Generation of Highly Repetitive Optical Pulses Based on Intracavity Four-Wave Raman Mixing

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An extremely long train of highly repetitive pulse (17 THz) is obtained by the rotational four-wave Raman mixing of molecular hydrogen in a resonator using a computer simulation. This highly repetitive pulse can be obtained only when the laser wavelength and the resonator are adjusted to specified values. This pulse train has potential for use in ultrafast data communication because of the accurately determined repetition rate or even as a frequency standard, since the frequencies of the emission lines can be stabilized and locked to the above values.

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Several strategies have been reported for the generation of ultrashort optical pulses which break the 1-fs barrier [1,2]. For example, a method which is based on the generation of high-order harmonics suggests the possibility of the generation of 0.2-fs pulses [3,4]. On the other hand, a method based on the generation of high-order Raman emissions by means of four-wave Raman mixing indicates the potential for generating 4.8-fs/3.8-fs pulses [5,6]. Although two major approaches for the generation of the ultrashort pulses have been reported, all these approaches are based on the "pulsed" method; i.e., either a single or several pulses are generated using a pulsed pump source. In a number of applications, a train of highly repetitive pulses, i.e., usually referred to as "continuous-wave" (cw) ultrashort pulses, is desirable, for example, for use in data communication.

Recently, a cw stimulated Raman emission has been generated using a cw pump source [7–10]. Since stimulated Raman scattering occurs at MW pump power levels, the use of a resonator which substantially increases the intracavity laser power using a pair of high-reflectivity mirrors is necessary. In this approach, the Raman wave is also designed to be resonant in the cavity, in order to enhance intracavity power. By using a pair of mirrors having reflectivities of $R = 99.995\%$, the threshold power of the pump laser for the generation of the vibrational Raman emission can be reduced to <1 mW [9,10].

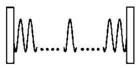
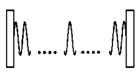
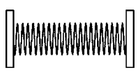
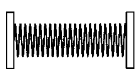
In this Letter, we propose a new approach for the generation of highly repetitive ultrashort optical pulses, based on cw four-wave Raman mixing in a cavity under specified conditions. A computer simulation indicates that several rotational Raman lines are generated using commercially available mirrors, providing a 17-THz pulse train for orthohydrogen. We report herein the advantages of this method and possible applications in science and technology.

The approach used in this study is briefly explained in Table I. In order to simultaneously generate numerous Raman lines, all the Raman waves (Stokes and anti-Stokes emissions) must be resonant in the cavity. The wavelength of the emission which is resonant in the cavity is given by $2l/(2n + 1)$, where l is the length of the cavity and $(2n + 1)$ is the number of half waves in the cavity. It should be noted that an odd number of half waves are present in the cavity and form a single ultrashort pulse at the center. On the other hand, if there are an even number of half waves in the cavity, two pulses are formed behind the front and back mirrors, producing a broader ultrashort pulse consisting of a pair of pulses with equal intensities. Thus, the wave number of the emission line is given by $(2n + 1)/2l$. The frequency separation of the emission lines between the n and $(n + 1)$ orders is specified to be 587 cm^{-1}, which is the rotational Raman shift frequency of molecular hydrogen. The cavity length, l, is, then, calculated to be 17.04 μm. This means that the fundamental and all the rotational Raman beams emitting at this "magic wavelength" are resonant in the cavity having this "magic length" and have one of the maxima at the center of the resonator. If these emission lines are superimposed on each other, only a single sharp peak appears at the center of the cavity and other peaks smear away.

High-order Stokes and anti-Stokes Raman emissions are known to appear through four-wave Raman mixing, in which all the Raman waves are coherently phased [11]. The phase locking in the resonator can also be explained by excitation using a periodic (57 fs) sine wave, which is the result of the superposition of the fundamental and the first-Stokes beams. This situation is similar to synchronous pump mode locking; a train of pulses separated by 57 fs, which corresponds to the round-trip time of the resonator, acts as an exciting beam, which is modulated by a mode locker operated at a frequency of 17 THz (= 1/57 fs). As a result, the Raman emissions are phase locked (mode locked), thus providing a short optical pulse at 57 fs intervals.

The evolution of the Raman spectrum and the temporal profile of the synthesized pulse by superposition of the fundamental and Raman emissions were calculated, the procedure for which has been reported previously [11]. A major difference is the low output power of the laser, which is achieved by the use of a pair of resonator mirrors having high reflectivities (99.995%). It should be noted that such a mirror is currently commercially

TABLE I. Raman waves resonant in a cavity: $2n + 1$: number of half waves in the resonator; λ_n: wavelength of the emission resonant in the cavity; l: length of the cavity; ν_n: wave number of the emission resonant in the cavity. Parameter λ and ν are the wavelength and the wave number of the emission when l is assumed to be 17.04 μm. The refractive index of the Raman medium is assumed to be 1 in this table.

