Spectral broadening and temporal reshaping of a femtosecond laser pulse due to a nonlinear optical interaction in a hydrogen gas

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Abstract. We observed spectral broadening and pulse shaping of a femtosecond laser pulse after passing the beam through a pressurized hydrogen gas. When the pump pulse width was 100 fs, the measured spectrum contained several peaks; these peaks are found to be rotational Raman lines from the dependence of the emission intensity on the laser beam polarization. The spectral structure indicates that both stimulated Raman scattering and self-phase modulation contribute to the spectral broadening. We also reveal that the waveform of a femtosecond laser pulse is strongly modulated after beam propagation through a hydrogen gas. The substantial compression of a pulse was observed when both rotational Raman emissions of para-hydrogen and ortho-hydrogen were generated. This intensity modulation can be explained by coherent superposition of a pump pulse and Stokes pulses. This explanation clarified that identity of carrier envelope phases between Stokes pulses will be needed for the generation of the compressed ultrashort pulse observed in our experiments. The present approach has a potential for development in a novel method for compressing a laser pulse.

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1 Introduction

Generation of an ultrashort pulse based on phase modulation using Raman coherence has received considerable attentions in a few years [1-7]. The phase modulation of an ultrashort pulse is caused by a time-varying refractive index of a media on which the Raman coherence is excited by two-color nanosecond lasers [1] or an intense ultrashort pulse [2,3]. This modulation provides high-order stimulated Raman sidebands [2,4] or negatively chirped spectral broadening [3]; the correction of the phase relationship among the Raman sidebands [5] or the addition of a positive chirp [3,6,7] gives rise to extremely shortening of a pulse duration to the extent of a few fs. Nowadays, it is expected that these techniques would provide the generation of a subfemtosecond laser pulse in the optical region.

We have demonstrated the compression of a femtosecond laser pulse without phase compensation as a result of nonlinear propagation of a femtosecond laser pulse in a pressurized hydrogen gas [8]. This phenomenon can be explained by the modulation of temporal and spectral intensity due to stimulated Raman scattering and four-wave Raman mixing thorough a Raman coherence excited by a femtosecond laser pulse itself. The notable point in this result is that the self-compression was achieved without an initial preparation of a Raman-active medium by other pump lasers. Therefore it has a possibility of providing an efficient compression method of a laser pulse only by passing it through a hydrogen gas, which is an easier method than a conventional one that has two stages consisting of a spectral-broadening stage and a subsequent phasecompensation stage.

In this report, we present the temporal and spectral characteristics of femtosecond laser pulses modulated by nonlinear polarization in a Raman-active medium. We show that the spectral broadening after passing a beam through a hydrogen gas depended on a pulse width, a sign of chirp, and polarization of input pulses. The observed results implied that not only self-phase modulation but also Raman process broadened the spectrum with an asymmetric...
structure. In addition, we observed the strong intensity modulation of the output pulse when the spectrum included Raman lines of ortho- and/or para-hydrogen. The pulse shape seemed to be determined by these Stokes emissions. The results of calculation suggested that the generation of a single compressed pulse needed to lock the phase among multiple Raman pulses. Our experimental results indicate that the phase-locking generation of Raman emissions would be achieved through an interaction of an ultrashort pulse with an excited Raman coherence of hydrogen molecules.

### 2 Experimental setup

In this study, a Ti:sapphire laser regenerative amplifier system (Thales, Concert) was used to generate ultrashort laser pulses at a center wavelength of 785 nm with a 1-kHz repetition rate. The pulse duration was changed from 100 fs (nearly transform-limited) to 3 ps by adding either a positive or negative chirp using a stretcher consisting of a pair of gratings. The maximum output energy was 1.5 mJ, and it was attenuated using a polarizer and a rotating waveplate. A Raman cell was filled with a hydrogen gas consisting of 25% of para-hydrogen and 75% of ortho-hydrogen at a pressure of 10 atm. The polarization of an input pulse was varied between linear and circular by rotating a quarter-wave plate placed just before the input window of the Raman cell. The laser pulse was focused into the Raman cell using lenses with an appropriate focal length to prevent unfavorable nonlinear effects induced by the silica windows with a 5-mm thickness. The output pulses were collimated and measured after cutting off the center part of the beam using an aperture with a diameter of 3 mm. The spectrum was measured with a multichannel spectrometer (Ocean Optics, USB2000), and the pulse width was estimated from an autocorrelation trace measured with a noncollinear multi-shot second harmonic autocorrelator (APE, Pulse Check).

