# **Carrier-Envelope Phase Stabilization of Modelocked Lasers**

Tara M. Fortier, David J. Jones, Scott A. Diddams<sup>\*</sup>, John L. Hall<sup>†</sup>, Jun Ye<sup>†</sup> and Steven T. Cundiff<sup>†‡</sup>

JILA, University of Colorado and the National Institute of Standards and Technology, Boulder, CO 80309-0440

# Robert S. Windeler Bell Laboratories, Lucent Technologies, Murray Hill, NJ 07733

## ABSTRACT

Carrier-envelope phase stabilization of few cycle optical pulses has recently been realized. This advance in femtosecond technology is important in both extreme nonlinear optics and optical frequency metrology. The development of air-silica microstructure fiber was an enabling technology for performing phase stabilization. The microstructure fiber provides a group-velocity zero within the spectral region that can be accessed with titanium sapphire lasers. The presence of a group-velocity zero allows the output spectrum of the laser to be broadened so that it exceeds one octave, which is necessary for our phase stabilization technique. We will present the basics of the technique and role played by the microstructure fiber. Measurement of the nonlinear phase shift in the fiber is also performed to determine the contribution of amplitude to phase noise conversion.

Keywords: ultrafast lasers, microstructured fiber, carrier-envelope phase, phase control, metrology

## **1. INTRODUCTION**

Over the past decade two seemingly disparate fields were developing with increasing rapidity: single frequency laser stabilization and ultrafast laser technology. Highly stabilized continuous wave (CW) lasers, with near delta function line shapes, are employed in high precision measurements of fundamental physics and spectroscopy. Ultrafast physics, on the other hand, takes advantage of the high peak powers of the near delta function time profiles of the pulses emitted by modelocked lasers to explore the world of nonlinear optical physics and observe physics on very short time scales. The subject of this paper is how two seemingly different fields have come together to impact optical frequency metrology and extreme nonlinear physics.

Optical frequency synthesis has been of great interest to the scientific community since the development of the first laser. An early use of stabilized lasers was an interferometric measurement of the speed of light.<sup>1</sup> Since then, the technology of laser frequency stabilization has been refined and simplified making it as an indispensable tool in modern optics labs. Many current projects continue to push the limits of measurement science such as gravitational wave detection, tests of special relativity and high precision spectroscopy.

One goal of the metrology community is to obtain absolute measurements of optical frequencies. Such a measurement is accomplished by comparison to the hyperfine transition in Cesium at 9.193 GHz. An optical transition, for example that of the HeNe laser transition, is at frequency of ~ 500THz. How does one make a comparison between an optical transition and a Cesium transition that differ by several orders of magnitude? One of the first methods to overcome this frequency gap was the measurement of secondary optical standards.<sup>1-6</sup> Such optical standards are measured using expensive, high maintenance, frequency chains that link a high harmonic of the microwave standard to an optical transition via several intermediate oscillators using phase locked loops. Synthesis of optical frequencies from secondary standards, however, is not a trivial process. Consequently, several techniques have been devised over the years to deal with this particular problem.<sup>7-11</sup>

<sup>&</sup>lt;sup>\*</sup> Current address: Time and Frequency Division, National Institute of Standards and Technology, Boulder, CO 80303

<sup>&</sup>lt;sup>†</sup> Staff Member, NIST Quantum Physics Division

<sup>&</sup>lt;sup>‡</sup> e-mail: cundiffs@jila.colorado.edu

The time domain pulse train of a femtosecond laser corresponds to a frequency comb in the frequency domain. The frequency spacing between adjacent comb lines, determined by the repetition rate of the laser, is on the order of ~100 MHz, a radio frequency (RF) that can be directly compared to a Cesium clock. If the frequency of one of the comb lines is determined absolutely and the comb spacing is stabilized, in essence one has created an optical ruler. This idea was presented in the 1970's.<sup>12</sup> Until recently, however, stabilization of such a comb required it to be referenced to a secondary standard. By employing a self-referencing technique for stabilizing the frequency comb emitted by a femtosecond laser, described below, it is possible to perform a direct microwave-to-optical comparison using a single optical oscillator and a microwave clock. This technique, apart from significantly simplifying the stabilization process, also reduces the uncertainty in the stabilization process, by eliminating the need for stabilized intermediate oscillators.

