Achieving the highest spectral resolution over the widest spectral bandwidth – Precision measurement meets ultrafast science

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Phase control of a single-frequency continuous-wave laser and the electric field of a mode-locked femtosecond laser has now reached the same level of precision, resulting in preservation of optical phase coherence over macroscopic observation times exceeding seconds. The subsequent merge of CW laser-based precision optical-frequency metrology and ultra-wide-bandwidth optical frequency combs has produced remarkable and unexpected progress in precision measurement and ultrafast science. A phase-stabilized optical frequency comb spanning an entire optical octave (> 300 THz) establishes millions of precise marks on an optical frequency "ruler" that are stable and accurate at the Hz level. Accurate phase connections among different parts of electromagnetic spectrum, including optical to radio frequency, are implemented. These capabilities have profoundly changed the optical frequency metrology, resulting in recent demonstrations of absolute optical frequency measurement, optical atomic clocks, and optical frequency synthesis. Combined with the use of ultracold atoms and molecules, optical spectroscopy, frequency metrology, and quantum control at the highest level of precision and resolution are being accomplished at this time. Parallel developments in the time-domain applications have been equally revolutionary, with precise control of the pulse repetition rate and the carrier-envelope phase offset both reaching the sub-femtosecond regime. These developments have led to recent demonstrations of coherent synthesis of optical pulses from independent lasers, coherent control in nonlinear spectroscopy, coherent pulse addition without any optical gain, and coherent generation of frequency combs in the VUV spectral regions. Indeed, we now have the ability to perform completely arbitrary, optical waveform synthesis, complement and rival the similar technologies developed in the radio frequency domain. With this unified approach in time- and frequency-domain controls, it becomes practical to pursue simultaneously coherent control of quantum dynamics in the time domain and high precision measurements of global atomic and molecular structure in the frequency domain. These coherent light-based precision measurement capabilities may eventually be extended to the XUV spectral region, where new possibilities and challenges lie for precise tests of fundamental physical principles.

1. Introduction

There has been a remarkable recent convergence of the fields of ultrafast optics, optical frequency metrology, and precision laser spectroscopy [1-3]. This activity arises from unprecedented advances in the control of optical phases ranging from ultrashort to macroscopic laboratory time scales. A single frequency continuous wave optical field can now achieve a phase coherence time exceeding 1 s [4, 5] and this phase coherence can be precisely transferred to the electric waveform of an ultrafast pulse train [6]. The consequence is revolutionary for both precision measurement and ultrafast control, resulting in key advances in absolute optical frequency measurement [7], optical atomic clocks [8, 9], optical frequency synthesizers [10], carrier-envelope phase stabilization [11, 12], united time-frequency spectroscopy [13, 14] and high-resolution quantum control [15] (using a train of phase-coherent ultrashort pulses), coherent pulse synthesis [16] and amplification [17], ultra-broad, phase coherent spectral generation [18], frequency comb generation in the VUV spectral region [19, 20], and broad bandwidth ultrasensitive molecular detections [21]. We now possess all the experimental tools required for *complete* control over coherent light, including the ability to generate pulses with arbitrary shape, and precisely controlled frequency and phase, and to synthesize coherent light from multiple sources. The combination of this ability to do complete arbitrary waveform synthesis in the optical region of the spectrum, with recently developed optical pulse measurement techniques, is analogous to the development of oscilloscopes and waveform generators in the early to mid 20^{th} century.

The intrinsic connections between the time-domain pulse train and the frequency-domain comb spectrum permit the determination of absolute frequencies of comb lines and the use of frequency-domain-based optical phase control techniques to exert time-domain effects [22, 23]. The ultra-stable field is characterized by the variety of high-resolution spectroscopy and high-precision measurements enabled by CW lasers that are best described by their near delta-function frequency spectra. In contrast, the field of ultra-fast phenomena studies femtosecond events utilizing laser pulses that approach the limit of time domain delta-functions. At present time these two fields share nearly the same fractional resolution "figure of merit" with frequency and temporal widths on the order of one part in 10^{15}. The connection between the ultra-stable and the ultra-fast arises from the fact that femtosecond lasers produce pulses in a periodic train via mode-locking, with a corresponding rigorous periodicity in the spectral domain. The frequency domain spectrum consists of a comb of discrete modes separated by the repetition frequency f_r. The existence of dispersion inside the laser cavity results in a phase slip (denoted by $\Delta\phi_{ce}$) between the "carrier" phase and the envelope peak for each of the successive pulses. In the frequency domain, $\Delta\phi_{ce}$ yields an offset of the mode comb from exact harmonics of the f_r by $f_o = \Delta\phi_{ce}\ f_r /2\pi$. Hence each comb frequency is given by $\nu_n = nf_r + f_o$, where n is an integer (10^5 - 10^6). If the optical comb spectrum is sufficiently broad, it becomes straightforward to directly measure and control both f_r and f_o, leading to a high precision femtosecond-laser-based optical comb for optical frequency metrology and for study and control of time-domain dynamics by directly controlling the electric field waveform. Indeed, the frequency comb (and its counterpart in the time domain – a long, phase-coherent train of ultrashort pulses), building on our newly acquired capabilities in preservation and

control of the optical phase ranging from ultrashort time scales (10^{-15}) to long time windows exceeding one second, has formed the foundation for precision optical frequency metrology, combined time- and frequency-domain spectroscopy, and high-resolution and ultrafast quantum control.

