Effect of hyperfine structure on atomic frequency combs in Pr:YSO

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Abstract

Quantum memory will be a key component in future quantum networks, and atomic frequency combs (AFCs) in rare-earth-doped crystals are one promising platform for realizing this technology. We theoretically and experimentally investigate the formation of AFCs in Pr$^{3+}$:Y$_2$SiO$_5$, with an overall bandwidth of 120 MHz and tooth spacing ranging from 0.1 MHz to 20 MHz, showing agreement between our calculations and measurements. We observe that the echo efficiency depends crucially on the AFC tooth spacing. Our results suggest approaches to developing a high-efficiency AFC quantum memory.

I. INTRODUCTION

Long-range quantum networks will require quantum repeaters to overcome transmission losses, and most current approaches to quantum repeaters require quantum memory [1]. A practical quantum memory would enable long-lived storage and high-fidelity, high-efficiency retrieval of single photons. One promising candidate implementation is the atomic frequency comb (AFC) protocol [2]. The central idea of this scheme is to store an optical pulse in a material with a frequency-comb absorption spectrum with tooth spacing $f_{\text{rep}}$ and then to recover an echo after a time $T_{\text{rep}} = 1/f_{\text{rep}}$ (for on-demand memory, control pulses can be used to pause and resume the state evolution). Here we show that AFC quality and echo efficiency depend on the relationship between $f_{\text{rep}}$ and the spacings of the hyperfine energy levels involved. Our results suggest a path to developing high-efficiency AFC quantum memory.

The rare-earth-ion-doped crystal Pr$^{3+}$:Y$_2$SiO$_5$ (Pr:YSO) is suitable for various optical storage techniques for reasons we discuss in the next section, and it was the first material used to demonstrate on-demand AFC memory [3]. It has been studied extensively, including work on photon echo [4], spectral-hole-burning techniques [5], and stimulated Raman adiabatic passage [6, 7]. The latter technique was used for selective retrieval of pulses [8], and further studies investigated the rephasing of these coherent populations [9] and on-demand retrieval based on spontaneous Raman scattering [10]. Storage of optical pulses has been demonstrated with stopped light using electromagnetically induced transparency [11, 12] and a memory has been realized at the single-photon level using stopped light in a spectral hole [13]. An AFC quantum memory has also been demonstrated at the single-photon
The AFC protocol has also been studied in other rare-earth-ion-doped crystals, which have similarly advantageous optical properties. Storage and retrieval of single photons has been demonstrated: time-bin qubits \[16, 17\] and entanglement with another photon \[18\] survive storage. Pulse storage and retrieval has also been demonstrated using superhyperfine levels \[19\] and hybridized electron-nuclear hyperfine levels \[20\].

One attractive feature of the AFC protocol highlighted in Ref. \[2\] is that in principle it enables retrieval efficiency arbitrarily close to unity. In practice, however, experimental demonstrations have achieved limited efficiency: Refs. \[21, 22\] used cavity enhancement to achieve efficiencies just over 50%.

### A. Hyperfine structure of Pr:YSO

The YSO crystal structure has two distinct Y sites where Pr impurities can occur \[23, 24\]. Here we study Pr ions at the site that has \({}^{3}H_{4-1}D_{2}\ (g-e)\) hyperfine transitions at 605.977 nm. The homogeneous linewidth of these optical transitions is on the order of 1 kHz and can be observed when the crystal is cooled to liquid helium temperatures to eliminate phonon broadening; the inhomogeneous broadening, arising from local variations in the crystal structure surrounding each Pr ion, is about 4.4 GHz \[25\]. In the absence of an external magnetic field, the hyperfine states are all two-fold degenerate, and the ground- and excited-state manifolds each have three distinct energy levels. Each Pr ion therefore has nine transitions labeled \(a-i\) in ascending order of energies; conversely, for any laser frequency within the inhomogeneous bandwidth, there are nine different classes of atoms \(A-I\) for which one of these transitions is resonant with the laser, with the laser addressing transition \(x\) for class \(X\), as shown in Fig. 1a.