Another consequence of this technique is phase control of femtosecond pulses in the time domain. Control of the carrierenvelope phase of a light pulse is control over its electric field. Most nonlinear processes are sensitive to the intensity of light pulses only. However, the advent of few-cycle pulses has made it possible for processes that are sensitive to the actual electric field of each pulse to be studied. These processes often display a threshold dependence on the electric field.

The method of self-referencing was first made possible with the introduction of the air-silica microstructure fiber.<sup>13</sup> Its unique dispersion characteristics allow for extreme external spectral broadening of short pulses from a Kerr lens modelocked Titanium:Sapphire (KLM Ti:S) laser. In this paper we will explain the role of the air-silica microstructure fiber in our phase control experiments. To understand the unique behavior of this fiber we will present a method for characterizing the nonlinear behavior of the fiber by measuring the nonlinear phase induced in the fiber as a function of laser intensity.

# 2. INFLUENCE OF THE CARRIER ENVELOPE PHASE ON THE SPECTRUM OF MODELOCKED LASERS

To understand how frequency domain techniques can lock the carrier-envelope phase, it is necessary to understand the correspondence between the time and frequency domain pictures of the pulses emitted by modelocked lasers.

### 2.1 Time and Frequency Domains

Modelocking is achieved by establishing a fixed phase relationship between each lasing longitudinal mode in a laser. For a laser to lase on multiple longitudinal modes it is necessary that each mode be on resonance within the cavity. The resonance condition imposed on the gain spectrum of the laser causes the laser spectrum to consist of discrete, equally spaced, frequencies. Because the cavity length is an integer number of a wavelength the frequencies making up the spectrum be integer multiples of the laser repetition rate, i.e.,  $f_n = n f_{rep}$  where  $f_{rep}$  is one over the round trip cavity time. (The cavity modes of a laser, because of dispersion, do not have equally spaced frequency components. However, once modelocking begins, the nonlinear processes within the gain medium act to equalize the mode spacing.) The coherent interference of these frequencies results in a periodic formation of a train of pulses in the time domain, separated in time by one over the repetition rate of the laser, i.e.,  $\tau = 1/f_{rep}$ . Each of these pulses can be described mathematically as a field with carrier frequency,  $\omega_c$ , amplitude modulated by an equal processing in  $(\alpha)$ . In general the carrier and the equal processing at different velocities.

amplitude modulated by an envelope, see Figure 1 (a). In general the carrier and the envelope travel at different velocities because of dispersion in the laser oscillator. This results in a shift of the pulse-to-pulse carrier phase with respect to the

envelope,  $\Delta \phi = \left(\frac{1}{v_g} - \frac{1}{v_p}\right) l_c \,\omega_c \, \text{mod} \, 2\pi$ . In the frequency domain, a pulse to pulse phase shift results in rigid shift of the

frequency comb by an amount  $\delta = f_{rep} \Delta \phi / 2\pi$ . Thus, the result is that dispersion in the cavity acts to shift the frequency of the entire comb such that each individual comb line can be expressed as an integer multiple of the repetition rate of the laser plus a frequency offset, that is  $f_n = \delta + n f_{rep}$ , see Fig. 1 (b).



Figure 1 (a) Time-domain picture of the pulses from a modelocked laser and (b) the corresponding frequency comb, where  $\delta$  is the comb frequency offset and  $f_{rep}$  is the comb spacing corresponding to the lasers' repetition rate.

#### 2.2 Phase stabilization using frequency domain techniques

Frequency measurements require stabilization of the entire frequency comb. For this, it is necessary to fix the comb spacing,  $f_{rep}$ , and the frequency offset of the comb,  $\delta$ . This is typically accomplished by locking a single comb line to a known optical frequency and locking the cavity length of the laser. This method would allow us to take advantage of the highly developed techniques used in single-frequency laser stabilization. However, the addition of another stabilized oscillator apart from complicating matters, adds another source of uncertainty. Also, carrier-envelope phase stabilization requires not only control of  $\delta$ , but also some method to measure it. The above method for stabilization fixes  $\delta$  but does not yield its value. A more elegant approach<sup>14</sup> that stabilizes and measures frequency offset using a single frequency comb is a self-referencing technique described here.