2. Precision optical frequency metrology

The historical development of optical frequency metrology is intimately related to precision spectroscopy, clock-signal generation, and frequency synthesis [24]. The outstanding spectral properties of optical frequency standards offer unprecedented resolution and precision and potentially the highest accuracy for physical measurements. Researchers have constructed optical-frequency synthesis chains that span the vast frequency gap between specific optical and microwave spectral regions, resulting in a number of important measurements including the determination of the speed of light, the refinement of the Rydberg constant, the Lamb shift, and the fine structure constant. However, until recently it was an overwhelming challenge to synthesize arbitrary, absolute optical frequencies. Wide-bandwidth optical frequency combs have changed all that.

Measurement of absolute optical frequency. Under the framework of the present definition of the SI unit of second, for an optical frequency measurement to be absolute, it must be referenced to the primary microwave standard. Measurement of f_r is straightforward. Measurement of f_o is more involved as the pulse-to-pulse–carrier-envelope phase shift requires interferometric measurement. When the optical spectrum spans an octave in frequency, measurement of f_o is greatly simplified using the "self-referencing" technique [25]. An important aspect of the frequency comb technology is its high degree of reliability, precision, and accuracy. The uniformity of the comb's mode spacing has been verified to a level below 10^{-18} [26]. The most accurate absolute frequency measurement results come from optical standards that are based on transitions with extraordinary quality factors. The recent work at NIST shows an absolute frequency measurement of an optical clock transition in a single trapped Hg^+ ion with an uncertainty of 1×10^{-15} [27], limited by the Cs fountain clock [28]. For neutral atoms the recent determination of the absolute frequency of the ultranarrow $^1S_0 - {}^3P_0$ clock transition in ^{87}Sr is now within 2.4×10^{-15} uncertainty [29, 30], limited by the instability of a hydrogen maser calibrated by the primary Cs atomic fountain [28].

Optical atomic clock. With the advent of wide-bandwidth–optical-comb technology, it is now possible to transfer the stability of the highest-quality optical frequency standards across vast frequency gaps to other optical and microwave spectral regions. Recent experimental demonstrations support the concept that the most stable and accurate frequency standards will be based on optical transitions [27, 29, 30]. The advantage of optical frequency standards stems from their extraordinarily high resonance quality factors [29]. To create an optical atomic clock, one uses an optical frequency standard to stabilize f_r of a femtosecond comb, thus transferring the optical-phase information to the microwave domain [8, 9]. Since the comb system has two degrees of freedom, f_r and f_o, one has to ensure that a direct and unambiguous phase

relation between the optical standard and f_r is established. This implies that either f_o is strictly known via self-referencing or the influence of f_o is eliminated [9, 31, 32].

Optical clocks based on neutral atoms tightly confined in optical lattices are starting to show promise as future time/frequency standards [29, 30, 33-35]. These optical lattice clocks enjoy a high signal-to-noise ratio from the large numbers of atoms, while at the same time allowing Doppler-free interrogation of the clock transitions for long probing times, a feature typically associated with single trapped ions. The unique atomic structure of alkaline earth atoms such as strontium permits studies of narrow line physics based on the forbidden $^1S_0$ - $^3P_0$ and $^1S_0$ - $^3P_1$ transitions, permitting laser cooling to sub-recoil temperatures and observation of discrete momentum packets [36]. With these ultracold bosonic ^{88}Sr in free space, precision spectroscopy has revealed important cold collision-related resonance lineshape broadening and frequency shifts [37]. The millihertz-wide $^1S_0$ - $^3P_0$ line at 698 nm in ^{87}Sr, made possible due to the nuclear spin of 9/2 for fermionic ^{87}Sr, is especially attractive for an optical atomic clock. Ultracold ^{87}Sr atoms are trapped in a one-dimensional optical lattice engineered to have exactly matched AC Stark shifts between the ground and the excited states [38].

^{87}Sr atoms are first cooled in a dual-stage magneto-optical trap to mK temperatures using the strong (32 MHz) $^1S_0$ - $^1P_1$ line and then to μK temperatures using the weak (7 kHz) $^1S_0$ - $^3P_1$ intercombination line. Approximately 10^4 atoms are loaded into a 1D ~300 mW standing-wave optical lattice. The lattice wavelength of 813 nm is chosen to zero the net Stark shift of the clock transition, thus also eliminating line broadening due to the trapping potential inhomogeneity. The atoms are confined in the Lamb-Dicke regime, and the recoil frequency (5 kHz) is much smaller than the axial trap frequency (50 kHz). The probe is carefully aligned along the lattice axis and spectroscopy is both Doppler-free and recoil-free [34]. In the transverse direction, the lattice provides a trapping frequency of about 150 Hz, which is smaller than the recoil frequency, but still much larger than the clock transition linewidth. Atoms can be held in the perturbation-free lattice for times exceeding 1 s, which is important for Hz-level spectroscopy.

The extremely narrow natural linewidth (1 mHz) of the clock transition in ^{87}Sr is probed with a cavity-stabilized diode laser operating at 698 nm. The high finesse cavity is mounted in a vertical orientation to reduce sensitivity to vibrations. To characterize the probe laser, two independent stable laser systems are built and an optical heterodyne beat experiment is performed between the two lasers. Figure 1 shows that this heterodyne comparison demonstrates a laser linewidths below 0.2 Hz for a 3 s integration time and ~2 Hz for a 30-s integration time (limited by nonlinear laser drift). Another characterization is also performed by comparing the 698 nm laser against a highly stabilized Nd:YAG laser [5]. This comparison is made possible using a phase-stabilized femtosecond frequency comb, which is tightly locked to the 698 nm diode laser, precisely transferring the diode laser stability to each of the million modes of the comb. A heterodyne beat signal of ~ 1 Hz linewidth is demonstrated between the sub-Hz Nd:YAG laser and the corresponding fs comb mode at 1064 nm. This measurement demonstrates the reality of frequency stability transfer at the 10^{-15} level between lasers of different colors.

