Relative Carrier-Envelope-Offset Phase Control Between Independent Femtosecond Light Sources

Yohei Kobayashi, Zhiyi Wei, Masayuki Kakehata, Hideyuki Takada, and Kenji Torizuka

***Abstract*—The authors studied the measurement and control of relative carrier-envelope-offset phases among femtosecond light pulses whose optical frequencies have a subharmonic relation. They report the fluctuation dynamics of the phases and their control of two types of passive pulse-timing synchronized systems: passively synchronized mode-locked Ti:sapphire and Cr:forsterite lasers and a femtosecond subharmonic optical parametric oscillator. These techniques will facilitate Fourier synthesizing among subharmonic pulses in order to realize subfemtosecond pulse-train generation in the visible and infrared regions.**

***Index Terms*—Carrier-envelope-offset phase, Fourier synthesis, optical frequency comb, optical parametric oscillator, ultrashort lasers.**

I. Introduction

REMARKABLE progress has been made in controlling the carrier-envelope-offset (CEO) phase in a Ti:sapphire laser oscillator [1]–[3]. Controlling the CEO phase corresponds to controlling the CEO frequency in the frequency domain. CEO frequency control can yield an ultrawide optical frequency comb. The CEO phase-control technique combined with laser-cooled atom or ion trapping can achieve an all-optical atomic clock [4]. In time-domain applications, the absolute phase affects the probability of tunneling ionization. The ion and photoelectron trajectories driven by a laser field can thus be controlled [5]. The high-order harmonics generation is also concerned with the absolute phase, and the pulse shape of the high-order harmonics in a few femtosecond region strongly depends on the absolute phase [6]. Attosecond pulse generation by high-order harmonics was demonstrated recently [7], [8]. A CEO phase-controlled high-intensity laser will be used to explore the phase-dependent application.

Attosecond pulse-train generation in the visible and infrared regions was also proposed by using Fourier synthesizing of subharmonic pulses [9], [10]. In order to generate attosecond pulses in this scheme, we must prepare timing and phase-locked subharmonic pulses. Fully active control of pulse timing and the CEO phase between two Ti:sapphire laser pulses was demonstrated [11], and they have been synthesized. Applying similar control techniques, researchers recently demonstrated the control of the CEO phase and pulse timing between Ti:sapphire and Er-doped fiber lasers [12]. Another approach to providing phase-coherent multicolor pulses is to apply CEO-phase control to passively synchronized femtosecond systems. The timing of a synchronized dual-wavelength laser consisting of Ti:sapphire and Cr:forsterite lasers [13] is a promising candidate for the CEO -phase-controlled light source because the optical frequency ratio between them is roughly 3:2. A femtosecond optical parametric oscillator (OPO) with an optical frequency ratio of 1:2:3 (idler: signal: pump) [14] is also an appropriate light source. In spite of these activities, controlling the CEO phase among multicolor pulses with sufficient precision and reliability to realize subfemtosecond pulse-train generation is still a challenging problem. Frequency conversions of the amplified pulses by an optical parametric amplifier could generate subharmonic pulses, and their phase relation is fixed when white continuum is used as seed pulses and the pulse energies are quite stable. They could also be used for subfemtosecond pulse train generation, although the repetition rate is limited. This scheme can be used for phase-sensitive highly nonlinear phenomena. On the other hand, the phase controlled subharmonic pulses generation in high-repetition rate can be used not only in frequency-domain applications described above but also time-domain applications such as coherent pulse addition [15] and coherent antistokes raman spectroscopy [16]. The high-repetition rate and phase relation-locked oscillators would be suitable for phase-dependent spectroscopy. Also, these light sources can be amplified by laser-gain mediums or optical parametric amplifiers.

In this paper, we study the CEO phase control for two types of passively synchronized femtosecond systems. The first one is passively synchronized Ti:sapphire and Cr:forsterite lasers. Superposing the second harmonic of the Ti:sapphire and the third harmonic of the Cr:forsterite laser, we can obtain a beat signal representing the relative CEO phase. The dynamics of the free-running fluctuation of the beat signal and control of the beat signal by an electronic feedback have been reported. The locked beat signal has a spectral width of 3 kHz. The second system is a femtosecond subharmonic OPO. The sum frequency between the pump and idler and the second harmonic of the signal have the same wavelength, thus we can obtain the phase relation by superposing them. The beat signal was locked using electronic feedback, and the locked spectrum width was 100 kHz. Here, the pulse timings in both systems were passively synchronized, and relative phases were controlled actively by changing the cavity length slightly while maintaining passive-timing synchronization. Finally, we also present a comparison of the two systems.

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Y. Kobayashi, M. Kakehata, H. Takada, and K. Torizuka are with the National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba 305-8568, Japan (e-mail: y.kobayashi@aist.go.jp).

Z. Wei was with the New Energy and Industrial Technology Development Organization, Tokyo 170-6027, Japan. He is now with the Laboratory of Optical Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100080, China.

