Coherent control of light and its broad applications

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ABSTRACT

Precise phase control of ultra-wide-bandwidth optical frequency combs has produced remarkable and unexpected progress in precision metrology and ultrafast science. The combination of the ability to do completely arbitrary, optical, waveform synthesis with recently developed optical pulse measurement techniques is analogous to the development of oscilloscopes and waveform generators in the early 20^{th} century. The development of ultra-stable optical frequency standards into optical atomic clocks and optical frequency synthesizers again complement and rival the similar technologies developed in the radio frequency domain.

The recent unification of ultra-precision laser technology with ultrafast lasers and nonlinear optics has enabled revolutionary progress in both areas that has significant potential for diverse application in the physical science and technology. Precision optical frequency measurements have been reduced to a simple task even while the highest level of measurement precision has been achieved. Precision optical clocks have now been established, using optical transitions of superior quality factors to provide radio-frequency clock signals with stability surpassing almost all conventional rf sources. An optical frequency synthesizer has also been demonstrated, with a high-power, widely tunable, single-frequency cw laser providing the desired optical frequency output on demand. Control of the carrier-envelope phase of ultrafast pulses has become possible. This advance in femtosecond technology is important for both extreme nonlinear optics and optical frequency metrology. Pulse trains from independent mode-locked lasers have been synchronized below the 1 fs level and their carrier waves phase locked, leading to coherent pulse synthesis. We now appear to have 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 perform 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 20th century.

Of course this explosion of interdisciplinary research activities comes to fruition only because of physical insight and mature technologies developed by many pioneers who have devoted decades of hard work in the independent fields of ultra-sensitive spectroscopy, ultra-stable lasers, ultra-fast lasers, and nonlinear optics. For example, commercially available solid-state Ti:Sapphire lasers provide simple and reliable solutions to the need of ultrafast laser technology for precision frequency metrology. The development of new optical fiber technologies allowed simple generation of octave-spanning bandwidth.

To understand the connection between the ultra-stable and the ultra-fast, we need to first point out each of their unique features. The ultra-stable field is typified by the variety of high-resolution spectroscopy and high-precision measurements carried out by continuous wave (CW) lasers that can be best described by their near delta-function frequency spectra. In sharp contrast, the field of ultra-fast phenomena encompasses the study of sub-picosecond events utilizing laser pulses that approach the limit of time domain delta-functions. In fact, at this point in time state-of-the-art laser sources from these two fields share nearly the same delta-function "figure of merit" with frequency and temporal widths on the order of a few parts in 10^{15} Hz and seconds, respectively. The connection between the ultra-stable and the ultra-fast arises from the fact that femtosecond (fs) laser oscillators produce pulses in a periodic train via mode-locking, with a corresponding rigorous periodicity in the spectral domain. In fact, the frequency domain spectrum consists of a comb of discrete modes separated by the repetition frequency $f_{rep}=1/\ \tau_{r.t.}$, where $\tau_{r.t.}$ is the cavity round trip time. The extent of the time domain pulse train provides the frequency resolution of individual comb components, while the total extent of the frequency domain mode comb is approximately limited to the inverse of the pulse duration. The generation of ultrashort pulses requires that the group velocity (v_g) dispersion inside the laser cavity is minimized across the pulse's frequency spectrum. This criterion is not directly related to the frequency comb spacing, since the individual mode frequencies correspond to eigenmodes of the phase-velocity (v_p) of the light. In general, we have $v_g \neq v_p$ due to laser cavity dispersion. This fact results in a pulse envelope function that is not fixed with respect to the underlying optical oscillation frequencies — there is a phase slip (denoted by $\Delta\phi$) between the "carrier" phase and the envelope peak for each of the successive pulses emitted by the laser. In the frequency domain, $\Delta\phi$ yields an offset of the mode comb from exact harmonics of the f_{rep} by the amount $f_{ceo} = \Delta\phi\, f_{rep}\ /2\pi$. Hence each optical comb frequency is effectively given by $f_n = nf_{rep} + f_{ceo}$. Here n represents an integer (on the order of 1 million) harmonic number of the optical comb line relative to the repetition rate, f_{rep} .

One sees how the frequency-domain control of both f_{rep} and f_{ceo} makes it possible to establish a fs-laser based optical comb at a high precision for optical frequency measurement and distribution. The bandwidth of a fs-laser comb already covers more than 100 nm, and it can be further extended to cover an entire optical octave using nonlinear optical effects enabled by microstructure fibers. With an octave-spanning spectrum it becomes possible to measure optical frequencies in a single step with a direct reference to the present realization of the SI unit of time, namely the cesium microwave standard. This is accomplished when the mode spacing (f_{rep}) of the femtosecond comb is locked with feedback control to an rf signal source that itself is referenced to cesium. The octave interval between the frequency f and its second harmonic $2f$ can be simply expressed as $2f - f = f = \mathrm{n}\, f_{rep}$. This strategy can be implemented in two equivalent ways. The first approach is to use the comb to measure the frequency gap between a CW laser and its second harmonic. And the second approach is to frequency double the infrared portion of the comb spectrum and to heterodyne it with the existing visible portion of the spectrum. The resulting beat frequency is the comb offset f_{ceo}. When the laser is controlled in such a fashion that both the radio frequencies f_{rep} and f_{ceo} are established with reference to the cesium standard, one then has an entire array of optical frequencies with precisely known frequencies $f_n = nf_{rep} + f_{ceo}$.

