

Available online at www.sciencedirect.com



Journal of Photochemistry Photobiology A:Chemistry

Journal of Photochemistry and Photobiology A: Chemistry 182 (2006) 220–224

www.elsevier.com/locate/jphotochem

# Generation of optical-field controlled high-intensity laser pulses

Masayuki Kakehata ∗, Hideyuki Takada, Yohei Kobayashi, Kenji Torizuka

*Photonic Research Institute, National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba Central 2, 1-1-1 Umezono, Tsukuba 305-8568, Japan*

Available online 11 July 2006

#### **Abstract**

We demonstrated control of electric fields of optical pulses by combining the pulse shaper with carrier-envelope phase (CEP) stabilized lasers. We employed spectral phase controller known as the pulse shaper for controlling the pulse envelope and the CEP. In order to evaluate the accuracy of the device, we measured the CEP shift, and delay shift by the spectral interferometry method and confirmed that the CEP and the delay were precisely controlled by the pulse shaper independently. We demonstrated control of the CEP and the relative spectral phase of amplified high-intensity laser pulses by installing the pulse shaper in a CEP stabilized amplifier system. The pulse shaper worked as an ideal CEP shifter because it controlled the phase of very broad spectrum precisely, and independent control of CEP was achieved. © 2006 Elsevier B.V. All rights reserved.

*Keywords:* Femtosecond lasers; Carrier-envelope phase; Pulse shaping; High-intensity lasers

#### **1. Introduction**

The carrier-envelope phase (CEP) is the relative phase between the peak of the pulse envelope and the electric field. Recently, methods to control the pulse-to-pulse shift of the carrier-envelope phase that is called carrier-envelope offset (CEO) phase, have been demonstrated [\[1,2\].](#page-4-0) These results opened a way to control the electric-field shape of optical pulses. Such laser sources are important for the metrology as well as for controlling laser–matter interactions. Using a carrier-envelope offset (CEO) phase stabilized few-cycle laser oscillator, dependence of the photoelectron emission on the CEP was investigated [\[3\].](#page-4-0) Furthermore, using CEP stabilized amplified laser pulses, control of high-order harmonic generation and attosecond pulse measurement were demonstrated [\[4,5\], a](#page-4-0)nd measurement of the electric field of optical pulse was demonstrated using the attosecond pulses [\[6\].](#page-4-0)

CEP stabilization in an amplified laser system was demonstrated in a chirped-pulse amplification (CPA) system composed of a material-based pulse stretcher and a prism-based pulse compressor [\[4\],](#page-4-0) and in a commonly used CPA system employing grating-based stretcher/compressor and regenerative/multi-pass amplifiers [\[7\]. T](#page-4-0)here are also passive CEP stabilization schemes

1010-6030/\$ – see front matter © 2006 Elsevier B.V. All rights reserved. doi[:10.1016/j.jphotochem.2006.05.025](dx.doi.org/10.1016/j.jphotochem.2006.05.025)

using an optical parametric amplifier [\[8\]](#page-4-0) or a difference fre-quency generation scheme [\[9\].](#page-4-0)

If one assumes a pump-probe experiment with CEP stabilized pulses, one has to prepare a set of pulses, one has fixed CEP and the other has controllable CEP without shifting the relative delay and without changing the pulse envelope shape. A CEP shifter that controls only the CEP will be required. The wellknown easiest way to control the CEP of a pulse is to change the optical path length in a dispersive material. A thin plate, however, shifts the relative delay of the pulse and it will change the pulse envelope shape for a very broad spectrum due to the group velocity dispersion [\[10\].](#page-4-0)