### 3 Results

#### 3.1 Spectral broadening of a femtosecond pulse after passing through a hydrogen gas

Figure 1 shows the observed spectra for a circularly, linearly, and elliptically polarized input pulse with a duration of 100 fs.
on the red side. In contrast, a linear-polarized input pulse expanded its spectral region on both the red side and blue side, and an elliptical-polarized pulse emphasized the peaks on the red side. This behavior exactly agrees with the dependence of a rotational Raman polarizability on the pump and seed beam polarization, and the conservation law of the angular momentum in Raman mixing process [9]. This result strongly suggests that the newly generated spectral components were resulted from rotational Raman scattering. It is noted that we observed rotational Stokes and anti-Stokes emissions that cannot be generated by a linearly polarized pump laser without a seed laser even though using an input pulse with linear polarization. This fact suggests that the spectral components broadened by self-phase modulation will serve as seed for four-wave Raman mixing. Additionally, the blue shift of the center wavelength of the input laser.

![Autocorrelation traces of output pulses](image)

**Fig. 3.** Autocorrelation traces of output pulses whose spectra include an intense Stokes emission. The dashed curve represents an autocorrelation trace of the input pulse.

shown in Fig. 1 may be caused by self-phase modulation affected by self-steeping that generally emphasizes the blue-side component [10].

Figure 2 shows dependence of the output spectrum on a pulse width of an input beam with circular polarization. One can see in this figure that output spectra produced by positively or negatively chirped input pulses were much different from that produced by a nearly transform-limited one. Figure 2(a)-(c) show output spectra when we used the input pulse with a positively chirp (Fig. 2(a) and (b)), or a nearly transform-limited pulse (Fig. 2(c)). Figure 2(d) shows an output spectrum pumped by an input pulse with the shortest pulse width that was 100 fs. Figure 2(e) and (f) show output spectra produced by a negatively chirped input pulse. The spectrum of the input pulse is shown in the inset. In Fig. 2(a) and (b), the new spectral components were created almost symmetrically on both the red side and blue side around the center wavelength of the input laser. This spectral broadening is a typical behavior of a self-phase modulated ultrashort pulse [10]. On the other hand, Fig. 2(c) and (d) show an asymmetrical structure with a peak on the red side apart from the input laser wavelength. The wavelength of the newly created peak was 819 nm, which corresponds to that of an Stokes emission of ortho-hydrogen. As shown in Fig. 2(a)-(d), when we continuously changed a positively chirped input pulse into a transform-limited one, we found that the spectral component on the red side gradually collapsed and the peak at 819 nm was rapidly growing. This indicates that the spectral broadening is caused by not only self-phase modulation but also Raman process in this input pulse-width limit. Further adding a negative chirp resulted in reducing the emission intensity of the 819-nm peak and narrowing a spectral width of the output pulse. This narrowing in frequency domain will be explained by the fact that the phase modulation provided by a Kerr effect compensates the negative chirp of the input pulse in the time domain.

It is noted that, as mentioned in the following subsection, we observed the strong reshaping of the waveform of the output pulse when the Stokes emission appeared as shown in Fig. 2(c) and (d). We believe that this reshaping of the waveform would crucially affect the spectral structure through nonlinear optical effects strongly depending on the waveform of a pump pulse.
3.2 Pulse shaping accompanied by Raman emissions