				$l = 17.04\ \mu$m	
n		λ_n	$\nu_n = \frac{1}{\lambda_n}$	λ [nm]	ν [cm^{-1}]
1		$\frac{l}{\frac{1}{2}} = 2l$	$\frac{1}{2l}$	34 080	293.4
2		$\frac{l}{\frac{3}{2}} = \frac{2}{3}l$	$\frac{1}{\frac{2}{3}l}$	11 360	880.2
3		$\frac{l}{\frac{5}{2}} = \frac{2}{5}l$	$\frac{1}{\frac{2}{5}l}$	6816	1467
n		$\frac{l}{\frac{2n+1}{2}} = \frac{2}{2n+1}l$	$\frac{2n+1}{2l}$	...	...
$n + 1$		$\frac{l}{\frac{2(n+1)+1}{2}} = \frac{2}{2(n+1)+1}l$	$\frac{2(n+1)+1}{2l}$	...	...
20		$\frac{l}{\frac{2\times 20+1}{2}} = \frac{2l}{41}$	$\frac{41}{2l}$	831.2	12 030
21		$\frac{l}{\frac{2\times 21+1}{2}} = \frac{2l}{43}$	$\frac{43}{2l}$	792.6	12 620
22		$\frac{l}{\frac{2\times 22+1}{2}} = \frac{2l}{45}$	$\frac{45}{2l}$	757.4	13 200

available and has been used for the generation of vibrational Raman emission [7–10]. The dispersion of the hydrogen gas is taken into account in the calculations reported here.

Figure 1 shows the spectrum and the temporal profile of the rotational Raman emission generated in the resonator. In calculation 1(A), the laser source emitting at two different wavelengths (792.321 and 830.973 nm; 0.05 W each) is assumed to be focused into a resonator (17.034 μm). This two-frequency laser beam could be obtained either by the superposition of two independent Ti:sapphire or diode laser beams or, more directly, by rotational stimulated Raman scattering in a cavity using a single-frequency laser beam. In Fig. 1(A), only the first anti-Stokes and the second Stokes beams appear. However, numerous rotational lines are apparent at high pump powers. A beatlike temporal profile is observed at 0.05 W, since the induced Raman emissions are weak. The pulses are substantially shortened at high pump powers. Highly repetitive pulses (17 THz), consisting of short pulse widths (8 fs), are obtained at a pump level of 0.5 W. In this calculation, the reflectivity of the mirror is assumed to be flat over the entire spectral region. It is, however, difficult to obtain such a broadband high-reflectivity mirror. Figure 2 includes the result obtained using commercially available mirrors, the reflectivity curve of which is shown in the figure. Numerous rotational Raman lines are observed, providing a 15-fs pulse train. With increasing pump power, an irregular spectral pattern is obtained as the result of the limited spectral region of the coating. However, a pulse train is clearly evident in any case, and the result remains essentially unchanged to that in Fig. 2(B).

Unfortunately, it is difficult to manufacture a high-reflectivity mirror in which the coating bandwidth exceeds 100 nm. However, it might be possible to manufacture a mirror which has a periodic high reflectivity every 587 cm^{-1}, since such an etalonlike mirror has already been manufactured using a single-layer coating. As has been demonstrated using a femtosecond laser, it might be possible to generate numerous rotational Raman lines extending over the entire visible region by increasing the laser power, although a dispersion-compensated, i.e., "chirped" mirror would be desirable. This approach may be potentially useful for the generation of a train of subfemtosecond pulses.

There are several variations in this approach. The cavity length might be too short, if the cavity length of 17.04 μm is compared with those used in the generation of vibrational Raman emission ($l = 73$–77 mm). For example, the gain is proportional to the geometrical factor of $\tan^{-1}(l/b)$, where b is the confocal parameter. In order to retain the gain, the curvature of the mirror