The pulse-shaping technique [\[11\]](#page-4-0) that controls the relative spectral phase allows us to control the shape of the pulse envelope. The technique has been widely used to compensate for the residual dispersion [\[12\], t](#page-4-0)o optimize the laser–matter interaction [\[13\],](#page-4-0) and to control spectral distribution of HHG [\[14\].](#page-4-0) In these experiments, however, the relative phase between the pulse envelope and the carrier field was not stabilized. The shape of the pulse envelope was controlled precisely, however, the electric field inside the pulse envelope was not stabilized. If we apply the pulse-shaping technique for the CEP stabilized pulses, we can fully control the electric field shape of pulses. We proposed and demonstrated an active CEP-shifting device employing a pulse shaper, which is an active CEP shifter with a calibrated phase shift that can shift the CEP without affecting the delay and the shape of the pulse envelope [\[15,16\].](#page-4-0) We here report a

<sup>∗</sup> Corresponding author. Tel.: +81 29 861 5155; fax: +81 29 861 3349. *E-mail address:* [kakehata-masayuki@aist.go.jp](mailto:kakehata-masayuki@aist.go.jp) (M. Kakehata).

<span id="page-1-0"></span>

Fig. 1. Relation between the spectral phase and the carrier-envelope phase and delay.

chirped-pulse amplification system providing electric-field controlled optical pulses by combining the CEP stabilized laser system with the pulse shaping devices that works not only as a pulse envelope shaper but also as a CEP shifter.

## **2. Theory**

Fig. 1 schematically shows the relation between the spectral phase and the CEP and the time delay. The left shows the spectral phase as a function of the angular frequency and the right shows the optical waveforms in time domain. We here assume a transform-limited pulse that has a flat spectral phase (constant value or a linear function of the frequency). In the spectral domain, the CEP is defined as the intercept of the spectral phase at zero frequency, and the slope of the spectral phase corresponds to the delay of the pulse [\[17\]. A](#page-4-0)n ideal CEP shifter that shift only the CEP without changing the pulse timing (delay) and the pulse envelope should give constant phase shift to all the frequency component.

In some experiments, insertion of dispersive material into the beam path is used as CEP shifting method. The dispersive material gives both the delay shift and the phase shift to the pulse. Because the delay shift is determined by the group velocity and the phase shift is determined by the phase velocity, propagation by  $L = 1/(dn/d\lambda)$  results in CEP shift of  $2\pi$  from the difference of the two velocities. For example, propagation of  $L = 1/(dn/d\lambda) = 58 \mu m$  for fused-silica plate at 800 nm. This method is easy and work well for 10 fs pulses around 800 nm, however, for pulses having much broader spectrum, the effect of the higher-order term of the spectral phase can not be neglected. The higher-order term changes the pulse envelope shape and will result in error in the CEP shift [\[10\]. T](#page-4-0)he method using the pulse shaper can control the phase of a broad spectrum [\[12\], t](#page-4-0)herefore, it can be an ideal CEP shifter.

The schematic of the optical-field controlled laser system is shown in Fig. 2, which is a combination of a CEP stabilized laser and a pulse shaper. Such system allows us to control both the optical phase inside the pulse envelope and the shape of the pulse envelope.

#### **3. Experimental**

The pulse shaper is composed of a liquid-crystal spatial light modulator (SLM, SLM-S640/12 Jenoptic) placed at the Fourier transform plane of a 4*f* optical configuration, two cylindrical mirrors  $(f = 224 \text{ mm})$ , two prisms (SFL03) and folding mirrors as shown in Fig. 2. [Fig. 3](#page-2-0) shows the diagram of the laser system. We installed the pulse shaper in a CEP stabilized CPA system [\[7\].](#page-4-0) The amplifier system is composed of a CEO stabilized oscillator, a pulse-selection system, a grating-based pulse stretcher, a regenerative amplifier, a multi-pass amplifier, and a grating-based pulse compressor. The CEO beat frequency *f*<sub>ceo</sub> was stabilized to  $f_{\text{osc}}/8$  with phase-locked-loop electronics. The pulses possessing the same CEP are selected by a Pockels cell in the regenerative amplifier. The pulse energy is 3.5 mJ after the four-pass amplifier, and the compressed pulse is about 1 mJ.



