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# Fast, long-scan-range pump-probe measurement based on asynchronous sampling using a dual-wavelength mode-locked fiber laser

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**Abstract:** A simple, fast and long-scan-range pump-probe scheme is experimentally demonstrated using a dual-wavelength passively mode-locked fiber laser. The pulse trains from the dual-wavelength laser have a small difference in their repetition frequencies inherently determined by the intracavity dispersion. This enables the realization of the asynchronous sampling scheme with a tens-of-nanosecond-long delay range and a picosecond scan step at a millisecond scan speed. Instead of two synchronized ultrafast lasers in the traditional asynchronous sampling scheme, just one fiber laser is needed in our scheme, which could significantly simplify the system setup.

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**OCIS codes:** (140.4050) Mode-locked lasers; (300.6500) Spectroscopy, time-resolved; (320.7120) Ultrafast phenomena; (320.7090) Ultrafast lasers.

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## 1. Introduction

Optical pump-probe spectroscopy techniques have become a powerful tool to investigate the fast transient dynamics of numerous physical phenomena [1]. By using a strong pump pulse and a weaker probe pulse and measuring the changes in the probe at different delays between the two pulses, the transient changes in the optical properties induced by the pump could be recorded as a function of time with a very high temporal resolution. The scanning scheme that generates the varying time delay between the pump and probe, thus, plays a critical role in the measurement speed, temporal range, and temporal resolution of the pump-probe techniques. While femtosecond pulses are mostly used and femtosecond to picosecond delay time resolution is necessary in many studies, the time constants of the transient events could vary from femtoseconds to tens of nanoseconds [2–4], depending on the physical phenomena involved. Simultaneously realizing such a long delay range in the pump-probe setup with a fast scan rate and a high temporal resolution is very challenging. Most of the conventional pump-probe spectroscopy setups use mechanical delay lines to sweep the delay between the pump and probe pulses. Yet, high-precision, linear step-motor stages are too slow for the applications with more demanding scanning speed requirements. Relatively fast scanning delay lines are often realized by a retro-reflector mounted on an electro-mechanical vibrator [5, 6]. However, the scanning range of the temporal delay is limited to tens of picosecond in order to realize a scanning rate of tens of hertz. Specially designed mechanical structures such as special rotary reflectors [7, 8] enable a nanosecond delay range at a scan rate of several hundreds Hz. A fast-swinging, oscillating optical delay line [9] could also realize relatively long delay range (e.g. ~a few nanoseconds) at relatively fast scan rates (e.g. hundreds of Hz). However, besides the complexity of these structures, the high-speed movements of the mechanical components could pose serious problems to the signal quality, linearity, and system stability. Further significant increase in the maximum delay is difficult to achieve, limited by the size of the already bulky moving parts. Also the vastly different beam paths at the different delays could also result in different beam divergence in free space that is difficult to deal with [4]. Stretching a fiber spool could be a more compact alternative scheme [10], but the picosecond-level scanning range is too limited for many applications.

Asynchronous optical sampling has emerged as a promising technique based on a very different working principle, which enables ultrafast time domain spectroscopy without a mechanical delay line [11]. In the currently studied implementations, it requires two mode-locked lasers of slightly different pulse repetition rates, so that the pulses from different lasers automatically walk-off from each other in the time domain. It easily circumvents the challenges faced by the conventional optical delay lines, and could simultaneously realize

high scanning speed and large scan range with high resolution. Using two mode-locked solid-state femtosecond lasers with  $\sim 1$  GHz repetition rates and an 11 kHz repetition rate difference, high-speed asynchronous optical sampling pump-probe measurements over a 1-ns time delay were realized [12]. Schemes based on similar dual-laser, dual-comb systems have also found very interesting applications in THz domain spectroscopy [13], fast, high-precision distance measurement [14] and multi-heterodyne spectroscopy [15]. Nevertheless, the dual-laser system involved is often quite complicated where the frequency difference of two mode-locked lasers is carefully controlled by an active feedback system. To avoid the complexity of the above dual-laser systems, optical sampling by cavity tuning was proposed by using just one ultrafast laser with a dynamically tunable and more complicated cavity [16, 17]. However, mechanical scanning inside the special laser cavity is still required in this scheme.

