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A new approach for the generation of ultrashort optical pulses

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A short optical pulse is essential in studies on ultrafast phenomena and nonlinear spectroscopy. Laser pulses as short as 6 fs were generated in 1987, but there has been no breakthrough since. In this paper we propose a new approach to generate a shorter optical pulse at terawatt levels based on a two-color stimulated Raman effect using a simple optical system.

1. Introduction

A wide frequency domain is essential for the generation of a short optical pulse, as being realized from the uncertainty principle,

$$\Delta t \Delta \epsilon \geqslant h/2\pi , \qquad (1)$$

where Δt is the pulse width, $\Delta \epsilon$ is the energy (frequency) width, and h is Planck's constant. Additionally, the optical wave consisting of equidistant frequencies must be phase-locked and coherently superimposed, as being verified mathematically by Fourier transformation. Practically, an optical modulator is inserted in the resonator to lock phases of the longitudinal modes of the laser. Optical pulses as short as 27 fs have been generated by this mode-locking technique [1]. This pulse is further focused into an optical fiber to induce self phase modulation to increase the spectral bandwidth, and the expanded pulse in a time domain by optical components with positive group velocity dispersion is compressed to 8 fs by pairs of gratings with negative quadratic dispersion [2] and yet to 6 fs by additional pairs of prisms with negative cubic dispersion [3].

2. Limitations

A short optical pulse has currently been generated by a liquid or solid laser due to a wide spectral bandwidth, e.g. by a dye laser or a titanium sapphire laser. However, further pulse shortening is difficult as long as we use a liquid or a solid as a laser medium, since higher-order group velocity dispersion must be compensated carefully, but it is difficult practically.

On the other hand, a gas laser usually provides monochromatic emission, due to a discrete transition of an atom or molecule and to small perturbation from the buffer gas. Exceptionally, an excimer laser produces a short optical pulse, since the excimer has a repulsive potential in the ground state and has a wide spectral bandwidth. However, the pulse width is still restricted to ~ 100 fs, due to a limited spectral bandwidth.

Thus it is essentially difficult to generate a shorter optical pulse by straightforward extension of current laser technology.

3. Reported proposal

Only a proposal is published so far to solve the above problem [4]. This paper reports a possibility to generate femtosecond optical pulses by coherent superposition of precisely equidistant frequencies generated by sum- and difference-frequency mixing in nonlinear optical crystals; the induced group velocity dispersion is compensated dynamically in a millisecond time region for each emission line by using a computer-controlled phase shifter. However, this approach has an inevitable limitation; a contin-

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uous wave laser with a very narrow linewidth (~kHz) must be used and the peak power generated is much less than 1 W. It is noted that this laser pulse can not be amplified due to group velocity dispersion induced in a laser amplification medium. Then, a high-power, e.g. a kilowatt, radiation field may not be generated.

4. Two-color stimulated Raman effect

In 1987, we found a colorful laser beam emitting from the deep-ultraviolet to the near-infrared, when a dye laser beam is focused into pressurized hydrogen to convert its frequency by a stimulated Raman effect [5]. Later, it is noticed that many rotational and vibrational stimulated Raman lines were generated simultaneously. This result was attributed to the fact that the dye laser unexpectedly emitted at two different frequencies separated by 587 cm^{-1} due to defect and misalignment of the laser. Thus rotational lines were generated by the two-color stimulated Raman effect, i.e. a sort of four-wave mixing; a hydrogen molecule was excited stepwise by two photons to an imaginary level and the third laser emission acted as a seed beam, generating the fourth laser emission. This process substantially reduced the threshold for the generation of higher-order rotational stimulated Raman emission. By optimizing this hydrogen pressure, the beam polarization, and the focusing condition, either vibrational rotational lines could be generated selectively [6]. Thus this two-color stimulated Raman emission provides a coherent beam consisting of equidistant frequencies, like the longitudinal modes of a laser. However, the frequency domain concerned is extremely large and extends to more than several thousands cm^{-1}.

In this report, we present the potential performance and characteristics of an ultrashort optical pulse generated by this two-color stimulated Raman effect. We propose a simple and realistic method for the generation of a femtosecond pulse at terawatt levels using an instrument commercially available.

