Generation of two-color femtosecond pulses by self-synchronizing Ti:sapphire and Cr:forsterite lasers

Zhiyi Wei*

New Energy and Industrial Technology Development Organization, 3-1-1 Higashi-Ikebukuro, Tokyo 170-6027, Japan

Yohei Kobayashi, Zhigang Zhang,[†] and Kenji Torizuka

National Institute of Advanced Industrial Science and Technology, 1-1-1 Umezono, Tsukuba, Ibaraki 305-8568, Japan

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We report a novel technique for the synchronization of two different femtosecond solid-state lasers by crossing of both laser pulses in a Kerr medium. Stable dual-wavelength femtosecond pulses at central wavelengths of 820 and 1250 nm have been obtained. The tolerance of cavity-length mismatch is ~0.6 μ m, where the pulse widths of the Ti:sapphire and the Cr:forsterite lasers are 18 and 40 fs, respectively, at average powers of 600 and 110 mW. The typical timing jitter derived from the cross correlation is less than 3 fs. © 2001 Optical Society of America

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The invention of the Kerr-lens mode-locked (KLM) femtosecond Ti:sapphire laser has revolutionized research and development in ultrafast science.¹ This technique has subsequently been successfully extended to many solid-state lasers, such as Cr:forsterite (Ref. 2) and Cr:YAG.³ These femtosecond lasers with different wavelengths supply spectroscopy researchers a choice of instruments for various purposes. Achievement of synchronization among lasers at different wavelengths has been an important objective. Although a timing-lock system with electrical feedback enables us to synchronize two different lasers,⁴⁻⁶ the problems of relatively large timing jitter and random relative time delay remain.

A self-synchronized two-color Ti:sapphire laser with a single laser crystal was developed during the past several years.⁷⁻¹¹ Unfortunately, the two trains of laser pulses in the schemes mentioned above can be operated only at similar wavelengths, because they share the same gain spectrum, and this limits the tuning range and further applications in spectroscopy.

In this Letter we propose and demonstrate a novel scheme for achieving a two-color femtosecond laser system based on two different gain media. This system consists of two independent lasers, one Ti:sapphire and the other Cr:forsterite. We obtain stable synchronous laser pulses with central wavelengths near 820 and 1250 nm by crossing both laser beams in one Kerr medium. We believe that this is the first report of synchronizing two kinds of ultrashort pulse lasers with different gain media. With our approach we achieve synchronization between pulses with a much wider frequency difference than with the conventional scheme. This result will open new applications, such as subfemtosecond pulse-train generation by Fourier synthesis,¹² for a two-color laser.

Our proposal for self-synchronizing of two mode-locking lasers was motivated by considerations in previous reports of the empirical properties of a single Ti:sapphire crystal. In those two-color mode-locked lasers, two laser beams have to overlap in the gain medium for synchronization to occur. However, if the overlap volume is too large, gain competition can make the laser unstable. Therefore the two beams should be partially separated in the gain region for stable synchronization. Thus we have arrived at two conclusions: First, a perfectly collinear overlap of the two beams is not necessary for synchronization. Therefore the experiment will be greatly simplified because ultrabroadband mirrors will not be necessary for construction of a coupled laser cavity with two types of gain media. Second, we could potentially synchronize completely off-resonant (i.e., long-wavelength) light in a gain material. The synchronization is usually explained as a being due to a fast-response third-order optical nonlinearity (i.e., an electronic Kerr effect or cross-phase modulation). The nonlinear coupling coefficient for the off-resonant light would be much smaller than that for lights within a gain band. But we would have a chance to enhance it by close spatial coupling of the two beams. Although these are only qualitative considerations, they provided a guideline for the following experiment.

