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High-performance iodine fiber frequency standard

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We have constructed a compact and robust optical frequency standard based around iodine vapor loaded into the core of a hollow-core photonic crystal fiber (HC-PCF). A 532 nm laser was frequency locked to one hyperfine component of the R(56) 32-0 $^{127}\text{I}_2$ transition using modulation transfer spectroscopy. The stabilized laser demonstrated a frequency stability of 2.3×10^{-12} at 1 s, almost an order of magnitude better than previously reported for a laser stabilized to a gas-filled HC-PCF. This limit is set by the shot noise in the detection system. We present a discussion of the current limitations to the performance and a route to improve the performance by more than an order of magnitude. © 2011 Optical Society of America

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The performance of optical frequency standards based on laser-cooled atoms and ions now surpasses the best microwave clocks [1,2]. However, there is a great deal of interest in developing simpler optical frequency standards for practical applications. One of the earliest optical frequency standards, based on iodine vapor, continues to inspire a great deal of work due to its simplicity [3–7]. Optical transitions of iodine continue to be included in the Bureau International des Poids et Mesures recommended wavelengths for the practical definition of the meter [8,9], with recent work demonstrating fractional frequency stabilities of $2\text{--}5 \times 10^{-14}$ at 1 s [5–7] and 4.6×10^{-13} at 1 year [7]. The approaches outlined in Refs. [5–7] are limited to the laboratory because they use large, fragile glass iodine cells. In contrast, an approach based on HC-PCFs could deliver the same long interaction region between gas and laser in a compact, robust, and portable format. Existing work on HC-PCF gas cells has largely focused on hydrogen-, acetylene- and rubidium-loaded fibers for spectroscopy [10–13]. Frequency standards based on gas-filled HC-PCFs were first demonstrated in 2005 [10]; to our knowledge the current state of the art is an acetylene-loaded HC-PCF with a fractional frequency stability of 1.2×10^{-11} at 1 s [14]. We demonstrate an iodine-loaded HC-PCF as the core of a laser frequency stabilization system with a performance nearly an order of magnitude better than that earlier result.

A simplified outline of the experimental system is shown in Fig. 1. A 1.3 m long Kagome structure HC-PCF with 25 μm core diameter [15] is mounted so that each end is close (~ 1 mm) to a window inside a vacuum system, while the bulk of the HC-PCF sits outside the vacuum. A teflon fitting compresses a rubber cone onto the outside of the HC-PCF to create a vacuum seal around the fiber. The vacuum system includes a reservoir of solid iodine with a vapor pressure of 36 ± 1 Pa (vapor pressure of iodine at room temperature $\sim 23.5^\circ\text{C}$) [16] in a separate valved section of the vacuum system.

Counterpropagating pump and probe beams are coupled into opposite ends of the HC-PCF through 4 $\times$ microscope objectives. The polarizations of the pump and probe are set orthogonal so that they can be easily

separated at the output of the fiber. Because of unwanted reflections from the fiber, together with a small birefringence, residual pump light still falls on the photodiode that detects the transmitted probe signal. An acoustic optical modulator (AOM) shifts the pump beam by 200 MHz to reduce effects of this interference on the photodiode. This AOM is used to apply a frequency modulation to the pump beam to generate an error signal for the frequency locking system (see below). A 10 cm long traditional iodine gas cell at room temperature, with an independent pump–probe beam pair, is used as a reference.

We lock a 532 nm Innolight Nd:YAG laser to the a1 hyperfine component of the R56(32,0) manifold. This line exhibits a 4.8 ± 0.5 MHz linewidth (full width at half-maximum) in the cell, and 15 ± 2 MHz in the HC-PCF under operational conditions. The cell width arises principally from iodine–iodine collisions, whereas the fiber width is due to iodine–background gas collisions (~ 3 MHz) and transit time broadening (~ 3 MHz) [11,17] together with power broadening from the high intensities (~ 9 MHz). Using the R(56) 32-0 on-resonance absorption we estimate the iodine pressure in the fiber as 0.8 ± 0.3 Pa, which is achieved after a ~ 15 min loading period.

Figure 2 shows the electronic locking system. The pump undergoes frequency modulation (FM) using the

Fig. 1. (Color online) Simplified overview of the optical system, with counterpropagating pump (green) and probe (blue) beams. MO, microscope objective; AOM, acoustic optical modulator; PD, photodiode; HWP, half-wave plate; V, vacuum valves. The counterpropagating beams are separated for ease of visualization.

Fig. 2. (Color online) Simplified overview of the electronic locking system. Insets show the experimental data obtained at the error point and directly on the photodiode.

AOM, which produces amplitude modulation on the probe according to the frequency difference between the laser and the hyperfine feature. This modulation transfer lock avoids issues associated with unwanted AM produced in conjunction with the FM. The peak FM deviation was chosen to produce suitable error signals (4 MHz for the fiber, 2 MHz for the cell). The bandwidth of this frequency control system is ~ 150 Hz: the frequency at which the magnitude of the laser free-running fluctuations equals the noise floor of the control system.

High intensities in the fiber produce substantial Stark shifts of the iodine energy levels [18]. Fluctuations in the in-coupled power associated with vibration and mechanical drift of the alignment between the beams and fiber core cause flicker frequency fluctuations at the level of $\sim 3 \times 10^{-11}$. To overcome this we have incorporated active alignment for both the pump and the probe beams, which reduces the intensity fluctuations by a more than a factor of 5 at 1 Hz [19]. This additional stabilization reduced the power-driven fluctuations so that the standard now exhibited a white frequency limit consistent with detection shot noise.

