped Raman generation/amplification and compression of fundamental and rotational Raman lines or coherent superposition of the rotational stimulated Raman emissions generated by using a transform-limited pulse, represent a potentially useful approach to the generation of a single, ultrashort, and highly intense laser pulse.

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