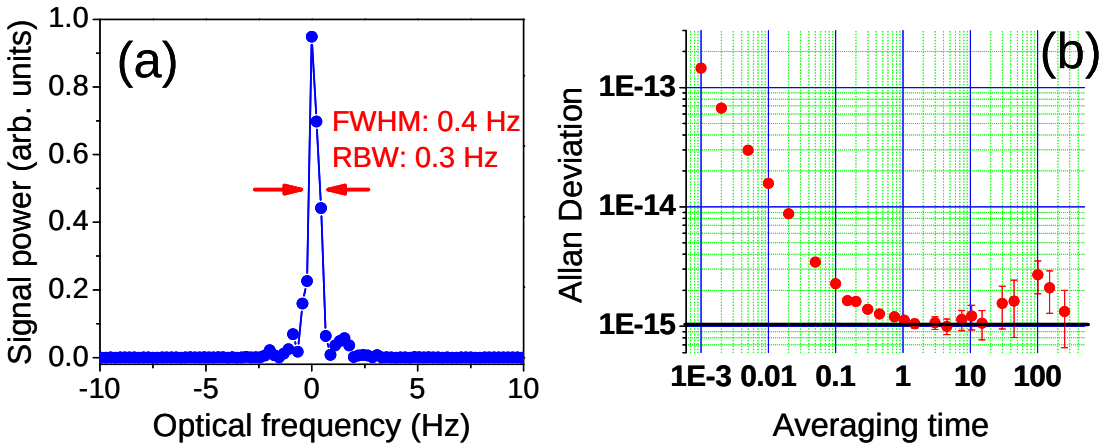

Fig. 1 Measurement of probe laser (698 nm) linewidth by optical heterodyne comparison between two independent laser systems. (a) A measurement of laser linewidth from the heterodyne beat. (b) The corresponding Allan deviation (fractional frequency instability) of the stabilized laser. The solid line at a fractional frequency stability of 1×10^{-15} denotes the thermal noise stability limit of the passive optical cavity.

For absolute frequency measurements of the clock transition, we frequency count the probe laser against a hydrogen maser signal calibrated by the NIST primary Cs fountain clock. A self-referenced octave spanning frequency comb is locked to the probe laser and its repetition rate is counted. The instability of this frequency counting signal is $2.5 \times 10^{-13}/\sqrt{\tau}$, where τ is the integration time. This is the limitation on frequency counting statistics. The measured clock frequency is 429,228,004,229,874.96(1.16) Hz [30].

When Zeeman sublevels of the ground and excited clock states are degenerate (nuclear spin $I = 9/2$ for $^1S_0$ - $^3P_0$) linewidths of < 5 Hz (resonance quality factor $Q \sim 10^{14}$) are achieved. This spectral resolution greatly facilitates evaluation of systematic effects below the 10^{-15} level. The differential magnetic moment between the ground and excited states leads to a first-order Zeeman shift of the clock transition. This can lead to shifts or broadening from stray magnetic fields, depending on the population distribution among the magnetic sublevels. By varying the strength of an applied magnetic field in three orthogonal directions and measuring the spectral linewidth as a function of field strength, the uncertainty of the residual magnetic field has been reduced to < 5 mG for each axis. The resulting net uncertainty for magnetically-induced frequency shifts is now <0.2 Hz ($<5 \times 10^{-16}$). Understanding and controlling the magnetic shifts is essential for the ^{87}Sr optical clock since the accuracy of all recent measurements has been limited by the sensitivity to magnetic fields.

Reduction of other systematic uncertainties (due to lattice intensity, probe intensity, and atom density) is straightforward with the high spectral resolution. Recent results in JILA indicate an overall systematic uncertainty of 9×10^{-16} for the Sr lattice clock. Using the hydrogen maser as the frequency reference, the averaging times necessary

to achieve 10^{-15} uncertainties for all systematic effects are still long. A more effecive approach to studying most systematic effects is thus to make frequency measurements under several values of the same systematic parameter within a time interval sufficiently short that the mode frequency of the ultrastable optical cavity used as a reference does not drift over the desired level of uncertainty. In the future, direct comparison among various optical clocks, such as Sr against single trapped ion-based optical atomic clocks (Hg^+ and Al^+) at NIST, will help to fully evaluate the system stability and accuracy.

The clock transition is probed at an unprecedented level of spectral resolution. With the nuclear spin degeneracy removed by a small magnetic field, individual transition components allow exploring the ultimate limit of our resolution by eliminating any broadening due to residual magnetic fields or light shifts. Figure 2(a) shows a sample spectrum of the $^1S_0$ ($m_F = 5/2$) - $^3P_0$ ($m_F = 5/2$) transition, where m_F is the nuclear spin projection onto the lattice polarization axis. The linewidth is probe time limited to ~ 1.8 Hz, representing a line Q of ~ 2.4 x 10^{14}. This Q value, the highest ever achieved in any form of coherent spectroscopy, is reproduced reliably, with some scatter of the measured linewidths in the 1-3 Hz range. The hertz level linewidths allows one to resolve all hyperfine components of the clock transition (nuclear spin is I=9/2 for ^{87}Sr), and measure the differential ground-excited g-factor that arises from hyperfine mixing of $^3P_0$ with $^3P_1$ and $^1P_1$. This measurement yields an experimental determination of the $^3P_0$ lifetime.