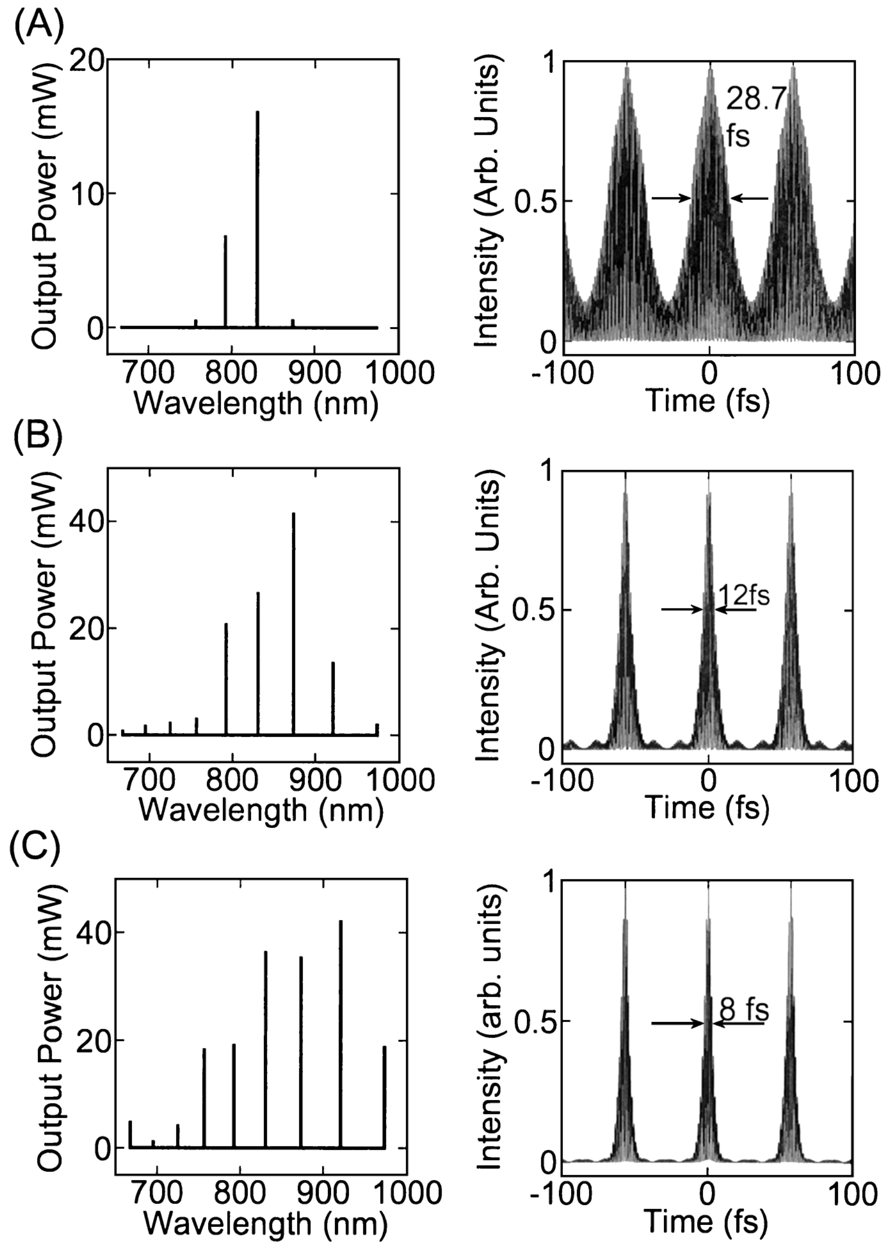


FIG. 1. Evolution of the spectrum and the temporal profile of the emission, consisting of fundamental and Raman beams. Pump laser power: (A) 0.05 W; (B) 0.3 W; (C) 0.5 W. Pump laser wavelengths: 792.321 and 830.973 nm. The mirror reflectivity is assumed to be flat and is 99.995%. Mirror curvature, 5 m; cavity length, 17.034 μm.

should be in the order of microns. It would, however, be possible to multiple the cavity length, e.g., 400 times ($l = 6.8$ mm). We were able to confirm, by a computer simulation, that the number of waves in the resonator is simply multiplied by a factor of 400, and the generated pulse has exactly the same repetition rate. Thus, the situation remains essentially unchanged. The use of a long cavity is desirable, since the number of beam reflections by the cavity mirrors is decreased, reducing the loss of the pump and Raman beams, thus allowing the use of a low-power pump laser. Of course, the cavity length can be multiplied further, e.g., by a factor of 10, to obtain a size ($l = 68$ mm) similar to those used in the generation of vibrational Raman emissions [7–10].

