A 10 THz ultrafast function generator—generation of rectangular and triangular pulse trains

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Abstract. We report the synthesis of a user-specified, shaped ultrafast pulse train by manipulating the spectral phases of Raman sidebands with a wide frequency spacing line-by-line. Trains of rectangular and triangular pulses are stably produced at an ultrahigh repetition rate of 10.6229 THz, reminiscent of an ultrafast function generator.

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1. Introduction

The generation and measurement of a user-specified, shaped ultrafast pulse train (SUPT) have had a significant impact in relation to the coherent control of chemical reactions [1, 2] and optical communications [3]–[5]. Line-by-line control, in which spectral lines are resolved and manipulated individually, leads to a fundamentally new regime for SUPT generation [4, 6]. Adiabatically generated Raman sidebands (Raman comb) [7]–[10] are a suitable source for implementing line-by-line control because they have discrete characteristics typically greater than THz frequency spacing and good mutual coherence. By using such Raman sidebands, a synthesized SUPT can have an ultrahigh repetition rate corresponding to the wide frequency spacing of the Raman sidebands. Here, we report the synthesis of 10 THz ultrahigh repetition rate SUPT using rotational Raman sidebands ($J = 2 ← 0$) in parahydrogen. We show that trains of rectangular and triangular pulses are stably produced, similar to an ultrafast function generator.

Before proceeding, we briefly review previous studies on the adiabatic Raman technique in relation to ultrafast technology. The potential for generating single cycle pulses was discussed theoretically [11] and shown experimentally [12, 13]. The generation of high-quality ultrashort pulses was reported based on a direct time-domain measurement [14]. The control of the carrier–envelope offset frequency was demonstrated with a cross correlation measurement [15] and the $f − 2f$ self-referencing technique [16]. Arbitrary optical amplitude waveform generation was discussed theoretically [17]. Carrier–envelope phase control was realized by developing a build-up scheme of Raman sidebands [18]. Recently, a train of Fourier-transform-limited pulses was generated by a glass-plate-based line-by-line phase controller [19].

2. Numerical simulation: production of an effective target for shaped ultrafast pulse train (SUPT) generation

SUPT generation was performed in two stages. The first stage was numerical SUPT generation on a computer. We employed the same spectrum as that in the actual SUPT generation using Raman sidebands, and determined the optimal spectral phase for a target pulse shape. In the second stage, we generated Raman sidebands experimentally and synthesized the SUPT by employing the numerically obtained optimal spectral phase as an effective target. In this section, we describe the process in the first stage for determining the optimal spectral phases (effective target) on a computer.

The gray bars in figure 1(a) represent seven discrete frequency components, designated $\Omega_{_1}$ to $\Omega_7$. The spectral intensity distribution and the frequency spacing (10.6229 THz) were set the same as those employed for the actual SUPT generation using Raman sidebands. (Experimental details are provided in section 3.) Numerical SUPT generation was carried out by manipulating the spectral phases of these seven frequency components iteratively based on the hill-climbing algorithm [20] to realize a target pulse shape. In this process, we defined a cost function, $C$, as the square of deviation of a shaped pulse from a target pulse shape, which is expressed as

$$ C = \int |I_{\text{tar}}(t) - I(t)|^2 \, dt. \quad (1) $$

$I_{\text{tar}}(t)$ and $I(t)$ are the target and recurring pulse shape in the time domain, respectively. The iteration process was initiated from a pulse shape in which all the spectral phases were zero.
Then, the cost function, \( C \), was minimized by changing the phase of each sideband line-by-line based on the hill-climbing algorithm. A new phase change was accepted if the new cost function was smaller than that of the last accepted phase change. The iteration was stopped automatically when the cost function was saturated. The optimal spectral phases (effective target spectral phase) and the corresponding pulse shape (effective target pulse shape) were thus identified for the ideal target pulse shape, \( I_{\text{tar}}(t) \).

A typical example when the target pulse shape, \( I_{\text{tar}}(t) \), was set to an ultrahigh repetition rate train of the rectangular pulses (gray line in figure 1(b); one cycle time: 94.1363 fs (10.6229 THz); duty cycle: 50%) is shown by the blue line with dots (effective target spectral phase) in figure 1(a), and by the blue curve (effective target pulse shape) in figure 1(b). The distortion of the effective target pulse shape from the flat top of the ideal target pulse shape is attributed to the fundamental limitation imposed by the number (seven) of frequency components, which was restricted by the detection sensitivity in the present experimental system.