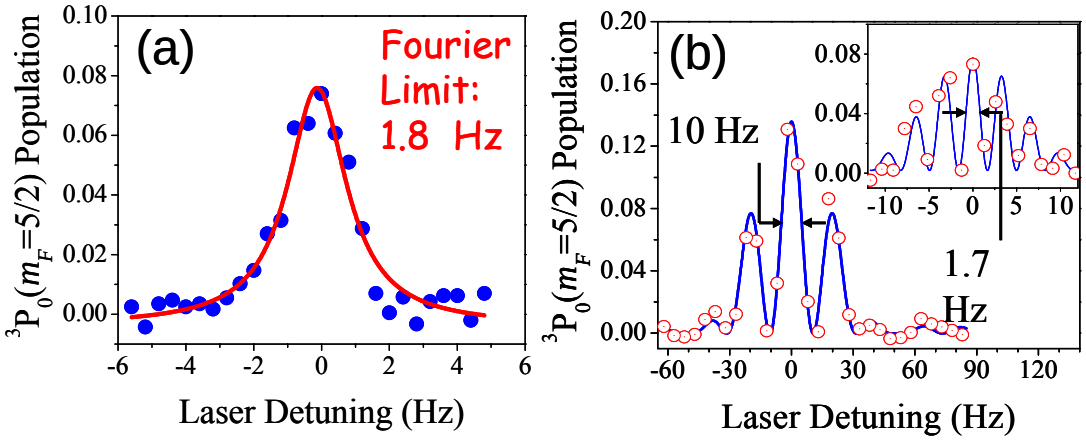

Fig. 2 (a) A typical spectrum of the $^1S_0$ - $^3P_0$ clock transition, exhibiting a line quality factor Q ~2.4 x 10^{14}. The linewidth shown is 2.1(2) Hz, in good agreement with the probe time limit of 1.8 Hz. One such trace takes approximately 30 seconds to collect (since the atom trap must be reloaded for each data point), and involves no averaging or normalization. (b) Ramsey spectra of the $^1S_0$ - $^3P_0$ clock transition. The preparation and probe pulses are 20 ms, the evolution time is 25 ms. Fringe width is 10.4(2) Hz. In the inset, the preparation and probe pulses are 80 ms, the evolution time is 200 ms. Fringe width is 1.7(1) Hz.

Besides the single-pulse spectroscopy of the clock line, two-pulse optical Ramsey experiments can also be performed on an isolated Zeeman component. When a system is limited by the atom or trap lifetime, the Ramsey technique can yield higher spectral resolution at the expense of signal contrast. An additional motivation for Ramsey spectroscopy in the Lamb-Dicke regime is the ability to use long interrogation pulses,

which results in a drastically narrowed Rabi pedestal compared to free-space atoms. The reduced number of Ramsey fringes facilitates the identification of the central fringe. Figure 2(b) shows a sample Ramsey spectrum, where the preparation and probe pulses are 20 ms and the free evolution time is 25 ms, yielding a pattern with a fringe width of 10.4(2) Hz, as expected. Figure 2(b) inset shows the same transition with the preparation and probe pulses of 80 ms and the evolution time of 200 ms. Here the width of the central fringe is reduced to 1.7(1) Hz. Both spectra exhibit no degradation of the fringe contrast. However, the quality of the spectra deteriorated at longer evolution times. Our inability to increase the resolution as compared to single-pulse Rabi spectroscopy suggests that the linewidth is not limited by the atom or trap lifetime, but rather by phase decoherence between the light and atoms, most likely due to nonlinear laser frequency fluctuations during the scan. This is supported by Rabi spectroscopy where the the laser stability appeared to limit the linewidth repeatability at the probe time limit near 0.9 Hz.

Optical frequency synthesizer. A future goal of ultrafast technology is to demonstrate arbitrary pulse synthesis in the time domain, including the capability of phase-coherent stitching of distinct optical bandwidths. This time-domain capability would complement the goal of constructing an optical frequency synthesizer that allows access in the frequency domain to any optical-spectral feature with a well-defined optical carrier wave. Such a capability would greatly simplify precision-laser spectroscopy. The frequency comb provides an optical frequency grid with lines repeating every f_r over an octave optical bandwidth and with every line stable at the 1-hertz level. This capability creates the basic infrastructure to construct a highly stable frequency synthesizer for both rf and optical spectral domains. In a traditional rf synthesizer, the output is a single-frequency rf-"delta" function that can be tuned to any desired frequency on demand. Realization of such a frequency synthesizer in the optical domain is facilitated by a wide-tunable cw laser that covers a significant portion of the visible spectrum. The frequency of the cw lasers is controlled by the optical frequency comb and is directly linked to the absolute time/frequency standard. Such a system with continuous, precise frequency tuning and arbitrary frequency setting on demand is now in existence in our laboratory [10].