A schematic of the experimental layout is shown in Fig. 1. A standard KLM Ti:sapphire laser¹³ and a modified Cr:forsterite laser were used. The Ti:sapphire rod was 4 mm long, and the length of the Cr:forsterite rod was 7 mm. Both were Brewster cut. The temperatures of the crystal holders were kept at 11 and 1°C, respectively. We purged the Cr:forsterite with flowing nitrogen to keep the surface dry. To introduce an overlap for beam coupling in the Ti:sapphire crystal, we inserted an additional pair of concave mirrors (M3 and M4) into the ordinary Cr:forsterite laser cavity. To ensure that the two beams were close enough to interact inside the gain medium, we half-cut the concave mirrors (M1–M4) about the Ti:sapphire crystal and set them parallel less than 1 mm apart by using independent mirror mounts. Both cavity lengths were set at 1.95 m, and the shorter arms from the Ti:sapphire rod were adjusted to be as much alike as possible. A pair of



Fig. 1. Schematic of the two-color Ti:sapphire and Cr:forsterite lasers. M1–M7 (CVI), mirrors with radii of curvature of 10 cm. Two pairs (M1 and M2, M3 and M4) were half-cut from two 12.7-mm round mirrors. M8, mini mirror of \emptyset 10 × 1 mm size. Mirrors M1, M2, and M8 are coated at 850 nm; M3–M7 are coated at 1300 nm. P1, P2, fused-silica prisms; P3, P4, SF6 prisms; PDs, fast photodiodes.

fused-silica prisms (P1 and P2) and a pair of SF6 prisms (P3 and P4) were used to compensate for the dispersion inside the respective cavities. We mounted end mirror M8 upon a piezoelectric transducer (PZT) to tune the cavity length in the Ti:sapphire laser and used a broadband semiconductor saturable-absorber mirror¹⁴ (SESAM) for self-starting of Kerr-lens mode locking in the Cr:forsterite laser.

The pump sources were an all-line argon laser and a diode-pumped 1.06-µm Nd:YVO₄ laser (Spectra-Physics T-40Z-106C). With the pump power of a 4-W argon laser and a 9-W 1.06- μ m laser, we obtained stable KLM powers of 600 and 110 mW from the Ti:sapphire and the Cr:forsterite lasers, respectively. We used two frequency counters (Hewlett-Packard HP 53132A) to monitor the repetition rates of both KLM lasers, which were tuned to 78.8487 MHz. Covering the lasers with a closed box to prevent air turbulence and acoustic noise reduced the repetition-rate drift of both lasers to less than ± 1 Hz within 1 min. To verify the synchronization between the two pulses, we also used an oscilloscope (Hewlett-Packard HP 54616B) to observe the pulse trains of both lasers; triggering in one pulse train allowed the other pulse train to be viewed. The other pulse train could not be clearly displayed on the oscilloscope before successful synchronization. Even if the cavity lengths were tuned to the same wavelength, slips of the trace were always observed, possibly as a result of existing environmental perturbations.

The pulse trains were quickly locked after careful alignment of the beam overlap in the Ti:sapphire crystal, and a stable trace was shown on the oscilloscope when we tuned the repetition rates to be nearly equal by varying the voltage on the PZT. Then both pulse trains were locked, and the repetition rate was maintained with an accuracy of ~ 1 Hz, even if we tuned the PZT continuously, until driving the PZT further collapsed the synchronization and the two pulse traces were independent again. Once beam overlap was achieved and two cavity lengths were properly matched by tuning of the PZT, the lasers could be kept synchronized for more than 10 min.

Figure 2 shows the behavior of the repetition rate of the Ti:sapphire laser $(f_{\rm RT})$ and its difference $(\Delta f_{\rm RT})$ from the repetition rate of the Cr:forsterite laser. The horizontal scales are the displacements of the end mirror. By either shortening [Fig. 2(a)] or lengthening [Fig. 2(b)] the Ti:sapphire cavity length, we found that synchronization began only when the difference in the repetition rates was ~ 5 Hz and exhibited similar behavior in both directions, which reveals that the Cr:forsterite laser was forced to match the repetition rate of the Ti:sapphire laser and further that slaving of the repetition rate is determined by the Ti:sapphire laser's cavity length. The relation of master-slaving seems reasonable in view of the higher intensity of the Ti:sapphire laser than of the Cr:forsterite. Beyond the tolerance of the cavity-length mismatch, which was found to be $\sim 0.6 \ \mu m$, the locking of the pulse trains was lost and the frequency difference was beyond 15 Hz. In addition, we noticed that the central wavelength of the Ti:sapphire (Cr:forsterite) laser would shift to the red (the blue) as its cavity length is shortened in the synchronized regime. Lengthening the cavity would shift the wavelengths in the opposite direction. Both the maximum wavelength shifts of the Ti:sapphire and of the Cr:forsterite lasers were less than 5 nm.