Fig. 2. Schematic of the laser system generating optical-field controlled pulses.

<span id="page-2-0"></span>

Fig. 3. Experimental setup of the optical-field controlled high-intensity laser pulse generation.

The amplified pulse is 20 nm FWHM, which corresponds to a 50 fs FWHM transform-limited pulse. The relative CEP of the amplified pulse was measured by the self-referencing spectral interferometry method [\[18,19\]](#page-4-0) and the relative spectral phase was measured by the Spectral phase interferometry for direct electric field reconstruction (SPIDER) method [\[20\].](#page-4-0)

## **4. Results and discussion**

## *4.1. Control of CEP shift by a pulse shaper*

Fig. 4 illustrates the spectral interferometry setup to test the feasibility of a pulse shaper as a CEP shifter. The pulse-train from a CEP unstabilized Ti:sapphire laser oscillator (80 MHz, 35 fs FWHM) was split into two pulses, a reference pulse and a probe pulse. The probe pulse passes through a pulse shaper. We measured the spectral interference (SI) between the reference pulse and the probe pulse. Because we measured the relative CEP shift and relative delay shift in this experiment, the CEP of the input pulse  $(\phi_0)$  was not stabilized.

First, we measured the spectral interferometry (SI) to obtain the phase difference  $\phi_1(\omega) = \phi_{\text{orb},0}(\omega, t = 0) - \phi_{\text{ref}}(\omega)$  after giving a certain initial spectral phase by SLM. Second, we applied periodic phase modulation by SLM and measured the time-dependent phase difference  $\phi_2(\omega, t) = \phi_{\text{prb}}(\omega, t) - \phi_{\text{ref}}(\omega)$ . We then calculated the relative phase shift  $\phi_s(\omega, t) = \phi_2(\omega, t)$  $t$ ) –  $\phi_1(\omega)$ , and fit a linear function to  $\phi_s(\omega, t)$  in the  $\omega$ - $\phi$  domain and derived the relative delay shift (slope of the linear function) and the relative CEP shift (intersect of the linear function) as shown in [Fig. 1.](#page-1-0)

[Fig. 5](#page-3-0) shows the relative CEP shift and delay shift for different experimental conditions: (a) without phase modulation, (b) sinusoidal delay shift by a PZT, and (c) sinusoidal CEP shift by the pulse shaper. [Fig. 5\(a](#page-3-0)) gives us information on the stability of the interferometer and the accuracy of the measurement; the standard deviation was 0.1 rad rms and 0.06 fs rms in 20 s. We



Fig. 4. Experimental setup to test the pulse shaper as a CEP shifter. CM, cylindrical mirror ( $f = 242$  mm) and prisms are made of SFL03.

<span id="page-3-0"></span>

Fig. 5. Change of the CEP shift and delay shift. Measured (a) without modulation, (b) with sinusoidal delay modulation applied by the delay line in the interferometer, and (c) with sinusoidal CEP modulation by the pulse shaper.

found that the stability depends on the environmental noise and that the smallest value can be about one-half of the presented results. Fig. 5(b) demonstrates that we can separately measure the relative delay shift and the relative CEP shift. The measured delay shift of 6.4 fs, agreed with the applied delay shift 7.2 fs, within the accuracy of the PZT actuator. CEP shift was achieved by shifting the phase so that the  $\phi_s(\omega, t)$  shift vertically. Fig. 5(c) confirms that we can control the relative CEP shift without shifting the relative delay. The measured CEP shift of 4.1 rad agreed with the applied shift of 4.0 rad within the experiment accuracy. The results demonstrate that we can clearly distinguish the CEP shift from the delay shift and that the pulse shaper works as an active CEP shifter without changing the relative delay [\[15,16\]. T](#page-4-0)he CEP shifter is a kind of an achromatic phase shifter as reported using the gratings and mobable optics [\[21\]. P](#page-4-0)recise measurements of both the phase shift and the delay shift have not been reported for the achromatic phase shifters so far.

