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Optical flywheels with attosecond jitter

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It has been known for some time that the steady-state pulse propagating inside a mode-locked laser is the optical equivalent of a mechanical flywheel. By measuring the timing error spectrum between phase-locked optical pulse trains emitted from two nearly identical 10 fs Ti:sapphire lasers, we demonstrate a record low integrated timing error of less than 13 as, measured from d.c. to the Nyquist frequency of the pulse train, which is 41 MHz. This corresponds to the lowest high-frequency phase noise ever recorded of −203 dBc Hz^{-1} (assuming a 10 GHz carrier) for offset frequencies greater than 1 MHz. Such a highly uniform train of pulses will enable the synchronization of pump–probe experiments that measure the evolution dynamics of chemical[1,2] and atomic processes[3,4] evolving on femtosecond and attosecond timescales. The ultralow timing jitter of such pulse trains will also allow photonic analog-to-digital conversion of mid-infrared waveforms with a resolution of 6 bits[5].

In the last 10 years, the advent of femtosecond laser frequency combs[6] has revolutionized frequency metrology, and with it a wide variety of disciplines ranging from astrophysics and the search for exoplanets[7–9] to precision spectroscopy[10,11] and, most recently, attosecond science[12,13]. Optical atomic clocks have also been realized with fractional frequency instabilities below 1×10^{-16} achieved in only a few seconds of averaging[14,15]. In each of these examples, the mode-locked laser derives its long term (>1 s) phase and frequency stability from an external reference. In this Letter, we demonstrate the fundamental stability of the pulse train from Kerr lens mode-locked Ti:sapphire lasers on short timescales (<100 μs). The existence of a well-defined frequency comb is due to the pristine temporal periodicity of the pulse envelope and well-defined carrier-phase evolution from pulse to pulse (although the latter is not important for this work). Here, we have established an upper limit for the timing jitter of a pulse train and potential phase noise of microwave signals extracted from a pulse train emitted by a mode-locked Ti:sapphire laser, on short timescales less than 10 μs. Note that we refer only to the pulse envelope and not the underlying optical carrier when discussing phase noise.

As with any oscillator, the translation invariance of the steady-state oscillation with respect to pulse position or carrier phase leads to diffusion of the pulse position and phase in the presence of noise. It can be shown[16] that the contribution to pulse position drift (due to the fundamental spontaneous emission noise of the gain medium compensating for output coupling and internal losses of a Kerr lens mode-locked laser generating soliton-like pulses) scales as

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle \Delta t^2 \rangle = \frac{\pi^2}{6}\tau^2 \frac{h\nu}{E_\mathrm{P}\tau_\mathrm{c}} \qquad (1)$$

where Δt is the offset of the pulses from their nominal time locations, τ is the full-width at half-maximum pulse width divided by 1.76, E_P is the intracavity pulse energy, τ_c the cavity decay time, h Planck's constant and ν the optical frequency. The lowest phase noise oscillators demonstrated to date have been microwave systems based on large (~5 cm), precisely grown sapphire crystals operating at 10 GHz to take advantage of the anomalously low absorption of electromagnetic energy in sapphire at that frequency[17]. As a reference for the current state of the art, a single-sideband (SSB) phase noise of −190 dBc Hz^{-1} at 1 kHz offset for a 10 GHz carrier is predicted for the next generation of cryogenically cooled sapphire loaded microwave cavity oscillators[18]. (Throughout this Letter we will compare phase noise values as if all oscillators operate at 10 GHz to enable a direct comparison of results.) Although it is possible to extract radiofrequency (RF) harmonics of the pulse repetition rate of mode-locked lasers through direct photodetection of the output to generate electronic signals at frequencies including and exceeding 10 GHz, here we measure directly the timing jitter of the pulse stream in the optical domain and compute the corresponding RF-signal phase noise for a 10 GHz harmonic generated from the pulse train under ideal conditions.