5. Computer simulations

Equidistant multi-frequency laser emission can be generated by a few methods, e.g. by the vibrational stimulated Raman effect. Figure 1 shows the temporal profile of the laser pulse obtained by computer simulation, assuming that a Fourier-transform-limited optical pulse is focused into hydrogen and ten vibrational lines are generated and phase-locked. Laser pulses as short as 1.4 fs are generated every 7.9 fs. However, it is practically difficult to construct a laser producing a high-power transform-limited pulse (<8 fs) even by state-of-the-art laser technology; a 10 fs, <0.5 μJ pulse is generated even in the best case [7]. It is possible to use a longer optical pulse as long as it is shorter than the dephasing time (1.1 ns for the vibrational Raman and 0.46 ns for rotational Raman with 5 atm hydrogen [8]), though a pulse train is obtained. However, the restriction "transform-limited" must be kept rigorously within an error of a few fs. Thus the generation of one femtosecond pulse is theoretically possible by using the vibrational stimulated Raman effect, but the requirement is far beyond the performance obtained by present laser technology. It is noted that an ultrashort dye laser pulse amplified by an electron-beam-pumped XeF (C–A transition) excimer laser has a potential to be used for the present purpose, because of its very wide spectral bandwidth (68 nm at 470 nm) [9].

Figure 2 shows the result of a computer simulation using rotational stimulated Raman emission. Assuming that ten rotational lines are generated simultaneously using ortho-hydrogen as has already been demonstrated with a 1 MW dye laser pulse in our laboratory, laser pulses as short as 6.4 fs are gen-

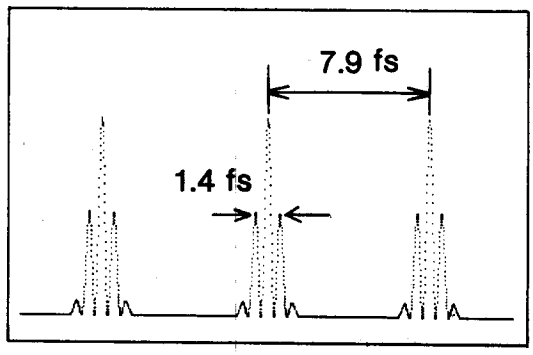

Fig. 1. Laser pulse obtained by a focusing dye laser (wavelength, 385 nm; linewidth, 0.01 nm; number of longitudinal modes, 69) into hydrogen based on the vibrational stimulated Raman effect.

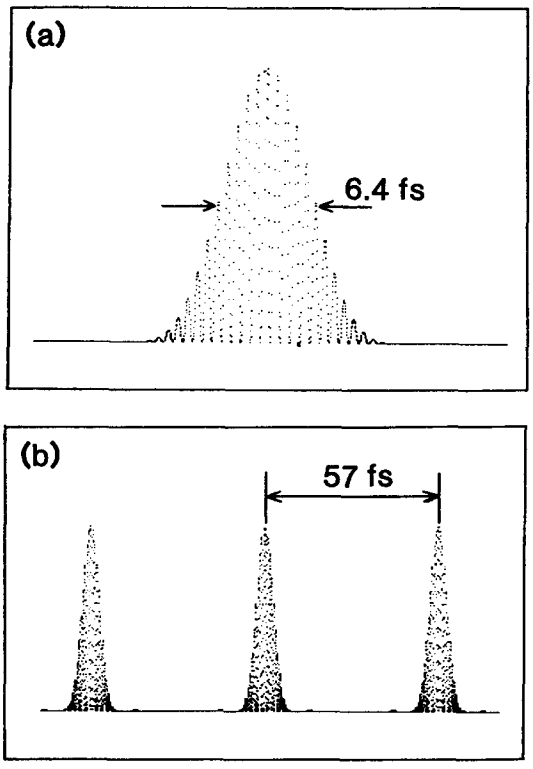

Fig. 2. Laser pulse obtained by focusing the dye laser specified in fig. 1 into ortho-hydrogen based on two-color stimulated Raman effect, (a): expanded part of (b).

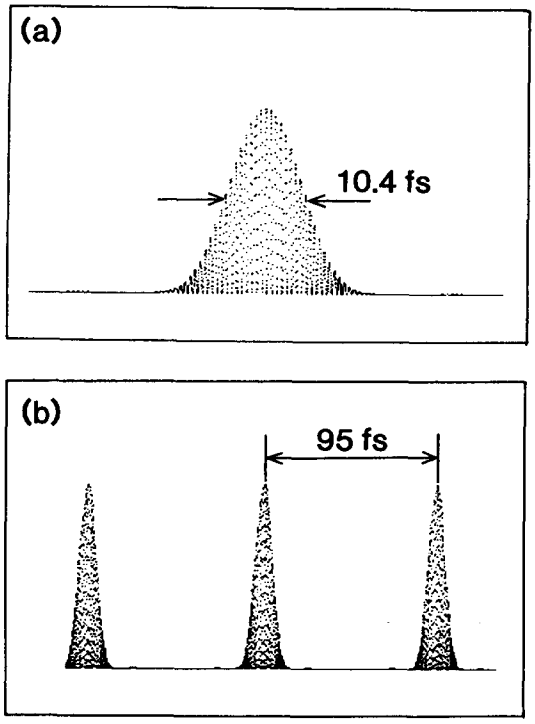

Fig. 3. Laser pulse obtained by focusing the due laser specified in fig. 1 into para-hydrogen based on two-color stimulated Raman effect, (a): expanded part of (b).

