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Laser Spectroscopy and Frequency Combs

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Abstract. The spectrum of a frequency comb, commonly generated by a mode-locked femtosecond laser consists of several hundred thousand precisely evenly spaced spectral lines. Such laser frequency combs have revolutionized the art measuring the frequency of light, and they provide the long-missing clockwork for optical atomic clocks. The invention of the frequency comb technique has been motivated by precision laser spectroscopy of the simple hydrogen atom. The availability of commercial instruments is facilitating the evolution of new applications far beyond the original purpose. Laser combs are becoming powerful instruments for broadband molecular spectroscopy by dramatically improving the resolution and recording speed of Fourier spectrometers and by creating new opportunities for highly multiplexed nonlinear spectroscopy, such as two-photon spectroscopy or coherent Raman spectroscopy. Other emerging applications of frequency combs range from fundamental research in astronomy, chemistry, or attosecond science to telecommunications and satellite navigation.

There could be no laser spectroscopy without the laser. In 1964, the Physics Nobel Prize recognized the seminal work on masers and lasers by Townes, Basov, and Prokhorov. Lasers soon became intriguing tools for nonlinear spectroscopy. Laser spectroscopy really took off with the advent of widely tunable and highly monochromatic dye lasers [1] in the early seventies. The first ICOLS conference was held in Vail, Colorado, in 1973, organized by Richard. G. Brewer and Aram Mooradian. Since then, the field has attracted many talented scientists, and today we count no fewer than 17 additional Nobel Prize winners in our community.

Sometimes, relatively simple concepts can create new tools that open new doors. One example is the laser frequency comb [2], which arrived about 15 years ago, as a tool for measuring optical frequencies of 100's or even 1000's of THz. By providing a phase-coherent link between the optical and the radio-frequency regions, a frequency comb makes it easy to count the frequency of a laser wave. Before, this task had remained close to impossible. A frequency comb is produced by a mode-locked femtosecond laser. Such lasers have been developed since the late 1960s, and they were in widespread use as sources of short flashes of light for the study of ultrafast phenomena in molecules, liquids, and solids. Researchers interested in precision spectroscopy were using a different kind of laser, operating on a single resonator mode, and perfected to achieve very high stability. Since then, frequency comb techniques have greatly enriched and extended the frontiers of both fields. A mode locked laser is a source of a million perfectly evenly spaced comb lines or longitudinal laser modes, all

oscillating continuously, and interfering so as to produce short pulses of light. During the dark time between pulses, the laser modes cancel each other by mutual interference.

Today, laser frequency combs are commercially available as ultra-precise spectroscopic measuring tools. A compact fiber-based frequency comb for space applications will be launched with a Texus sounding rocket in the Fall of 2013. The frequency range of laser combs is being extended to new regions from the THz regime to the extreme UV. Quite a few laboratories are exploring new approaches to create highly miniaturized comb generators, based on cascaded four-wave mixing in toroidal micro-resonators [3,4].

The original motivation for the invention of the frequency comb technique in our laboratory has been precision laser spectroscopy of the simple hydrogen atom, which permits unique confrontations between experiment and quantum electro-dynamic theory. The frequency of the particularly sharp 1S-2S two-photon resonance has recently been measured with an uncertainty of 4.2 x 10^{-15} [5,6]. Increasing control over systematic uncertainties may make it feasible for an atomic hydrogen optical clock to compete with ion clocks or optical lattice clocks, so that the second as unit of time could be redefined in terms of the simplest atom. Laser spectroscopy of additional transitions in hydrogen could help to resolve the "proton size puzzle" which has been revealed by laser spectroscopy of the 2S-2P Lamb shift in muonic hydrogen [7,8]. In the near future, laser spectroscopy of magnetically trapped antihydrogen should become feasible, so that one can search for conceivable differences between matter and antimatter.