While most of the previously studied mode-locked lasers operate at one center wavelength, it has been demonstrated that dual-wavelength femtosecond lasers can be realized under certain cavity configurations [18–20]. It is demonstrated that in a passively mode-locked fiber ring laser using a carbon nanotube modelocker, stable subpicosecond pulse trains can be simultaneously generated in two separate wavelength bands around the 1550 nm window [20]. It is observed that the repetition rates of the pulse trains at different center wavelengths possess a small difference ( $\Delta f$ ), due to the intracavity group velocity dispersion (GVD). It is proportional to  $D_{ave} * L * \Delta\lambda$ , where  $D_{ave}$  is the average GVD of the cavity,  $L$  is the cavity length and  $\Delta\lambda$  is the difference of the center wavelengths of the output. Cavities designed with different amount of intracavity dispersion and  $\Delta\lambda$  can possess different  $\Delta f$ , which leads to different temporal sampling steps. Drifts in the cavity length that is common to free-running lasers would lead to nearly the same changes in the repetition rates at two wavelengths. This would result in negligible changes in the dispersion-determined frequency difference. Therefore, even free-running, dual-wavelength lasers with relatively long cavities under less stringent environmental control could be used to realize asynchronous optical sampling. On the other hand, changing the wavelength separation of these two operating wavelengths can significantly alter  $\Delta f$ , when the total cavity dispersion is fixed for a specific laser. It could be leveraged to realize functionality similar to the dual-laser systems previously demonstrated. Here, we present a novel pump-probe scheme based on asynchronous sampling using a dual-wavelength fiber laser. Without any external delay lines and using only one femtosecond laser with no intracavity dynamic tuning, two-color pump-probe measurements are realized with  $\sim 70$  ns scan range and  $\sim 500$  Hz scan rate. Unlike the conventional two-color pump-probe scheme where tight synchronization of two laser sources with a very low relative timing jitter are required [6], our scheme could be realized with a very compact, low cost, and simple setup.

## 2. Experimental setup

The pump-probe setup based on the dual-wavelength mode-locked laser is shown in Fig. 1. The dual-wavelength mode locked fiber laser is based on an Erbium-doped fiber (EDF) ring laser configuration with a single-wall carbon nanotube SWNT modelocker [20]. The laser cavity consists of  $\sim 4.5$  m EDF (INO Er105) and  $\sim 8.6$  m SMF. An isolator is placed in the cavity to ensure uni-directional propagation of light. An optical attenuator is used to adjust the gain profile to enable dual-wavelength operation of the laser, and a sheet of SWNT-doped polycarbonate is sandwiched between two FC/APC connectors as the mode-locker. Through adjusting the intracavity loss, thus the gain profile, dual-wavelength operation of the laser can be achieved. After their powers are boosted by an optical amplifier, the pulses at different wavelengths are separated by a bandpass-filter splitter into the pump and probe. The splitter has passbands of 1528.5–1536.5 nm and 1554.2–1562.2 nm, roughly matching the center wavelengths of the dual-wavelength output of the laser. The pump and probe can then be individually controlled by adjusting their power and state of polarization using a tunable attenuator and polarization controllers, before they are re-combined by a 2\*2 3-dB fiber coupler. The re-combined pump and probe are launched into the device-under-test (DUT). In

our current experiments, we use the widely studied semiconductor optical amplifier (SOA) as an example of DUT, to demonstrate the principle of operation. At the exit of the DUT (Covega SOA-4589), an optical filter that has a passband of 1554.2-1562.2 nm is used to block the pump pulses and let the probe pass. The probe pulses are then detected by a photodetector with a bandwidth of  $\sim 100$  MHz connected to a 500-MHz-bandwidth real-time oscilloscope (Agilent MS07054). The measured trace of the oscilloscope can then be used to retrieve the pump-probe information. The oscilloscope, we note, can be replaced with high-speed analog-to-digital data acquisition systems in future applications [14].

Due to the slight repetition rates difference ( $\Delta f$ ) between the pump and probe pulse trains, the probe pulse automatically and periodically walks through the pump pulse with a time delay step of  $\Delta f / f^2$ , which decides the temporal resolution for the asynchronous sampling scheme [11].  $f$  is the repetition rate of the probe pulses. The scan time to obtain one pump-probe measurement trace is  $1 / \Delta f$  in the real measurement time. The relationship between the real measurement time ( $\tau_r$ ) and the effective delay time  $\tau$  is  $\tau = \tau_r * \Delta f / f$ . Because of the huge difference between  $\Delta f$  and  $f$ , very high temporal delay resolution can be realized while much slower sampling speed is needed in the measurements. The maximal delay range is  $T = 1 / \Delta f$ , which is directly related to the length of the laser cavity.

In our scheme, with only one passively mode-locked fiber laser, the overall experimental setup is very simple and can be rather compact. As both the pump and probe pulses go through almost the same path, except for the consideration of excessive fiber dispersion and pulse distortion, no special care is needed in tailoring the length of the fibers in the setup, which is a key advantage of the asynchronous sampling method.