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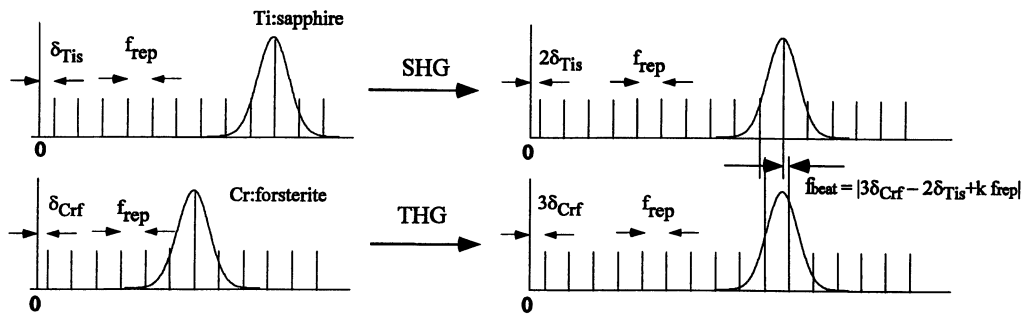

Fig. 1. Schematic of CEO frequency-relation measurement in a dual-wavelength laser in the frequency domain. Comb spacings $(f_{\rm rep})$ are the same because the Ti:sapphire laser and Cr:forsterite laser are passively synchronized. Spectra of the frequency-doubled Ti:sapphire and the frequency-tripled Cr:forsterite laser overlap at 415 nm, and a beat signal can be observed. SHG is second-harmonic generation and THG third-harmonic generation.

II. CEO Phase Control Between Ti:Sapphire and Cr:Forsterite Lasers

The dual-wavelength laser outputs timing-synchronized two-femtosecond laser pulses at the center wavelengths of 830 and 1245 nm. The power of the Ti:sapphire and Cr:forsterite lasers are typically 400 and 200 mW with pulsewidths of 50 fs. The spectra of the third harmonic of the Cr:forsterite and the second harmonic of the Ti:sapphire laser overlap at 415 nm. Thus, we can obtain the beat signal by superposing them. The schematic of the comb relation is shown in Fig. 1. Here, the comb frequencies of the Ti:sapphire $(f_{\rm Tis})$ and the Cr:forsterite $(f_{\rm Crf})$ lasers are defined as $f_{\rm Tis} = \delta_{\rm Tis} + mf_{\rm rep}$ and $f_{\rm Crf} = \delta_{\rm Crf} + nf_{\rm rep}$, where $\delta_{\rm Tis}$ and $\delta_{\rm Crf}$ are the offset frequencies of Ti:sapphire and Cr:forsterite pulses, $f_{\rm rep}$ is the repetition frequency, and m and n are natural numbers. The second harmonic of the Ti:sapphire $(f_{\rm TisSH})$ and the third harmonic of the Cr:forsterite $(f_{\rm CrfTH})$ lasers can be written as $f_{\rm TisSH} = 2\delta_{\rm Tis} + m'f_{\rm rep}$ and $f_{\rm CrfTH} = 3\delta_{\rm Crf} + n'f_{\rm rep}$, where m' and n' are natural numbers. The observable beat frequencies $(f_{\rm beat})$ can then be expressed by $f_{\rm beat} = |(3\delta_{\rm Crf} - 2\delta_{\rm Tis}) + kf_{\rm rep}|$, where k is an integer. The beat frequency can also be written by using the cavity length difference between the two lasers. When the cavity length changes slightly during passive synchronization, the offset frequency relation changes to maintain a constant mode spacing. The phase relation is changed by changing the cavity length difference because the optical phase after one round trip depends on the cavity length, although the round-trip time of the pulse envelope is always the same. The beat frequency change $(\Delta f_{\rm beat})$ can, thus, be written using cavity length difference (Δl) as $\Delta f_{\rm beat} = 4|\Delta l|/\lambda_{\rm Tis} \times f_{\rm rep}$, where $\lambda_{\rm Tis}$ is the wavelength of the Ti:sapphire laser.

Fig. 2 illustrates the experimental setup. The dual-wavelength laser produces 75.4-MHz femtosecond pulse trains with less than 1-fs jitter [17]. Their typical pulsewidths were 50 fs. The third harmonic of the Cr:forsterite pulse was generated by using two successive BBO crystals, and the second harmonic of the Ti:sapphire pulse was obtained by using one BBO crystal. These pulses were superimposed in time and space and detected by a photomultiplier tube (Hamamatsu; H6780-06). The typical beat signal signal-to-noise ratio was 30 dB in 100-kHz resolution bandwidth (RBW). The beat signal was amplified by stages of amplifiers (Hamamatsu; C5594, Mini-Circuit; ZFL-1000LN) and passed through low-pass filters to select one-beat frequency and eliminate the repetition frequency. The beat frequency was compared with the reference frequency by a digital-phase-frequency comparator. The reference frequency was produced by dividing the repetition frequency of lasers by four. The displacement of the end mirror in the Ti:sapphire laser cavity can be changed by a piezo-electric transducer (PZT) in accordance with the error signal produced by the comparator. An electro-optical modulator (EOM) and polarized beam splitter were inserted in the pump light source to change the pump power of the Ti:sapphire laser. The error signal also drove the EOM to rapidly compensate phase error. When the pumping power in a Ti:sapphire crystal changes, the linear refractive index and the nonlinear refractive index change because they depend on the circulating pulse energy. Those changes introduce a beat shift that depends on the effective optical path length changes in the Ti:sapphire and Cr:forsterite laser cavities.