3. Experimental

The optimal spectral phases obtained numerically were employed as the effective target in the actual SUPT generation stage using Raman sidebands. In this section, we describe the details of the experimental configuration (figure 2) and the procedure for convergence with the effective target in the experimental system.

3.1. Detailed configuration of the experimental system

Raman sidebands were generated by adiabatically driving the rotational Raman process \( (J = 2 \leftarrow 0) \) in parahydrogen \(^{14},^{16}\). The density and temperature of parahydrogen were set at \( 3 \times 10^{20} \text{cm}^{-3} \) (interaction length: 15 cm) and 77 K (liquid-nitrogen temperature), respectively. The two pump laser radiations, \( \Omega_0 \) and \( \Omega_{-1} \), were produced with a dual-frequency injection-locked nanosecond pulsed Ti:sapphire laser \(^{21},^{22}\). They were both Fourier-transform-limited 6 ns pulses (spectral width \( \sim 50 \text{MH} \)) and completely overlapped each other temporally.
Figure 2. Experimental setup. LN cryostat, liquid nitrogen cryostat; G, grating; FS, fused-silica plates; FM, folding mirror; CM, concave mirror; PC, personal computer; DAC, digital-to-analogue converter.

and spatially. The frequencies of $\Omega_0$ and $\Omega_{-1}$ were set at 382.4280 THz (783.9186 nm) and 371.8050 THz (806.3162 nm), respectively, and were slightly detuned by $\delta = +990$ MHz from the Raman resonance to satisfy the adiabatic condition [7, 14]. The two pump laser beams were then loosely focused in the parahydrogen with an $f = 800$ mm lens. The peak intensity was estimated to be 5.7 GW cm$^{-2}$ (total pulse energy: 5.4 mJ).

The sidebands were generated in the forward direction and then introduced into the spectral line-by-line phase controller (SLLPC) after being collimated with an $f = 300$ mm lens. The SLLPC was a spatial phase modulator consisting of 48 fused-silica plates. The system was configured in a folded 4f configuration [23]. The incident Raman sidebands were dispersed with a grating (600 grooves mm$^{-1}$) and each single sideband was focused onto a single glass plate placed in the Fourier plane by using a concave mirror ($f = 0.5$ m). The change in the spectral phase of each sideband was accomplished by changing the angle of the corresponding fused-silica plate through a bimorphous-type piezo actuator. The focused beams were reflected by the folding mirror positioned just behind the glass plates, and all the sidebands were finally recombined into one beam in space.

The phase-controlled Raman sidebands and the corresponding pulse shape in the time domain were characterized by using modified spectral-phase interferometry for a direct electric-field reconstruction system for discrete spectra (SPIDER–DS) [24]. This system identified
the relative spectral phases among the sidebands on the basis of the interference with a pair of sum frequencies, which were produced from the Raman sidebands and the two-frequency pump beams [24]. The SPIDER–DS system operated rapidly and generated an error signal (the deviation from the effective target spectral phase) on a LabVIEW-based personal computer. Finally, the error signal was fed back to the SLLPC system through a set of 48-channel digital-to-analogue converters and amplifiers (maximum applicable voltage: ±60 V).

3.2. Process of convergence with an effective target

The concrete process for convergence with an effective target spectral phase was as follows. First, we measured the sideband phase change caused by an applied voltage to a bimorphous-type piezo actuator in the SLLPC, as a prerequisite of quantitative phase manipulation. We scanned the applicable voltage, $v_n$, to the actuator over the full range (60 to –60 V; minimum step: 1 V), to give the periodic phase change, $\phi_n(v_n)$, at the $n$th sideband. This periodic phase change also induced a periodic intensity change in the related interfered sum-frequency, $I_{n,n+1}$, in the SPIDER–DS [19]. We normalized this periodic intensity change to $-1$ to 1 (designated by $I_{n,n+1}$), and then reduced the relation: the relative spectral phase, $\phi_{n+1}(v_{n+1}) - \phi_n(v_n) = \cos^{-1}(I_{n,n+1})$, versus the applied voltage, $v_n$, where $v_{n+1}$ was fixed. The same procedure was executed line-by-line for all the sidebands.

We can employ the above relation to directly set the spectral phases of the sidebands to obtain an effective target. This process, however, provided only coarse adjustment, since the SLLPC system included the hysteresis inherent in the piezo actuator. To ensure that the spectral phases converged sufficiently with the effective target, the process was followed by iterative phase manipulation based on the precise phase measurement provided by the SPIDER–DS. We produced the error signal from the SPIDER–DS measurement together with the above relation, fed this error signal back to the SLLPC, and then evaluated the spectral phases again with the SPIDER–DS. We repeated this set of procedures until the spectral phases of the sidebands reached the effective target within the minimum resolution of the SLLPC ($\pm 0.1$ radians).

