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**Title:** Probing intra-soliton-molecular jittering on attosecond timescale

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This document provides supplementary information to “Attosecond timing jitter within a temporal soliton molecule,” https://doi.org/10.1364/OPTICA.397897. We present here supplementary material describing the design of home-made Ti:sapphire laser, the route to the soliton molecular operation as well the soliton molecular output characteristics; The full experimental setup of balanced optical cross-correlator (BOC) for intra-soliton-molecular timing jitter characterization and the time stretch dispersive Fourier transform (TS-DFT) setup for molecular relative phase measurement; An independent intra-molecular PSD measurement that covers the full Nyquist frequency based on a large bandwidth photodetector; The data of the measured relative intensity noise (RIN) power spectral density (PSD) of the Ti:sapphire laser under the soliton molecular state; The procedure for the calculation of photodetection shot noise limit of the BOC system; Intra-soliton-molecular timing jitter measurement at a different 125 fs binding separation; BOC-based PSD measurements for vibrational states. The above material helps interested readers on the detailed experimental design and data analysis.

1. Ti:sapphire Laser setup
A home-made Kerr-lens mode-locked Ti:sapphire laser is used to generate temporal soliton molecules. The layout of the Ti:sapphire laser is shown in Fig. S1(a) and the photo of the laser is shown in Fig. S1(b). The pump laser is an all-solid frequency doubled Nd:YVO4 laser (Spectra-Physics, Millennia) delivering 12 W of maximum continuous-wave (CW) power at 532 nm. A spherical lens (L) with focal length of 75 mm focuses the pump power into a Brewster-cut Ti:sapphire crystal, which has 23 mm thickness and 0.25 wt. % doping concentration. The Ti:sapphire laser cavity is based on a standard Z-fold resonator design consisting of two folding mirrors (C1 & C2), which are curved mirrors with the same radius of curvature of 75 mm. The laser beams collimated by C1 and C2 are bounded by a 7% output coupler (OC) and a high-reflective end mirror (EM), respectively. The length of the two linear arms are ~ 470 mm and ~ 690 mm, respectively. The longer arm contains a chirped mirror pair (CM1 & CM2, Layertech) and a fused silica prism pair (P1 & P2) for group velocity dispersion compensation. The coating design of CM1 and CM2 can cancel each other’s oscillation of group delay dispersion (GDD). The entire laser cavity is built on a low expansion invar alloy breadboard and enclosed in an aluminum case. The fundamental repetition rate is 120 MHz. The output laser pulses are p-polarized since the Ti:sapphire crystal acts as a Brewster plate.

Fig. S1. (a). Design of the Ti:sapphire laser; C1 & C2: curved mirror pair; CM1 & CM2: chirped mirror pair; EM: end mirror; L: lens; M1: folding
plane mirror; OC: output coupler; P1 & P2: prism pair; (b) Photo of the Ti:sapphire laser system; (c) The pulse train of bound state solitons after photo-detection; (d) The first harmonic of the radio frequency spectrum for the photo-detected bound state laser pulse train.

Single pulse CW mode-locking is routinely initiated at ~ 2.8 W green light pump power by slightly shaking the end mirror (EM) when the laser is operating at its inner border of one stability zone. Adjust the insertion of prism P1 such that the laser works in a stretched-pulse mode-locking regime with ~ 40 nm FWHM spectral width centered at ~ 800 nm. When the pump power is further increased to ~ 3.4 W, bound state mode-locking can be achieved by finely tuning the amount of insertion of P1. The output average power at bound state is 400 mW. The pulse train (c) and radio frequency spectrum (d) after photo-detection show high stability of bound state mode-locking.

2. Full experimental setup on intra-soliton-molecular timing jitter and phase characterization

The full experimental setup is shown in Fig. S2. The entire apparatus including the Ti:sapphire laser is built on a floating optical table (Newport). The soliton molecular pulse train output from the Ti:sapphire is pre-chirped by six bounces in a chirped mirror pair (CMP1, Layertech). Each bounce provides ~60 fs² group delay dispersion. Then, the output pulse train is directed to the Michelson interferometer (M1), where the laser power is firstly divided by a 50:50 fused silica broadband plate beamsplitter (BS, Thorlabs: BSW11R). Then, the transmissive and reflective laser beams are retroreflected by silver coated retroreflection mirrors (RM, Thorlabs: HR1015-P01) and combined at the BS again. One RM is mounted on a piezo transducer (PI, P840.1) and then placed on a motorized linear translation stage (Suruga Seiki). A half waveplate (HWP) is inserted into one arm of M1 in order to rotate beam polarization by 90°.

