Supporting Online Material

Materials and Methods

For our experimental tests, two of the femtosecond laser synthesizers, NIST-BB1 and NIST-BB2, were built at the National Institute of Standards and Technology (NIST). The other two systems were constructed at the Bureau International des Poids et Mesures (BIPM, Sevres) and the East China Normal University (ECNU, Shanghai) and are named BIPM-C2 and ECNU-C1, respectively. All four synthesizers are based upon passively mode-locked, high repetition rate (800-1000 MHz) femtosecond lasers that utilize independently-pumped titanium-doped sapphire as the gain medium. Beyond that common feature, significant differences exist. Most notably, NIST-BB1 and NIST-BB2 employ four-mirror ring lasers that directly emit a broadband spectrum spanning ~620-1000 nm (S1), while BIPM-C2 and ECNU-C1 employ six-mirror ring lasers (S2) whose outputs are spectrally broadened in a nonlinear microstructure optical fiber to span ~530-1100 nm (S3). Both BIPM-C2 and ECNU-C1 are entirely contained in sealed aluminium boxes with dimensions of 69 × 54 × 23 cm³. In contrast, only some of the components of NIST-BB1 and BB2 are enclosed, but the majority of the parts were in the open laboratory.

The frequency of a reference laser $f_L$ relative to mode N of the femtosecond laser synthesizer is given by $f_L = f_{c0} + N \times f_{rep} + f_0$ (see Fig. S1). Several reports (S4-S6) have described our scheme for the measurement and control of $f_{c0}$ and $f_{rep}$. Here we present some details that are most relevant to the present experiments. For the BIPM-C2 and ECNU-C1 systems, ~150 mW of the output of a mode-locked Ti:sapphire laser (S1) is coupled into a microstructure optical fiber (S3) that is ~30 cm in length. The spectrum emitted from each fiber spans the octave from about 530 nm to 1100 nm. We use the self-referencing technique (S7) to determine $f_{c0}$ by frequency-doubling the
infrared components and heterodyning them with the visible components. Because the Ti:sapphire femtosecond lasers employed in the NIST-BB1 and BB2 systems emit a very broad continuum directly, additional broadening in nonlinear optical fiber is not required. Instead we can measure $f_{\text{ceo}}$ for these lasers by frequency-tripling light emitted near 960 nm and heterodyning it at 320 nm with frequency-doubled light from 640 nm ($S5,S6$). Once it has been measured, we then phase-lock $f_{\text{ceo}}$ for each system to a stable radio frequency synthesized from a hydrogen maser (instability $\sim 2 \times 10^{-13} \tau^{-1/2}$, $\tau$ in seconds), using the pump laser's power as the actuator ($S8$). With $f_{\text{ceo}}$ fixed in this manner, we control the other degree of freedom ($f_{\text{rep}}$) of each comb by measuring and phase-locking the heterodyne beat ($f_b$) between one element of the comb at 456 THz and a cavity-stabilized diode laser having frequency $f_L$. In this case, a piezo-mounted mirror inside the laser cavity is used as the actuator. We continuously monitor the phase-locked signals $f_{\text{ceo}}$ and $f_b$ with high-resolution frequency counters ($S9$) to verify that the performance of the phase-locks does not limit the synthesizer performance. Using this phase-locking scheme, the fluctuations in $f_L$ should dominate the noise in the output of the femtosecond laser synthesizer ($S6$).

**Supporting Text**

Experiments with femtosecond lasers confirmed that $f_{\text{rep}}$ of a mode-locked laser frequency comb extending over 44 THz could be uniform at the level of a few parts in $10^{18}$ ($S10,S11$). Recently, it was further shown that frequency combs created by a nonlinear optical process (difference frequency generation) could be uncertain at a level $<10^{-20}$ relative to the initial frequency comb from which it was produced ($S12$). However, these experiments employed only a single femtosecond laser and did not verify that the actual frequency position of the modes of the comb could be controlled or reproduced relative to a frequency standard or reference oscillator at this level. In order to minimize the possibility of unknown systematic effects, a better test of the
mode-locked laser frequency comb is the comparison of several independent devices. A few tests have been performed using microwave standards to reference femtosecond laser-based synthesizers, which were subsequently compared in the optical domain (S8,S13,S14), resulting in uncertainty limits as low as $5 \times 10^{-16}$. Referencing the femtosecond laser-based synthesizer to an optical standard provides improved stability, and when two such devices were compared agreement was found at the level of $4 \times 10^{-17}$ (S15). In a different approach (S16), Stenger et al. introduced the concept of using the femtosecond laser as a “transfer oscillator.” In this scheme, a clever choice of frequency mixings effectively eliminates the noise properties of the unstabilized femtosecond laser when it is used to determine the ratio of widely separated optical frequencies. This technique has been tested with a single femtosecond laser comb that measures the ratio of the fundamental frequency of a CW laser to its second harmonic with an uncertainty of $7 \times 10^{-19}$. 
**Figure S1:** A. The femtosecond laser synthesizers BIPM-C2 and ECNU-C1 are based on mode-locked Ti:sapphire lasers that have their outputs spectrally broadened in a nonlinear optical fiber to span an octave (red curve). In contrast, the NIST-BB1 and NIST-BB2 synthesizers employ specially designed Ti:sapphire lasers that directly emit a broad continuum (blue curve) that spans ~70% of an octave. B. Beneath the broad envelope, the output spectrum from the synthesizer consists of a comb of optical frequencies with spacing $f_{\text{rep}}$. The offset of the comb from harmonics of $f_{\text{rep}}$ is designated as $f_{\text{ceo}}$. Two phase-lock loops control the position of the comb elements relative to a stable continuous wave (CW) laser having frequency $f_L$. 

![Figure S1](image-url)
References