Other applications of laser frequency combs have been evolving rapidly. Astronomers can use laser combs as calibration sources [9] for highly resolving spectrographs to search for extra-solar planets or to observe the postulated accelerating expansion of space in the universe. The control of the electric field of ultrafast laser pulses with frequency comb techniques has become [10] a key for attosecond science. Amongst the unforeseen applications of frequency combs, broad spectral bandwidth linear and nonlinear spectroscopy holds much promise: we can study complex molecular spectra in a highly multiplexed manner by using a hundred thousand or a million comb lines all at once.
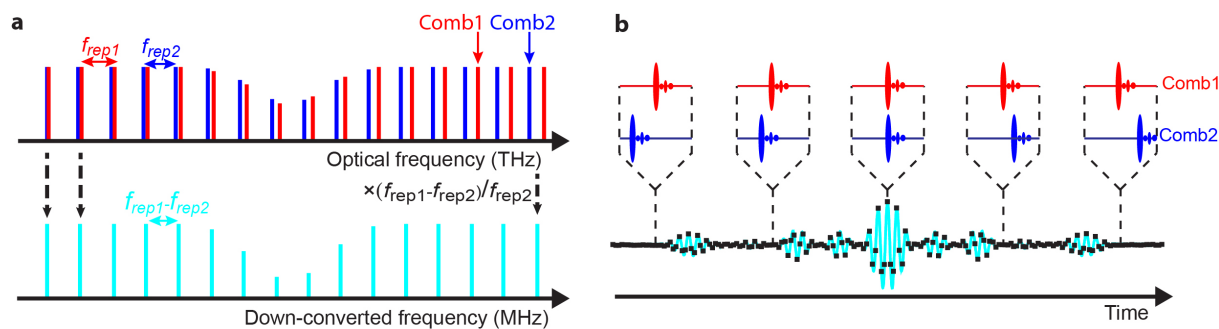

Figure 1. a) Frequency domain picture of the physical principle of dual-comb spectroscopy. Due to interference between pairs of optical comb lines, the optical spectrum is effectively mapped into a radio-frequency comb. The frequencies of the optical spectrum are down-converted to $\Delta f_{ceo} + n (f_{rep1} - f_{rep2})$, where Δf_{ceo} is the difference of carrier-envelope slippage frequencies of the two combs and n an integer. b) Time domain picture of the physical principle of dual-comb spectroscopy. The pulses of two combs with slightly different repetition periods interfere. The pulses from each comb overlap at linearly increasing time delays, mimicking a scanning interferometer.

Fourier Transform Spectroscopy [11] is a known method of highly multiplexed spectroscopy. This technique is in widespread use in many laboratories. The basic approach has not changed much since this technique emerged in the early seventies [12]. A Fourier spectrometer uses a (incoherent) light source, a moving-arm Michelson interferometer, and a single photo-detector connected to the data

acquisition electronics of a computer. The autocorrelation function of the light, as filtered by an absorbing sample, the "interferogram", is Fourier-transformed to reveal the spectrum.

The incoherent light source can be replaced by a broadband laser frequency comb source of repetition frequency f_{rep1}, and the light reflected of a moving mirror can be replaced by that of a second laser frequency comb, tuned to a slightly different pulse repetition frequency f_{rep2}. This type of dual-comb spectroscopy offers several important advantages. First it uses coherent laser light rather than incoherent light. Secondly it does not require any moving parts. By simply dialling a knob, one can simulate a mirror moving at 10 km/s, the escape velocity from earth. In this way, 1/f noise in the detector signal can be greatly reduced, and a million-fold improvement in the acquisition speed of a spectrum has been demonstrated.

In the frequency domain (Fig.1a), dual-comb spectroscopy can be understood by considering the two frequency combs of slightly differing line spacing. Each pair of optical lines, one from each comb, produces a radio frequency beat note on the detector. These beat signals form a frequency comb in the radio frequency regime, spaced by $f_{rep1}-f_{rep2}$. Optical frequencies are thus converted into radio frequencies with a conversion factor $(f_{rep1}-f_{rep2})/f_{rep2}$.

In the time domain (Fig.1b), dual-comb spectroscopy can be described in the language of asynchronous optical sampling. A first pulse excites the molecules and a second pulse observes the free induction decay by interferometric sampling. As in a sampling oscilloscope, a periodic waveform, sampled asynchronouly, appears stretched in time by a factor $f_{rep2}/(f_{rep1}-f_{rep2})$, so that the signal can be recorded electronically and digitally processed.