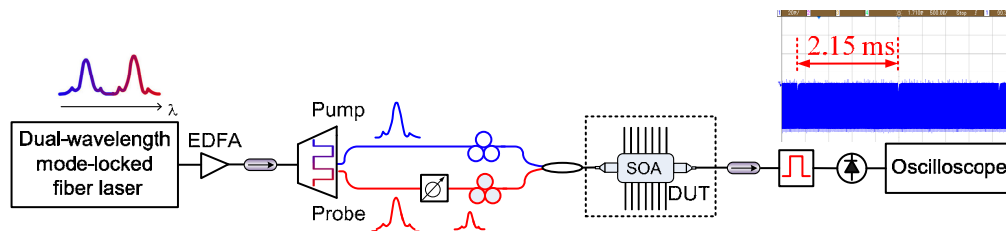


Fig. 1. Schematic of the proposed pump-probe measurement system.

### 3. Experimental results and discussions

Through tuning the cavity loss of the dual-wavelength mode-locked fiber laser, the gain spectral profile of the EDF can possess two peaks with similar gains under certain pump and signal conditions, which lead to the dual-wavelength operation of the laser. Figure 2(a) shows the output optical spectrum of the dual-wavelength laser under the dual-wavelength mode. Two mode-locked wavelength peaks are present, one centered at  $\sim 1532.4$  nm and one at  $\sim 1556.0$  nm. The 1556 nm-wavelength output has a repetition rate ( $f$ ) of 15.752704 MHz and the one at 1532.4 nm has a repetition rate ( $f + \Delta f$ ) of 15.753170 MHz, when measured by a 1-GHz-bandwidth photodetector (New Focus 1611) and an RF spectrum analyzer (Agilent N9320B), as shown in Fig. 2(b). The difference in their repetition rates  $\Delta f$  is 466 Hz, due to the group velocity dispersion in the fiber cavity. We note that, when this free-running laser is operated under unregulated ambient environment, the repetition rates of the pulse trains at the two lasing wavelengths could drift by as much as tens of Hz in tens of minutes, due to the changes in the cavity length. Yet, the changes in  $\Delta f$  can still remain no larger than the resolution of our measurement instrument ( $\pm 1$  Hz). The timing jitter of the 1532.4 nm pulses is measured as  $\sim 1.2$  ps using the method described by Linde [21], while the jitter at 1560 nm is estimated to be  $< 300$  fs. The jitters are believed to be significantly affected by the wavelength stability besides other common source of jitters, and they would determine the temporal resolution of our setup [13].

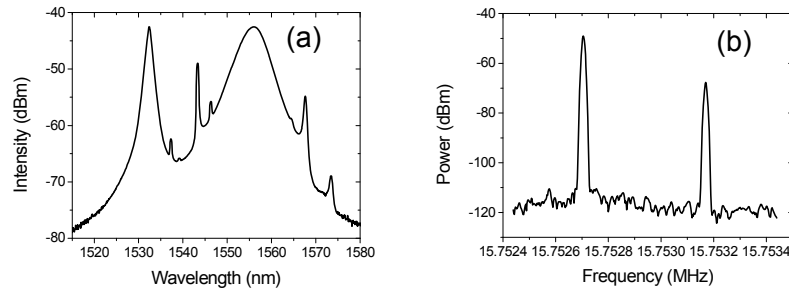


Fig. 2. (a) Output optical spectrum of the dual-wavelength laser; (b) the corresponding RF spectrum.

The dual-wavelength output is amplified to an average power of 3.5 mW using an EDFA and is then filtered with the bandpass filter splitter. The pulses at the center wavelength of 1532.4 nm are set as the pump, and the ones at 1555.1 nm as the probe in our experiments. The average powers of the pump and probe beam that are injected into the SOA are adjusted to 504  $\mu$ W and 29  $\mu$ W, respectively. The pulse shape is measured by a home-built autocorrelator. The pulse widths of the pump and probe pulses are 1.56 ps and 1.15 ps, respectively, if a  $\text{sech}^2$ -shape pulse is assumed.