The two outputs from the transmissive and reflective ports of BS serve as input to balanced optical cross-correlator (BOC). Each input is focused onto a type II-phase matched beta barium borate (BBO) crystals with 0.4 mm thickness by a silver coating parabolic mirror with focus length of 25.4 mm. The extraordinary axes of the two BBOs are placed with orthogonal orientation. After filtering out the 800 nm fundamental frequency laser with optical short-pass filters (Thorlabs, FES0450), the generated SFG optical signals are detected by a balanced photo-detector (Newfocus, Model 2107) with a transimpedance gain of 626 kV/A. Given that the common-mode rejection ratio of balanced photodetection is sensitive to phase difference between the two signals, a linear transition stage is used to carefully balance the optical length difference between the two optical cross-correlation paths. The timing resolution of BOC is very sensitive to pulse duration and pulse chirp. To this end, it is very important to only use metal-coating mirrors for beam folding and focusing. We specifically choose very thin transmissive components (BS and HWP) to make sure that the GDD can be well compensated by CMP1.

For MI arm length stabilization, the photo-detected voltage signal from BOC is firstly low pass filtered (Mini circuits, SLP-10.7+) and directed to a loop filter (Newport, LB100S). The output from loop filter is voltage amplified (PI, E617) and then actuates the PZT in the MI, thus close the loop. After phase-locking, power spectral analysis on the BOC output outside the locking bandwidth is conducted. A fast Fourier transform network analyzer (Stanford Research Systems, SR770) and an RF spectral analyzer (RIGOL, DSA815) are used to measure the power spectral density (PSD) at < 100 kHz Fourier frequencies and > 100 kHz Fourier frequencies, respectively. The measurement results are connected and divided by the square of timing discrimination slope, resulting in timing jitter spectral power density.

Figure S2 also shows the time stretch dispersive Fourier transform (TS-DFT) setup, which consists of a 500-m-long single mode fiber (Nufern, 780-HP), a high speed photodetector with 30 ps rise time (Newport, 818-BB-45FS), as well as an 8-GHz real-time oscilloscope (Rohde & Schwarz, RTP). For a TS-DFT measurement, ~5 mW output power from the Ti:sapphire laser has been coupled to the single mode fiber. The temporal stretched soliton molecules are received by the photodetector, and recorded by the oscilloscope in real time. The large GDD introduced by the long fiber effectively maps optical spectrum of each soliton molecule into the overlapped linearly chirped laser pulse pair. Therefore, sequence of single-shot optical spectrum can be obtained and plotted as an evolving graph.

Fig. S2. Detailed experimental apparatus. BBO: beta barium borate crystal; BOC: balance optical cross-correlator; BS: beam splitter; CMP: chirped mirror pair; FM: flip mirror; HWP: half waveplate; PD: high speed photo-detector; PM: parabolic mirror; PZT: piezo transducer; RM: retroreflection mirror; SMF: single mode fiber; TS-DFT: time stretch dispersive Fourier transform.

3. Timing jitter PSD measurement up to the Nyquist frequency

For a complete characterization of intra-molecular timing jitter PSD, measurement up to the Nyquist frequency of 60 MHz (half of fundamental repetition rate) is required. However, the measurement is cutoff at 1 MHz by the bandwidth of BPD (Newfocus, 2017). To extend the measurement bandwidth, an independent measurement has been conducted by using another BPD (Thorlabs, PDB420A). The RF output bandwidth is as high as 75 MHz, while the BOC-based measurement is thermal noise limited due to a lower timing discrimination slope at setpoint A.

To partly solve this problem, we block one photodetector and conduct the experiment with the optical cross-correlation (OC) method. Even though the OC-based measurement is easily affected by laser intensity noise, it is still effective to provide an upper estimate for the timing jitter of a mode-locked laser [S1, S2]. The measurement result is shown in Fig. S3. The discriminator slope at setpoint A is 100 mV/ fs. The measurement covers the Nyquist...
frequency. No additional noise structures from soliton molecules are found from 1 MHz to 60 MHz.

The measurement from 1 MHz to 60 MHz is still limited by photodetector thermal noise, and the noise floor of $\sim 1 \times 10^{-10}$ fs$^2$/Hz is about one order higher than the shot-noise limited measurement in Fig. 3 of the main manuscript. The integrated rms timing jitter is 75 as [1 MHz, 60 MHz], see Fig. S3(b). By combining the data from 100 Hz to 1 MHz, and by excluding the integrated timing jitter introduced by the Michelson interferometer, an upper estimate of intra-molecular timing jitter is 60 as rms [100 Hz, 60 MHz].

It is well known that laser intensity noise can cause a fluctuation of pulse timing via self-steepening effect [54]. Timing jitter coupled by RIN has also been confirmed in a sub-10 fs Ti:sapphire laser [55]. Similar phenomenon is not observed here, for a soliton-molecular mode-locking state. The reason is that the laser intensity noise is common mode for the two solitons that composes a molecule. Given that the timing change induced by self-steepening effect is identical for the two solitons, the contribution to intra-molecular timing jitter (relative timing jitter between the solitons) is negligible.

Fig. S3. (a) Timing jitter PSD measurements with optical cross-correlation (OC) method by using a high bandwidth BPD (Thorlabs, PDB420A). Both the measurement results at setpoint A&B as well as the respective noise floor are provided. The inset shows the cross-correlation trace and the output voltage of the cross-correlator at Setpoint A. (b) Enlarged view of (a) from 1 MHz to 60 MHz. The integrated jitter is also provided.