4. Results

The SUPT generated using the Raman sidebands were examined for two cases: an ultrahigh repetition rate train of rectangular pulses and an ultrahigh repetition rate train of triangular pulses. We show the results in this section.

Figures 3(a)–(c) show the results when the target pulse shape was set as a train of rectangular pulses (gray line in figure 3(c); duty ratio: 50%). The initial spectral phases of the Raman sidebands are shown by the black dotted line in figure 3(a), together with their power spectrum (gray bars: $\Omega_{-4}$ (339.936 THz) to $\Omega_2$ (403.674 THz); identical to those in figure 1(a)). The blue line in figure 3(a) is the effective target spectral phases obtained numerically in advance as described in section 2, and the blue curve in figure 3(c) is the corresponding effective target pulse shape in the time domain. We employed the process for converging the spectral phases of sidebands with the effective target and realized the spectral phases shown by the red line in figure 3(a) at the end of the iterative phase manipulation. The obtained spectral phases precisely matched the effective target (blue line).

An example of this convergence process is shown for the $\Omega_1$ sideband in figure 3(b). Iteration number 0 indicates the initial phase. At the first coarse adjustment, which is reali
by directly applying the relation of the relative spectral phase versus the applied voltage, this phase reached the value at the iteration number 1. Then, the adjustments based on the precise phase measurement by the SPIDER–DS were applied iteratively, and the phase (red square) was finally set at the target (blue line) within ±0.1 radians after six successive trials.

The red curve in figure 3(c) shows the pulse shape realized in the time domain. A precise match with the effective target pulse shape (blue curve) was also found in the time domain. Note that this pulse shape was reduced for measuring the spectral phases of the sidebands with nanosecond pulsed envelopes, and they showed fine stability (< ±0.1 radians) for several hours. This shows that the ultrahigh repetition rate (10.6229 THz) train of the rectangular pulse shape was stably produced with more than 10 000 successive pulses.

Next we examined the case where the target was set at a train of triangular pulses. The results we obtained are shown in figures 4(a)–(c), in a similar way to those in figure 3. SUPT generation was also successfully achieved for this train of triangular pulses at an ultrahigh repetition rate of 10.6229 THz.

Here, one of the sideband phases, $\Omega_2$, was $\sim$ 0.3 radians from the effective target. As described above, the present system offers the ability to control the spectral phases with ±0.1
Figure 4. Generation of an ultrahigh repetition rate train of triangular pulses. (a) Spectral phases (blue line, effective target; black dotted line, initial phase; red line, achieved phase) and power spectrum (gray bars) of the Raman sidebands. (b) An example of the convergence process of the spectral phase to the target (blue line), shown for the $\Omega_{-3}$ sideband. (c) A train of triangular pulses: target pulse shape (gray line), effective target pulse shape (blue line) and achieved pulse shape (red line).

radians resolution. However, in some cases, we encountered degraded convergence behaviour for a spectral phase with an unknown cause. This was such a case. However, we could confirm that here the obtained pulse shape matched the effective target pulse shape sufficiently well at this stage, and thereby halted the iterative phase manipulation.

5. Conclusion

We have described the synthesis of SUPT using Raman sidebands with a wide frequency spacing, which were generated by adiabatically driving the rotational Raman process ($J = 2 \leftarrow 0$) in parahydrogen. Trains of rectangular and triangular pulses were produced at an ultrahigh repetition rate of 10.6229 THz as if the system were an ultrafast function generator. The trains were stably constituted over a nanosecond time scale with more than 10 000 shaped pulses.

Note that in the frequency region beyond 1 THz, various elementary excitations, such as phonon, magnon, superconductor gap and so on, emerge. The key idea for potential applications of the present ultrahigh-repetition-rate SUPT source would be manipulations of light–matter interaction...
interactions on the basis of a resonance between an ultrahigh-repetition rate and characteristic frequencies of such elementary excitations.

Acknowledgments

We are grateful to T Suzuki for valuable advice. We thank K Midorikawa and A Suda for useful discussions. We also thank N Yamazaki, N Sawayama and A J Kiran for their technical help with the experiment.

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New Journal of Physics 13 (2011) 023030 (http://www.njp.org/)