As mentioned above, the frequency of each comb line is given by  $f_n = \delta + n f_{rep}$ . The field of each comb line can be expressed as  $\exp[i(kz + \omega_n t + \phi_o)]$ , where  $\omega_n = 2\pi(\delta + n f_{rep})$  (For the present discussion the phase term contributed by kz and  $\phi_o$  will be ignored because they represent an overall time independent phase). We may experimentally determine  $\delta$ by measuring the heterodyne beat between a comb line on the high frequency extreme of the spectrum and the second harmonic of a comb line at the low frequency end of the spectrum. The resulting beat frequency is  $|f_{2n} - 2f_n| = \frac{1}{\delta} - 2nf_{n-1} = \delta$ 

$$\left| (\delta - 2n f_{rep}) - 2(\delta - n f_{rep}) \right| = \delta .$$

Once  $\delta$  is measured its stabilization is enabled through control of the pulse group delay in the cavity. Understanding how this is done requires a basic knowledge of the layout of a Ti:S laser. For modelocking to be possible, dispersion in the cavity needs to be minimized. Since the lasing medium is a source of positive dispersion it is necessary to introduce negative dispersion, usually in the form of a prism pair, for compensation. In a Ti:S laser the prism pair is placed between one of the curved mirrors and the end mirror of the laser. After light has traversed both prisms the spectrum of the light is spatially dispersed on the end mirror. Tilting of this mirror introduces a time delay to each longitudinal mode resulting in phase that is linear in frequency. This can be understood by consider the following: each comb line has a phase term given by  $\omega t$ . A

shift in frequency can be obtained by introducing a time delay, in other words  $\omega_n(t+a) = \omega_n t + a\omega_n = \omega_n t + \phi$ . If it is possible to change the cavity length of the laser in such a way that each modelocked frequency experiences an equal phase shift, then we can control the group delay.

Stabilization of the frequency offset has an important consequence in the time domain: stabilizing the carrier-envelope phase. Recall that  $2\pi\delta = f_{rep}\Delta\phi$ , so that in fixing  $\delta$  we are also fixing  $\Delta\phi$ . Once  $\delta$  is measured it is stabilized using phaselocked loops that compare its value to a synthesized RF frequency. The synthesizer to which we lock  $\delta$  has an external input from a photodiode measuring the laser's repetition rate. From this input the synthesizer derives fractional multiples of  $f_{rep}$ , that is  $\delta$  is locked to a synthesized frequency  $(N/16) f_{rep}$ , where N is an integer that can be selected anywhere from 1 to 16. This allows us to lock the pulse-to-pulse carrier envelope phase slip,  $\Delta\phi = 2\pi N/16$ , at sixteen different positions between 0 and  $2\pi$ . For example, if N = 4 then every fourth pulse has the same phase.

## 2.3 External Broadening

The self-referencing technique can only be realized if the bandwidth of the frequency comb spans an optical octave, i.e. the high frequency components have twice the frequency as the low frequency components. Previously, generation of an optical octave incurred a lot of amplitude and phase fluctuations. KLM Ti:S lasers currently exist that can produce an optical octave straight out of the laser oscillator.<sup>15, 16</sup> The necessary technology, however, is quite involved and not easily accessible. A more common technique that is employed to externally broaden the spectra of a modelocked laser is self phase modulation (SPM) in optical fibers. An important detail to note is that external broadening of a frequency comb via SPM preserves the periodicity and pulse-to-pulse phase information of the incident frequency comb.

Self phase modulation occurs in a medium with a nonlinear index of refraction, i.e. a third order optical nonlinearity. SPM broadens the spectrum of an incident frequency comb, which can be understood in the frequency domain as four-wave mixing between the frequency comb components producing new wavelengths. For any nonlinear process, high intensities are needed. The advantage of optical fiber is that the pulse is confined to a small mode field diameter over an extended interaction range. Therefore, femtosecond pulses can maintain high intensities over the length of the fiber as long as dispersion does not cause excessive broadening of the incoming pulse. Recently, the development by Lucent Technologies<sup>\*</sup> of air-silica microstructured fiber has



Figure 2 Initial pulse from a KLM Ti:S laser and the output generated by broadening in air-silica microstructure fiber.

<sup>\*</sup> Lucent Technologies is a trade mark name, used here for identification purposes only and does not constitute an endorsement by the authors or their institutions.

made it possible to produce an octave of bandwidth with an ordinary KLM Ti:S laser. This is achieved by dispersion shifting the zero velocity dispersion point of the fiber to a shorter wavelength such that it lies at 780 nm, as opposed to ~1300 nm in ordinary silica fiber. The fiber also supports single-mode propagation. As a result the pulse can propagate with significantly less dispersion, thereby maintaining higher peak intensities over longer distances than were possible in ordinary fiber. A typical input spectrum from a KLM Ti:S laser and the continuum generated by SPM in air-silica microstructure fiber are shown Figure 2.