As a result of the hyperfine structure, spectral-hole burning at even a single frequency results in multiple features (Fig. 1b-c) \[24\]. The large transmission peak at zero detuning occurs because all 9 classes become transparent at this frequency. As an example, the transmission peak at 4.7 MHz occurs because 3 of the 9 classes that absorb at this frequency are depopulated. Due to power broadening, our measurement cannot distinguish features separated by less than 0.2 MHz, but such closely-spaced peaks may appear as broader peaks or shoulders, as seen at 5.35 MHz.
FIG. 1: (a) Each Pr atom has nine transitions a-i (left); any laser frequency within the 5 GHz inhomogeneous line addresses nine atom classes A-I (right). (b) Transition frequencies for all atom classes that have a transition at zero detuning. Rows are grouped by common ground states. (c) Effect of spectral-hole burning at zero detuning: measured transmission spectrum (blue), and simulated population assuming equal probabilities for all transitions (red). The baselines for both correspond to the unburned crystal. The burn laser addresses transition $x$ for atom class $X$, depleting the corresponding ground-state population and reducing absorption on transition $x$ and two other transitions as well (highlighted cells in (b)); the population is transferred to the other two ground states, increasing absorption on the other 6 transitions.
The excited-state spacings in Pr:YSO are fortuitously similar, leading to a comb-like response to spectral-hole burning at a single frequency. This occurs because (1) the transparency for classes $A, D, G$ at 4.58 MHz detuning overlaps with the transparency for $B, E, H$ at 4.84 MHz, and (2) an additional transparency occurs for $A, D, G$ at 9.42 MHz, further extending the comb-like response (similar reasoning for classes $C, F, I$ and $B, E, H$ explains the peaks at $-9.42 \text{ MHz}, -4.84 \text{ MHz,}$ and $-4.58 \text{ MHz}$). This observation suggests that the material has a naturally periodic spectrum, which motivates this work.

II. EXPERIMENT

We use our experimental setup for two different measurement protocols, a comb-measurement protocol and a pulse-echo protocol, each of which proceeds in two stages. In the first stage of both protocols, we prepare an AFC. In the second stage, we either probe the transmission spectrum (the comb-measurement protocol), or observe the transmission and echoes of a pulse sent to the AFC (the pulse-echo protocol).

We use a tunable diode laser at 1212 nm, which is amplified in a fiber Raman system and then doubled, producing a beam at 606 nm. To stabilize the laser frequency, we use Pound-Drever-Hall feedback [26] to lock the 1212 nm light to a reference cavity with a Zerodur spacer, held in a vacuum chamber. We use a wavemeter to monitor the doubled light and to tune to the Pr transition. This beam is modified using double-pass acousto-optic modulators (AOMs) to prepare the three different beams used in our measurements. A burn beam is produced by applying a frequency-modulated (FM) sine wave to the AOM: this signal has a frequency-comb structure, with 60 MHz bandwidth (twice the FM deviation) and tooth spacing equal to the modulation frequency. After the double pass, this results in an optical frequency comb with 120 MHz bandwidth and tooth spacing equal to the modulation frequency. A probe beam is produced by (slowly) sweeping the AOM frequency, resulting in a frequency-swept beam covering the same bandwidth as the burn beam. Alternatively, a 10 ns pulse is generated by applying a pulse to the AOM.

In our setup (Fig. 2), we use a Pr:YSO crystal with 0.05% substitution of Pr for Y, housed in a cryostat maintained below 4 K. Two slightly displaced, parallel beams are focused to overlapping beam waists inside the crystal. Beam #1 is the burn beam described above: we vary the tooth spacing of this optical frequency comb in 0.1 MHz steps, from 0.1 MHz.
FIG. 2: (a) We prepare an AFC by burning the crystal for 30 seconds with an optical frequency comb (beam #1). We pick off some of this beam to beat against a local oscillator for a balanced heterodyne measurement of the optical spectrum: this is the spectrum with which we drive the crystal. (b) Timing diagrams for the two measurements (not to scale).

