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A Tunable Ultrafast Source by Sum-Frequency Generation between Two Actively Synchronized Ultrafast Lasers *

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We demonstrate an experimental setup of a tunable ultrafast laser source by sum-frequency generation (SFG) between a mode-locking Ti:sapphire laser and a Nd:YVO_4 laser. The generated wavelength by SFG is tunable from 450 nm to 480 nm with timing jitter no more than 1 ps. The average output power is over 20 mW and the maximum is about 30 mW at 457 nm. This ultrafast laser is a simple and easy tuning source applied to some pump-probe spectroscopy and ultrafast dynamics experiments.

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Ultrafast laser sources at wavelength between 450 nm and 500 nm have been applied to pump-probe experiments of biology and chemistry such as ultrafast pump-probe imaging spectroscopy of β-carotene,[1−3] coherent anti-Raman scattering[4] and ultrafast dynamics of excited-state evolution in some materials.[5] On account of the limited gain media that could not cover all wavelengths of ultrafast lasers in both femtosecond and picosecond, it is an interesting research topic to generate them in an indirect way such as optical parametric amplification and oscillation (OPO), second harmonics generation (SHG), and sum-frequency generation (SFG). The tunable range is wide for OPO and OPA, but the efficiency and stability, which are the advantage of SHG, did not show a good performance. However, the SHG is restricted by the fundamental ultrafast sources (900–1000 nm) to generate lasers from 450 nm to 500 nm.

In the past decade, we have developed techniques of passively and actively synchronizing between femtosecond or picosecond lasers.[6−9] A passively synchronized femtosecond laser with sub-femtosecond timing jitter is obtained based on the cross-phase modulation in a Kerr medium between a Ti:sapphire and a Cr: forsterite laser or two Ti:sapphire lasers,[6,7] which could be applied to SFG experiments. On the other hand, SFG between an actively synchronized femtosecond Ti:sapphire laser and a Nd:YVO_4 laser is realized by using a precise phase-locked loop (PLL), indeed showing a possible shortcut to extend ultrafast laser wavelengths by SFG.

Although the timing jitter of passively synchronized lasers have reached attosecond time scale,[6,7] the passive synchronization needs an accurate adjustment of the two laser cavities and its mismatch tolerance is no more than 10 µm. Furthermore, in the experimental setup of SFG mentioned above, the difference of time duration between two lasers (femtoseconds and picoseconds) is up to three orders, leading to a difficulty of active synchronization by the PLL. Particularly, it indulges with a good solution of precise electronics. In addition, the tunable range of the above lasers is narrow and limited by the two branches of the synchronized lasers.

In this Letter, we demonstrate a tunable SFG experiment between two actively synchronized picosecond lasers. The experimental setup is shown in Fig. 1. One of the two synchronized independent lasers is a commercial tunable Ti:sapphire laser (Tsunami, Spectra Physics Inc.), running at 800 nm; the other one is our home-made Nd:YVO_4 picosecond laser, running at 1064 nm. The time duration of the Tsunami and Nd:YVO_4 laser is ∼60 ps and 10 ps, respectively. The repetition rates of both are 80 MHz.

The phase locking method, which is called the "active synchronization", follows the same scheme as carried out before.[9] One cavity mirror of the Nd:YVO_4 oscillator mounted on a piece of piezoelectric transducer (PZT) plays a major role of the synchronization, which is depicted in Fig. 1. To detect the phase information, two photodiodes obtain the repetition rates from the laser signal reflected onto them by two pieces of glass. The repetition rate of Tsunami and the Nd:YVO_4 oscillator is the reference signal (RF) and the local oscillator (LO) of the mixer respectively; they are filtered by low-pass filters (100 MHz) and mixed in the fundamental frequency 80 MHz. The error signal from the intermediate frequency (IF) of the mixer is the controlling signal to the proportional-integration (PI) circuit of the phase-locked loop. Because the PZT changes its dimension by the high-voltage on account of the piezoelectric effect, it is

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necessary to use a high-voltage amplifier to amplify the controlling signal to drive the PZT for synchronization. What is more, a delay line adjusts the overlapping and a periscope makes the polarization of the Tsunami and Nd:YVO_4 laser to be the same for the phase matching condition of the β-BaB_2O_4 (BBO) crystal located after a mirror M4 (HT@800 nm, HR@1064 nm). A convex lens (L1, f = 50 mm) focuses two laser beams into the BBO crystal. The BBO crystal is $8 \times 8 \times 14\,\mathrm{mm}^3$ in dimensions with cut-angle $\theta = 25.7°$.