To achieve a better stability, it is advantageous to use an optical frequency standard instead of a microwave reference to stabilize an entire optical comb. In doing so one can in fact derive a microwave signal from the optical standard, leading to a so-called optical atomic clock. For this purpose it is important to establish an optical comb with excellent phase coherence among its individual components. The phase stability needs to exceed that of the optical standards. With this capability, we will be able to transfer the stability of a single optical oscillator to the entire comb set over its vast bandwidth, and also derive clock signals in the microwave/rf domain without any stability compromise. Optical standards based on single ions and cold atoms promising potential stability around 1 x 10^{-16} at 1 s and potential accuracy at 1 x 10^{-18} may very well become future national standards, but such systems would require elaborate designs. On the other hand, excellent candidates in cell-based optical frequency standards do exist, such as an iodine stabilized Nd:YAG laser, that would offer compact, simple, and less expensive system configurations, albeit at the cost of performance degradation by perhaps two decades. Along with optical combs, a competent laboratory would be able to realize a network of microwave and optical frequencies at a level of stability and reproducibility that surpasses the properties of basically all normal commercially available frequency sources, but with a reasonable cost. Easy access to resolution and stability offered by optical standards would greatly facilitate application of frequency metrology both to precision experiments for fundamental physics and to practical devices.

An optical frequency grid with stable lines over a large optical bandwidth is useful for a number of applications. However, often times we desire a single-frequency optical-"delta"-function that can be tuned to any preferred frequency position on demand. Realization of such an optical frequency synthesizer (analogous to its radio-frequency counterpart) will add a tremendously useful tool for modern optics-based experiments. One can foresee an array of diode lasers, each covering a successive tuning range of ~ 10 – 20 nanometers and emitting some reasonably useful power, that would collectively cover most part of the visible spectrum. Each diode laser frequency will be controlled by the stabilized optical comb, and therefore related to the absolute time/frequency standard, while the setting of the optical frequency will be done through computer control to any desired value. For the first step, we have constructed an electronic control system that allows a widely tunable diode laser to tune through an targeted spectral region at a 10 MHz step while maintaining reference to the stabilized optical comb. Suitable software allows the laser to be scanned on an arbitrary but exact pattern anywhere within its tuning range.

The frequency-domain-based laser control techniques have had a profound impact to the time-domain carrier-envelope phase stabilization. Precision control of the time-domain carrier-envelope phase has been a highly desirable and yet elusive goal in the ultrafast science since the advent of a few cycle pulses. Normally the absolute phase of an optical wave's electric field is not relevant, in that any shift in this phase has no measurable effect. However, within the context of few-cycle optical pulses, the electric field does not have this invariance. This condition is due to the reference provided by the few-cycle width of the pulse envelope and the value of the electric field's phase (relative to the envelope) drastically alters the optical character of a few-cycle pulse. One example where the pulse-shape matters is coherent x-ray (or high harmonic) generation. In addition to x-ray generation, other examples of extreme nonlinear optics or strong field processes where the absolute phase is critical include: attosecond pulse generation and strong-field ionization.

Our motivation of working with separate ultrafast lasers stems from the desire of creating an arbitrary light wave-form generator, with the capability of synchronizing and phase-locking arbitrary, separate mode-locked lasers of distinct optical properties. The ability to generate coherent light with ultra-broad bandwidths is essential for many applications in ultrafast science and technology. In the past ten years, advances in this area have led to broad-bandwidth, ultrashort-pulse, laser systems that routinely generate sub-10 femtosecond pulses. Moreover, coherent bandwidths can be extended over very broad spectral regions using nonlinear frequency conversion techniques such as white-light continuum generation, parametric amplification, molecular phase modulation, etc. However, for some applications these techniques can suffer from poor efficiency and lack of flexibility, in particular for applications where arbitrary pulse synthesis at very different wavelengths is required. Synthesis of arbitrary pulses by coherently combining the output fields of two or more separate lasers would enable the generation of optical waveforms with distinct optical properties in distinct regions of the spectrum with potentially high powers. Techniques we developed along these lines have already made a strong impact to the field of nonlinear-optics based spectroscopy and nanoscale imaging, showing significant improvements in experimental sensitivity and spatial resolutions.