The rate at which mode-locked lasers emit pulses is related to the inverse of the optical cavity length. As a result, phase noise in the emitted pulse train can be generated not only by spontaneous emission, but also by environmental perturbations of the cavity length including mirror vibrations, as well as slight laser cavity misalignments leading to intracavity power fluctuations. These power fluctuations, in addition to those caused by relative intensity noise (RIN) from the pump laser, are transformed into phase noise through the self-steepening effect[19]. The Gordon–Haus effect[20] may also lead to increased timing jitter through centre frequency fluctuations of the optical spectrum of the laser pulse, which are transformed into phase fluctuations via the intracavity dispersion. Extremely short pulse lasers may be able to minimize the contribution of Gordon–Haus jitter by using the entire gain bandwidth of the Ti:sapphire crystal, greatly restricting changes in the centre of gravity of the pulse spectrum. Assuming careful elimination of such noise sources, spontaneous emission from the gain medium directly driving the pulse timing diffusion according to equation (1) will remain. Theoretical estimates of the timing jitter of pulse trains from solid-state mode-locked lasers are often expressed in terms of phase noise of an extracted microwave signal at a harmonic of the laser repetition rate. For a laser with an 80 MHz pulse repetition rate, it has been predicted that the harmonic at 10 GHz will show phase noise below −190 dBc Hz^{-1} for offset frequencies greater than 1 kHz (refs 16,21). However, until the present study, such low levels of phase noise at high offset frequencies have never been confirmed.

Measurement of phase noise in optical pulse trains can be difficult if the optical pulse train has been converted to a RF signal[22,23], as low noise conversion of optical pulse trains into microwave signals is a challenging problem[24]. RF measurement techniques are limited by thermal noise in the terminating resistors as well as

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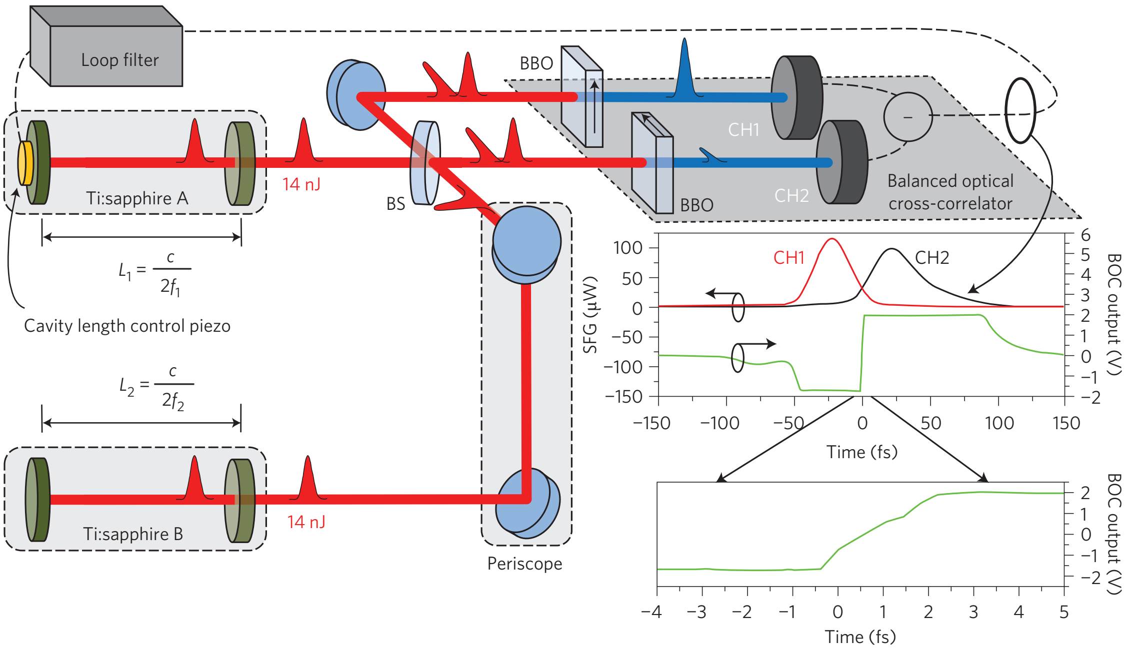

Figure 1 | Experimental apparatus. The outputs from two nearly identical Ti:sapphire lasers (A and B) are combined using a beamsplitter (BS), with the polarization of one pulse train rotated using a periscope composed of silver mirrors before the beamsplitter. Orthogonal orientation of the extraordinary axis of the two beta barium borate BBO crystals (indicated by arrows on the crystals) ensures that equal upconversion efficiency occurs only when the two pulses exactly overlap at the entrance to both crystals. The resulting error signal is characterized by a phase error-to-voltage conversion of 22.1 kV rad^{-1} (±1.0 kV rad^{-1}) for a 10 GHz carrier in a linear region with a range of just over 2 fs. BOC, balanced optical cross-correlator.

the signal level available for input to the phase noise measurement system, and have a sensitivity of about −192 dBc with discriminator slopes of nearly 110 V rad^{-1} (refs 17,25). Direct optical techniques can achieve higher sensitivities by avoiding the photodetection process. Balanced optical cross-correlation[26–28] (Fig. 1), allows direct measurement of the optical pulse train timing jitter, which can be rescaled as phase noise. Equivalent phase error discrimination slopes usually exceeding 1 kV rad^{-1}, or 100 mV fs^{-1} of pulse arrival time error, can be achieved.