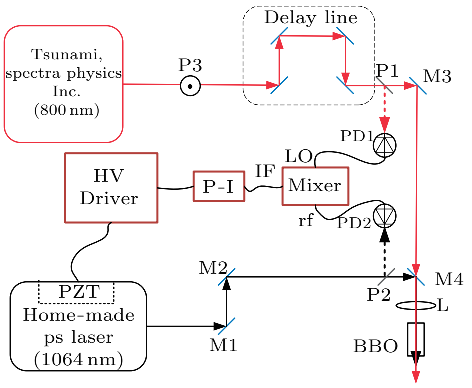

Fig. 1. SFG experimental setup. M1,M2: 45° reflecting mirrors, HR@1064 nm; M3: 45° reflecting mirror, HR@800 nm; M4: dichroic mirror, HR@1064 nm, HT@800 nm; P1, P2: reflecting pieces of glass; P3: periscope; PZT: piezo transducer; HV Driver: high-voltage driver.

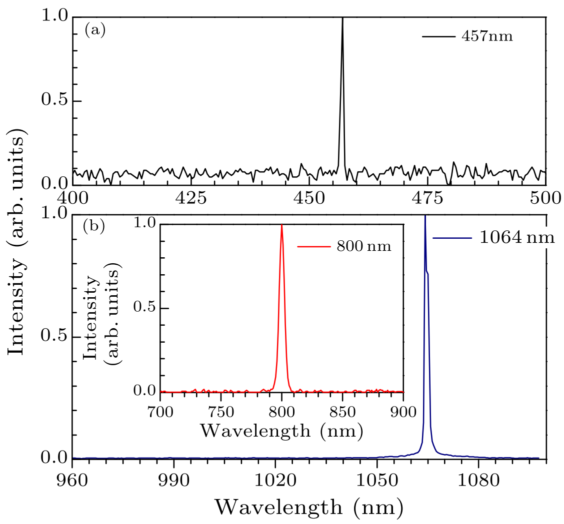

Fig. 2. (a) Spectrum of SFG signal wavelength at 457 nm, corresponding to the pump at 800 nm and the idler at 1064 nm. (b) Spectrum of the pump and idler wavelengths (800 nm, 1064 nm) for generating 457 nm.

Experimentally, the Tsunami laser first runs at 800 nm and the Nd:YVO_4 laser runs at 1064 nm. After the PLL closed and tuning the delay line carefully, two laser pulses arrive at the BBO crystal at the same time and sum-frequency generation occurs as shown in Fig. 2(a). The SFG signal wavelength is at 457 nm (Fig. 2(b)), corresponding to the calculation result applying pump (800 nm) and idler (1064 nm) wavelengths to the formula

$$\frac{1}{\lambda_p} + \frac{1}{\lambda_i} = \frac{1}{\lambda_s}.$$

As the Tsunami laser is tunable in a wide range (700–1000 nm), we tune the pump wavelength away from 800 nm, during which the loop is still closed. Rotating the angle of BBO crystal adapts to different pump wavelengths for phase matching. During this process, wavelengths of SFG are also obtained in a wide tunable range from 455 nm to 479 nm. In Fig. 3(a), we depict this range in some wavelengths: 455 nm, 463 nm, 467 nm, 469 nm, 473 nm, 476 nm, and 479 nm. The corresponding pump wavelengths are 794 nm, 821 nm, 833 nm, 840 nm, 851 nm, 863 nm, and 870 nm, as shown in Fig. 3(b). Nevertheless, the commercial Tsunami laser is tunable from 700 nm to 1000 nm. Moreover, the calculated difference of phase matching angles of the BBO crystal for pump wavelengths between 700 nm and 1000 nm is only 3.8°, that is to say, SFG could occur in Tsunami's tunable range (700–1000 nm) in the BBO crystal by rotating the phase matching angle experimentally. Thus, the SFG signal wavelength could cover from 422 nm to 515 nm, corresponding to the pump wavelength from 700 nm to 1000 nm.

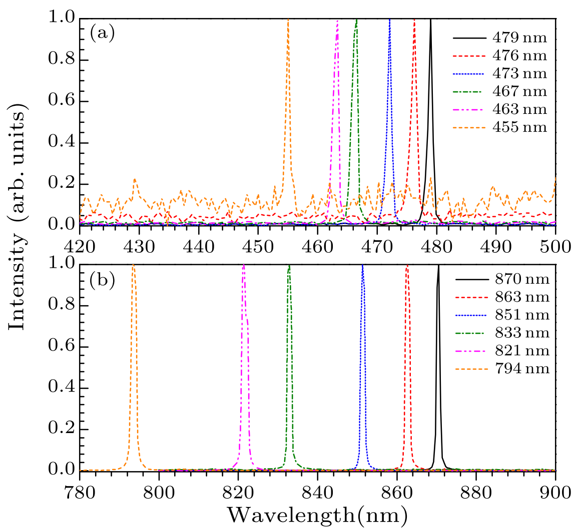

Fig. 3. (a) Experimentally tunable wavelengths of SFG from 455 nm to 479 nm while tuning the Tsunami laser. (b) Tunable pump wavelengths of the Tsunami laser from 794 nm to 870 nm.

