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Precision stabilization of femtosecond lasers to high-finesse optical cavities

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We report on direct stabilization of a femtosecond laser by a high-finesse passive optical cavity. Detailed comparison of two distinct stabilization schemes leads to new understanding of the optimum conditions for cavity stabilization and limitations on the ability to transfer the frequency stability of the cavity to the microwave domain. The stability of the frequency comb is explored in both the optical and the radio frequency domain. With an independent, stable cw laser, we verify that the linewidth and stability of the optical comb components, respectively, reach below 300 Hz and 5×10^{-14} at 1 s averaging time, both limited by the reference cw laser.

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Stabilization of femtosecond (fs) lasers has had a revolutionary impact on the fields of optical frequency metrology [1–3] and ultrafast science [4,5]. Improving the precision in which the repetition rate and carrier-envelope phase of ultrashort pulses can be controlled will also aid studies in coherent light–matter interactions [6]. Decades of research concentrating on the stabilization of cw lasers to ultrastable optical cavities has greatly improved spectroscopic resolution [7] and helped advanced the entire field of precision measurement. Previous work has also focused on the precision locking of FM sidebands to stable cavities in efforts to link the cavity free-spectral range (FSR) to optical frequencies (e.g., [8–10]). Recently published results have extended these ideas to cavity stabilization of fs lasers [11,12]. The additional complexity arising from simultaneous stabilization of the two degrees of freedom of an fs laser requires thorough considerations of the control loop designs and the restrictions due to dispersive phase shifts inside the reference cavity. It is under such an extensive and exciting context of scientific topics that we carefully analyze the performance of a fs laser directly stabilized to a high finesse, passive optical cavity. The broader goal of this work is to not only elevate the stability of ultrafast lasers to levels comparable with state-of-art cw lasers, but also enable a number of applications based on external "fs enhancement cavities" to provide attractive field-enhancement effects and sensitive detection capabilities in nonlinear optics and spectroscopy [13,14].

To develop a deeper understanding on the fundamental issues of cavity-fs pulse interaction, we have taken two different but related approaches to stabilize an fs laser to a high-finesse optical cavity. The comparison between the respective performances, as detailed in this paper, highlights fundamental limitations on the ability to detect fluctuations of the frequency comb's spacing directly from the external cavity. Conditions for optimizing the signal-to-noise ratios (S/N) of the recovered cavity error signals in the presence of the dispersive phase shifts from standard quarter-wave dielectric mirror coatings are described.

The absolute frequency of any single component of the "fs comb" can be expressed as $\nu_m=mf_{\mathrm{rep}}+f_{\mathrm{ceo}}$, where ν_m is the optical frequency of the mth comb component, f_{rep} is the repetition frequency of the pulse train, and f_{ceo} is the offset frequency resulting from the evolving pulse-to-pulse carrier-envelope offset phase [15]. To characterize and stabilize the entire fs comb necessitates sensitive detection and control of two different parameters related to f_{rep} and f_{ceo}. The schematics shown in Fig. 1 depict two cavity-locking configurations explored in this paper and their associated spectral regions (shaded areas) used to acquire two error signals (e_1 and e_2) for locking the fs comb. A mode-locked Ti:sapphire laser producing <20 fs pulses was constructed to match the length of the 39.5 cm reference cavity ($f_{\mathrm{rep}}=380$ MHz) consisting