## **3. EXPERIMENT**

This section describes the experimental details of frequency-domain phase stabilization of femtosecond pulses using self-referencing and the techniques used in time-domain verification of this stabilization.

#### 3.1 Laser

The central element in our experiments is a passively modelocked KLM Ti:S laser that generates a 90 MHz pulse repetition rate. The spectral bandwidth of the pulses generated is approximately 70 nm and is centered at 800 nm, corresponding to a transform-limited pulse width of ~10 fs. The baseplate of the laser oscillator is temperature controlled and the laser itself is enclosed in an airtight box to isolate it from external perturbations. Stabilization of the repetition rate is achieved by mounting the output coupler of the laser on a piezoelectric actuator. The actuator is then controlled by a phase-locked loop that compares the repetition rate, as measured by a fast silicon photodiode, to an external microwave clock.

Stabilization of  $\delta$  is achieved by placing the end mirror of the laser on an actuator that tilts about the horizontal. The tilting actuator is controlled by an independent phase-locked loop that compares the measured value of  $\delta$  to a synthesized RF frequency. A schematic is represented in Figure 3.



Figure 3 Experimental setup for measuring the absolute position of the femtosecond comb via a self-referencing technique.

#### 3.2 Frequency Domain Self-Referencing Techniques For Carrier-Envelope Phase Stabilization

The phase measurement we use to determine  $\delta$  is accomplished using an interferometer that compares the frequency between the doubled low frequency end of the comb to the high frequency end of the comb. Light from the laser is used for phase control measurements and for cross correlation measurements that are described below. In the self-referencing arm of the experiment, about 100 mW of power are injected into an air-silica microstructure fiber with approximately 50% coupling efficiency. No external dispersion compensation is used to recompress pulses before they are injected into the fiber. With an input bandwidth of 70 nm we generated an output bandwidth of greater than 550 nm, sufficient for the self-referencing technique. Once the light exits the fiber it is split by a dichroic beamsplitter that transmits 1060 nm and reflects 530 nm. The reflected green light at 530 nm is put through an acoustooptic modulator (AOM) that shifts the frequency of the entire comb by  $f_{AOM}$  such that the comblines have frequencies  $f_n = \delta + n f_{rep} + f_{AOM}$ . The transmitted 1060 nm light is focused into a 1 cm long  $LiB_3O_5$  (LBO) doubling crystal that generates ~ 3mW of green light. Its comb lines are given by  $f_m = 2\delta + mf_{rep}$ , where m is an integer. (Note that the comb spacing between adjacent comb lines is still  $f_{rep}$ . This is because frequency doubling of a comb is really four-wave mixing between comb lines.) The doubled 1060 nm arm and the shifted 530 nm arm are then recombined at a polarizing beamsplitter. Before this beamsplitter a translating mirror is used match the optical path length of each arm. Once the two arms are spatially and temporally overlapped they traverse a 10 nm green bandpass filter and are heterodyned in an avalanche photodiode (APD). The resulting radio frequency (RF) beats are equal to  $\pm(\delta - f_{AOM})$ , where  $f_{AOM}$  is generated to be 7/8 of the repetition rate. This beat signal is then fed to a tracking oscillator. The pure RF signal generated from the tracking oscillator is then compared against a synthesized frequency. The servo generates an error signal that feeds back to the actuator on which the end mirror of the laser is attached. Once the offset is locked the signal from the tracking oscillator is counted on a frequency counter to measure to how accurately we are locking the offset frequency. We have stabilized  $\delta$  with a variance of 3.6 Hz with a maximum excursion of 5.6 Hz that was measured with 0.1 second gate time. This corresponds to stabilization of the carrier-envelope phase to 0.19 mrad with a maximum excursion of 0.56 cycles/s.



#### 3.3 Time Domain Verification of Carrier-Envelope Phase Stabilization

Figure 4 Schematic of the cross-correlator used time domain verification of phase control.