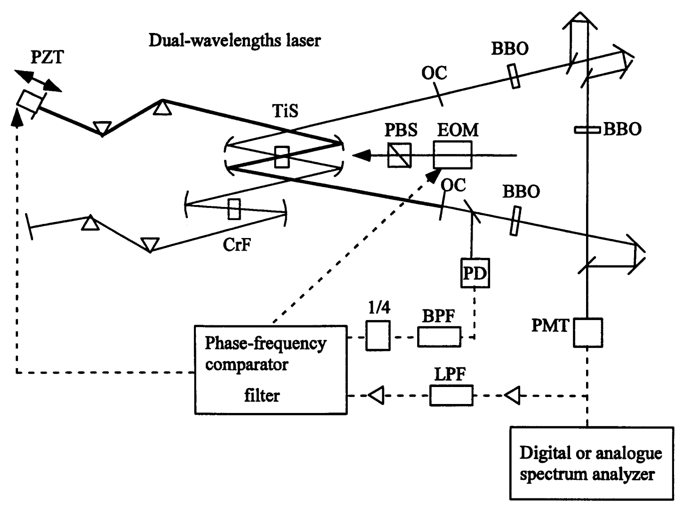

Fig. 2. Experimental setup for CEO frequency-relation control in a dual-wavelength laser. Solid lines indicate the optical paths, and dashed lines represent the electronic paths. Bold lines show the optical paths in the Ti:sapphire laser cavity. TiS, Ti:sapphire crystal; CrF, Cr:forsterite crystal; OC, output coupler; PBS, polarized beam splitter; PD, photo diode; PMT, photo-multiplier tube; LPF, low-pass filter; "1/4", frequency divider.

The dynamics of the initial fluctuation of the beat frequency is important to lock the beat signal to the reference signal. The beat frequency moves 75.4 MHz (repetition frequency) when the cavity-length difference changes to 0.21 μm (one-fourth of Ti:sapphire wavelength). We measured the dynamics of the beat

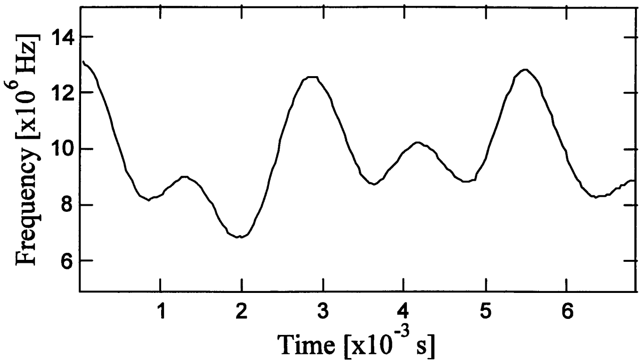

Fig. 3. Beat signal dynamics in free-running operation measured by a digital spectrum analyzer. Horizontal axis represents time, and the vertical axis, frequency. Frequency and time spans are 10 MHz and 6.8 ms. Beat frequency moves more than 5 MHz within 1 ms.

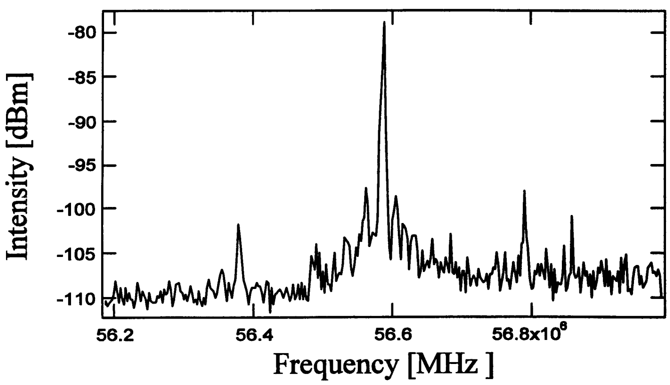

Fig. 4. Locked beat signal obtained by a spectrum analyzer with an average of ten traces. Beat frequency is locked to three-fourths of the repetition rate. Span is 1 MHz with an RBW of 3 kHz, and observed spectrum width is also the RBW.

signal using a digital spectrum analyzer (Techtronics; RSA230). Fig. 3 presents the observed beat movement. The beat dynamics was measured for 6.8 ms with a 10-MHz frequency range at a center frequency of 10 MHz. The beat frequency changed more than 5 MHz within 1 ms. The effective cavity length fluctuates for many reasons such as mechanical vibrations, air turbulence, temperature fluctuations, change of the laser pumping power, and change of circulating pulse energies. The cavity length has to be adjusted by at least a few tens of nanometers within 1 ms to lock the beat frequency. This measurement only shows the relatively slow movement of the beat signal. Faster fluctuation cannot be measured by this method. Normally, faster fluctuation has smaller amplitude. Thus, combining the PZT and the EOM would achieve appropriate locking.