erated every 57 fs. This result is in contrast to those obtained using para-hydrogen as shown in fig. 3; pulses as short as 10.4 fs are generated every 95 fs. By the analogy of modelocking, the pulse width is determined by the frequency domain available, which is limited by the laser power used at present. Then, the pulse width can be further reduced, e.g. to 1 fs or less, by using a higher-power laser. Note that the restriction "transform-limited" must be kept within an error of a few tens fs, which is less rigorous than that in the case of the vibrational stimulated Raman effect. In modelocking, the time period between repetitive pulses is determined by the reciprocal of the frequency separation between the longitudinal modes, i.e. $c/2L$, where c is the speed of light and L is the length of the laser cavity. Contrarily, the time period between the pulses in this study is determined by the reciprocal of the Raman shift frequency, i.e. ω. Major differences are the wide spectral range and the large frequency separation, allowing the generation of much shorter and highly-repetitive pulses. A compression ratio, defined as the ratio of the time period to the pulse width, is determined by the number of rotational lines generated.

6. Requirements

In order to generate ultrashort laser pulses according to the above method, the four following requirements must be satisfied. First, a high-power transform-limited pulse is necessary; a $\sim$100 fs pulse is needed for the generation of a single optical pulse. Second, a two-color laser beam linearly polarized is desired, whose frequency separation must be adjusted to the energy for rotational transition of the Raman medium. Third, the multi-frequency laser beam must be phase-locked without using any liquid or solid optics. Fourth, no phase change must occur by other nonlinear effects.

A schematic diagram proposed for the generation of an ultrashort pulse is shown in fig. 4. A high-power transform-limited pulse with a ~100 fs pulse width has been generated by a combination of a quenching dye laser, a short cavity dye laser, a distributed feedback dye laser, a frequency doubler and an excimer amplifier [10]. Such a system is already commercially available and produces a <150 fs, 20 mJ, 248 nm (~100 gigawatt) pulse [11].

After being passed through a $\lambda/4$ wave plate to obtain a circularly polarized beam, the laser beam may be focused into a Raman cell to generate a rotational line. The laser beam is further collimated and passed through a $\lambda/4$ wave plate to obtain a linearly polarized beam. The two-color laser beam consisting of fundamental and rotational Raman emission may be passed through pairs of prisms with negative velocity dispersion to correct a positive group velocity dispersion induced by the optics placed before and after the Raman cell, if necessary (not shown in fig. 4). The laser beam is focused into hydrogen injected from a pulsed slit nozzle into a chamber to generate a multi-frequency beam. A schematic explanation of the phase-locking is shown in fig. 5. When the laser wavelength is adjusted, e.g. to 247.54 nm, a rotational line appears at 249.73 nm for para-hydrogen; the spectral purity may be improved by passing them through an etalon to ensure a transform-limited pulse after the Raman cell, if necessary (not shown in fig. 4). Each emission has 114 and 113 cycles every 28.219 μm, respectively. These two waves have maxima simultaneously spaced by this distance, and other emission lines generated by four-wave or multi-wave mixing must have maxima at these positions and are eventually phase-locked; a stimulated Raman-gain effects starts occurring at the highest radiation field. Thus phase-locking is automatically achieved without using any special optical, which is similar to the optical Kerr effect occurring in a mode-locked titanium sapphire laser. However, the present phase-locking is performed in hydrogen at a moderate gas pressure, where group velocity dispersion is negligibly small. To our best knowledge, no other nonlinear effect such as self phase modulation has been found at present power levels, as long as a hydrogen gas is used as a Raman medium. Then, the final requirement may be also satisfied.

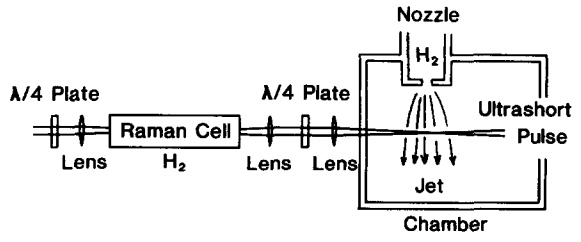

Fig. 4. Schematic diagram of laser system proposed for the generation of ultrashort optical pulse.