#### *4.2. Control of CEP of amplification pulses*

We measured the relative CEP of the amplified pulses by the self-referencing spectral interferometry (SI) method, with modulating the CEP by the pulse shaper to test whether the relative CEP is really controlled by the pulse shaper. The "relative CEP" means that the CEP measured by the



Fig. 6. Self-referencing SI fringe measured with applying sinusoidal CEP modulation by the pulse shaper at 2 Hz.

self-referencing SI method has a constant phase shift relative to the true value at the measurement point [\[18,19\].](#page-4-0) After the spectrum broadening by self-phase modulation in a  $150 \,\mu m$ inner-diameter Kr filled hollow fiber, the second harmonic (SH) was generated by passing a 300  $\mu$ m BBO crystal. We measured the SI between the SH and fundamental of the same pulse. The exposure time of the spectrometer CCD was set 21 msec, and about 12 pulses were exposed to one trace  $(f_{\text{amp}} = 555 \text{ Hz})$ . The self-referencing SI fringe position, which indicate the relative value of the CEP, was observed around 420 nm. Fig. 6 shows the temporal evolution of the self-referencing SI signal (95 traces in 2 s). The applied CEP modulation at 2 Hz was clearly observed in the fringe, which clearly shows that the CEP of amplified pulse was controlled by the pulse shaper.

## *4.3. Control of relative spectral phase of amplified pulses by the pulse shaper*

We measured the relative spectral phase of the amplified pulses by the SPIDER method [\[20\]](#page-4-0) with applying parabolic



Fig. 7. (a) Measured spectral phase (grey dots) and fitted second order dispersion (solid curve) and the measured SPIDER signal shifted to the fundamental frequency (thin solid curve) for applied dispersion of  $-500$  fs<sup>2</sup>. (b) Plots of the measured second order dispersion as a function of applied dispersion. Dotted line corresponds to measured value equal to the applied value.

<span id="page-4-0"></span>spectral phase by the pulse shaper. The measured spectral phase was fitted with a second order function of the angular frequency to determine the second order dispersion as shown in [Fig. 7\(a](#page-3-0)) for which  $-500 \text{ fs}^2$  dispersion was applied. We measured the spectral phase for different value of applied second order dispersion, and plotted as in [Fig. 7\(b](#page-3-0)). The measured value agreed well with the applied dispersion.

#### **5. Conclusion**

We demonstrated an active carrier-envelope phase shifter and pulse envelope shaping using a 4*f*-pulse shaper. A pulse shifter was employed with the CEP stabilized CPA system to actively control the relative CEP and the pulse envelope of amplified pulses. These results clearly demonstrate the relation between the spectral phase and the electric field shape of an optical pulse. The method allows us to control the CEP precisely without affecting the pulse delay and the pulse envelope shape, which can not be achieved by using dispersive material such as a thin fused-silica plate. We also demonstrated that a combination of a pulse shaper and a CEP stabilized CPA system allows us to design electric fields of optical pulses.

#### **Acknowledgements**

The authors are grateful to K. Nishijima, H. Takamiya, and D. Yoshitomi for their technical assistance and fruitful discussions. A part of this study was financially supported by the Budget for Nuclear Research of the Ministry of Education, Culture, Sports, Science, and Technology, based on screening and counseling by the Atomic Energy Commission, Japan.