Fig. 4 depicts the beat signal locked using PZT and EOM. The RBW was 3 kHz with 1-MHz span. Ten traces were averaged, and the data acquisition time was about 0.2 s. The reference frequency was $f_{\rm rep}/4 = 18.8$ MHz in this experiment. The beat frequency observed here was $f_{\rm rep} - f_{\rm rep}/4 = 3f_{\rm rep}/4$. The measured beat frequency was centered at three-fourths of the repetition frequency (56.6 MHz). The obtained spectrum width of the locked beat was 3 kHz (RBW). The sideband 200 kHz from the beat signal corresponds to the oscillation of the EOM, thus the bandwidth of the EOM feedback would be about 200 kHz. When only the PZT locked the beat signal, the locked spectrum width was about 100 kHz.

Here, we controlled the beat frequency to one-fourth of the repetition frequency. Under this condition, the CEO phase recovers every four pulses. If we select every fourth Ti:sapphire and Cr:forsterite laser pulses, we will be able to synthesize them with a constant phase relation. The controlled beat frequency is the offset frequency relation of $f_{\rm beat} = |3\delta_{\rm Crf} - 2\delta_{\rm Tis}|$ in this experiment. We previously reported measuring the CEO phase relation by using photonic-crystal fiber [16]. The white continuum produced by the Ti:sapphire laser was superimposed with the Cr:forsterite laser pulse, and the beat signal was obtained. The comb frequency of the white continuum is $f_{\rm Tis} = \delta_{\rm Tis} + mf_{\rm rep}$, and that of the Cr:forsterite laser is $f_{\rm Crf} = \delta_{\rm Crf} + nf_{\rm rep}$. The obtained beat frequency ($f'_{\rm beat}$) was $f'_{\rm beat} = |\delta_{\rm Crf} - \delta_{\rm Tis}|$ at that time. By combining these two kinds of beat measurements, we can determine both offset frequencies ($\delta_{\rm Crf}$ and $\delta_{\rm Tis}$) at the same time.

III. CEO Phase Control of the Pump, Signal, and the Idler in OPO

Femtosecond subharmonic pulses can also be obtained by using OPO. We previously reported subharmonic OPO by using a KTiOPO4 (KTP) crystal with Ti:sapphire laser pumping [14], [19]. The wavelengths were 850 (pump), 1275 (signal), and 2550 nm (idler). The signal and the idler are the subharmonic pulses of the pump. When we define the optical frequency of the pump as 3ω, the optical frequency of the signal is 2ω, and that of the idler, ω. The nonphase-mating nonlinear processes in the KTP crystal produce the second harmonic of the signal ($4\omega_1 = 2 \times 2\omega$), sum-frequency between the pump and idler ($4\omega_2 = 3\omega + \omega$), sum frequency between the pump and signal ($5\omega = 3\omega + 2\omega$), and the second harmonic of the pump ($6\omega = 2 \times 3\omega$). The phase relation of the subharmonic pulses can be easily observed by using these two 4ω pulses. Fig. 5 schematically illustrates the beat measurement in the OPO. We define the comb frequencies of the pump, signal, and idler as $f_{\rm pump} = \delta_{\rm pump} + lf_{\rm rep}$, $f_{\rm signal} = \delta_{\rm signal} + mf_{\rm rep}$, and $f_{\rm idler} = \delta_{\rm idler} + nf_{\rm rep}$, where $\delta_{\rm pump}$, $\delta_{\rm signal}$, and $\delta_{\rm idler}$ are the offset frequencies; l, m, n are natural numbers; and $f_{\rm rep}$ is the repetition frequency. The second harmonic of the signal ($f_{\rm signalSH}$) and the sum frequency between the pump and the idler ($f_{\rm pump+idler}$) can be expressed as $f_{\rm signalSH} = 2\delta_{\rm signal} + m'f_{\rm rep}$ and $f_{\rm pump+idler} = (\delta_{\rm pump} + \delta_{\rm idler}) + l'f_{\rm rep}$, where m' and l' are natural numbers. Therefore, the observable beat frequency ($f_{\rm beat}$) can be expressed as $f_{\rm beat} = f_{\rm signalSH} - f_{\rm pump+idler} = |2\delta_{\rm signal} - (\delta_{\rm pump} + \delta_{\rm idler}) + kf_{\rm rep}|$, where k is an integer. The parametric generation has the frequency relation $\delta_{\rm pump} = \delta_{\rm signal} + \delta_{\rm idler}$, thus the beat frequency is described as $f_{\rm beat} = |(3\delta_{\rm signal} - 2\delta_{\rm pump}) + kf_{\rm rep}|$. The observable beat signal in the OPO is similar to that of a dual-wavelength laser. The offset frequencies of the signal and pump correspond to those of the Cr:forsterite and Ti:sapphire lasers.

