Accurate measurement of large optical frequency differences with a mode-locked laser

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We have used the comb of optical frequencies emitted by a mode-locked laser as a ruler to measure differences of as much as 20 THz between laser frequencies. This is to our knowledge the largest gap measured with a frequency comb, with high potential for further improvements. To check the accuracy of this approach we show that the modes are distributed uniformly in frequency space within the experimental limit of 3.0 parts in 10^{17}. By comparison with an optical frequency comb generator we have verified that the mode separation equals the pulse repetition rate within the experimental limit of 6.0 parts in 10^{16}. © 1999 Optical Society of America

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Following the dramatic progress made in recent years in the development of optical frequency standards based on trapped ions[1] or narrow atomic resonances such as the hydrogen $1S-2S$ transition,[2] precise and reliable optical-to-radio-frequency conversion has become the missing clockwork for the construction of a future optical clock. These clocks possess a potential accuracy far beyond the accuracy of the current state-of-the art cesium clocks. In the past, phase-coherent comparisons between optical and radio frequencies were performed with harmonic frequency chains that created successive harmonics from a well-known radio frequency provided, for example, by a cesium atomic clock. Only a few frequency chains that reach all the way to the visible or the UV have been built so far.[2−4]

The use of a mode-locked laser in combination with optical frequency interval dividers[5,6] promises to have the potential to provide an optical clockwork that has the capability to operate reliably for an extended period of time. A phase-locked optical frequency interval divider can reduce an arbitrarily large frequency difference between two input lasers by phase coherently locking a third laser to the precise center of the gap (Fig. 1). If n divider stages are used in cascade, a large optical frequency gap can be reduced by a factor of 2^n until it becomes accessible to radio-frequency counting techniques. Such a chain of divider stages may be used in the future to measure absolute optical frequencies by determining the gap between laser frequencies and their nonlinear conversion products. To measure the laser frequency f this could be the difference between f and its second harmonic $2f$ (which yields $2f − f = f$) or the difference between an auxiliary frequency f_a and the sum frequency $f_a + f$.[5,6] Such a new type of frequency chain allows one to choose a path from the optical region to a beat note in the radio-frequency domain without ever leaving the optical region.[6] The frequencies of the laser oscillators may be chosen such that diode lasers can be used exclusively. However, to reach the radio-frequency domain in this way would require a large number of interval divider stages, unless the largest directly measurable beat frequency is increased. Although the application of optical-frequency comb generators[7] (OFCG's), which

are highly efficient electro-optic modulators, for this purpose has been proposed,[8] we show here that mode-locked lasers make it possible to measure even larger frequency gaps.

The spectrum of a mode-locked laser may be thought of as being due to strong amplitude modulation of some carrier frequency. The modulation frequency is the pulse repetition rate $v_g/2L$, which is the inverse round-trip time in a cavity of length L at a group velocity v_g. Therefore we expect a uniform comb of sidebands or laser modes. Because the frequency of each mode is determined by its phase velocity, which is in general different from v_g, the mode frequencies are expected to be offset from being exact harmonics of the pulse repetition rate. Although femtosecond lasers with nearly quantum-noise-limited timing jitter have been demonstrated,[9] the accuracy and stability of the frequency comb of longitudinal laser modes have so far gone largely unexplored. We report on experiments to check the accuracy of the comb of frequencies emitted by a mode-locked Ti:sapphire laser. For
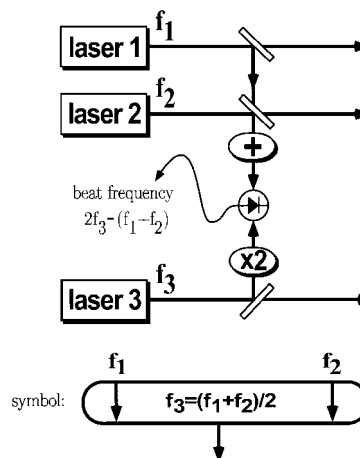

Fig. 1. An optical frequency interval divider receives two input laser frequencies, f_1 and f_2. The frequency of a third laser, f_3, can be determined (or set with a phase-locked loop) accurately relative to the midpoint $(f_1 + f_2)/2$ by detection of the beat frequency between the second harmonic $2f_3$ and the sum frequency $f_1 + f_2$. The symbol is used in Figs. 2 and 3.

this purpose we phase lock two laser diodes as much as 20 THz [=2.9 times the spectral width (FWHM)] apart. We employ a commercial laser (Coherent, Inc., Model Mira 900) with a pulse length of 73 fs as derived from an autocorrelation trace. Assuming a sech pulse shape, the time–bandwidth product of this laser is 1.5 times the Fourier limit. The application of mode-locked lasers for the measurement of optical frequency intervals was demonstrated in elementary form more than 20 years ago,[10] and similar ideas of absolute optical frequency measurements have been presented in Ref. 11. This technique was also used for a recent determination of the absolute frequency of the cesium D_1 line[12] as well as for the first phase-coherent UV-to-radio-frequency link.