The signal lasers of SFG after the loop closed is more stable and brighter than that before the loop closed. This is due to the active synchronization by the PLL. Figures 4(a) and 4(b) show the difference of SFG laser trace before and after the loop closed acquired by an oscilloscope (Tektronix, TDS 3052B). Clearly, at the same trigger level of the oscilloscope, the laser trace is much better with nearly no shaking shown on the oscilloscope when the loop is closed. Specifically, tuning the delay line, we obtain the typical cross-correlation by SFG as shown in Fig. 5. It is obvious that the FWHM of the cross correlation is ∼20 ps according to the Gaussian fit (in red) of the measured data. The standard deviation of the intensity fluctuation of SFG at a fixed delay is less than 0.1, indicating that the RMS timing jitter between the pump and idler lasers is less than 1 ps.

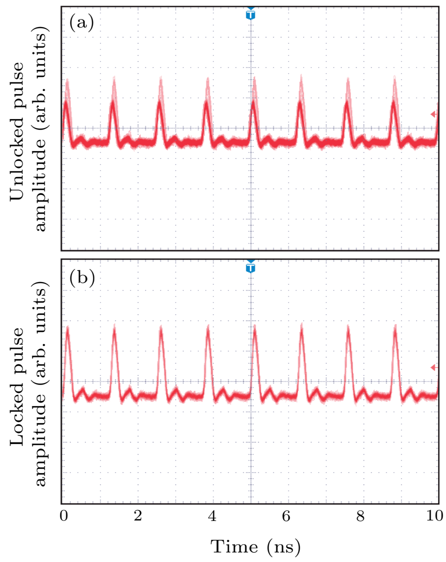

Fig. 4. (a) Unstable SFG laser trace before the PLL closed. The laser pulses fluctuate at the peak. (b) Stable SFG laser trace after the PLL closed.The laser pulses is stable and with no fluctuation.

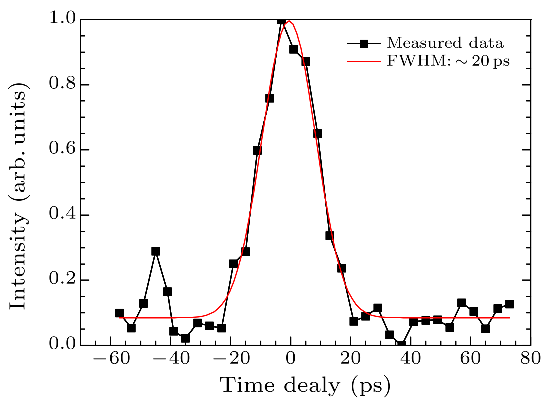

Fig. 5. Typical cross-correlation measurement and the Gaussian fit between the pump and idler lasers.

We measured the average power at different wavelengths as shown in Fig. 6, referring to the SFG wavelengths in Fig. 4(a), while the power of the pump (Tsunami) and idler ($Nd:YVO_4$) is at the same value of 1 W, respectively. From Fig. 6, the output power of SFG is almost above 20 mW and the maximum is at 457 nm with the power of 28 mW. These results are better than those we have performed before on account of the synchronized lasers in picoseconds time duration, of which the maximum is about three times larger than that in Ref. [9].

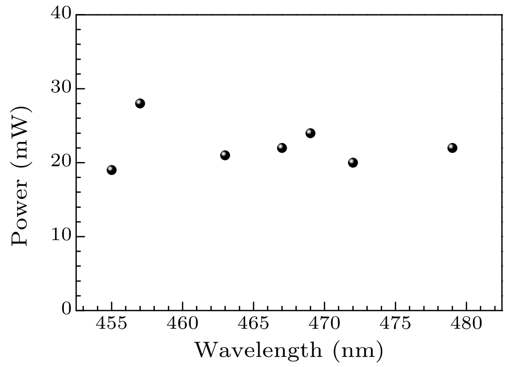

Fig. 6. Average output powers at tunable different SFG signal wavelengths from 455 nm to 479 nm.

In conclusion, we have demonstrated a tunable ultrafast source by sum-frequency generation between two actively synchronized independent lasers with an uncomplicated phase-locked loop solution for pump-probe experiments. The wavelength is widely tunable from 455 nm to 479 nm and has the potential to be tuned in a larger range from 422 nm to 515 nm. The synchronization is conveniently achieved and stable with timing jitter less than 1 ps while the cross correlation of the two lasers is ∼20 ps. The typical output powers of the SFG are higher than 20 mW, with the maximum 28 mW at 457 nm, which is much higher than that which we reported before. This tunable ultrafast laser is a simple and stable source for pump-probe imaging spectroscopy and coherent anti-Raman scattering imaging, etc.

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