The behavior of the beat-signal change when the cavity length varies in OPO is identical to that in a dual-wavelength laser. The comb spacing of the signal and the idler are subject to that of the pump laser. When the cavity-length difference changes slightly keeping the oscillation, only the offset frequency relation changes. The change of the beat signal ($\Delta f_{\rm beat}$) with

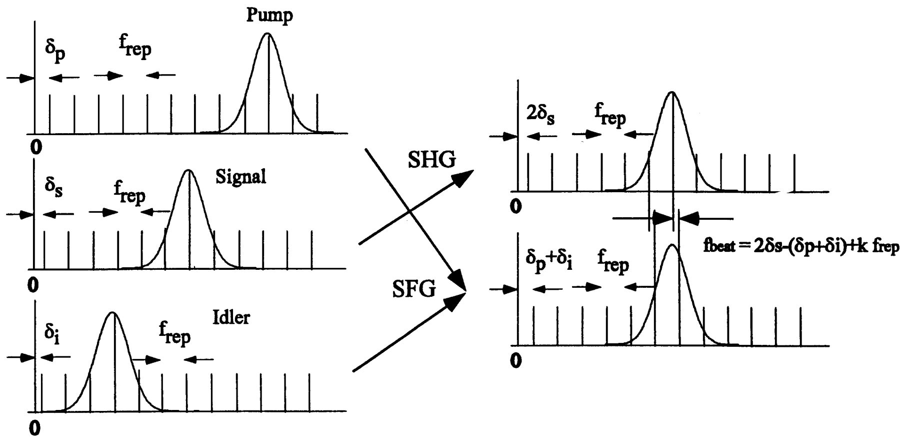

Fig. 5. Schematic of the CEO-frequency relation measurement in subharmonic OPO in the frequency domain. Optical frequencies of the doubled signal and the sum frequency between the pump and the idler overlap. Beat signal can thus be obtained by superposing them. SFG is sum-frequency generation and SHG, second-harmonic generation.

the cavity length change (Δl) can be expressed as $\Delta f_{\rm beat} = 6|\Delta l|/\lambda_{\rm signal} \times f_{\rm rep}$, where $\lambda_{\rm signal}$ is the signal wavelength. The dynamics of the initial beat fluctuation is similar to that in a dual-wavelength laser. Reducing the initial fluctuation is significant in controlling the phase relation. In [19], the controlled beat signal had a width of 500 kHz. The Ti:sapphire oscillator and OPO exceeded 1 m at that time. A larger size implies larger fluctuation of the cavity length because of the mechanical vibrations, air turbulences, and temperature fluctuations. The lower beam height in the oscillator has less fluctuation in the mirror position. Based on the above, we have made compact Ti:sapphire laser and OPO.

Fig. 6 presents the experimental setup. The Ti:sapphire laser consists of a 4-mm-thick Ti:sapphire crystal and prism pair (Lak14). The repetition frequency is 82 MHz at the center wavelength of 850 nm with a 20-nm spectrum width. The output power is 1 W with 7-W pumping (Coherent; Verdi X). A PZT is attached to one reflective mirror (10-mm diameter and 1-mm thick) to adjust the cavity length. The beam height is 25.4 mm from the base plate, and the cavity is 380×190 mm. The cavity is protected from external noise by a metal box, breadboard, rubber cushion, and optical table with air damper. The cavity-length fluctuation during free-running operation can be observed by measuring timing jitter. We measured the stability of the free-running cavity. The pulse train detected by a photo diode was measured by a vector signal analyzer (Agilent Technology; 89 441A). Fig. 7 depicts the power spectrum density and calculated pulse-timing jitter without control. The pulse-timing jitter from 10-Hz to 100-kHz integration was 3.2 ps, and that from 1-Hz to 100-kHz integration was 29 ps. No 50-Hz component was observed in this experiment, although 50 Hz and its harmonic fluctuations due to mechanical vibration or air flow are generally large because the electric power frequency is 50 Hz.