3. Wide-bandwidth and high-resolution spectroscopy

The advent of precision femtosecond optical combs brings a new set of tools for precision atomic and molecular spectroscopy. For example, ultrafast lasers are now being used not only for time-resolved spectroscopy on fast dynamics, but also for precision spectroscopy on structural information. Indeed, coherent control of dynamics and precision measurement are merging into a joint venture. The ability to preserve optical phase coherence and superior spectral resolution over a wide spectral bandwidth permits detailed and quantitative studies of atomic and molecular structure and dynamics. The spectral analysis can be performed over a broad wavelength range, allowing precise investigations of minute changes in atomic and molecular structure over a large dynamic range. For example, absolute frequency measurement of vibration-overtone transitions and other related resonances (such as hyperfine splitting) can reveal precise information about the molecular potential-energy surface and relevant perturbation effects.

I_2 hyperfine interactions and clocks. With the development of an optical frequency synthesizer, we have performed high-resolution and high-precision measurement of hyperfine interactions of the first excited electronic state (B) of molecular iodine (I_2) over an extensive range of vibrational and rotational quantum numbers towards the dissociation limit. Experimental data demonstrate systematic variations in the hyperfine parameters that confirm calculations based on *ab initio* molecular potential energy curves and electronic wave functions derived from a separated-atomic-basis set. We have accurately determined the state-dependent quantitative changes of hyperfine interactions caused by perturbations from other electronic states and identified the respective perturbing states [39]. Analysis of various perturbation effects leads to precise determination of molecular structure over a large dynamic range [40, 41]. The work on I_2 near the dissociation limit is also motivated by the desire to establish a cell-based, portable optical molecular clock [42, 43].

Substantial progress is being made in the control of molecular degrees of freedom, with the goal of preparing molecules in a single quantum state for both internal and external degrees of freedom. Cold molecules provide a new paradigm for precision tests of possible time-variations of fundamental constants. For example, when both electronic and vibrational transitions are probed precisely, one would be comparing clocks built from two fundamentally different interactions, one of the origin of quantum electrodynamics (characterized by the fine structure constant α), the other of strong interaction (characterized by the electron/proton mass ratio). Optical frequency combs will not only aid in the production of ultracold molecules but also allow studies of molecular spectroscopy and quantum control with unprecedented precision and resolution.

Unite Time Frequency spectroscopy. With precise control of both time- and frequency-domain properties of a pulse train, we have combined these two applications in a spectroscopic study of ultracold Rb atoms, achieving united time-frequency spectroscopy for dynamics and global structure [13, 14]. Precision spectroscopy of global atomic structure is achieved with a direct use of a single, phase-stabilized femtosecond optical comb. The pulsed nature of excitation allows real-time monitoring and control capabilities for both optical and quantum coherent interactions and state transfer. It is a synthesis of the fields of precision spectroscopy and coherent control: at short time scales we monitor and control the coherent accumulation and population transfer, at long times we recover all the information pertinent to the atomic level structure at a resolution limited only by the natural atomic linewidth, with a spectral coverage spanning hundreds of terahertz. This powerful combination of frequency domain precision and time domain dynamics represents a new paradigm for spectroscopy.

The spectroscopic resolution and precision is not compromised by the use of ultrafast pulses, since they are associated with a phase-stabilized, wide-bandwidth femtosecond comb. In other words, phase coherence among successive ultrashort pulses supports spectroscopic resolution limited only by the length of the overall pulse train. Phase coherence among various transition pathways through different intermediate states produces multi-path–quantum-interference effects on the

resonantly enhanced two-photon-transition probability. The transition spectrum is analyzed in terms of f_r and f_o. Both are stabilized to high precision. Prior knowledge of atomic transition frequencies is not essential for this technique to work, indicating that it can be applied in a broad context. These results show the significant advantage of the direct frequency comb spectroscopy (DFCS) that the comb frequencies may be absolutely referenced, for example to a Cesium atomic clock, enabling precision spectroscopy over a bandwidth of several tens of nanometers. Another unique feature of the wide-bandwidth optical comb is that it allows all relevant intermediate states to resonantly participate in the two-photon excitation process. This participation, in turn, permits phase coherence among different comb components to induce a stronger transition rate through quantum interference. The resonant interaction with the intermediate states also makes it possible to explore population transfer dynamics and the mechanical consequences of light-atom interactions. Furthermore, spectral phase manipulation can now be combined with the time-domain optical phase coherence to enable quantum coherent control at resolutions limited only by the natural linewidths [15]. Indeed, the field of coherent control of atomic and molecular systems has seen advances incorporating high power femtosecond laser sources and pulse shaping technology. This has allowed for demonstrations of robust coherent population transfer via adiabatic passage techniques, coherent control of two-photon absorption, resolution enhancement of coherent anti-Stokes Raman scattering, and progress towards cold-atom photoassociation [44]. It is in this exciting context that we can now combine the femtosecond comb and spectral phase manipulation with the aim towards coherent control at the highest possible spectroscopic resolution.

Properties of the transient coherent accumulation effect, including spectral phase manipulation, are explicitly demonstrated via a finite number of phase-stabilized femtosecond pulses. Specifically, we can measure atomic transition linewidth and population transfer versus the number of applied phase-coherent femtosecond pulses and as a function of linear frequency chirp. The basic idea of multi-pulse coherent accumulation is that the transition amplitude for excitation of a specific atomic energy level may be increased significantly with the number of femtosecond pulses, and can be done such that other non-resonant states remain unexcited, thus enabling high state selectivity. Tuning the comb leads to constructive or destructive quantum interferences of the two-photon transition amplitude only observable in this multi-pulse context and may be useful for eliminating nonlinear absorption. Positive and negative linear frequency chirp, as well as spectral phase manipulation via a spatial light modulator, can be applied to the comb modes and thus the two-photon transition amplitudes. It is shown that the role of chirp in the single pulse case is no longer necessarily applicable in these multi-pulse experiments since the atomic coherence persists between femtosecond pulses. The combination of pulse shaping with the femtosecond comb is shown to increase the signal of a two-photon transition at a specific chirp while maintaining high resolution.