To check whether the frequency comb emitted by a mode-locked laser can satisfy the exceptional demands of an all new optical clock we performed a series of experiments. All of them make use of an optical frequency interval divider, as illustrated in Fig. 1. Second-harmonic and sum frequencies are generated in $KNbO_3$ crystals.

Our optical interval dividers are based on extended cavity laser diodes[13] that can be phase locked to a mode of the mode-locked laser by stabilizing their radio-frequency beat signals to a precisely known reference frequency,[14] the so-called local oscillator.

A low-noise beat signal between a particular mode and the laser diode is obtained with the help of an optical grating that preselects some of the modes in the vicinity of the laser diode frequency. Even though the signal-to-noise ratio achieved is typically near 30 dB in a bandwidth of 1 MHz, it is possible that single optical

cycles are lost by the phase-locked loop. To prevent such cycle-slipping events from entering our data we continuously measure the in-lock beat frequencies with a bandwidth (10-MHz bandpass) that differs from the input bandwidth of the phase-locked loop (40-MHz low pass). If the deviation from the given local oscillator frequency is larger than some threshold we do not include this data point in the evaluation, even though some of the rejected data points are probably not connected with cycle-slipping events.

In the first experiment, as sketched in Fig. 2, we phase locked two laser diodes at 822.8 and 870.9 nm to two modes of the mode-locked laser separated by more than 20 THz. The local oscillator frequencies were 20 MHz, with one laser having a positive and the other a negative frequency offset. If the number of modes between the two diode laser frequencies happens to be an odd number, we expect another mode of the frequency comb precisely at the center between the two laser diodes. A third laser diode is then phase locked at 20 MHz below the center mode of the frequency comb. With the help of an optical interval divider we can verify that the central mode is at the expected position in frequency space, confirming the uniform distribution of the modes in the frequency comb. Because of the frequency conversions used for setting up the divider stage we expect a beat frequency of twice the local oscillator frequency. We measured this frequency with a radio-frequency counter (Hewlett Packard Model 53132A) by using gate times of 1, 10, and 100 s, which yield resolutions of 1 mHz, 0.1 mHz, and 10 μHz, respectively. The radio-frequency counter and the local oscillators were all referenced to the same local cesium clock. In combination with the 1-s gate time, a cycle-slipping threshold of 0.5 Hz seemed to be sufficient; the result did not change significantly when this value was decreased. For the other measurements it was then appropriate to reduce the threshold in proportion to the inverse of the gate time, as a possible cycle slip is averaged over this period. The results are summarized in Table 1. The weighted average of the results obtained with the various gate times is a -0.59 ± 0.48 mHz deviation from the expected 40 MHz. This verifies the uniform distribution of the modes within a relative precision of 3.0×10^{-17}. The statistical uncertainty in the measured average does not follow the expected dependence on the inverse square root of the number of readings and the inverse of the counter gate time. The analysis shows that the scatter of data points consists of a nearly Gaussian distribution, whose width indeed follows the expected dependence, and a few outlying points that increase in number with the gate
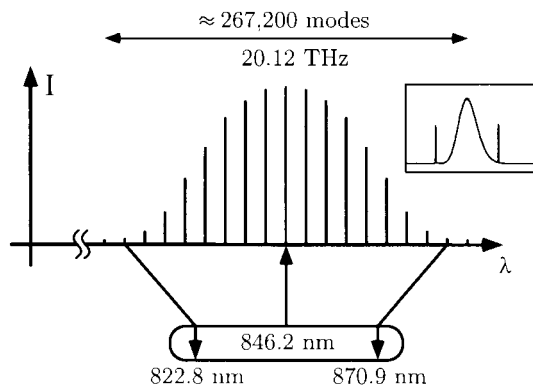

Fig. 2. The uniform distribution of the modes of a mode-locked laser is verified by comparison with an optical frequency interval divider. Inset, measured spectrum of the frequency comb together with the 822.8- and the 870.9-nm laser diodes drawn into it.