There are many applications for the present method of generating highly repetitive optical pulses by a superposition of highly coherent cw laser emission, since the linewidth of each emission is estimated to be <8 kHz and can be reduced to 10–100 Hz, as has been reported for vibrational Raman emission [9]. This suggests that an extremely long train of highly repetitive pulse could be generated; i.e., only 8×10^3 pulses of every 4×10^{14} pulses are missing. Therefore, this procedure might be advantageous of ruse in ultrafast data communications, although

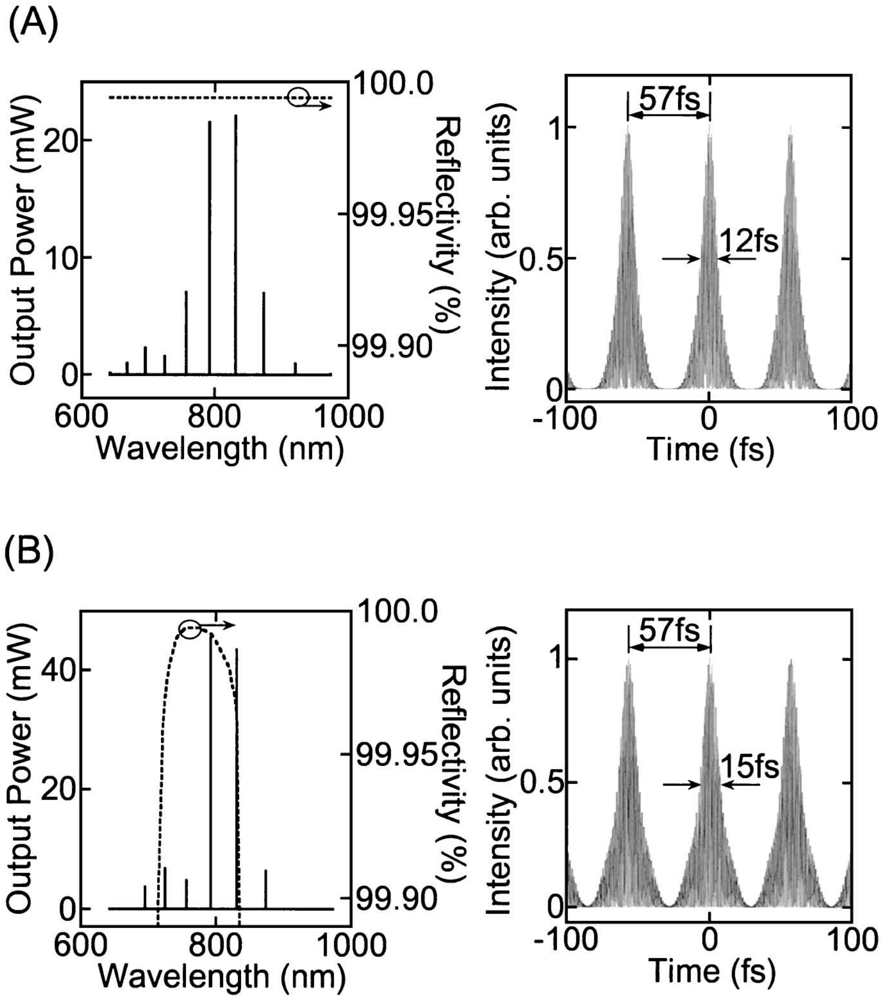


FIG. 2. Effect of mirror reflectivity on the evolution of the spectrum and the temporal profile of the emission, consisting of fundamental and Raman beams. (A) A mirror is assumed to have flat reflectivity (broken line, 99.995%); (B) a commercially available mirror having a reflectivity curve (broken line) is assumed to be used. Pump laser power: (A) 0.1 W and (B) 0.2 W. Other conditions are the same as those in Fig. 1.

ancillary technology must be developed. It should be noted that the laser frequency, i.e., the repetition rate, is determined by the physical parameter of molecular hydrogen, i.e., the rotational Raman shift frequency, and, as a result, it is absolutely fixed. In other words, lasers which are manufactured independently in different places oscillate at precisely the same frequency and repetition rate.

Similar work might be possible using parahydrogen (Raman shift frequency, 354 cm^{-1}), the pulse separation of which is 95 fs (10 THz). This alternative might be useful in vibrational four-wave Raman mixing using molecular hydrogen or even other Raman media such as CH_4 or SF_6. In the case of hydrogen (Raman shift frequency, 4155 cm^{-1}), the generation of 125-THz pulses (an 8-fs pulse train) is possible. The spectral region is very wide, and the dispersion can be easily and precisely compensated due to the small number of very narrow emission lines. Therefore, it may be potentially useful to break the 1-fs barrier using visible photons. This ultrashort pulse could be of value in fundamental studies, including investigations of ultrafast phenomena.

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