4. Carrier-envelope phase coherence and Time-domain applications

Femtosecond-comb technology has also made dramatic advances in time-domain experiments as it has in optical frequency metrology. Stabilization of the “absolute” carrier-envelope phase at a level of tens of milliradians has been demonstrated and

maintained over many minutes, laying the groundwork for electric field synthesis. The capability of precisely controlling pulse timing and the carrier-envelope phase allows one to manipulate pulses using novel techniques and achieve unprecedented levels of flexibility and precision.

Timing synchronization and phase lock of independent mode-locked lasers. To establish phase coherence among independent ultrafast lasers, it is necessary to first achieve synchronization among these lasers so that the remaining timing jitter is less than the oscillation period of the optical carrier. Detecting timing jitter should be carried out at a high harmonic of f_r to attain much-enhanced detection sensitivity. This approach has enabled tight synchronization between two independent mode-locked lasers with a residual rms-timing jitter < 1 fs [45-47]. The second step in achieving phase locking of separate femtosecond lasers requires effective stabilization of the phase difference between the two optical carrier waves. Phase locking demands that the spectral combs of individual lasers be maintained exactly coincident in the region of spectral overlap so that the two sets of combs form a continuous and phase-coherent entity. We detect a coherent heterodyne-beat signal between the two mode-locked lasers, yielding information about the difference in the offset frequencies, which can then be controlled, resulting in two pulse trains with nearly identical phase evolution. The established phase coherence between the two lasers can be revealed via a direct time-domain analysis. When the two femtosecond lasers are phase locked, autocorrelation reveals a clean pulse that is shorter in apparent duration and larger in amplitude than the individual original pulses. A successful implementation of coherent light synthesis has therefore become reality: the coherent combination of output from more than one laser where the combined output can be viewed as a coherent, femtosecond pulse being emitted from a single source [16].

Extend phase-coherent femtosecond combs to the mid-IR spectral region. Being able to combine the characteristics of two or more pulsed lasers working at different wavelengths provides a more flexible approach to coherent control. This may be particularly important in previously unreachable spectral regions. Two stabilized mode-locked Ti:sapphire lasers are employed to enable difference-frequency generation (DFG), which is tuned from a few to tens of microns [18]. The ultimate goal of this work is to make an optical-waveform synthesizer that can create an arbitrary optical pulse on demand and to use the novel source to study and control molecular motion. For precision molecular spectroscopy in the IR region, the DFG approach produces an absolute frequency-calibrated-IR comb. One of the important spectral regions is 1.5 μm, where compact, reliable, and efficient mode-locked lasers exist and there is rich information on molecular spectra. Distribution of optical frequency standards over optical fiber networks is also important [48]. We have achieved tight synchronization and coherent phase locking between the 1.5 μm mode-locked lasers and a visible femtosecond frequency comb [49].

Femtosecond lasers and external optical cavities. As reported earlier, coherent optical spectroscopy has now led to the recovery of a record-high quality factor ($Q > 2.4 \times 10^{14}$) for a doubly-“forbidden” natural resonance observed in a large ensemble of trapped ultracold Sr atoms. This unprecedented spectral resolving power impacts fields ranging from precision frequency metrology to quantum optics and quantum

information science. Ultrastable lasers, together with optical frequency combs, can now maintain optical phase coherence beyond 1 s and transfer this stability across hundreds of terahertz. As it becomes increasingly challenging to maintain phase coherence beyond multiples of seconds, it is natural that we look beyond the visible domain and consider speeding up the "wheel of precision measurement" to the next level of carrier frequency. We have thus pursued two related experimental directions to address this vision. One is the generation of phase coherent frequency combs in the VUV (50 - 200 nm) spectral domain [19, 20]. In parallel, as described in the previous section, we have also pursued direct frequency comb spectroscopy to ready ourselves for quantum optics and precision spectroscopy once phase coherent sources become available in VUV.

Both experiments benefit from the use of femtosecond enhancement cavities [50]. These are passive optical cavities with high finesse and low dispersion over a large spectral bandwidth such that incident femtosecond pulse trains can be efficiently coupled inside [17]. Pulse energies can be enhanced by three orders of magnitude to >10 μJ while the original pulse repetition frequency is maintained. This capability permits phase coherent high harmonic generation process to take place at enhanced average efficiency [19]. In addition to the power enhancement aspect, femtosecond cavities effectively increase the interaction length between matter and light, allowing direct frequency comb spectroscopy to acquire linear or nonlinear atomic and molecular signals with dramatically increased sensitivity [21].