The short pulses on either side of the frequency sweep are used to demarcate the sweep duration and help automate data analysis; to minimize their impact on the measurements, they are tuned to a frequency 20 MHz outside the AFC and sweep bandwidths.

to 20 MHz. The first stage of both measurement protocols consists of 30 seconds of nearly continuous burning with beam #1, aiming to imprint the optical frequency comb on the atomic transmission spectrum and establish an AFC.

In our comb-measurement protocol, beam #2 is the probe beam described above. During the 4 ms frequency sweep of the probe beam, we acquire the transmission spectrum of the crystal. In the pulse-echo protocol, beam #2 is the 10 ns pulse described above. We monitor emission from the crystal to detect the transmitted pulse and its echoes. Due to the different natures of the output signals from the two measurements, detectors with different response bandwidths are required.

In the second stage (of both measurement protocols), the 1 s measurement cycle is synchronized to an electrical pulse indicating the cryostat cycle, in order to minimize effects of
vibrations. We repeat the measurements for approximately 30 cycles [27] and average the results. In addition to the 30 s burn in stage 1, in stage 2 of each cycle, 930 ms is spent burning the crystal to reinforce the AFC.

III. THEORY

Here, we develop a theoretical model to describe time evolution of the atomic populations of the 9 hyperfine levels. We apply this model to predict the transmission spectra produced in our experiment.

A. Density Matrix Formulation

We use the density matrix as the core of our theoretical model of the system. Alternative descriptions include the rate equation and the stochastic Schrödinger equation [28]. The rate equation is appropriate when it is possible to distinguish clearly between resonant and non-resonant transitions. Here, because the atom classes form a continuum, in the sense that there are no energy gaps larger than the homogeneous broadening, there are always some atom classes which are intermediate between resonant and non-resonant, which do not have an obvious treatment using the rate equation. Such classes give us a description of the widths of the resonance. In principle, the widths will depend on a parameter, the Rabi frequency scale \( \Omega_0 \), which links the signal strength to the Hamiltonian.

An average of the Stochastic Schrödinger equation (SSE) is formally equivalent to the density matrix [29], although the SSE has advantages for single-particle dynamics [28]. Ref. [29] suggests that the density matrix is favored algorithmically for small matrices, and the SSE is favored for large ones. Ref. [29] describes a cross-over when the dimension of the Hamiltonian exceeds 200. Here, the Hamiltonian is only of dimension 6, suggesting the density matrix formalism will be more efficient. This is true, but it understates the case. Because our Hamiltonian is periodic, it is possible to obtain a solution in the logarithm of the number of periods. We were not able to find a comparable algorithm for the SSE, making the density matrix the better method for this application.

The quantum Liouville equation for the density matrix \( \rho \) is

\[
\frac{d\rho}{dt} = -i[H, \rho] + \frac{d\rho}{dt}_{\text{relax}},
\]  

(1)
where \( H \) is the Hamiltonian and \( \frac{d\rho}{dt}\big|_{\text{relax}} \) describes the relaxation. We assume the relaxation is purely due to spontaneous emission and use the form given by Ref. [30].

The further development of this equation requires an explicit form for the Hamiltonian, which we discuss next. Lovrić et al. experimentally determined the Hamiltonian parameters, including the orientation of the ground- and excited-state manifolds with respect to the crystal axes [31]: however, they report incorrect Hamiltonian parameters that would result in an oscillator-strength matrix with two columns swapped when compared with earlier results [5] and even with their own work [31], as has been pointed out earlier [32].

