Measurement of the Shot-to-Shot Carrier-Envelope Phase Slip of Femtosecond Laser Pulses

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We measure the shot-to-shot slip of the carrier-envelope phase (CEP) of mode-locked femtosecond laser pulses by using a 2f-to-f self-referencing interferometer. The 2f-to-f interferometer, composed of a one-octave spectrum generator and a nonlinear interferometer, yields the beat signal between the 2f-component and the second harmonic of the f-component generated in a nonlinear crystal. The CEP slip between two successive laser pulses is successfully measured from the beat frequency with a phase jitter of 2.7 mrad. We also control the shot-to-shot CEP slip from 0 to ∼π by adjusting either the intracavity prism insertion or the pumping power of the laser oscillator. This measurement offers basic information for the design of a feedback control loop to stabilize the shot-to-shot CEP slip of mode-locked laser pulses.

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I. INTRODUCTION

With the advancement of sub-10-fs pulse generation techniques [1], the description of an optical pulse based on the slowly varying envelope approximation has come to face a new aspect. Unlike many-cycle pulses, the few-cycle pulses are significantly modified in their shape by the phase offset between the optical carrier peak and the envelope peak, called the absolute phase or the carrier-envelope phase (CEP). It has been theoretically shown that the spectrum of a femtosecond pulse propagating in a nonlinear medium depends on the CEP of the laser pulse [2] and that the conversion efficiency of high-order harmonic X-ray generation, which is an extreme nonlinear process, using sub-10-fs pulses can be affected by the CEP [3]. Paulus et al. [4] experimentally observed that the photoionization process driven by 2-cycle optical pulses depended upon the CEP of the pulses. These results show that light-matter interactions become sensitive to the CEP of the pulse as the laser pulse duration gets shorter than a few optical cycles.

For coherent control of nonlinear optical processes and light-matter interactions driven by few-cycle pulses, the random fluctuation of the CEP between successive pulses should be suppressed through a stabilization of the CEP.

Even though active control of the CEP value of laser pulses has not been reported until now, the measurement and the stabilization of shot-to-shot CEP change of mode-locked laser pulses has been intensively investigated using nonlinear interferometry schemes [3,5–7]. The spectral interferometry method was also proposed as a tool for single-shot CEP measurement [8] and was utilized to measure shot-to-shot CEP change [9,10]. Especially, Baltuška et al. [10] demonstrated that an optical parametric amplifier seeded by a white-light continuum could be a self-stabilized source of few-cycle pulses wherein the same CEP was reproduced in each laser shot, even though active control was still not achieved.

As in time-domain applications, such as nonlinear optics and light-matter interactions, the CEP of few-cycle laser pulses plays a very important role in frequency-domain applications. The stabilization technique of the shot-to-shot CEP slip of a mode-locked laser has had a great impact on optical frequency metrology because it offers a stabilized optical frequency comb in the hundreds of THz region, which functions as a simple, but highly precise, frequency standard [6]. The frequency-domain application has now been extended to ultrahigh-resolution spectroscopy [11,12] and optical comb synchronization of two independent lasers [13].

In this paper, by using nonlinear interferometry, we demonstrate the measurement and the control of the shot-to-shot CEP slip of femtosecond optical pulses gen-
Thus, measure the time-domain quantity $\Delta \phi$ where

$$\Delta \phi = 2\pi \delta / f_{\text{rep}},$$

(2)

where $f_{\text{rep}}$ is the repetition rate of the laser. We can, thus, measure the time-domain quantity $\Delta \phi$ by probing the frequency-domain quantity $\delta$. The measurement of $\delta$ can be easily realized if we have a one-octave spectrum that covers from a frequency $f$ to its doubled frequency $2f$. The frequency of the $n$th optical comb is expressed, in terms of the repetition rate and the frequency offset, as

$$f_n = nf_{\text{rep}} + \delta.$$  

(3)

The second-harmonic frequency of the $n$th optical comb is given by $f_{\text{SHG}} = 2f_n = 2(nf_{\text{rep}} + \delta)$, whereas the $2n$th optical comb has the frequency $f_{2n} = 2nf_{\text{rep}} + \delta$. The two frequencies, $f_{\text{SHG}}$ and $f_{2n}$, cannot be discriminated in the spectrum, but by interfering $f_{\text{SHG}}$ with $f_{2n}$, we can get a beat frequency of $f_{\text{SHG}} - f_{2n} = \delta$, the offset of the frequency comb.