To measure the timing jitter in the pulse train of mode-locked lasers, we phase-locked the pulse trains (envelope arrival phase, not carrier wave phase) from two Ti:sapphire lasers and measured the residual timing jitter between the pulse trains outside the bandwidth of the phase-locking feedback loop. To detect the timing jitter between the pulse trains we used balanced optical cross-correlation[26–28] (Fig. 1), which performs sum-frequency generation (SFG) between orthogonally polarized pulses from two femtosecond lasers, with the difference in power of the upconverted signals as a measure of the temporal overlap between the two pulses. The phase error, or equivalently the timing jitter spectrum between the two pulse trains, is measured by dividing the balanced detector's output voltage spectrum by the slope of the balanced detector's phase error discriminator. The slope of the phase error discriminator was determined both before and after the two pulse trains were phase-locked together, as well as while the phase-lock feedback loop was closed. Before and after the loop was closed, measurement of the discriminator slope was carried out by detuning the repetition rates of the lasers slightly (3 Hz) to allow the pulses to slowly pass through one another, performing a repeating cross-correlation, and recording the output of the balanced detector using an oscilloscope. When the pulse trains were phase-locked together, we injected a small controlled time delay to one of the pulse trains before the beamsplitter and coherently detected the output of the balanced detector using a network analyser. The mean value determined from these three measurements was 22.1 kV rad^{-1} (±1.0 kV rad^{-1}) or 1.432 V fs^{-1} (±65 mV fs^{-1}) (see Supplementary Information), resulting in a measurement uncertainty due to uncertainty in the cross-correlator slope of ±0.4 dBc Hz^{-1} and ±0.6 as for the resulting phase and integrated timing error measurements, respectively (Fig. 2).

The residual timing jitter spectrum from the phase-locked pulse trains is plotted in Fig. 2, together with the integrated pulse arrival time error. This phase error spectrum represents an upper limit to the phase noise or pulse timing noise spectrum of one of the pulse trains. The bandwidth of the feedback loop for phase-locking the two lasers can be estimated from the servo peak to be 30 kHz. Below 30 kHz, the feedback loop is suppressing the phase error spectrum, which is dominated mainly by environmental cavity length perturbations as well as pump laser intensity noise. Offset frequencies above 30 kHz show the intrinsic phase noise spectrum between the two lasers up to 1 MHz, where the phase noise spectrum falls below the noise floor. The resulting integrated timing error, which covers the entire Nyquist region (for the 80 MHz pulse repetition rate) from 100 Hz to 41.5 MHz, is 12.3 ± 0.6 as with a noise floor contribution of 8.1 ± 0.4 as, where the uncertainty in the integrated values arises from uncertainty in the cross-correlator slope, as discussed above. The large noise spur at 190 kHz is a result of RIN of one of the pump lasers, which is converted into phase noise, with the slight difference in centre frequency of the spur being due to drift in the RIN spectrum. The dependence of timing jitter on pump laser RIN was confirmed experimentally by modulating both pump lasers in turn and coherently detecting the resulting timing jitter using the cross-correlator. This measurement yielded conversion factors of $1/f$ ns W^{-1} and $0.3/f$ ns W^{-1} (see Supplementary Information) for each laser for modulation frequencies f, below the upper state lifetime of the Ti:sapphire gain, typically 300 kHz. Using these conversion factors and the measured RIN spectra from both pump lasers, we generate the dotted black curve in Fig. 2, which shows the estimated contribution to the timing jitter from pump laser RIN. The