Since we were unable to find the Hamiltonian elsewhere in the literature, we derived a Hamiltonian that is consistent with the hyperfine level structure and oscillator strengths (which are readily available in the literature) and that takes the conventional form \( \frac{1}{2} I \cdot Q \cdot I \), where \( I \) is a vector of length 3 representing the spin-5/2 nuclear spin states and \( Q \) is a rank-2 traceless symmetric tensor giving the orientation of these spins in a given manifold relative to the crystal axes. There are 10 degrees of freedom to be determined: the ground- and excited-state manifolds each have 2 energy spacings (Fig. 1) and 3 Euler angles indicating their orientation with respect to the crystal axes. However, it is actually sufficient to know only the relative orientation of the manifolds, so we only need to determine 3 Euler angles.

The 3\( \times \)3 oscillator-strength matrix, given in Table I, provides four constraints on these Euler angles. We use a least-squares fit and find Euler angles \( \alpha = 10.3(15)\degree \), \( \beta = -164.4(15)\degree \), and \( \gamma = -130.7(15)\degree \) (digits in parentheses, here and throughout, are total uncertainties at one standard error, and represent the uncertainty of the least significant digits).

Returning to the Liouville equation, we proceed to a rotating frame

\[
\rho_{k\gamma,\ell\delta} = \sigma_{k\gamma,\ell\delta} e^{-i[E_{\gamma\delta} + (\gamma - \delta)\Delta]t}.
\]

(2)

Here, \( \gamma \) and \( \delta \) index the optical manifold (with 1 for \( g \) and 2 for \( e \)), \( k \) and \( \ell \) index the hyperfine level within a given manifold, and \( \Delta \) is the detuning relative to the \( |g, \pm 1/2\rangle \rightarrow |g, \pm 1/2\rangle \) transition for a reference atom class. Whereas \( \rho \) varies with an optical frequency (roughly 500 THz), \( \sigma \) varies with the hyperfine splitting, a radio frequency (RF) of order 10 MHz, as shown in Fig. 1h. The change of variables allows the optical frequencies to be removed analytically and allows the program to solve numerically on the RF time scale. A time step near 1 ns is chosen, with a small adjustment often required to let the time step be
TABLE I: Transition probability amplitudes between the \( ^3H_4 \) and \( ^2D_1 \) (g and e) manifolds. (The manifolds g and e are not to be confused with the transitions g and e.) State labels are those of Fig. 1. Standard uncertainties are based on the propagation of the standard uncertainties of the Euler angles given in the text.

| \( |e, \frac{1}{2}\rangle \) | \( |e, \frac{3}{2}\rangle \) | \( |e, \frac{5}{2}\rangle \) |
|-----------------|-----------------|-----------------|
| \( |g, \frac{1}{2}\rangle \) | 0.753(23) | -0.602(28) | -0.265(24) |
| \( |g, \frac{3}{2}\rangle \) | -0.634(27) | -0.772(22) | -0.048(08) |
| \( |g, \frac{5}{2}\rangle \) | -0.176(17) | 0.204(19) | -0.963(07) |

commensurate with the period of the laser.

\[
\frac{d\sigma}{dt} = \mathcal{L}\sigma, \tag{3}
\]

defining the Lindblad matrix \( \mathcal{L} \) from context. This equation is solved in a straightforward manner by discretizing time and assuming that the Lindblad matrix is piecewise-constant in time. In each time step, the solution is

\[
\sigma(t + \delta t) = \exp(\mathcal{L}\delta t)\sigma(t). \tag{4}
\]

In practice, the matrix exponential is found through its Taylor series in the form used in Horner’s method, namely

\[
\exp(M) = 1 + M(1 + \frac{M}{2}(1 + \frac{M}{3}(...))). \tag{5}
\]

for any matrix \( M \). The terms are evaluated from the innermost parenthesis, starting from \( \frac{M}{30} \). This method proves to be more accurate than evaluating the eigenvalues and eigenvectors with LAPACK and taking the exponential, stabilizing the solutions.