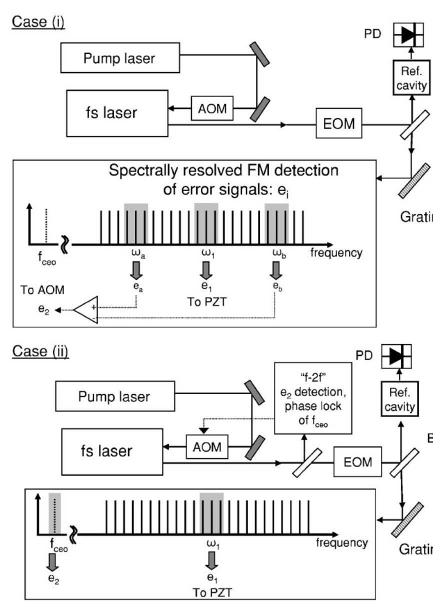

FIG. 1. Stabilization of a femtosecond laser to a passive reference cavity by (i) "all-cavity locking," in which both degrees of freedom of the fs frequency comb are detected and stabilized to the cavity, and (ii) with cavity locking assisted by independent detection and phase locking of f_{ceo} using microstructure fiber for spectral broadening and an "f-$2f$" interferometer for detection of the beat note. Shaded regions indicate spectral positions at which the fs comb is locked in each case. e_i, error signal recovered at spectral region ω_i; BS, beam splitter; PD, photodiode plus spectrometer.

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of an ultra-low-expansion (ULE) spacer with two ULE substrate-based low-loss mirrors optically contacted on both ends. The finesse of the cavity at 800 nm is $\sim$3000. The cavity is mounted inside a vacuum chamber for thermal and vibration isolation. The laser output is mode-matched to the cavity after passing through an electro-optic phase modulator (EOM) placing sidebands on each comb component. Both f_{rep} and f_{ceo} need proper adjustments to optimize frequency overlap between the comb components and the corresponding cavity modes [12]. The light reflected from the cavity is spectrally dispersed with a grating and error signals from the cavity are obtained at different spectral regions resonant with the cavity (ω_i) using standard Pound-Drever-Hall techniques [16]. Each error signal from the cavity is composed of over $\approx$2000 frequencies detected over a $\approx$2 nm region.

The finite (S/N) in the discriminator of any feedback loop, ultimately limited by fundamental shot noise in the photodetection process, limits the precision with which one can lock a particular frequency. The expected fractional frequency instability at an optical frequency ν under servo control can be expressed as $\sigma_y=(\Delta\nu_c/\nu)(S/N)^{-1}$, where $\Delta\nu_c$ is the detected cavity linewidth at ν. In both cases (i) and (ii), a group of frequencies near 815 nm are used to obtain the first error signal e_1. This signal is sent through a loop filter and fed back to a piezoelectric transducer (PZT) attached to an end mirror of the laser for control of the laser cavity length, locking the average optical frequency of the comb components near ω_1 to the corresponding collection of cavity modes. Fluctuations not resolved at e_1 [denoted as $\epsilon_1 \equiv \Delta\nu_c(S/N)^{-1}$] can be expressed in terms of fluctuations in f_{rep} and f_{ceo} as $m_1\delta f_{\mathrm{rep}}+\delta f_{\mathrm{ceo}}=\epsilon_1$ where m_1 is the mode number of the frequency comb centered at ω_1. The primary difference between cases (i) and (ii) is the method in which the second degree of freedom of the fs comb is obtained. In the "all-cavity" locking configuration of case (i), the second error signal is generated by detecting differences in signals e_b and e_a: $e_2\equiv(m_bf_{\mathrm{rep}}+f_{\mathrm{ceo}})-(m_af_{\mathrm{rep}}+f_{\mathrm{ceo}})-\Delta f=\Delta m f_{\mathrm{rep}}-\Delta f$, where $\Delta m=m_b-m_a$, $\Delta f=(\omega_b-\omega_a)/(2\pi)$, and we make a somewhat idealized assumption of perfect common mode rejection of f_{ceo}. To first order, e_2 only detects changes in f_{rep}. This signal is sent through a second loop filter to an acousto-optic modulator (AOM) for control of the laser pump power, thereby locking f_{rep} to the cavity. More details on this can be found in previous papers using a similar stabilization scheme [11,12]. It should be noted that in principle only two spectral regions need be detected in case (i), with the sum and difference frequencies providing the two error signals. The use of a third detector was chosen to provide a more direct comparison between cases (i) and (ii), and to better isolate the two error signals in case (i) so as to minimize coupling between the independent feedback loops. The noise (ϵ_2) remaining in e_2 under locked conditions permits f_{rep} to fluctuate as $\Delta m\times\delta f_{\mathrm{rep}}=\epsilon_2$. In the "$f_{\mathrm{ceo}}$-assisted" cavity locking of case (ii), e_2 is generated by measuring f_{ceo} directly using a microstructure fiber for spectral broadening [17] and an "f-$2f$" interferometer to detect the beatnote in the standard way [2]. A stable frequency synthesizer is used as a reference to phase lock f_{ceo}, again by controlling the pump laser power with the AOM. The residual noise seen at e_2 is $\delta f_{\mathrm{ceo}}=\epsilon_2$.