The proper control of $\delta$ allows the generation of CEP-controlled optical pulses as long as the change in the repetition rate is negligible. When $\delta$ is stabilized to be constant, the frequency of each optical comb is exactly defined, and the CEP difference between two successive pulses is the same as that of the previous two. Especially, in the case of zero $\delta$, all the pulses have the same CEP, and the absolute phase is fixed to an unknown value $\phi_0$. The $\phi_0$ value can be determined with another method, such as high-order harmonic generation or photo-ionization. The physical factors that affect $\delta$ in a Kerr-lens mode-locked laser oscillator are the intracavity dispersion and the Kerr nonlinearity. The difference between the group velocity and the phase velocity of a laser pulse depends on the path length of the intracavity dispersive materials and can be controlled, practically, by adjusting the insertion depth of a prism installed for intracavity dispersion compensation. The Kerr nonlinearity, which induces an intensity-dependent change in the group velocity, can be controlled by adjusting the intracavity pulse energy through pumping power control.

III. MEASUREMENT AND CONTROL OF SHOT-TO-SHOT CEP SLIP

The setup of the $2f$-to-$f$ self-referencing interferometer consists of a white-light continuum generator and a nonlinear interferometer, as illustrated in Fig. 3. The
Fig. 3. Experimental setup for the measurement of shot-to-shot CEP slip. The laser source is an MDC mode-locked Ti:sapphire laser capable of delivering 3-cycle (sub-8-fs) optical pulses (ASF: air-silica microstructure fiber; DBS: dichroic beamsplitter; KTP: KTP crystal; 40×: 40× objective lens; BS: beamsplitter; BPF: band-pass filter at 532 nm; PBS: polarizing beamsplitter; P: linear polarizer; APD: avalanche photodiode).

conventional spectrum of a prism-dispersion-controlled Ti:sapphire laser [14] covers the region from 700 nm to 900 nm (or 750 nm to 950 nm), and that of a mirror-dispersion-controlled (MDC) laser [15] covers the region from 650 nm to 1000 nm, which is still not sufficiently broad to cover one octave. The most common technique to obtain an ultrabroadband spectrum is continuum generation by means of the self-phase modulation (SPM) effect of a high-intensity laser pulse in a solid or an optical fiber. Recently, Ell et al. [16] succeeded in generating a one-octave spectrum directly from a Ti:sapphire laser by installing an additional SPM arm in the cavity, but they employed specially designed, not commercially available, double-chirped mirrors for intracavity dispersion compensation. So far, continuum generation outside the laser cavity has been common because it does not require neither special coatings nor careful intracavity dispersion control. In this work, we use an air-silica microstructure fiber [17] to generate a super-continuum with a one-octave spectrum. This fiber significantly reduces the laser intensity required for the wide spectral broadening caused by SPM. It prevents the laser pulse from being temporally broadened during the SPM process and enhances the spectral broadening during the propagation because the dispersion at the center wavelength of Ti:sapphire lasers, 800 nm, is negative. Additional nonlinear processes, such as four-wave mixing and Raman scattering, are excited in a long fiber, and, as a result, a flattened super-continuum spectrum is obtained. The offset of the optical combs does not change during those nonlinear processes. We focus 30-fs, 3-nJ laser pulses generated from a mode-locked Ti:sapphire laser into a 12-cm-long air-silica microstructure fiber by using a 40× objective lens and recollimate the generated super-continuum by using another objective lens. The laser oscillator is capable of delivering three-cycle (7.8-fs) optical pulses, as shown in the inset of Fig. 3, by means of external compression, but the uncompressed (negatively chirped) 30-fs pulses are enough for the nonlinear effect in the air-silica microstructure fiber even with consideration of the additional pulse broadening in the objective lens. The coupling efficiency of the two objective lenses and the fiber is 30%. Figure 4(a) shows the input laser spectrum on a logarithmic scale, and Figs. 4(b) and 4(c) shows the generated super-continuum spectra in logarithmic and linear scales, respectively. The generated super-continuum covers from 450 nm to 1150 nm, which is sufficiently broad to cover one octave. We choose 1064 nm as the frequency \( f_n \) and 532 nm as \( f_2n \) or \( 2f_n \) because Nd:YAG laser optics are conventionally available.

The super-continuum is divided into the infrared (IR) and green parts by using a dichroic beamsplitter, as shown in Fig. 3. The IR part containing \( f_n \), 1064 nm, is focused into a 1-mm-thick KTP crystal by use of a 10× objective lens to generate its second harmonic frequency, \( 2f_n \), at 532 nm, and is interfered with the green part containing \( f_2n \), 532 nm. The delay arm in Fig. 3 ensures temporal overlap of the two \( 2f(2f_n \text{ and } f_2n) \)-pulses. Since both beams have broad spectra around 532 nm, they are filtered by using two color filters centered at 532 nm with a bandwidth of 1 nm. The second-harmonic \( (2f_n) \) beam has s-polarization (vertical to the optical table) whereas the laser pulse and the generated continuum, including \( f_n \) and \( f_2n \), have p-polarization. The \( 2f_n \) and \( f_2n \) beams are combined in a polarizing beamsplitter to be spatially overlapped. Since the polar-
izing of one beam is orthogonal to that of the other, the interference should occur on a plane moderately rotated between the vertical and horizontal axes, which can be realized by inserting a linear polarizer. The angle of the polarizer should be properly selected to enhance the beat signal by equalizing the signal levels of two 532-nm beams because the intensity of the $f_{2n}$ beam is much stronger than that of the $2f_n$ beam. Finally, the beat signal is detected using an avalanche photodiode (APD) and is displayed either on an oscilloscope or on an rf spectrum analyzer.