Linear absorption dual-comb spectroscopy (Fig. 2a) requires two frequency comb sources, a single fast photodetector and a computer. It has been used [13-23] to demonstrate very short acquisition times on the order of microseconds, extreme sensitivity when used with cavity enhancement, and low to extreme resolution. Extreme accuracy is possible, the technique can reveal absorption (Fig. 2b) and dispersion, and it can be applied anywhere from THz frequencies to the VUV. By recording signals of some seconds, individual comb lines (Fig.3) are resolved [22]. A molecular spectrum is thus only sampled at discrete frequencies, but frequencies that can be known with the accuracy of an atomic clock. The comb spectrum can be scanned or interleaved for more complete coverage.
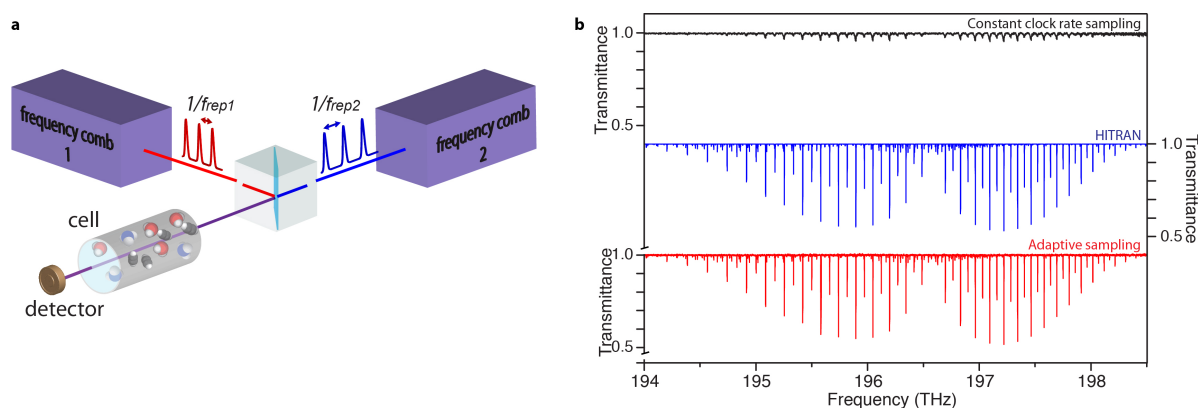

Figure 2. a) Dual-comb absorption spectroscopy. Two frequency combs, 1 and 2, have slightly repetition frequencies. Their beams are combined and are transmitted through the cell. They beat on a fast photodiode. b) Experimental dual-comb absorption spectra of $^{12}C_2H_2$ and $^{12}C^{13}CH_2$. Two experimental spectra are compared to a spectrum (blue) computed from the line parameters available in the 2008 HITRAN database. Both experimental spectra are measured with free-running fs lasers in identical conditions except for the interferometric real-time signal processing data acquisition clock. Two hundred consecutive spectra with an apodized resolution of 1.1 GHz, each measured within 467 μs, are averaged, resulting in a total measurement time of 93 ms and a total experimental time of 580 ms. When sampled at the constant clock rate of the digitizing data acquisition board, the spectrum

(black) is strongly distorted by the relative short-term fluctuations of the repetition frequency and carrier envelope phase slips of the fs lasers. This lowers the sensitivity and scrambles the spectral features. When compensating for the laser instabilities in real-time, the agreement of the experimental "adaptive" spectrum (red) with the computed spectrum is excellent and confirms the suitability of our technique for measurements with Doppler-limited resolution.