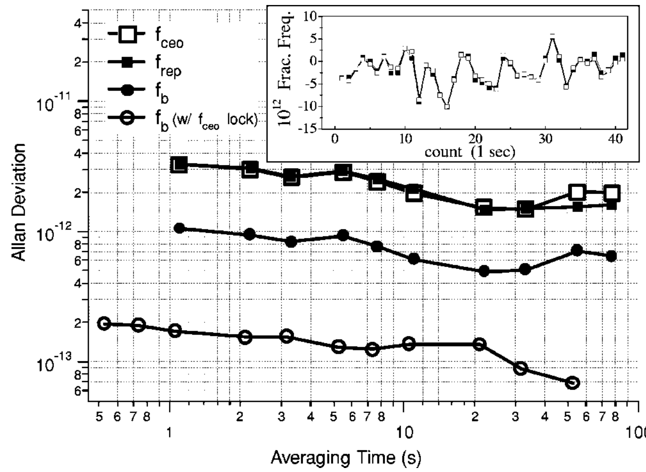

FIG. 2. Measured Allan deviations for the cavity-stabilized fs comb. Open (filled) squares show measurement of $f_{\mathrm{ceo}}(f_{\mathrm{rep}})$ under case (i). The inset shows a portion of simultaneous counts of fractional frequency fluctuations for f_{rep} and $-f_{\mathrm{ceo}}$. Circles correspond to measurements of the beat note between the fs comb and an iodine stabilized cw laser at 1064 nm under case (i) (filled) versus case (ii) (open).

The fs comb stability in cases (i) and (ii) is compared in both optical and rf domains by evaluating heterodyne beatnotes against other relevant standards. Approximately 2 mW from the fs laser is incident on the reference cavity. The overall instability for both cases can in principal be reduced by simply increasing the total optical power. The beat signals are characterized in terms of the resulting linewidth and the frequency stability that is evaluated by determining Allan deviations from the frequency counting records. Without loss of information about the stabilization result, we have removed a slow linear drift associated with the reference cavity (typically 30 Hz/s). The Allan deviation results are shown in Fig. 2. For case (i), the stability of the entire fs comb is assessed by counting both f_{rep} (filled squares) and f_{ceo} (open squares) directly. In order to count f_{rep} with sufficient resolution, it is first mixed down to $\approx$10 kHz using an independently characterized, stable rf reference before being sent into a frequency counter. For these measurements, $\Delta f=25$ THz(52 nm). The inset of Fig. 2 shows a portion of the counts made simultaneously of $\delta f_{\mathrm{rep}}/f_{\mathrm{rep}}$ and $-f_{\mathrm{ceo}}/(m_1f_{\mathrm{rep}})$ from which the Allan deviation is determined. These two quantities establish the stability and precision with which any single mode of the fs comb can be measured and accessed. Of course, the actual fractional frequency instability of any particular optical comb component can be better than either rf quantity alone, depending on correlations in f_{rep} and f_{ceo}. This can be seen by counting a third beat note in the optical domain between a single mode of the fs comb and an iodine-stabilized cw laser at 1064 nm. The fractional frequency instability of this beat note (filled circles) is less than that measured for either f_{rep} or f_{ceo}. The iodine spectrometer consists of a frequency doubled cw Nd:YAG laser locked to the $R(56)$ 32-0:a10 component of I_2 at 532 nm. This "molecular iodine clock" has been described in detail previously and was shown to be stable to better than 5 parts in 10^{14} at 1