4. Relative intensity noise measurement

The relative intensity noise (RIN) PSD of the Ti:sapphire laser under the soliton molecular mode-locking state has been characterized and shown in Fig S4. RIN has been defined as laser mean square intensity fluctuation divided by square of average optical power. The laser RIN can be characterized following a standard procedure in [S3]. To this end, ~0.5 mW of laser power has been directed to a low noise photodetector (Thorlabs, PDA36A), and the PSD ($S_{\text{RIN}}$) has been characterized by FFT analyzer (< 100 kHz) and rf spectral analyzer (> 100 kHz). The RIN PSD is obtained following:

$$S_{\text{RIN}} = \frac{S_{\text{RIN}}}{V_{\text{avg}}^2}, \quad (S1)$$

where, $V_{\text{avg}}$ is the average output voltage from the photodetector measured by a voltmeter. The laser intensity fluctuation is 0.2% rms integrated from 100 Hz to 1 MHz. The Ti:sapphire laser intensity noise has been mainly coupled from the pump laser power fluctuations. The pattern of intensity noise distribution in Fig. S4 is entirely different with intra-soliton-molecular timing jitter PSD shown as Fig. 3(i) in the main text. This proves that the common mode rejection ratio (CMRR) of the balanced photodetector in the BOC system is high enough to cancel out the impact of laser intensity noise during timing jitter measurement.

$$S_{\text{RIN}} = \frac{4eR P_{\text{SG}} G^2}{K_a^2}, \quad (S2)$$

where $e$ is the electron charge, $R$ is the detector responsivity, $P_{\text{SG}}$ is the absorbed SFG power by each photo-detector, $G$ is the detector transimpedance gain, and $K_a$ is the timing discriminator of the BOC. In our experiments, at setpoint A, the parameters are given by $R = 0.2A/W$, $G = 626kV/A$, $P_{\text{SG}} = 64\mu W$, $K_a = 368mV/fs$. Thus, the calculated shot noise level is $S_{\text{RIN}} = 2.37 \times 10^{-13}$ fs$^2$/Hz.

5. Shot noise floor calculation

The timing resolution of the BOC system (by using Newfocus 2107) is limited by photodetection shot-noise floor. The shot-noise limited timing jitter PSD $S_{\text{shot}}$ [S6] for balanced photodetection can be calculated by

$$S_{\text{shot}} = \frac{4eR P_{\text{SG}} G^2}{K_a^2}, \quad (S2)$$

6. Intra-soliton-molecular timing jitter measurement at 125 fs binding separation

During the experiment, soliton molecule with a binding separation of 125 fs was frequently observed in the meantime. The BOC-based measurement result (by using Newfocus 2107) is shown in Fig S5. The timing jitter PSD of the soliton molecule with 125 fs pulse separation is similar with that of 184 fs pulse separation in the main manuscript, where the intra-molecular timing jitter PSD (setpoint A) is limited by technical noise of Michelson interferometer in the low Fourier frequency and detection shot noise in the high Fourier frequency. The peak close to 1 MHz is related with laser intensity noise (The common-mode rejection...
ratio (CMRR) of the balanced photo-detector decreases at higher Fourier frequency, that’s why the impact of intensity noise emerges at ~1 MHz). The integrated intra-molecular timing jitter, ranging from 100 Hz to 1 MHz, is 32 as rms. This number is higher than that of the molecule with 184 fs pulse separation presented in the main text. The reason is that the shot-noise limited noise floor is ~4 times higher than that of Fig. 3 in the main manuscript. The higher shot-noise level is mainly due to the lower timing discriminator $K_n$ which is 194 mV/ fs, while this number is 368 mV/fs in the main manuscript.

Fig. S5. Timing jitter spectral density of soliton molecule with a binding separation of 125 fs. (a). Optical spectrum and BOC trace. (b). Timing jitter spectral density and the integrated timing jitter.

7. Vibrational states

Besides the stationary soliton molecules, vibrating states are also frequently observed in experiment. One example is shown in Fig. S6. Fig. S6(a) shows the optical spectrum and the obviously blurring of fringe contrast is characteristic of an oscillating state. The spectral modulation period indicates a 182 fs pulse separation. Fig. S6(b) shows the timing jitter PSD obtained by the BOC method. Timing jitter PSD measured at setpoint A shows a singular peak at 380 kHz, which confirms the oscillation of pulse separation for this molecule. The inset shows another oscillating state where the vibration frequency is continuously drifting (also, refer to visualization). The absolute level of timing jitter PSD is higher than data in the main text because a balanced photo-detector (Thorlabs, PDB 420A) with lower transimpedance gain is used for this measurement. This photo-detector is used because it covers a broader measurement bandwidth (>60 MHz).

Fig. S6. Timing jitter spectral density of a vibrating soliton molecule. (a). Optical spectrum and BOC trace. (b). Timing jitter spectral density measured with the BOC method.

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