No observable changes were found in either mode-locking power or duration relative to those in the unlocked mode. Although it is possible for the Ti:sapphire laser to produce pulses shorter than 11 fs when the prism's location is changed, our initial experiment showed that typical pulse durations (sech²) of 18 and 40 fs for the Ti:sapphire and the Cr:forsterite lasers, respectively, ensured better synchronization. Figure 3 shows an interferometric autocorrelation trace of each laser at this synchronized mode. The spectra were centered about 820 and 1250 nm, with bandwidths of approximately 42 and 43 nm. The time-bandwidth products of the two were 0.34 and 0.33, respectively, close to the transform limit in either case.

To measure the cross correlation and estimate the timing jitter, we interchanged the output coupler and mirror M8 of the Ti:sapphire laser and placed an orange glass filter behind the coupler to block the remaining pump laser. We also ignored extracavity



Fig. 2. Difference in repetition rates ($\Delta f_{\rm RT}$, filled circles) of Ti:sapphire and Cr:forsterite lasers versus mirror displacement from (a) decreasing and (b) increasing cavity lengths of the Ti:sapphire laser. The synchronization occurs in the range where $\Delta f_{\rm RT}$ remains 0. The repetition rate of the Ti:sapphire laser ($f_{\rm RT}$, open squares) is also plotted for reference. We subtracted 76 848 700 Hz from the exact repetition rate to easily scale the ordinate.



Fig. 3. Fringe-resolved autocorrelation traces of (a) Ti:sapphire and (b) Cr:forsterite lasers.



Fig. 4. (a) Cross-correlation trace and (b) intensity fluctuation of the SFG between two pulse trains. The cross-correlation FWHM is 74 ± 2 fs; the timing jitter is <3 fs.

dispersion compensation for both laser pulses, to simplify the cross-correlation configuration. After these changes, the pulse widths of the Ti:sapphire and Cr:forsterite lasers became 48 ± 2 and 58 ± 1 fs, respectively, with the assumption of a Gaussian shape. Then we crossed both beams in a β -barium borate crystal after the Ti:sapphire laser pulses had passed through a delay line. Two alternate sum-frequency signals (SFGs) at a central wavelength of $\sim 495 \text{ nm}$ were observed when we adjusted the delay line, because we could make the synchronization jump from one locking regime to another by tapping the table. We then overlapped both regimes by precisely adjusting the length of the short arm in the Ti:sapphire laser. Figure 4(a) shows a typical trace of a SFG when the delay was swept. The FWHM of the cross-correlation trace was 74 ± 2 fs. Within experimental accuracy, this value coincides exactly with the theoretical estimate of 75 ± 2 fs based on the pulse widths of the Ti:sapphire and Cr:forsterite lasers. In addition, measuring the intensity fluctuations of the SFG at a fixed delay also enables us to know the rms timing noise. Figure 4(b) is the time record of the SFG at 1-kHz bandwidth over 5 s, resulting in a timing jitter of 2.8 fs.

We believe that the observed synchronization is dependent on the nonlinear coupling between the two beams and their propagation effect. It has been pointed out that the combined effects of cross-phase modulation and group-delay dispersion are responsible for synchronization of the two-color Ti:sapphire laser.¹⁵ In our experiments the synchronization tolerance depends on the spatial overlap of the two beams, the pulse energies, and the pulse widths. On decreasing the overlap by gradually moving the Ti:sapphire crystal, we observed a reduction in tolerance from 0.6 μ m to 0, after which no synchronization occurred. In addition, we found that synchronization was impossible when the Kerr-lens mode locking of the Cr:forsterite laser became pure semiconductor saturable-absorber mirror mode locking, in which the pulses were running at picosecond duration.

In conclusion, we have demonstrated, for the first time to our knowledge, self-synchronization of two independent mode-locked lasers with different gain media. Stable two-color laser pulses were obtained at central wavelengths of 820 and 1250 nm.

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*Permanent address, Laboratory of Optical Physics, Institute of Physics, Beijing 100080, China.

[†]Present address, Ultrafast Laser Laboratory, University of Tianjin, Tianjin 300072, China.

Note added in proof: By optimizing the overlap between the Ti:sapphire and the Cr:forsterite lasers, we achieved a cavity-length mismatch tolerance of more than 1 μ m. The synchronization was maintained continuously for many hours.

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