#### **References**

- [1] D.J. Jones, S.A. Diddams, J.K. Ranka, A. Stentz, R.S. Windeler, J.L. Hall, S.T. Cundiff, Science 288 (2000) 635–639.
- [2] A. Apolonski, A. Poppe, G. Tempea, C. Spielmann, T. Udem, R. Holzworth, T.W. Hänsch, F. Krausz, Phys. Rev. Lett. 85 (2000) 740-743.
- [3] A. Apolonski, P. Dombi, G.G. Paulus, M. Kakehata, R. Holzworth, Th. Udem, Ch. Lemell, K. Torizuka, J. Burgdörger, T.W. Hänsch, F. Krausz, Phys. Rev. Lett. 92 (2004) 073902.
- [4] A. Baltuška, Th. Udem, M. Ulberacker, M. Hentschel, E. Goullelmakis, Ch. Gohle, R. Holzwarth, V.S. Yakovlev, A. Scrinzi, T.W. Hansch, F. ¨ Krausz, Nature 421 (2003) 611–615.
- [5] R. Kienberger, E. Goulielmakis, M. Uiberacker, A. Baltuska, V. Yakovlev, F. Bammer, A. Scrinzi, Th. Westerwalbesloh, U. Kleinberg, U. Heinzmann, M. Drescher, F. Krausz, Nature 427 (2004) 817– 821.
- [6] E. Goulielmakis, M. Uiberacker, R. Kienberger, A. Baltuška, V. Yakovlev, A. Scrinzi, Th. Westerralbesloh, U. Kleineberg, U. Heinzmann, M. Drescher, F. Krausz, Science 305 (2004) 1267–1269.
- [7] M. Kakehata, H. Takada, Y. Kobayashi, K. Torizuka, H. Takamiya, K. Nishijima, T. Homma, H. Takahashi, K. Okubo, S. Nakamura, Y. Koyamada, Opt. Express 12 (2004) 2070–2080.
- [8] A. Baltuška, T. Fuji, T. Kobayashi, Phys. Rev. Lett. 88 (2002) 133901–133904.
- [9] T. Fuji, A. Apolonski, F. Krausz, Opt. Lett. 29 (2004) 632–634.
- [10] P. Dombi, A. Apolonski, Ch. Lemell, G.G. Paulus, M. Kakehata, R. Holzwarth, Th. Udem, K. Torizuka, F.J. Burgdörfer, T.W. Hänsch, F. Krausz, New J. Phys. 6 (2004) 39.
- [11] A.M. Weiner, D.E. Leaird, J.S. Patel, J.R. Wulllert, Opt. Lett. 15 (1990) 326–328.
- [12] K. Yamane, Z. Zhang, K. Oka, R. Morita, M. Yamashita, A. Suguro, Opt. Lett. 28 (2003) 2258–2260.
- [13] A. Assion, T. Baumert, M. Bergt, T. Brixner, B. Kiefer, V. Seyfried, M. Strehle, G. Gerber, Science 282 (1998) 919–922.
- [14] R. Barterls, S. Backus, E. Zeek, L. Misoguti, G. Vdovin, I.P. Christov, M. Murnane, H.C. Kapteyn, Nature 406 (2000) 164–166.
- [15] M. Kakehata, H. Takada, Y. Kobayashi, K. Torizuka, K. Nishijima, H. Takamiya, T. Homma, H. Takahashi, Ultrafast Phenomena XIV, vol. 79, Springer Series in Chemical Physics, 2005, pp. 88–90.
- [16] K. Nishijima, M. Kakehata, H. Takamiya, H. Takada, Y. Kobayashi, T. Homma, H. Takahashi, K. Torizuka, IEEJ Trans. EIS 125 (2005) 1686–1693 (in Japanese).
- [17] A.W. Albrecht, J.D. Hybl, S.M. Gallagher Faeder, D.M. Jonas, J. Chem. Phys. 111 (1999) 10934–10956.
- [18] M. Kakehata, H. Takada, Y. Kobayashi, K. Torizuka, Y. Fujihira, T. Homma, H. Takahashi, Opt. Lett. 26 (2001) 1436–1438.
- [19] M. Kakehata, H. Takada, Y. Kobayashi, K. Torizuka, Y. Fujihira, T. Homma, H. Takahashi, Appl. Phys. B74 (Suppl.) (2002) S43– S50.
- [20] C. Iaconis, I.A. Walmsley, Opt. Lett. 23 (1998) 792-795.
- [21] A.V. Zvyagin, D.D. Sampson, Opt. Lett. 26 (2001) 187–189.