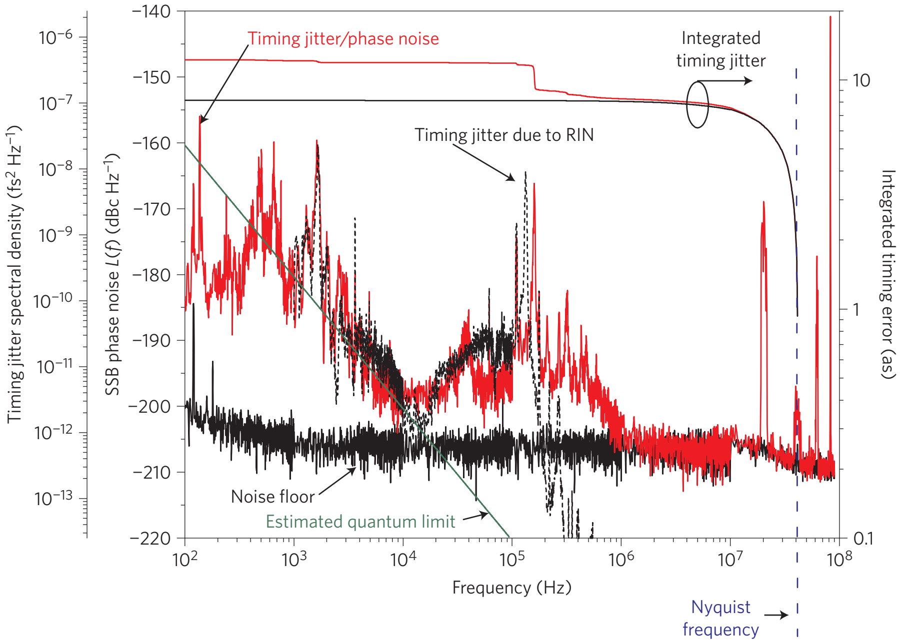

Figure 2 | Timing jitter spectral density and rescaled single-sideband phase noise between the phase-locked optical pulse trains from two mode-locked Ti:sapphire lasers, as measured by the optical cross-correlator. All traces have been scaled from the fundamental 80 MHz pulse rate of the Ti:sapphire lasers to 10 GHz as a common basis for comparison. The result of the optical cross-correlator measurement is plotted in red, and the noise floor of the optical cross-correlator measurement determined by measuring the output of the balanced detector when the input is blocked is plotted in black. The estimated contribution to the timing jitter spectrum due to pump laser RIN is plotted as a dotted black line. The estimated spontaneous emission-limited phase noise for the lasers measured here (green) was determined using the Fourier transform of equation (1).

conversion of pump laser RIN into pulse train phase noise is a serious challenge, although recent results demonstrate that it can be adequately suppressed by locking to an optical reference cavity[29].

Although the phase error for pulse trains from mode-locked lasers typically decreases at a rate of –20 dB/decade (ref. 30), the level of –200 dBc Hz^{-1} at 10 kHz offset predicted for the current laser configurations is still too low to allow the detection of phase noise caused by spontaneous emission noise. A noise floor no higher than –240 dBc Hz^{-1} at a 1 MHz offset appears to be necessary for direct observation of phase noise due to spontaneous emission noise in the laser cavity. Assuming Gaussian pulses, the maximum discriminator slope, described in terms of optical power on the balanced detector, is given by

$$\partial P/\partial t = 2\sqrt{2}P_c/\tau_p e^{1/4} \qquad (2)$$

where P_c and τ_p are the peak power and full-width at half-maximum duration of the upconverted pulse. Assuming a shot noise limited detection, the noise floor of the balanced optical cross-correlation technique will decrease as $1/P_c$ from –193 dBc Hz^{-1} for a 10 fs pulse with 1 μW converted power, requiring nearly 60 mW converted power for a –240 dBc Hz^{-1} measurement floor.

Optical pulse trains from femtosecond mode-locked lasers exhibit the lowest high-frequency phase noise of any type of oscillator. Here, we have established an upper limit on the pulse train phase noise at high offset frequencies of –203 dBc Hz^{-1} referenced to 10 GHz, resulting in an integrated timing jitter of less than 13 as considering the entire Nyquist frequency of the source laser (41 MHz). The estimated spontaneous emission limited phase noise level for a laser similar to those measured here, but with an optical spectrum corresponding to a 5 fs pulse, 1% output coupling, 125 nJ intracavity pulse energy and no noise contribution from Gordon–Haus jitter, would be –200 dBc Hz^{-1} at a 1 kHz offset frequency. Because active suppression of phase noise at high offset frequencies is extremely challenging, the low level of phase noise demonstrated here is a promising indicator that more advances are possible with respect to the construction of high-quality oscillators. Ultimately, oscillators based on mode-locked lasers are likely to benefit extremely high-speed and high-precision optical analog-to-digital conversion, timing and synchronization systems, as well as ultra-broadband and secure communications systems. This result is equally important for timing distribution and synchronization for the next generation of X-ray free-electron lasers (FELs) such as seeded X-ray FELs. In such facilities, the critical laser systems for seed and probe generation are solid-state laser-based, and the high-frequency jitter result demonstrated here indicates that there are no fundamental limitations in the jitter of laser oscillators that may limit synchronization above the sub-100 as range. Such precision would make ground-breaking optical pump and X-ray-probe experiments possible, measuring the evolution dynamics and structural changes of chemical[1,2] and atomic processes[3,4] evolving on femtosecond and potentially even attosecond timescales. The current measurement is only a first step in this direction. The jitter of optical timing distribution systems as well as drifts in the optical amplifier systems following the laser oscillators must be controlled to a similar level, both of which are formidable tasks.