Table 1. Results from the Setup of Fig. 2 with Statistical Uncertainties Derived from the Data[a]

Gate Time (s)	Mean Deviation (mHz) from 40 MHz	Relative Deviation	Approved Reading	Cycle-Slip Threshold	Number of Cycle Slips
1	-0.6 ± 2.4	1.2×10^{-16}	8442	0.5 Hz	202
10	-1.93 ± 0.73	9.5×10^{-17}	2936	50 mHz	257
100	0.54 ± 0.67	3.4×10^{-17}	338	5 mHz	179

[a]One additional point has been removed from the 100-s data set that was 825 mHz off but was not detected as a cycle slip.
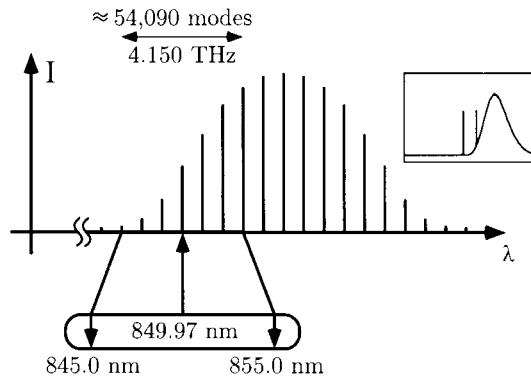
Fig. 3. Verifying the uniform distribution of the modes of a mode-locked laser on one side of its spectrum. Inset, measured spectrum of the frequency comb together with the 845- and 855-nm laser diodes drawn into it.

time. We believe that these points, which lead to the increased statistical uncertainty, are due to cycles that were lost by the counter connected to the divider stage. Unlike the phase-locked beat signals, the beat signal provided by the divider stage was not phase locked but was measured with only one bandwidth (10-MHz bandpass).

The (worst-case) specified systematic uncertainties of the frequency counter are 4 mHz, 0.4 mHz, and 40 μHz for gate times of 1, 10, and 100 s, respectively. Possible drifts of the ambient temperature may cause Doppler shifts by slowly changing the mirror separation by $\sim 20\lambda/K$ at a distance of 1 m. In our experiment this effect may cause a systematic shift of 6 mHz, provided that the ambient temperature changes by 1 K within a measurement time of 1 h.

In a second experiment we reduced the frequency difference between the laser diodes to 4.15 THz and locked them asymmetrically with respect to the spectrum of the frequency comb, as shown in Fig. 3. The measurement is performed in the same way as with the 20-THz frequency gap but with a gate time of 10 s only. With 1703 remaining frequency readings after rejecting 326 suspected cycle-slipping events we find a frequency deviation from 40 MHz of -0.70 ± 0.61 mHz.

To prove that optical frequency combs emitted by mode-locked lasers are useful tools for the precise determination of large optical frequency differences, it is not sufficient to verify the comb spacing. In addition, one has to show that the mode separation can be measured or stabilized to the precision needed. To verify that the mode separation equals the pulse repetition rate, which can easily be measured or phase locked, we performed an actual frequency-difference measurement. The setup used for this purpose is similar to the one shown in Fig. 3 but with the peak of the spectrum of the mode-locked laser tuned to the center between the 845- and 855-nm laser diodes and the local oscillators set with the same sign. We use 328 modes of an OFCG separated by the modulation frequency $f_{mod} = 6.321\,4$ GHz to phase lock the centered laser diode precisely $328 \times f_{mod} - 100$ MHz $= 2.0733192$ THz apart from the 855-nm laser diode (to 849.974 nm). By lock-

ing the 845-nm diode laser frequency at 54,205 modes of the mode-locked laser above the 855-nm laser diode (to 845.007 nm), we expect a beat signal of 44.1 MHz at the divider stage if the pulse repetition rate is set to 76.5 MHz with a phase-locked loop. The short-term stability of the mode-locked laser cavity helps to avoid the increase in phase noise that is due to the large frequency-multiplication factor.[15] To reduce the multiplication factor we detected the 100th harmonic of the pulse repetition rate with a fast detector and phase locked it to 7.65 GHz provided by a synthesizer (Hewlett-Packard Model 8360). With a total of 1859 nonrejected readings and 166 suspected cycle slippings at a counter gate time of 10 s we found a frequency deviation from 44.1 MHz of 2.2 ± 2.5 mHz. This confirms that the pulse repetition rate equals the mode separation with an accuracy of at least 6.0×10^{-16}. The fourfold larger statistical uncertainty compared with the result of Table 1 may be due to additional phase fluctuations caused by the OFCG.

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