We verify control of  $\Delta \phi$  in the time domain by second-order interferometric cross-correlation. Correlation of pulse *i* with pulse *i* + 2 is achieved using the cross-correlator shown in Fig. 4. A multi-pass cell is used to achieve the 20 ns delay needed to correlate one pulse with a second pulse two cavity round trips later. The cell is designed such that its two curved mirrors' radii of curvature and separation were chosen to mode match the output from both arms. Because of the large difference in the correlator's arm lengths, the entire correlator is held under vacuum at a pressure less than 300 mTorr. To ensure that an equal phase delay is imparted in each arm all the mirrors were coated in the same coating run and there are an equal number of mirror bounces in each arm. A 2 µm thick pellicle beamsplitter is used to minimize dispersion. The second order correlation was obtained using a windowless GaAsP photodiode. The bandgap of GaAsP is such that it yields a pure quadratic intensity response at 780 nm. A typical cross-correlation is shown in Fig. 5 (a).

To determine the pulse-to-pulse phase slip the fringe peaks of a correlation are fit to a Gaussian envelope. Note that a crosscorrelation does not exhibit the symmetry of an autocorrelation. In a correlation measurement of this type the fringes are the result of interference between the electric fields of two pulses. In an auto-correlation, because the phase of both pulses being correlated is the same, the highest peak is lined up at the peak of the fit envelope. In a cross-correlation the phase difference between the two pulses results in a shift of the peaks of the fringes with respect to the envelope. The relative carrier-envelope phase is measured by fitting the fringe peaks of the correlations to a Gaussian envelope and determining the fit parameters. A plot of experimentally determined values of relative phase for different frequency offsets,  $\delta = (N/16) f_{rep}$ , and a linear fit to

the results is given in Fig 5 (b). The results show a small offset from the theoretically expected relation  $\Delta \phi = 4\pi \delta / f_{rep}$  (a

factor of two is a result of correlating pulse *i* with pulse i + 2 as opposed to adjacent pulses). This offset is attributed to an imbalance between the two arms of the correlator despite our efforts to avoid this. Also the precision to which we can stabilize  $\delta$  tells us that we should be able to stabilize the envelope-carrier phase to a less than a milliradian.



Figure 5 (a) Typical cross-correlation, solid line, and the fit to a Gaussian envelope, dashed line,. (b) Experimentally determined values of pulse-to-pulse carrier-envelope phase for different frequency offsets and a linear fit to the results.

The error bars on our data do not demonstrate control to this precision. The disparity between our experimental and expected results, we believe, is due to mechanical noise in the cross correlator. Even though the correlator is held under vacuum, which helps to isolate it from acoustic noise, mechanical vibrations are easily transmitted from the optical table to the correlator.

An important detail is that in all these cases we have fixed the relative pulse-to-pulse phase slip not the overall "absolute" phase of each pulse, i.e.,  $\phi_0$ . Control of the absolute phase of a short pulse is of interest to the ultrafast community. Recently the development of few cycle pulses allow for the possibility of extreme nonlinear optical processes such as x-ray generation and above threshold ionization, whose efficiencies are highly dependent on the peak electric field intensity. Vice versa, experiments have been proposed to take advantage of a highly nonlinear process with such dependence to determine the absolute phase.<sup>17</sup>

In a modelocked laser the carrier slips under the envelope as the pulse circulates through the cavity. Once the pulse exits the cavity we no longer have active control of this carrier slip. Outside the laser the carrier-envelope phase for the pulse is determined by the dispersive elements that the pulse encounters external to the laser. The phase shift due to these elements is the same for each pulse. Therefore, when we speak of stabilizing the carrier envelope phase, we mean the relative phase from pulse to pulse taken at one point in space.

## 5. INTERFEROMETRIC MEASUREMENTS OF PHASE NOISE INDUCED IN AIR-SILICA MICROSTRUCTURE FIBER

One issue concerning the stabilization of the frequency comb offset is phase noise. If phase noise is generated in the microstructure fiber it manifests on delta and is fed back to the laser. Fiber phase noise can result from the intensity-dependent nonlinear index of refraction,  $n = n_o(\omega) + In_2$ , that allows for the extreme broadening of our pulses. Where *I* is the intensity of the light and  $n_2$  is the nonlinear index of refraction. Since SPM in the fiber is highly nonlinear, amplitude to phase noise conversion can occur. To determine if this conversion is significant, it is necessary to measure the amount of nonlinear phase induced per mW fluctuation in laser power.



Figure 6 Schematic of the interferometer used to measure the nonlinear phase induced in air-silica microstructure fiber.