The size and beam height of the OPO are identical to those of a Ti:sapphire cavity. In addition, most parts, such as mirror holders and stages, are also the same as those of the Ti:sapphire cavity. The cavity fluctuation of the OPO should thus also be similar to that of the Ti:sapphire laser cavity. A KTP crystal is suitable for subharmonic generation with a Ti:sapphire laser pump. We selected phase matching in the x–z plane at $\theta = 71.5$ deg. The OPO cavity produced a 20-mW signal from a 1% output coupler and other subharmonic pulses output through a concave mirror. The five subharmonic pulses were almost collinear and were collimated by a lens. Two 4ω pulses were separated from other pulses by a prism and slit and fed into a delay line. The polarization of the doubled signal $(4\omega_1)$ was rotated 90° by a waveplate to align the polarizations. Combined two-4ω pulses were detected by an Si-avalanche photodiode (APD: Hamamatsu S5343). Low-pass filters and amplifiers blocked the high-frequency component and selected one beat frequency. The beat signal was compared with the refer-

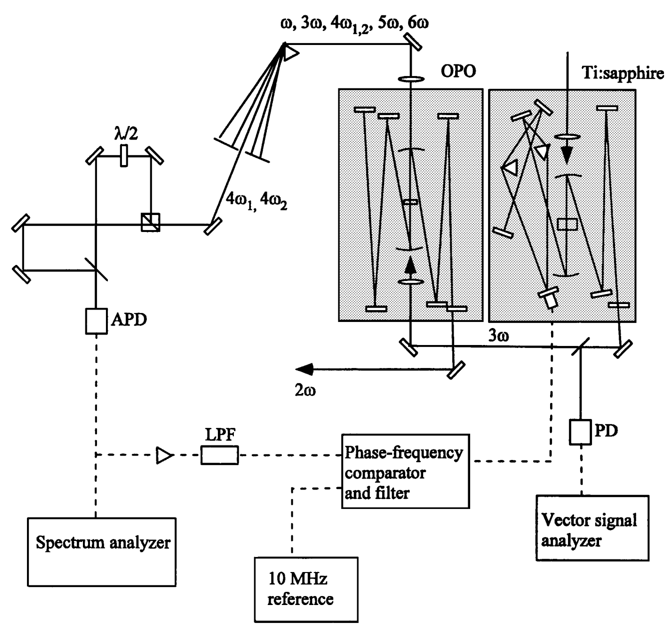

Fig. 6. Experimental setup for CEO frequency-relation control in the subharmonic OPO. Ti:sapphire laser produces 1-W output for an OPO pump with 82-MHz repetition frequency. Initial cavity fluctuation of a Ti:sapphire laser was measured by a vector signal analyzer. OPO produces not only the signal and the idler, but also sum frequencies and second harmonics of subharmonic pulses. Two -4ω pulses are combined after polarization alignment and detected by an APD. Beat signal is compared with the reference signal and the error signal drives the PZT in the Ti:sapphire laser cavity.

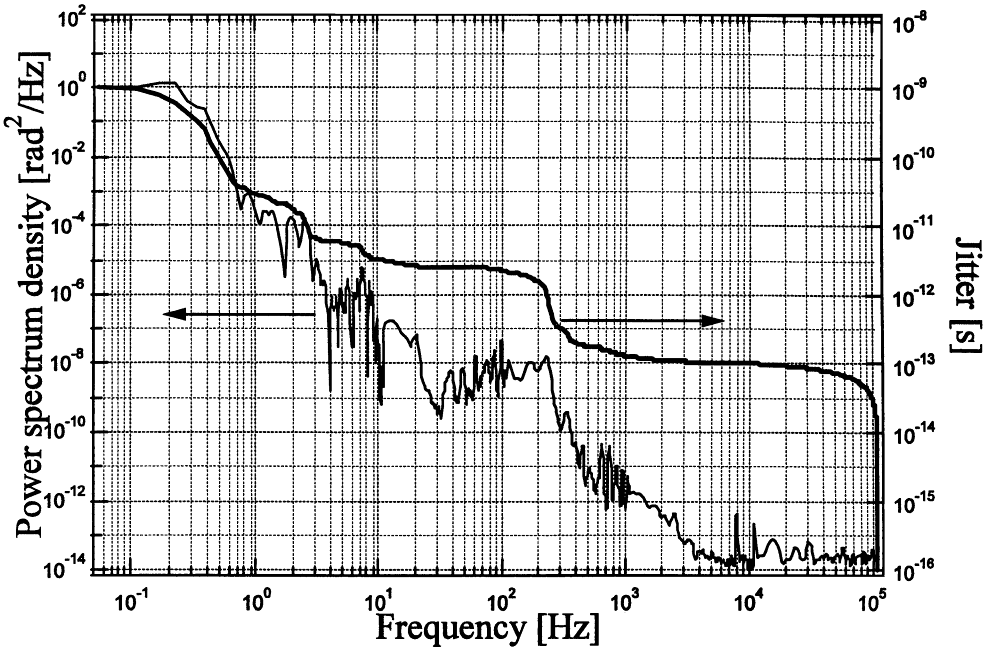

Fig. 7. Power spectrum density and calculated timing jitter of the Ti:sapphire laser without control.