Methods

The balanced optical cross-correlator[26–28] used here is presented in Fig. 1. Detection of changes in the temporal overlap of pulses from the two lasers was measured as the difference in optical power of the upconverted signals from each arm of the device. In the current experimental apparatus, we aligned the extraordinary axis of the two 400-μm-long beta barium borate (BBO) crystals in the two arms of the cross-correlator to different input polarizations. Because of the different orientations of the two crystals, a pulse from each laser experiences a different group velocity in each of the two arms of the cross-correlator. As a result, the two arms of the cross-correlator require opposite initial time offsets between the orthogonally polarized pulses for optimal conversion (Fig. 1, inset). Using the output of the cross-correlator and the balanced detector (Thorlabs PDB120A, modified for 36 kΩ trans-impedance gain and using Hamamatsu S5712 photodiodes) as the phase error discriminator in a phase-locked loop, the voltage output of the balanced detector was measured when the loop was locked to determine the relative phase error between the two optical pulse trains. Peak conversion from each arm of the cross-correlator was typically 100 μW, which was collected by a balanced detector to cancel intensity fluctuations of the two lasers. Ideally, the measurement floor of a balanced optical cross-correlation measurement is determined by the shot noise of the incident pulse train converted into apparent phase error by the trans-impedance amplifier and discriminator slope; however, here, the input current noise of the balanced detector limited the noise floor to –203 dBc Hz^{-1}.

For measurement of femtosecond laser pulse-train timing jitter, two nearly identical, Kerr lens mode-locked Ti:sapphire lasers were constructed, each pumped by a separate, single-frequency, diode-pumped, solid-state (DPSS) laser (Coherent Verdi V6). Double-chirped dispersion compensating mirrors were used to construct folded linear cavities with 83.6 MHz pulse repetition rates. Both lasers used output couplers that featured loss profiles matched to the gain spectrum of the Ti:sapphire crystal and $\sim$8% output coupling; however, no special steps were taken to reduce the expected pulse-train timing jitter by selecting a particular laser operating point. Each laser produced optical spectra corresponding to 10 fs transform-limited pulses (see Supplementary Information), with up to 120 mW of average output power.

In determining the integrated timing error between the pulse trains of the two mode-locked lasers in Fig. 2, there were four technical noise spurs at 80, 60, 40 and 20 MHz, none of which was counted towards the integrated timing error of the pulse train. The noise spur at 80 MHz is the pulse repetition rate of the lasers, f_R. The spurs at 20, 40 and 60 MHz are heterodyne beat signals between three different fields: the unsuppressed second harmonic generation from each individual pulse train and the sum-frequency field resulting from interaction of the two pulse trains. In addition, when calculating the integrated timing error from the Nyquist frequency (41.3 MHz for the 82.6 MHz pulse rate) to 100 Hz, the falling response of the detector beyond 20 MHz was corrected to give a white noise floor. The estimated spontaneous emission limited phase noise for the lasers measured here, plotted in green, was determined using the Fourier transform of equation (1). The corresponding spontaneous emission limited integrated timing error did not exceed 1 as until $\sim$200 Hz offset frequency.

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Author contributions

F.X.K. initiated the project. J.G.F. helped with the detection circuit. A.J.B. designed the experimental set-up, acquired the phase noise data and performed the initial data analysis. A.J.B., F.X.K. and J.G.F. interpreted the data.

Additional information

The authors declare no competing financial interests. Supplementary information accompanies this paper at www.nature.com/naturephotonics. Reprints and permission information is available online at http://www.nature.com/reprints. Correspondence and requests for materials should be addressed to F.X.K.