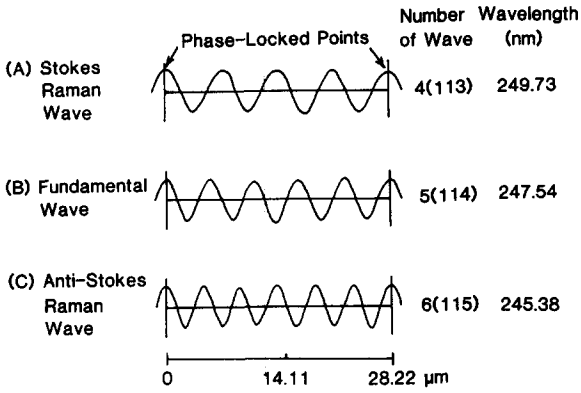

Fig. 5. Schematic explanation for phase-locking by four-wave mixing. The number of wave in 28.22 μm is reduced in the figure for explanation. The real number of wave is indicated in the parenthesis. Phase-locking of higher-order stimulated Raman emission are similar and are omitted in this picture.

In order to measure the pulse width of the laser generated, it is necessary to use a correlation technique based on a nonlinear optical phenomenon in the same chamber, since the laser pulse is broadened by passing it through an optical component such as a window. For example, a two-photon fluorescence/ionization technique might be applied to a gas sample injected from a different pulsed nozzle into the same chamber. Needless to say, a similar windowless experiment is essential in the application of this ultrashort pulse to avoid pulse broadening.

7. Characteristics

The wavelength of the ultrashort laser pulse generated is determined by the wavelength of the fundamental laser. Since only a few cycles are present in a femtosecond pulse, the fundamental beam must

have a higher frequency to obtain a wider frequency domain for the generation of a shorter optical pulse. Thus the laser emitting in the vacuum ultraviolet is preferential. Since hydrogen is transparent even in the vacuum ultraviolet, stimulated Raman emission has been generated to 130 nm [12]. Then, much shorter pulses may be generated by using a short-pulse ArF (193 nm) or F_2 (157 nm) laser as a fundamental beam. The time period between the pulses is determined by the rotational energy of the Raman medium. Then, the period can be increased by using a heavier molecule such as nitrogen or oxygen, though a higher pumping power is needed. Another approach to reduce the frequency separation might be the use of a fundamental beam consisting of phase-locked equidistant frequencies, whose separation is $587/n\ \mathrm{cm}^{-1}$ for ortho-hydrogen and $354/n\ \mathrm{cm}^{-1}$ for para-hydrogen, where n is an integer. The compression ratio is increased by a factor of n. In the extreme case, the laser pulse can be compressed from 1 ns (limited by the dephasing time) to 1 fs (limited by the frequency domain), if a complete transform-limited pulse is available.

8. Potentials

The approach proposed here used no liquid or solid optical component for the generation of an ultrashort optical pulse, so that no optical damage arises in this system; laser breakdown occurs at higher power levels in the gas phase. Thus a table-top laser producing a terawatt power, e.g. 10 fs/10 mJ, may be constructed by using a commercial laser. Moreover, a shorter optical pulse can be generated using a higher-power laser, e.g. a nuclear fusion driver. An ultrashort pulse with an extremely-high power obtained in this approach has a potential to open new scientific fields such as high-energy physics and chemistry as well as studies on ultrafast phenomena and nonlinear optical spectroscopy.

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References

[1] R.L. Fork, O.E. Martinez and J.P. Gordon, Optics Lett. 9 (1984) 150.
[2] W.H. Knox, R.L. Fork, M.C. Downer, R.H. Stolen, C.V. Shank and J.A. Valdmanis, Appl. Phys. Lett. 46 (1985) 1120.
[3] R.L. Fork, C.H. Brito Cruz, P.C. Becker and C.V. Shank, Optics Lett. 12 (1987) 483.
[4] T.W. Hänsch, Optics Comm. 80 (1990) 71.
[5] T. Imasaka, S. Kawasaki and N. Ishibashi, Appl. Phys. B 49 (1989) 389.
[6] S.W. Yoshikawa, S. Kawasaki, T. Imasaka and N. Ishibashi, Jpn. J. Appl. Phys. Lett. 30 (1991) 283.
[7] R.W. Schoenlein, J.-Y. Bigot, M.T. Portella and C.V. Shank, Appl. Phys. Lett. 58 (1991) 801.
[8] D.C. Hanna, D.J. Pointer and D.J. Pratt, IEEE J. Quantum Electron. 22 (1986) 332.
[9] T.E. Sharp, Th. Hofmann, C.B. Dane, W.L. Wilson Jr., F.K. Tittle, P.J. Wisoff and G. Szabo, Optics Lett. 15 (1990) 1461.
[10] S. Szatmári, G. Almási and P. Simon, Appl. Phys. B 53 (1991) 82.
[11] Laser-Laboratorium Göttingen e.V., Im Hassel 21, W-3400 Göttingen, Germany.
[12] H.F. Döbele, M. Hörl, M. Röwekamp and B. Reimann, Appl. Phys. B 39 (1986) 91.