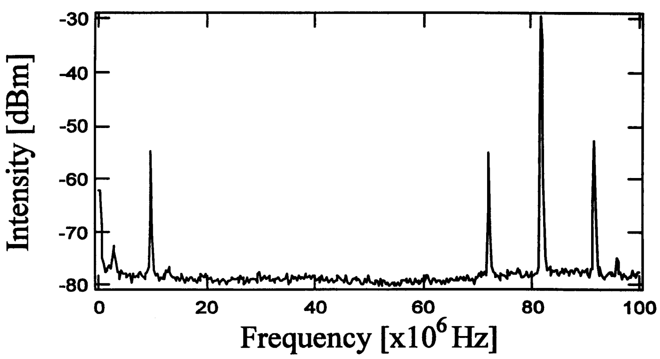

Fig. 8. Locked beat spectrum measured by a spectrum analyzer in 100-MHz span. Over 100 traces are averaged. RBW is 100 kHz and locked spectrum width is also RBW.

ence signal by a digital-phase frequency comparator. The error signal drove the PZT in the Ti:sapphire cavity.

Fig. 8 illustrates the beat signal locked to 10 MHz obtained by an analog spectrum analyzer (Agilent Technology; 4401B). The RBW is 100 kHz and span is 100 MHz. The peak at 82 MHz is the repetition frequency, and the beat signal is locked to 10 MHz. Beat signals at $82-10=72$ MHz and $82+10=92$ MHz were also observed. Over 100 traces were averaged. The spectrum width of the locked beat signal was RBW. The signal-to-noise ratio of the beat signal did not change when averaged. The locked beat line width and signal-to-noise ratio in this experiment were much better than those obtained in [19], although the size of the mirror attached to the PZT in order to adjust the cavity length in this experiment is larger. (The mirror in the previous experiment was 5 mm square and 0.3 mm thick.) This improvement results from less initial fluctuation of cavities and similar cavity shapes. The beat signal was locked to 10 MHz here, but it is easy to lock it to a subharmonic frequency of the repetition rate. When the beat frequency is locked to one-fourth of the repetition frequency, the CEO phase relation is recovered every four pulses. This is identical to CEO frequency control in a dual-wavelength laser.

IV. Comparison of Two Methods

We controlled the CEO phase relations of a dual-wavelength laser and an OPO. The controlled offset-frequency relation in a dual-wavelength laser is $|3\delta_{\rm Crf} - 2\delta_{\rm Tis}|$, and that in OPO is $|3\delta_{\rm signal} - 2\delta_{\rm pump}|$. The center wavelengths of the controlled pulses are almost the same in the experiments. The output powers of subharmonic pulses in the dual-wavelength laser (830 and 1245 nm) are higher than that of the OPO, and their pulsewidths can be reduced relative to those of an OPO. Basically, pulse durations of 10 fs in a Ti:sapphire laser and 20 fs in a Cr:forsterite laser can be achieved. For higher power and shorter pulse duration, a dual-wavelength laser would be appropriate for a phase-coherent subharmonic pulse generator. It is difficult to reduce the pulse duration of the pump in a femtosecond OPO because a high spectrum density is desired for OPO pumping. However, it would be feasible to generate a shorter pulse duration of signal and idler [20]. Higher signal and idler outputs would also be feasible in an OPO, although the output of the signal was 20 mW in this experiment. The main advantage of the OPO is simultaneous multisubharmonic pulse generation. Our OPO generates six subharmonic pulses at the same time, and no frequency conversion is necessary to observe the CEO phase relation. Furthermore, we can select wide signal and idler wavelength ranges. Fourier synthesis in any wavelength region would be feasible by using OPO. Next, we compare the stability of the two schemes. The dual-wavelength laser output timing synchronized Ti:sapphire laser and Cr:forsterite laser pulses for an entire day without any cavity control. The stability of the passive timing synchronization depends on the pulsewidths. In contrast, the oscillation of OPO can be maintained for only a few hours without feedback. The long-term stability of both passive synchronizations depends on the slow drift of the cavity length differences. Normally, it depends on the temperature fluctuation of the room. The tolerance of the cavity-length fluctuation of the dual-wavelength laser exceeds 3 μm, and that of the OPO is only 1 μm. The long-term stability of the uncontrolled cavities could be increased by temperature control. The short-term stability of the cavity length fluctuation can be estimated by the beat frequency change. In Fig. 3, the beat frequency fluctuation of the dual-wavelength laser in a few milliseconds was 10 MHz, which corresponds to the cavity length fluctuation of about 30 nm. In the case of the OPO, the cavity length fluctuation in the same time scale (milliseconds) was almost the same (20–30 nm), although long-term (hours) drift of it exceeds 1 μm. Their stabilities increased when the CEO frequency was controlled. Beat locking in both systems can be maintained for over one hour, and the OPO continues oscillating much longer. We cannot determine which is better with regard to a CEO frequency-controlled light source. In both systems, if the pulsewidths were about 10 fs, the coherently superimposed lights field would be single subfemtosecond pulse with about 80-MHz repetition rate, and they will be more ideal light sources to investigate subfemtosecond time resolution spectroscopy.