A typical interference profile obtained at the oscilloscope is shown as Fig. 5(a), where we can observe a 116-MHz pulse train with the amplitude modulation representing the beat between the $2f_n$ and the $f_{2n}$ beams. The measured rf spectrum is in Fig. 5(b), where we can observe three peaks at the frequencies of $f_{rep}$ and $f_{rep} \pm \delta$. The beat frequency $\delta$ manifests itself in the two side-peaks. The observed jitter in $\delta$ is $\pm 50$ kHz, indicating an inherent phase error in the CEP slip measurement of $\pm 2.7$ mrad. This jitter comes from the random noise of the unstabilized laser and the nonlinear phase noise in the air-silica microstructure fiber due to a fluctuation in the coupling energy [18]. Mechanical vibration of the optical table is the most serious factor that magnifies both noises. The thermal noise of the Ti:sapphire crystal and the flow of air were found to be negligible in the short-term stability.

After attaining the measurement tool, we controlled $\delta$ by changing the insertion depth of the intracavity prism and the pumping power. Figure 6(a) shows the prism-insertion dependence of $\delta$. The CEP slip occurring when the medium length is extended from $l_0$ to $l$ is given by $\Delta \phi = 2\pi \frac{dn}{d\lambda} (l - l_0)$, where $n$ is the refractive index. The intracavity prisms are made of fused silica whose $dn/d\lambda$ value at 800 nm is $-0.0173 \, \mu m^{-1}$. The fitting curve, calculated from the $dn/d\lambda$ value and the prism insertion depth, has a slope of $-0.307 \, rad/\mu m$ and shows good agreement with the experimental measurement. The intracavity power dependence of $\delta$ at a fixed prism insertion is shown in Fig. 6(b). The linear regression shows that the slope of the line is $-0.316 \, rad/nJ$. By finely tuning the two parameters, we could slow down $\delta$ to nearly zero, in which case the beat frequency changes at around the jitter level. Figure 6(c) depicts a 280-kHz pure beat signal in the time domain. We obtained it by removing the 116-MHz pulse trains from the original interference profile by using two additional APDs, while are represented by ADP2 and APD3 in Fig. 3, and a subtraction circuit. This low-frequency beat signal endures without dephase for several milliseconds and then fluctuates, implying that pulses with the same CEP can be generated every 3.6 $\mu s$ for several milliseconds in the unstabilized laser used here. When the beat frequency is further reduced below $\sim 280$ kHz, the sinusoidal beat signal was not regularly observed due to the jitter. Perfect isolation of the optical table and the experimental setup from the environment will significantly improve the CEP stability and allow a further reduction in the phase jitter. If a jitter-free $\delta$ and, ultimately, zero $\delta$ are to be obtained for a relatively long time (a few seconds), a laser stabilization process through feedback control of the prism insertion depth or pumping power is absolutely required. The quantitative characterization of shot-to-shot CEP slip of the laser pulses demonstrated in this work is a basic process for the fabrication of the required feedback loop and for the laser stabilization process.

IV. CONCLUSIONS

We measured the shot-to-shot CEP slip of a modelocked Ti:sapphire laser by using a $2f$-to-$f$ self-referencing interferometer. A 12-cm air-silica microstructure fiber was used to generate a one-octave spectrum, and a nonlinear interferometer was employed to observe the beat signal between the $2f_n$ and the $f_{2n}$
components. The measured beat frequency ($\delta$) had a jitter of $\pm 50$ kHz, which corresponded to a phase error of $\pm 2.7$ mrad in the shot-to-shot CEP slip measurement. We also controlled $\delta$ by changing the insertion depth of the intracavity prism and the laser pumping power, and by finely adjusting two parameters, we slowed down $\delta$ to nearly zero, close to the jitter level. Perfect isolation of the optical table and experimental setup from the environment is required to significantly reduce the jitter, and an active stabilization of the laser is necessary for jitter-free operation. The measurement of the parameter dependence of $\delta$ in this work will allow suitable design of a feedback control loop to stabilize $\delta$ to a certain frequency and to fix it to be zero for the generation of CEP-locked sub-10-fs laser pulses.

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