Figure 3 displays the fractional frequency stability of the free-running laser together with the performance when locked to a sub-Doppler feature in both the fiber and cell. All measurements were made by generating a beat note against a high-performance cavity-stabilized laser [20] whose performance is also displayed in Fig. 3. The frequency stability is in terms of the conventional

Fig. 3. (Color online) Fractional frequency stability of the free-running laser (red triangles), the laser locked to the 10 cm long iodine cell (blue crosses), the laser locked to the iodine-loaded HC-PCF (black squares), and the reference cavity (purple circles) against which these stabilities were measured. The limit to the frequency stabilization system for the fiber-stabilized laser (black line) is also displayed.

square root Allan variance measure [21]. The fiber results shows a white frequency characteristic with a fractional frequency stability of 2.3×10^{-12} at 1 s, which is almost an order of magnitude improvement over the best previous work using a gas-filled HC-PCF [14].

We have estimated the limits of the frequency control system by measuring the frequency error signal when the laser is tuned out of resonance with any iodine feature. Under these conditions the error signal will be sensitive to noise sources, such as any residual AM from the modulation locking scheme together with detector and shot noise; however, it will not be sensitive to effects that shift the iodine resonances (e.g., Stark, pressure and magnetic shifts). We see a white frequency noise floor, as shown in Fig. 3, which is consistent with the measured stability over 1 – 30 s integration range indicating that this detection noise is likely responsible for the performance in this range. We find further that this detection noise was associated with shot noise in the detection process with the measured current fluctuation at the output of the photodiode within 16% of the level calculated from first principles for the shot noise. For the cell-based iodine standard the input noise of the lock-in amplifier limited the performance a factor of 4 above the shot noise limit.

Under operational conditions (see Table 1), the slope of the frequency discriminator in the fiber is about half that of the cell, due to the FM deviation used as well as three physical reasons: (1) the fractional depth of the Doppler-free features are ~ 30 times higher in the fiber because of the high intensities and the excellent spatial overlap of the pump and probe beams; (2) the substantially lower fiber absorption (α_{a1}) because the iodine pressure is $\sim 1/45$ th that of the cell; (3) the linewidth of the Doppler-free features in the fiber that are ~ 3 times wider than that of the cell. This gives an expected fiber slope approximately equal to that of the cell, further reduced by a factor $\sim 2/3$ because the FM deviation used for the fiber lock was half the optimum value.

These observations show a route to substantially improving the short-term performance of the fiber frequency standard: one should have expected ~ 45 times less collisional broadening in the fiber because of the lower iodine pressure; however, this was not seen

Table 1. We List the Sub-Doppler Resonance Frequency Width, Measured Saturation Intensity, Operational Pump and Probe Intensities and Powers, the Doppler Broadened Absorption Coefficient at the Location of the a1 Hyperfine Component, and the Fractional Depth of the Hyperfine Component with Respect to the Doppler Broadened Absorption

Experimental Parameters		
	Cell	Fiber
Δ_{HF} (MHz)	4.8 ± 0.5	15 ± 2
I_{sat} (kW m^{-2})	42 ± 4	250 ± 30
I_{probe} (kW m^{-2})	1.6 ± 0.2	1500 ± 240
I_{pump} (kW m^{-2})	1.9 ± 0.3	400 ± 80
P_{probe} (μW)	670 ± 30	380 ± 40
P_{pump} (μW)	1500 ± 70	100 ± 10
α_{a1}	0.37 ± 0.01	0.035 ± 0.004
$\alpha_{\text{HF}}/\alpha_{\text{a1}}$	$\sim 0.3\%$	$\sim 9\%$

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because background gas (water/nitrogen/oxygen) is released during the iodine loading process. From the observed linewidth, we estimate a background pressure of ~ 150 Pa [16], along with a $\sim 7\%$ increase per hour in the hyperfine width due to continued outgassing. Refining the loading process to remove this background gas could reduce the low-intensity linewidth to that limited by transit time effects, which, due to the large mass of iodine, is ~ 2 MHz at low pressures [11,17]: a value 3 times below that one would obtain for acetylene. A decreased rate of collisions would allow operation at lower pump powers, for a given sub-Doppler signal strength, thereby alleviating Stark shift and power broadening effects. For maximum signal to noise on the discriminator one wishes the Rabi frequency of the pump to be of the same order as the intercollision frequency and under these conditions we calculate a possible reduction in linewidth from the current 15 MHz to 7 MHz with the removal of the background gas.

Furthermore, we currently do not have good control over the amount of iodine that can be loaded into the fiber. If we could reliably load an optimal density ($\alpha L \sim 1$), it would improve the discriminator signal by a factor of 9 from the results reported here. The combination of narrower linewidth, larger FM deviation and increased absorption depth could improve the frequency discriminator sensitivity by a factor of ~ 30 , allowing substantially improved short-term frequency stability. The compactness of the standard is likely to allow for effective temperature and magnetic shielding, although this will require the additional step of removing the fiber from the vacuum system [22]. Tight guidance of the pump and probe beams will minimize unwanted effects associated with fluctuations in their relative alignment. The combination of properties listed above suggest that long-term stability and reproducibility of the frequency standard can be good although this has not yet been directly tested.

A laser has been locked to the a_1 hyperfine component of the $R_{56}(32,0)$ transition in an iodine-loaded HC-PCF, with a fractional frequency stability of 2.3×10^{-12} at 1 s limited by shot noise in the detection system. Future work will improve the loading scheme so that it is possible to load a specific quantity of iodine as well as avoiding contaminants entering the HC-PCF. The predicted

performance of the frequency control system in this new standard should lie below 10^{-13} at 1 s.

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