V. Conclusion

We controlled the CEO frequency relation among femtosecond subharmonic pulses in two schemes. In one scheme, we controlled the relation using a dual-wavelength laser consisting of Ti:sapphire and Cr:forsterite lasers. We were able to control the offset frequency relation of $|3\delta_{\rm Crf} - 2\delta_{\rm Tis}|$ and obtain a 3-kHz locked beat signal. In the other scheme, we controlled the CEO frequency relation in a femtosecond OPO. Here, we were able to control the offset frequency relation of $|3\delta_{\rm signal} - 2\delta_{\rm pump}|$ and obtain a 100-kHz locked beat signal. These CEO-frequency-controlled subharmonic light sources will facilitate achieving Fourier synthesizing of multicolor femtosecond pulses such as subfemtosecond pulse-train generation or CEO-phase-sensitive applications.

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Yohei Kobayashi was born in Hokkaido prefecture, Japan, on May 28, 1970. He received the B.S., M.S., and the Ph.D. degrees in applied physics from University of Tokyo, Japan, in 1993, 1995, and 1998, respectively.

Since he joined Electrotechnical Laboratory in 1998, he has been working on the research of mode-locked lasers, optical parametric oscillators, and their applications. His current research interests include an optical frequency control of femtosecond lasers and an optical Fourier synthesis. In 2001, the laboratories were reorganized as National Institute of Advanced Industrial Science and Technology (AIST).

Dr. Kobayashi is a member of the Optical Society of America and the Japan Society of Applied Physics.

Zhiyi Wei received the Ph.D. degree in optics from the Xian Institute of Optics and Fine Mechanics, China, in 1991.

From 1991 to 1993, he continued his study at the Ultrafast Laser Spectroscopy Laboratory at the Zhongshan University, China, as a Post-Doctoral Fellow. During 1993 to 1997, he worked on ultrafast spectroscopy and technology of femtosecond laser pulses at the Rutherford Appleton Lab, U.K., the Chinese University of Hong Kong, the Hong Kong University of Science and Technology, and the University of Groningen, The Netherlands. In 1997, he moved to the Institute of Physics, Beijing, China, where he conducted research on generation and amplification of femtosecond laser pulses. During 2000 to 2002, he worked at the Institute of Photonics, Tsukuba, Japan, on a NEDO fellowship. He was promoted to Professor at the Institute of Physics in 1999. His current research interests include the development and application of multi-TW femtosecond laser, generation of attosecond pulse, and optical frequency metrology.

Masayuki Kakehata was born in Iwate prefecture, Japan, on January 9, 1966. He received the B.S., M.S., and the Ph.D. degrees in electrical engineering from Keio University, Yokohama, Japan, in 1988, 1990, and 1993, respectively.

He joined Electrotechnical Laboratory in 1993, where he worked on development of high-intensity Ti:sapphire laser systems and on field ionization of atoms and high-order harmonic generation and polarization control of high- intensity laser pulses. In 2001, the laboratories were reorganized as National Institute of Advanced Industrial Science and Technology (AIST), Japan. His current research interests include generation of high-intensity ultrashort laser pulses, control of carrier-envelope phase, and their applications to nonlinear optics.

Dr. Kakehata is a member of the Optical Society of America, the Japan Society of Applied Physics, and the Laser Society of Japan.

Hideyuki Takada was born in Tokyo, Japan, on March 22, 1966. He received the B.S. degree in electrical engineering and the M.S. degree in electrical and communication engineering from Tohoku University, Japan, in 1989 and 1991, respectively.

Since he joined the Electrotechnical Laboratory in 1991, he has been working on the research of mode-locked lasers and tunable solid-state lasers. Now, he is a Senior Research Scientist of the ultrafast laser group in National Institute of Advanced Industrial Science and Technology (AIST), Japan. His research interests include ultrashort pulse lasers and their applications.

Mr. Takada is a member of the Japan Society of Applied Physics.

Kenji Torizuka was born in Saitama prefecture, Japan, on July 5, 1957. He received the M.S. and Ph.D. degrees in physics from University of Tokyo, Japan, in 1982 and 1986, respectively.

Since he joined the Electrotechnical Laboratory in 1985, he has been working on the research of mode-locked lasers, tunable solid-state lasers, nonlinear optical materials, and ultrafast phenomena. Now, he is a Group Leader of the ultrafast laser group at the National Institute of Advanced Industrial Science and Technology (AIST), Japan. His research interests include ultrashort pulse lasers and their applications.

Dr. Torizuka is a member of the Japan Society of Applied Physics and the Optical Society of America.