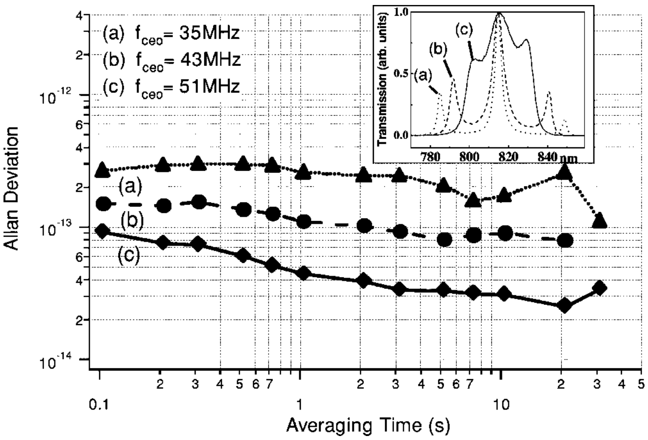

FIG. 3. Allan deviation of beat note at 1064 nm between an iodine-stabilized cw laser and cavity-stabilized fs comb with independent phase locking of f_{ceo} to three different values. The inset shows the spectrum of the fs laser transmitted through the reference cavity in each case.

second of averaging [3]. These measurements are consistent with the expected discriminator resolution (ϵ_i) described earlier. Rearranging the previous expression for ϵ_1 and dividing both sides by $m_1 f_{rep}$ we get $-\delta f_{ceo}/(m_1 f_{rep}) = (\delta f_{rep}/f_{rep}) - \epsilon_1/(m_1 f_{rep})$. By noting that $\delta f_{rep} = \epsilon_2/\Delta m$ and $\epsilon_1 \sim \epsilon_2$, we can assume $\epsilon_1/(m_1 f_{rep}) \ll \epsilon_2/(\Delta m f_{rep})$ and write $-\delta f_{ceo}/(m_1 f_{rep}) = \delta f_{rep}/f_{rep}$. This expression explains the strong (anti)correlation between the corresponding fractional instabilities of f_{ceo} and f_{rep} shown in the inset of Fig. 2. Although both error signals are tightly locked, the dominant noise is due to symmetric fluctuations of the fs comb centered at ω_1 that are not resolved by e_2. This also accounts for the lower instability measured at the optical frequency (282 THz) as it is spectrally located closer to ω_1 (2π 368 THz) than is f_{ceo}. The frequency fluctuation at an arbitrary mode number n of the comb is $\delta\nu_n = n\delta f_{rep} + \delta f_{ceo}$. From the above discussion we can replace δf_{ceo} and rewrite this as $\delta\nu_n = n\delta f_{rep} - (m_1 \delta f_{rep})$ or $\sigma_y = \delta\nu_n/\nu_n = (\delta f_{rep}/f_{rep})|n - m_1|/n$. This relates the fractional frequency fluctuations for the nth component of the fs comb to that in f_{rep}. For the measured beatnote at 282 THz, we would then expect a measured fractional frequency instability $\approx(368-282)/282 = 0.3$ times lower than that measured for f_{rep}, in good agreement with the observed results of Fig. 2.