As shown in Fig. 1, the oscilloscope trace shows a series of periodic dips where the magnitude of the detected probe pulse deviates from its average value, when it overlaps with the pump. This pump-probe signal occurs every 2.15 ms in the real measurement time, which equals  $1/\Delta f$ . A close-up view of the envelope of the probe pulse train when the SOA bias current is set to be 115 mA is given in Fig. 3 along with a fitted curve. Based on the repetition rates, the equivalent delay step in our current setup is 1.8 ps. The retrieved pump-probe signal shows a picosecond fast dip and nanosecond-long exponential recovery, which correspond to the ultrafast nonlinear gain dynamics and the slower carrier density recovery process of SOA, respectively [22, 23]. The details of the ultrafast picosecond response are shown in the inset of Fig. 3. Based on the pulsewidths of the pump and probe, the fast transient measured is likely limited by these finite pulsewidths. Further pulse compression and higher temporal resolution is needed to better resolve the subpicosecond effects.

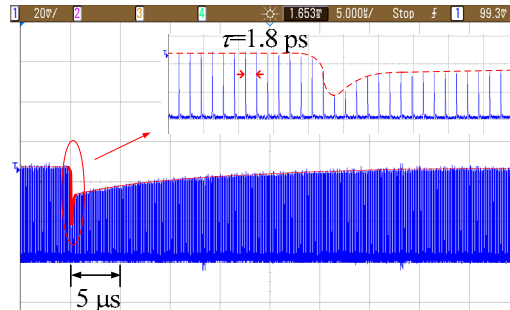


Fig. 3. Oscilloscope trace measured when the bias current of the SOA is 115 mA. Inset is the zoom-in of the fast transient of the curve.

By tuning the bias current, the SOA can operate in different regimes with different pump-probe characteristics. To validate our pump-probe method, the signals under different bias currents are measured, as shown in Fig. 4. When the bias is above the transparency current, the SOA shows gain saturation by the strong pump pulse as seen in Fig. 3. In contrast, the loss experienced by the probe pulse through the SOA is reduced when the pump current is below the transparency current, as the absorption of the pump pulse increases the carrier density. Therefore, at the bias current of  $\sim 35$  mA and 55 mA, the pump-probe traces show a peak with

a decay time constant of  $\sim 0.74$  ns for the slowly decaying term. In contrast, when the pump current is 95 mA and 115 mA, above the transparency current, a gain dip is observed. The slower recovery time constant is estimated to be 0.41 ns in effective time, showing a faster dynamics than the previous case. The transparency current of the SOA is  $\sim 75$  mA, where the ‘slower decay’ term almost vanishes. All the traces are normalized to the probe amplitude in the absence of the pump.

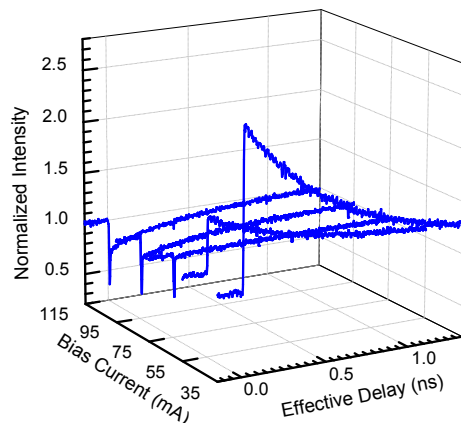


Fig. 4. Pump-probe signals under different bias conditions of SOA.

We note that, though two-color pump-probe experiments are done in this work, by utilizing nonlinear optical processes that would broaden or shift the center wavelength of one of the pulses, standard pump-probe measurement in the same spectral range are possible [24].

#### 4. Conclusions

A novel pump probe scheme is demonstrated by using a dual-wavelength femtosecond laser. Long scanning range ( $\sim$ tens of nanosecond) is realized at  $\sim 466$  Hz scanning rate with a picosecond resolution. Since there are no external mechanical delay line and no need for precise temporal adjustment of the setup, high-speed pump-probe measurements with very long scanning ranges could be realized in a rather simple system. It offers a simple and low-cost alternative approach to realize the asynchronous sampling scheme without using two mode-locked lasers. We note that the conventional two-laser asynchronous sampling systems have demonstrated very high temporal sampling resolution, excellent system stability and short pulsewidth meeting the needs of a wide range of applications. Our simple approach, though sufficient for the current application, still need much improvements to meet similar requirements. Further improvements in the dual-wavelength laser's performance, such as different combinations of  $f$  and  $\Delta f$  and better output stability and jitter, would help to enable pump-probe measurements with better resolution and accuracy. Such a dual-wavelength, dual-comb output could also be utilized to realize high-resolution distance measurements and other applications [24]. For the applications demands shorter, femtosecond pulses, it would be challenging to directly generate dual-wavelength pulses with very short pulsewidths in an EDF-based laser. Additional extracavity spectral broadening and pulse compression could be needed, with extra system complexity.

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