When the output pulse spectrum was broadened only by SPM, the autocorrelation trace of the pulse either exhibited no change or stretched due to a group velocity dispersion of the output window. In contrast, as mentioned in the preceded subsection, we found that the output pulse shape with Raman emissions underwent strongly intensity modulation as a result of an interaction with a hydrogen gas. Figure 3 shows autocorrelation traces ((a) and (c)) of output pulses whose spectra ((b) and (d)) include an intense Stokes emission. In this figure, the dashed carve represents an autocorrelation trace of the input pulse. We can see in Fig 3(a) that the autocorrelation trace of the output pulse with an intense Stokes emission of ortho-hydrogen (Fig. 3(b)) had a periodical structure within almost the same envelope as that of the input pulse. The cycle time of the periodical structure was 57 fs; this value was equal to the beat period derived from the multiplicative inverse of the deference of frequency between the pump and the Stokes emission of ortho-hydrogen, i.e. 587 cm$^{-1}$. This result allows us to interpret this periodical structure as the result of a coherent superposition of the pump pulse and the Stokes pulse; the superposition of two waves with a frequency deference, $\Delta \nu$, imposes a beat pattern with a periodical time of $1/\Delta \nu$ on the waveform [11]. Figure 3(c) shows an autocorrelation trace of a pulse whose spectra include Stokes emissions of both para-hydrogen and orthohydrogen (Fig. 3(d)). In this case, the pulse shape exhibited a single and compressed waveform without a periodical structure. The full width at half maximum (FWHM) of the trace was 45 fs; this value was almost one-third of that of the input pulse. The difference between the waveforms shown in Fig. 3(a) and (c) indicates that the Stokes emission of para-hydrogen crucially contributes to change the output waveform. In our experiments under various conditions of input beams, we observed the pulse compression as shown in Fig. 3(c) was always accompanied by both Stokes emissions of para- and ortho-hydrogen. We conclude, therefore, that the Stokes emission of para-hydrogen is essential for the generation of a single compressed laser pulse by this method. In order to discuss the effect of the Stokes emission of para-hydrogen on the output pulse shape, we calculated the temporal shape under the assumption that independent three pulses, a pump pulse, Stokes pulses of orthohydrogen and para-hydrogen, are coherently superposed, modulating the intensity shape of the output pulse. In this case, we need to take account of the relationship of carrier envelope phase (CEP) among these pulses. The electric filed of the laser is expressed using CEP, $\psi_j$, as follows:

$$
\epsilon(t) = E_j(t)\exp(i\psi_j) 
$$

(1)

$$
E_j(t) = \frac{1}{2\pi} \int I_j(\omega) \exp\left[i(\varphi_j(\omega) - \omega t)\right] d\omega 
$$

(2)

where $j$ represents pump, Stokes emissions of para- and ortho-hydrogen as $p$, $ps$, and $os$, respectively. $I_j(\omega)$ denotes spectral intensity, and $\varphi_j(\omega)$ is a spectral phase. In the following calculations, it was assumed that the phase of Stokes emissions follows that of the pump laser, i.e. $\varphi_j(\omega)$ is the same function in every pulses. Figure 4(b) shows an autocorrelation trace of a temporal waveform obtained from the spectra shown in Fig. 4(a) that includes a pump pulse and the Stokes emission of ortho-hydrogen. In this case, the autocorrelation trace shows a good agreement with the experimental results (Fig 3(a) and (b)), and the shape did not depend on the difference of CEP, $\Delta \psi = \psi_{os} - \psi_{ps}$. On the other hand, the temporal shape obtained from the superposition of three individual pulses strongly depended on $\Delta \psi$ (Fig. 4(c)-(e)). In the case of $\Delta \psi = 0$, the waveform has a compressed shape as shown in Fig 4(d), since three pulses constructively interfere. In contrast, $\Delta \psi = \pi$ provide no compressed...
In this report, we presented spectral broadening and pulse shaping of a femtosecond laser pulse after passing the beam through a pressurized hydrogen gas. When the input pulse width was 100 fs, the measured spectra contained several peaks; these peaks were found to be rotational Raman lines from the dependence of the emission intensity on the laser beam polarization. The spectral structure indicated that both Raman process and self-phase modulation contribute to the spectral broadening. The generation of rotational Raman emissions by a linearly polarized pump laser implied that the spectral components generated by self-phase modulation might serve as seed for four-wave Raman mixing. We also revealed that the waveform of a femtosecond laser pulse was strongly modulated after beam propagation through a hydrogen gas. The compression of a pulse width was observed when both rotational Raman emissions of para-hydrogen and ortho-hydrogen were generated. This intensity modulation could be explained by coherent superposition of a pump pulse and Stokes pulses. This explanation clarified that identity of carrier envelope phases between Stokes pulses will be needed for the generation of the compressed ultrashort pulse observed in our experiments. The result presented here is the first observation of femtosecond-pulse self-shaping caused by a nonlinear polarization based on the excitation of Raman coherence. We believe that the present approach has a potential for development in a novel method for compressing a laser pulse.

**References**