In case (ii), f_{ceo} is independently phase-locked to a stable frequency synthesizer. The tightly phase-locked loop shows no cycle-slips, leading to small residual errors ϵ_2 in f_{ceo}, or $\epsilon_2 \ll \epsilon_1$. At present, the resulting stability of f_{rep} is below the limit of rf reference standards used for its evaluation. An out-of-loop measurement of the comb stability in the optical domain is made by again counting the beatnote between the fs comb and the iodine stabilized cw laser at 1064 nm (see Fig. 2, open circles), showing a substantial improvement over that of case (i). The fractional frequency stability of every comb component, and therefore also f_{rep}, should be at the same level as measured at 1064 nm as shown in Fig. 2. The discrepancy in performance between cases (i) and (ii) is a result of the limited sensitivity in detection of e_2 when attempting to lock f_{rep} directly to the cavity. Given the relation $\epsilon_2 = \Delta m \delta f_{rep}$ for case (i), we can write a simple expression for the fractional frequency instability that can be expected when the reference cavity serves as the discriminator for f_{rep}: $\delta f_{rep}/f_{rep} \approx \epsilon_2/\Delta f$. To determine ϵ_2, we note that the stability of the comb components near ω_1 (at 1 s or longer) is roughly the same for both configurations in Fig. 1. Using the measured fractional frequency instability of $\sigma_y = 1.8\times10^{-13} = \delta\nu/\nu \approx 2\pi\epsilon_1/\omega_1$ yields an instability in locking point precision of 66 Hz at 1 s. If approximately the same imprecision is obtained at the different error points we can write $\epsilon_1 \approx \epsilon_a \approx \epsilon_b$. When both e_a and e_b are detection limited there is no correlation between them and $\epsilon_2 \approx \epsilon_1\sqrt{2}$. With $\Delta f = 25$ THz and using the above upper limit for the control loop's ability to discriminate against fluctuations in f_{rep} results in an estimated fractional frequency instability of $\approx 66\sqrt{2}$ Hz/25 THz $\approx 4\times10^{-12}$, in close agreement with the measured results. In general, when locking the fs comb to the cavity in case (i), the fractional frequency instability of f_{rep} will be approximately $\omega_1/(2\pi\times\Delta f)$ times greater than that of the optical frequencies near ω_1, relative to the cavity.

One could expect to improve the stability of f_{rep} by simply increasing Δf. However, the ultimate performance is limited by the cavity's dispersive properties. When multiple error signals need to be simultaneously detected, as in case (i), a trade-off arises in the optimization of a single error signal, say e_1, versus optimization of both e_1 and e_2. The dispersive properties of such mirrors can be estimated or measured directly (e.g., [10,18]) and are shown to exhibit a parabolic variation in the cavity FSR about some frequency ω_c, near the wavelength center of the mirror's coating. This characteristic variation in the FSR governs which regions of the equally spaced fs comb can be simultaneously resonant with the cavity. In order to detect three separate spectral regions in case (i), the central locking position ω_1 must be centered near ω_c. The inset in Fig. 3 shows the regions of the incident laser spectrum transmitted through the reference cavity when the fs comb is locked at $\omega_1 = \omega_c$ (central peak) for the three different values of f_{ceo} shown. Since the center of the fs comb is fixed at ω_1, each value of f_{ceo} corresponds to a unique value of f_{rep}. From curves (a)–(c) in the inset, it is evident that the position and separation of ω_a and ω_b will be determined by the value of f_{ceo}. Figure 3 shows the measured Allan deviation of the beatnote between the fs comb and the iodine stabilized YAG laser at 1064 nm for the three different values of f_{ceo} stabilization [case (ii)]. When f_{rep} closely matches the FSR of the reference cavity at ω_1 (curves c), the maximum bandwidth is transmitted through the cavity ($\approx$35 nm FWHM) and the measured fractional frequency instability of only $\sigma_y(1\text{ s}) = 4.5\times10^{-14}$ is limited by that of the iodine stabilized cw laser. Therefore, the best performance for the cavity stabilized fs laser is achieved when a single error signal at ω_1 is optimized (by properly adjusting f_{rep}) and used to stabilize the fs comb assisted by independently phase locking f_{ceo}. In contrast, when f_{ceo} is adjusted such that spectral regions separated by $\approx$55 nm are resonant with the cavity [curves (a)], the measured instability at 1 s is more than 6 times worse and the transmitted spectrum near ω_1 is much narrower ($\approx$4 nm) due to the local mismatch of f_{rep} with the cavity FSR at these positions. Curves (b) correspond