To evaluate the solution for a large number of frequency-modulated periods, we make use of the identity

\[
\int_0^{2T} dt \exp[\mathcal{L}(t)t] = \left[ \int_0^T dt \exp[\mathcal{L}(t)t] \right]^2, \tag{6}
\]

where \( T = T_{\text{rep}} \) is the period of \( \mathcal{L}(t) \). Iterating Eq. 6 allows the calculation of any period of the form \( 2^n T \) for positive integer \( n \) in a time proportional to \( n \). Since any integer may
be represented with binary digits, it is possible to calculate the propagator for any integer multiple $n$ of $T_{\text{rep}}$ in a time proportional to $n$, i.e., to the logarithm of the number of periods. Since the pulse duration is typically millions of times $T_{\text{rep}}$, this yields an enormous savings in computation time compared to propagating in small time steps for the duration of the burn beam.

In practice, evaluating the propagator for a time for a single period dominates over extending to millions of periods.

Finally, the Rabi overall coupling strength $\Omega_0$ is, in principle, a fitting parameter. In practice, we use a value determined empirically for the calculations at 4.8 MHz, by varying $\Omega_0$ over two orders of magnitude.

In principle, spatial effects play a role in spectral hole-burning. For example, along the burn beam propagation direction, transmission peaks will become progressively sharper as the beam is progressively filtered by the crystal. Here, we ignore all spatial dependence, which is justified since the observed transmission peaks are highly power-broadened, approximately two orders of magnitude broader than the homogeneous linewidth. However, our approach can be expanded to consider spatial effects.

B. Signal averaging

The theory requires knowledge of the periodic signal entering the crystal. Here, we describe how that periodic signal is obtained from the RF recorded after heterodyne mixing, shown in Fig. 2a. Signal averaging is used to extract the periodic signal from 20 $\mu$s samples of data discretized in 0.2 ns steps. The algorithm for averaging the signals is given in the Appendix.

Results for the case of a 4.8 MHz modulation frequency are shown in Fig. 3, including the data before and after the averaging procedure. The instantaneous phase shown in Fig. 3c may be compared to the design intent, which is a triangle-wave frequency modulation: the discrepancy is due to experimental limitations on producing a triangle-wave instantaneous frequency.

The quality of the averaged signal may be assessed by a comparison of its Fourier transform to the Fourier transform of the input signal, as shown for the example of 4.8 MHz in Fig. 4. The averaged spectrum is necessarily discrete. The experimental spectrum shows
FIG. 3: Input optical signals at 4.8 MHz optical-comb tooth spacing. (a) A portion of the output of the optical heterodyne signal showing the RF modulation of the optical input. (b) Real part of analytic signal (Fourier transform with negative frequencies removed) obtained by a stroboscopic average of the magnitude of the analytic signal and, separately, the instantaneous frequency with a small frequency shift to ensure periodicity. (c) Spectroscopic average of instantaneous frequency vs. time for the signal in part (b).

Strong peaks that are broadened in analogy with x-ray-diffraction Bragg peaks broadened through thermal motions of the atoms, as described by the Debye-Waller factor. The alignment of the discrete points to the peaks of the black curve validates the averaging process. Matches of similar quality were obtained for each of the 191 modulation frequencies from 1.0 MHz to 20.0 MHz in steps of 0.1 MHz. Calculations were not performed below 1.0 MHz.

IV. RESULTS

High-quality AFCs only occur for certain values of the modulation frequency (i.e., the tooth spacing imposed by the optical frequency comb burn beam): for a range of modulation frequencies close to the values of the excited-state spacings, we observe a marked improve-
FIG. 4: Magnitude of the Fourier transform of the input optical signal, a portion of which is shown in Fig. 3a (black) compared to its average shown in Fig. 3b (discrete magenta X), and a version with the smallest frequency shift required to achieve periodicity on the phase (discrete green points). Each set has been normalized to a maximum of 1.