In the discussion explaining the principles used in the self-referencing technique, the constant phase term, with contribution by kz was left out of the discussion. Previously we neglected any time variation of this term, which in most circumstances is the case since this term is a function of space not time. However, in a nonlinear medium where the index of refraction is a function of intensity, intensity fluctuations make it appear to the pulse as though the length of the medium is also fluctuating. This appears as a time variance in kz, which is a function of index of refraction  $\theta(t) = kz = \omega [n(I) z/c]$ . The noise resulting in time variations of kz are then introduced into our measurements of  $\delta$  via the heterodyne beat between  $f_n$  and  $f_{2n}$ .

We perform differential measurements of the nonlinear phase generated in air-silica microstructure fiber by comparing the phase at the output of the fiber with the phase from a reference laser using the interferometer in Fig. 6. The light from the laser is split by a beamsplitter, the light in one arm is put through the microstructure fiber, while the light in the other arm is used as reference and is shifted in frequency using an AOM. For a purely differential measurement both arms of the interferometer must experience the same phase shifts due to vibrations of reflecting surfaces. As a result both beams must reflect off the same mirrors. Where a beamsplitter and mirror are used for separating and recombining the two arms interferometer, both the mirror and beamsplitter were mounted on the same kinematic mount with a fixed spacer.

Amplitude modulation is introduced into the measurements by varying the intensity of the laser into the interferometer using a liquid crystal variable waveplate and a polarizer. In the reference arm, this results only in amplitude modulation of the reference field. In the arm containing the air-silica fiber, however, apart from amplitude modulation of the field this a time varying phase is introduced,  $\theta(t)$ , by amplitude-to-phase conversion by the fiber nonlinearity. This time varying phase is extracted by heterodyning the beams in the two arms of the interferometer in a silicon photodiode. The resulting RF heterodyne beat is equal to  $[\theta(t) - f_{AOM}t]$ , where  $\theta(t) = B \cos[\omega_m t]$  and  $\omega_m$  is the modulation frequency of the variable wave plate. A tracking filter is used to take out the amplitude modulation of the beat signal. The tracking filter produces a clean RF signal with constant amplitude, which is then mixed with  $f_{AOM}$  in a double balanced mixer. The mixed product is filtered leaving only the mixed-down low-frequency term corresponding to  $Cos(\theta(t) + \varphi)$ , where  $\varphi$  is the relative phase between the heterodyne beat and  $f_{AOM}$ . An RF delay line is used to set  $\varphi \approx \pi/2$  such that  $Cos[\theta(t) + \varphi] \approx \theta(t)$ . The amplitude of the nonlinear phase, B, is measured using a lock-in amplifier, locked to the modulation frequency of the variable waveplate. By comparing the ratio of the amplitude of the nonlinear phase generated from the intensity modulation we can determine the amount of phase modulation generated in the fiber per mW change in laser power. Preliminary measurements show this ratio to be ~ 0.05 rad /mW. This measurement was made at 780 nm with a total of 14 mW into the fiber with an input bandwidth of 100 nm broadened to ~500 nm. The total nonlinear phase resulting from an input laser power of 14 mW is ~ 0.6 radians, ten times smaller than is expected if the broadening is due to SPM in the absence of dispersion.<sup>18</sup> Therefore, an intensity fluctuation of ~ 10% results in the generation of about 50 mrad of phase. Considering that our laser fluctuates less than 1%, the phase noise introduced by this fluctuation is on the order of the noise generated by acoustic jitter of optic mounts. This is in agreement with our experimental ability to lock  $\delta$ .

One of the difficulties involved in this measurement is that the intensity of the light in the fiber is a function of the initial pulse parameters such as chirp and temporal pulse length. Because of this it is necessary that our pulses be well characterized. This is done using an autocorrelator that has the same dispersive elements as those used right up until the input of the fiber. Our intent is to obtain better quantitative measures of the phase noise induced per mW fluctuation in laser power. Our intent is to perform better phase measurement. Future measurements will be performed at different pulse chirps and as a function of the wavelength used for measuring the heterodyne beat.

## 6. CONCLUSIONS

In this paper we have demonstrated how to control and stabilize the frequency comb generated by the pulse train of a modelocked femtosecond Ti:S laser via self-referencing using air-silica microstructure fiber. We have demonstrated control and verification over the carrier-envelope phase of femtosecond pulses. We have also outlined a technique for nonlinear fiber measurement that will be used to obtain a quantitative value of the phase noise induced by pulse amplitude fluctuation in the microstructure fiber. Although we have presented only preliminary results here, we plan to use the experimental method described above to better characterize the behavior of the air-silica microstructure fiber.

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