We have recently accomplished several important studies on the interaction between a femtosecond-laser based optical frequency comb and a high-finesse, low-dispersion, passive optical cavity. We have achieved direct stabilization of a frequency comb to a high-finesse optical cavity [51]. The resulting frequency/phase stability between the frequency comb and the cavity modes demonstrates a fully coherent process of intracavity pulse buildup and storage. We have also developed a femtosecond comb-based measurement protocol to precisely characterize mirror loss and dispersion [52]. This technical capability has facilitated production of large bandwidth, low-loss and low-dispersion mirrors. In addition, we have studied the nonlinear response of intracavity optical elements, demonstrating their limitation on power scalability [53]. This study has led to the design of novel cavity geometries to overcome this limitation [54]. In short, we have achieved nearly three-orders of power enhancement inside a femtosecond buildup cavity within a spectral bandwidth of ~30 nm, resulting in an intracavity pulse train that (1) is completely phase coherent to the original comb from the oscillator, (2) has the original laser's repetition rate (~100 MHz), (3) has a pulse peak energy exceeding 5 μJ, (average power > 500 W), intracavity peak intensity > 10^{13} W/cm^2, and (4) is under 60 fs pulse duration. We also note that this enhancement cavity approach is compatible with a number of femtosecond laser systems, including mode-locked Ti:Sapphire and fiber lasers.

The coherently enhanced pulse stored in the cavity can be switched out using a cavity-dumping element (Bragg cell), resulting in a single, phase-coherent, amplified pulse [50]. The linear response of the passive cavity allows the pulse energy to build up inside the cavity until limited by cavity loss and/or dispersive pulse spreading. The

net cavity group delay dispersion over the bandwidth of the pulse has been minimized to maintain the shape of the resonant pulse. We have applied the coherent pulse-stacking technique to both picosecond [55] and femtosecond pulses [17], demonstrating amplifications > 500. An important application of these advanced pulse-control technologies is in the field of nonlinear optical spectroscopy and nanoscale imaging. Using two tightly synchronized picosecond lasers, one can achieve a significant improvement in experimental sensitivity and spatial resolutions for vibrational imaging based on Coherent Anti-Stokes Raman Spectroscopy (CARS) for acquisition of chemically selective maps of biological samples. Tight synchronization between the pump and Stokes beams tuned to a Raman-active vibrational mode eliminates background noise. The technologies of pulse synchronization and coherent pulse stacking have become ideal tools for this task of combining spectroscopy with microscopy [56, 57].

Massively parallel, highly sensitive, wide bandwidth, high resolution spectroscopy. With every optical comb component efficiently coupled into a respective high finesse cavity mode, we have established a network of parallel channels for ultrasensitive detection of molecular dynamics and trace analysis. This configuration provides an ideal spectroscopic paradigm suitable for the next generation of atomic and molecular measurements [21]. The approach presents simultaneously the following attractive characteristics: (i) A large spectral bandwidth allowing for the observation of global energy level structure of many different atomic and molecular species; (ii) High spectral resolution for the identification and quantitative analysis of individual spectral features; (iii) High sensitivity for detection of trace amounts of atoms or molecules and for recovery of weak spectral features; and (iv) A fast spectral acquisition time, which takes advantage of high sensitivity, for the study of dynamics.

We have developed cavity-enhanced direct frequency-comb spectroscopy utilizing a broad-bandwidth optical frequency comb coherently coupled to a high finesse optical cavity inside which atomic or molecular samples are located. Hundreds of thousands of optical comb components, each coupled into a specific cavity mode, collectively provide sensitive intracavity absorption information simultaneously across 100 nm bandwidth in the visible and near IR spectral region. By placing various atomic and molecular species inside the cavity, we have demonstrated real-time, quantitative measurements of the trace presence, transition strengths and linewidths, and population redistributions due to collisions and temperature changes. This novel capability to sensitively and quantitatively monitor multi-species molecular spectra over a large optical bandwidth in real-time provides a new spectroscopic paradigm for studying molecular vibrational dynamics, chemical reactions, and trace analysis. We will continue to develop state-of-the-art laser sources in the infrared spectral regions, possibly even covering the important 3 micron area, to further improve the system sensitivity.

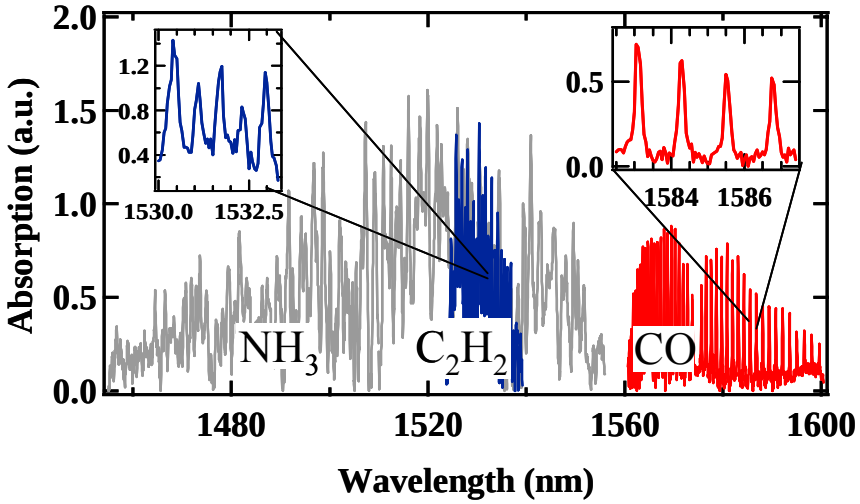

Fig. 3 Absorption spectra acquired by the cavity-assisted, massively parallel, frequency comb spectroscopy for 2 torr CO, 10 millitorr NH_3 and 1.5 millitorr C_2H_2, showing 150 nm of spectral information. The inset of the P-branch of the CO overtone spectrum show individually resolved rotational lines.