ment in AFC quality (Fig. 5). This effect is apparent in the transmission spectra, as well as the echo signals, where we see that high-quality AFCs give rise to the large-magnitude echoes. Here, we define AFC quality as the presence of a well-defined periodicity in the transmission spectrum. Therefore, to quantify statements about trends in the AFC quality, it is useful to calculate Fast Fourier transforms (FFTs) of the measured AFCs. The connection between AFC quality and echo magnitude is borne out across the full range of modulation frequencies (Fig. 5). We observe AFCs forming with tooth spacing equal to the modulation frequency up to approximately 6.7 MHz, corresponding to points on the line of slope 1 in the intensity maps. For larger modulation frequencies, the observed tooth spacing is still limited to less than 6.7 MHz, with the AFCs forming at subharmonics of the modulation frequency, corresponding to points on the lines of slope 1/2, 1/3, and 1/4. Across the entire range of modulation frequencies 0.1 MHz to 20 MHz, AFCs often form with a tooth spacing close to 4.7 MHz. Even when the modulation frequency is very different from 4.7 MHz, AFCs often form with this tooth spacing, if it is nearly a subharmonic of the modulation
FIG. 5: (a) AFCs prepared by spectrum burning using optical combs with tooth spacings ranging from 3.5 MHz to 5.5 MHz, a subset of our full data set ranging from 0.1 MHz to 20 MHz (spectra are shown in dB scale). High-quality AFCs occur for a range of modulation frequencies close to the excited-state hyperfine spacings (roughly 4.1 MHz to 5.1 MHz). (b) Experimentally observed echoes of stored pulses (solid blue lines) and fast Fourier transforms of the AFCs (dashed black lines). The solid black lines are hyperbolas indicating expected emission times at multiples of the inverse tooth spacing. The blue and dashed-black curves have been scaled to overlie each other.

There is a similar but weaker tendency for AFC tooth spacing at approximately half the excited-state-level spacing, 2.4 MHz.

The similarity between (a) and (b) of Fig. 6 may be understood as a consequence of the formalism of Afzelius et al. [2]. If we assume that all echo pulses are due to direct reemission of the absorbed source pulses (as opposed to pulses which are absorbed, then re-emitted, then re-absorbed, then re-emitted, or higher processes), then the pulse train predicted by Eq. (A8) of the reference is determined entirely by the time-domain number density of the AFC, which is itself the Fourier transform of the frequency-domain number density via
FIG. 6: These intensity maps summarize our data for all 200 modulation frequencies (a subset of which is shown in Fig. 5), using dark pixels to indicate (a) large Fourier components of the AFC and (b) large echo pulses. The vertical plots on the right show marginals obtained by summing horizontal rows of pixels, with red lines indicating 2.4 $\mu$s$^{-1}$ and 4.8 $\mu$s$^{-1}$. Towards the right of both intensity maps, the patterns are smeared out horizontally: this effect, as well as the split peak near 4.8 $\mu$s$^{-1}$ in the marginal plot in (a), is due to pixelation.

Eq. (A9) of the reference.

Our theoretical calculations yield transmission spectra, and as with the experimentally measured AFCs, we summarize these results by calculating FFTs of the spectra. Fig. 7 presents a direct comparison of the intensity map from Fig. 6a with the corresponding Fourier components of the theoretically predicted AFCs. There is good overall agreement, though the theory tends to overestimate the content of subharmonic Fourier components.
FIG. 7: Inverse storage time versus modulation frequency. The storage time is the time from the input pulse to an echo. In general, there can be several echoes. The peaks selected for display have a strength of at least 2.5% of the maximum for the experiment (solid blue circles) or the theory (hollow red circles). The strength of each peak is proportional to the area of each peak plus one unit corresponding to the smallest peaks. (The small increment was required to keep the smallest symbols from disappearing.) Both theory and experiment were obtained by Fourier transform of the absorption spectrum.

The theory was not calculated below 1 MHz.