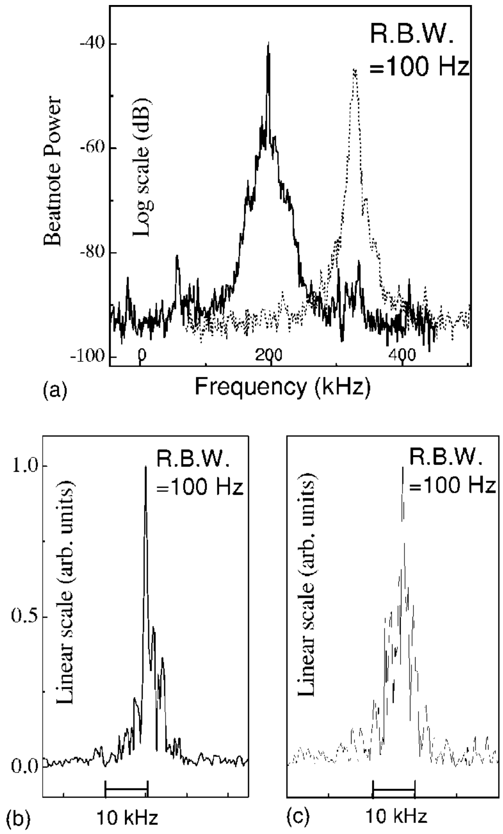

FIG. 4. Optical heterodyne beat notes between cavity-stabilized fs comb and iodine-stabilized cw laser at 1064 nm. Graph (a) shows beat notes when the fs comb is stabilized under case (i) dotted line, and case (ii) solid line. Graph (b) shows beat note case (ii) on a linear scale. More than 50% of the optical power is contained within a 300 Hz bandwidth. For reference, (c) shows the beat note between an independent cavity-stabilized cw laser and the second harmonic of the iodine stabilized cw laser. The linewidths in (b) and (c) are comparable, indicating that the true linewidth of the cavity-stabilized fs comb is not resolved by the iodine-stabilized laser.

to the conditions used for the data collected in Fig. 2. This variation in the measured stability is attributed to the degradation of the retrieved error signal e_1 when the fs comb modes are not optimally aligned with the cavity modes at ω_1.

The significance of these results for the all-cavity locking configuration is that by only increasing Δf to improve the stability of f_{rep} simultaneously degrades the quality of the error signals at e_1, e_a, and e_b. To first order these two effects cancel and the same fractional frequency instability of $\approx(3-4)\times 10^{-12}$ was measured for f_{ceo} and f_{rep}, regardless of the choice in Δf.

The linewidth of the cavity stabilized fs laser is measured at 1064 nm by mixing a single mode of the comb (after broadening the spectrum through a piece of microstructure fiber) with the fundamental of the iodine stabilized YAG laser. The recorded beat notes for cases (i) and (ii) are shown in Fig. 4(a). The linewidth of the beat note is clearly much narrower when the cavity stabilization is assisted with the f_{ceo} phase lock [case (ii)] as expected. Figure 4(b) shows a zoomed-in trace of the beat note for case (ii) on a linear scale.

In summary, the frequency stability of a high finesse reference cavity has been faithfully transferred to the entire fs comb using a single error signal from the cavity when f_{ceo} is independently phase locked to a stable rf source, which currently requires an octave-spanning spectrum. When locking both degrees of freedom of the fs laser directly to the reference cavity, the ability to simultaneously optimize error signals in different spectral regions is restricted by the parabolic variation of the cavity FSR, characteristic of the simple quarter-wave stack dielectric mirrors employed. Alternative configurations can be envisioned to perhaps mitigate this problem, but usually at the expense of considerable increase in complexity of the system. The current limitations demonstrated in this paper exemplify the need for dispersion controlled and low loss mirrors across large spectral bandwidths. Reference cavities incorporating these mirrors will provide a simple and effective means to tightly stabilize the fs laser to levels comparable with current state-of-art cw laser systems. The development of such “fs reference cavities” is also part of a larger effort to study the coupling and enhancement of ultrashort pulses in external high finesse cavities to access high field strengths with pulses directly from the oscillator [13].

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