Extreme nonlinear optics. To extend the coherent frequency comb structure and related precision measurement capabilities into the deep UV spectral region, we have recently demonstrated high-harmonic generation (HHG) in noble-gas ionization experiments at 100 MHz repetition rates enabled by a femtosecond enhancement cavity [19]. HHG provides a coherent source of vacuum-ultraviolet to soft x-ray radiation. HHG has traditionally relied on high-energy, low repetition rate amplified laser systems to provide the peak intensities needed for ionization of the gas target. The small conversion efficiency of the process, combined with the low repetition rate of amplified laser systems, results in low average powers in the XUV generation. Furthermore, the use of these sources as precision spectroscopic tools is limited, as the original laser frequency comb structure is lost in the HHG process. Using a femtosecond laser coupled to a passive optical cavity, coherent frequency combs in the XUV spectral region are generated via high-harmonics of the laser without any active amplification or decimation of the repetition frequency. We can thus significantly improve the average power conversion efficiency and reduce the system cost and size, while dramatically improving the spectral resolution. Since little of the fundamental pulse energy is converted, a femtosecond enhancement cavity is ideally suited for HHG as the driving pulse is continually recycled after each pass through the gas target. The presence of the frequency comb structure in the XUV and its extreme spectral resolution will enable similar revolutions in precision measurement, quantum control, and ultrafast science as in the visible region.

The enhancement cavity builds up the pulse energy from 8 nJ to 4.8 μJ, while maintaining the original pulse width of 60 fs and f_r of 100 MHz. The peak intracavity intensity of $>3 \times 10^{13}$ W/cm^2 is obtained at the intracavity focus. The single-shot efficiency of high harmonic generation using this technique is comparable to traditional amplifier-based systems at similar intensity levels. This demonstrates the dramatic increase in high-harmonic power that can be accessed using a high repetition

rate. High precision measurement of phase/frequency fluctuations in the high-harmonic generation process has also been performed for the first time. Two sets of the frequency combs at 266 nm that represent the third harmonic of the fundamental IR comb are brought together for beat detection. The rf spectrum of the beat note shows the clear presence of the comb structure in the UV. The resolution bandwidth-limited 1 Hz beat signal (Fig. 4) demonstrates that the full temporal coherence of the original near-IR comb has been precisely transferred to high harmonics.

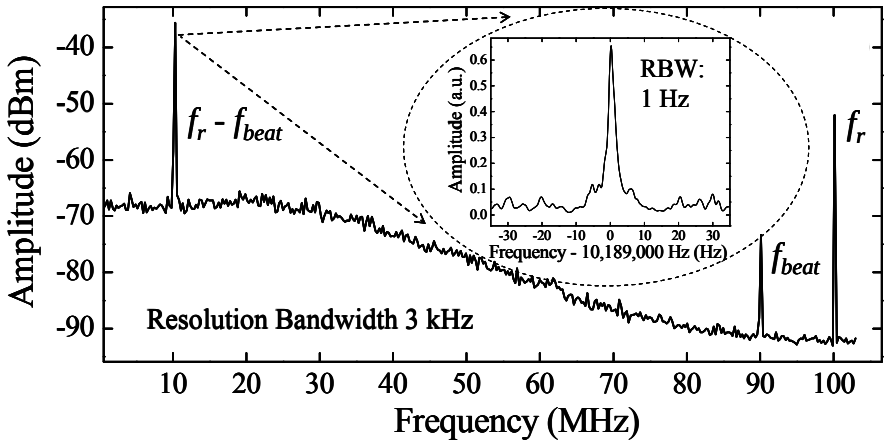

Fig. 4 Coherent heterodyne beat signal is detected between the HHG in Xe gas and bound optical nonlinearities in BBO. These two frequency combs (both at 3rd harmonic) spectrally overlap and provide the optical heterodyne beat signal at an offset radio frequency introduced by an acousto-optic modulator placed in one arm of the interferometer. The two corresponding pulses trains are overlapped in time. The coherent beat signal and repetition frequency detection are shown, demonstrating the HHG comb is phase coherent with respect to its parent comb from the laser, with the coherence limited only by the observation time. The linewidth shown in the inset is resolution bandwidth limited at 1 Hz.

To couple the HHG light out of the cavity, a thin sapphire plate is placed at Brewster's angle (for the IR) inside the cavity. However, the nonlinear response of the intracavity Brewster plate has so far limited the power scalability of the system. To solve this problem, we have designed novel enhancement cavity configurations that will allow us to use more powerful lasers [54]. One of the focusing cavity mirrors has a 200-μm hole drilled in the middle. By using a higher order cavity mode such as TEM01, we are still able to build up sufficient peak power inside the cavity for HHG to work. The generated VUV comb, however, leaks out of the mirror hole due to the significantly smaller diffraction angles enjoyed by the shorter wavelength light beam. This cavity geometry will allow a larger buildup power inside the cavity without any intracavity optics.

5. Summary

Recent developments in phase control of coherent light, ranging from cw lasers to ultrafast femtosecond lasers, have enabled breakthroughs in optical frequency metrology, optical frequency synthesis, optical atomic clocks, coherent control, extreme nonlinear optics, and sub-optical cycle physics. This revolution is continuing, with many exciting results emerging at this time or being expected to come in the near

future. We are indeed witnessing a wonderful period of amazing light! Of course, all of these revolutions would not have been possible without the coherent light source – the invention of LASER from Charlie Townes.

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