V. CONCLUSIONS

Pr:YSO is one of the most prominent candidate materials for implementing the AFC protocol, which is one of the most promising techniques for broadband quantum memory. Here, we have shown that AFC formation in Pr:YSO depends strongly on the tooth spacing being imposed; furthermore, we have shown that good AFC formation occurs near 4.7 MHz tooth spacing and presented evidence that this behavior is related to the spacings of the excited-state energy levels. Both experiment and theory indicate that, for small modulation frequencies, large AFC Fourier components occur at approximately 5 MHz: the smallest AFC tooth spacings we observe are approximately 1 MHz, giving a 1 µs upper bound on storage time (Fig. 7). Similarly, the AFCs are limited to tooth spacings less than approximately 6 MHz, so that the shortest possible storage time is approximately 0.15 µs. If a magnetic field is applied, the level spacings, and hence the allowed storage times, can be modified. This is a promising direction for future study.
On the theoretical side, we have derived the Hamiltonian parameters from the functional form of the hyperfine term using the transition matrix and energy spacings for Pr:YSO. We have developed a density-matrix code to calculate the population shifts and hence the transmission spectra as a function of the optical-comb tooth spacing, where the calculation time is logarithmic in the time that the burn beam is on. The calculated and measured spectra match show good agreement. Finally, we have shown that the Fourier transform of the absorption spectrum gives the photon echoes in the approximation where the reabsorption of photons is neglected. Although developed for Pr:YSO, similar techniques could be applied to other rare-earth systems.

In the AFC protocol, the tooth spacing, and thus the timing of the echo is not central to the operation of the memory. For on-demand operation, control pulses are required, and these can be used to extend storage to far longer times, in principle limited by the ground-state coherence times. However, the tooth spacing is important from an engineering perspective: for example, the echo time delay must be long enough to allow for the time required to apply the control pulses. Furthermore, a pre-programmable AFC storage without control pulses may prove useful for various applications. Our results are relevant to the design of such technologies. To avoid the restrictions on AFC formation at arbitrary tooth spacing, there is the possibility of applying a static magnetic field to lift the degeneracy of the hyperfine levels and change the spacings of the excited-state levels. Such a technique could enable the formation of high-quality AFCs with tooth spacings that cannot be achieved in the system with no applied magnetic field.

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APPENDIX

The signal averaging algorithm has the following steps:

1. The mean is subtracted from the sample and the sample is smoothed with a 7-point
symmetric binomial filter.

2. Locations of zero crossings (i.e., sign switches in adjacent smoothed data points) are found. If there are two zero crossings in consecutive time steps, the pair is not considered in the next step.

3. The instantaneous frequency \( f \approx \frac{1}{2\pi} \frac{\Delta \phi}{\Delta t} \) is calculated by assigning a phase shift of \( \Delta \phi = \pi \) to adjacent zero crossings which also give the time interval \( \Delta t \).

4. The instantaneous frequency is averaged stroboscopically given the known driving period.

5. The envelope is obtained by extracting the analytic signal from the smoothed data of step 2 above. The analytic signal is obtained through discrete FFT followed by zeroing out the negative frequencies, followed by the inverse FFT. The envelope is the absolute value of the analytic signal.

6. The envelope is averaged stroboscopically to create a periodic function.

7. The periodic phase is obtained by integrating the averaged instantaneous frequencies obtained in step 3 over a period. A small linear adjustment in the phase is made to enforce strict periodicity of both amplitude and phase.

8. The signal, a complex quantity, is formed as the product of the envelope from step 6 and the phasor of the periodic phase from step 7. (The phasor of a phase \( \phi \) is \( e^{i\phi} \).)

9. The signal is shifted in the Fourier domain to remove the central modulating frequency. This corresponds physically to incorporating the central frequency of the AOM into the optical reference signal. The effect is to create a function that may be sampled with more widely spaced points. In practice, there is a 5:1 downsampling at this step from 0.2 ns to near 1 ns. The step sizes are not exactly equal for different repetition rates because each time step must be an integer divisor of the corresponding period.

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