Despite its intriguing potential for the measurement of molecular absorption spectra spanning tens of nanometers within tens of microseconds at Doppler- limited resolution, the development of dual-comb spectroscopy is hindered by the extremely demanding requirements on the short-term stability of the laser combs. We have overcome this difficulty and experimentally demonstrated a straightforward concept of real-time dual-comb spectroscopy (Fig. 2b, [22,23]), which compensates for laser instabilities by electronic signal processing. It only uses free-running mode-locked lasers without any phase-lock electronics, a posteriori data-processing, or the need for expertise in frequency metrology.
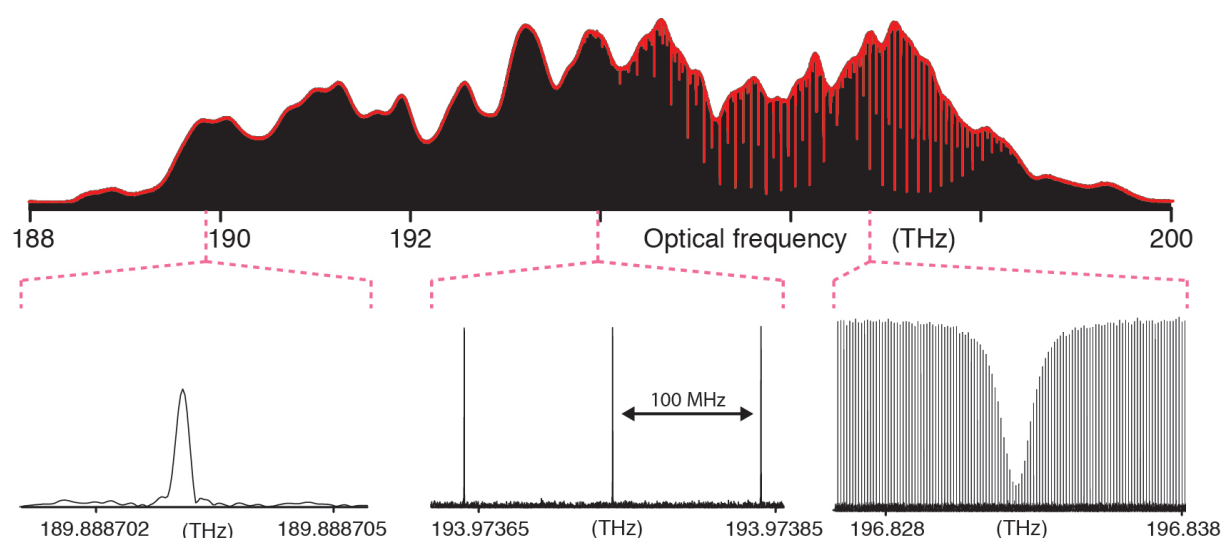

Figure 3. Near-infrared dual-comb spectrum with resolved comb lines. The spectrum across the entire domain of emission of Erbium femtosecond fiber lasers exhibits 120 000 well-resolved individual comb lines. Several acetylene absorption profiles shaping the discrete comb line intensities. The measurement time is 2.7 s and the optical instrumental linewidth is 200 kHz. In the lowest row, different degree of zooms in three different spectral regions well apart from these two frequencies isolate the individual comb lines and prove that the coherence between the two combs is maintained over the time of the measurement.

Since laser frequency combs involve intense ultrashort laser pulses, nonlinear interactions can be harnessed such as four wave mixing, intensity-dependent refractive index changes, light induced birefringence and dichroism, or other coherent transient phenomena including photon echoes.

Direct frequency comb Doppler-free spectroscopy of two-photon resonances has already been demonstrated in our first frequency comb experiments [24,25] with J. Eckstein and A. Ferguson at Stanford around 1976. As pointed out by E.V. Baklanov and V.P. Chebotaev [26], many pairs of comb lines with the same sum frequency contribute to the excitation of an atomic resonance, so that the excitation probability can be the same as for a resonantly tuned cw laser of the same average power. However, any observed resonance repeats with the line spacing of the comb, so that this type of direct frequency comb spectroscopy is only suitable for simple spectra composed of very few transitions.

Recently, we have demonstrated [27] the new technique of dual-comb two-photon spectroscopy, which identifies each transition uniquely by the modulation imparted by the interfering excitations. It can combine sub-Doppler resolution with a free spectral range only limited by the spectral bandwidth of the laser frequency combs. Proof-of-principle demonstration has been achieved with the 5S-5D two-photon transitions of rubidium (Fig. 4).
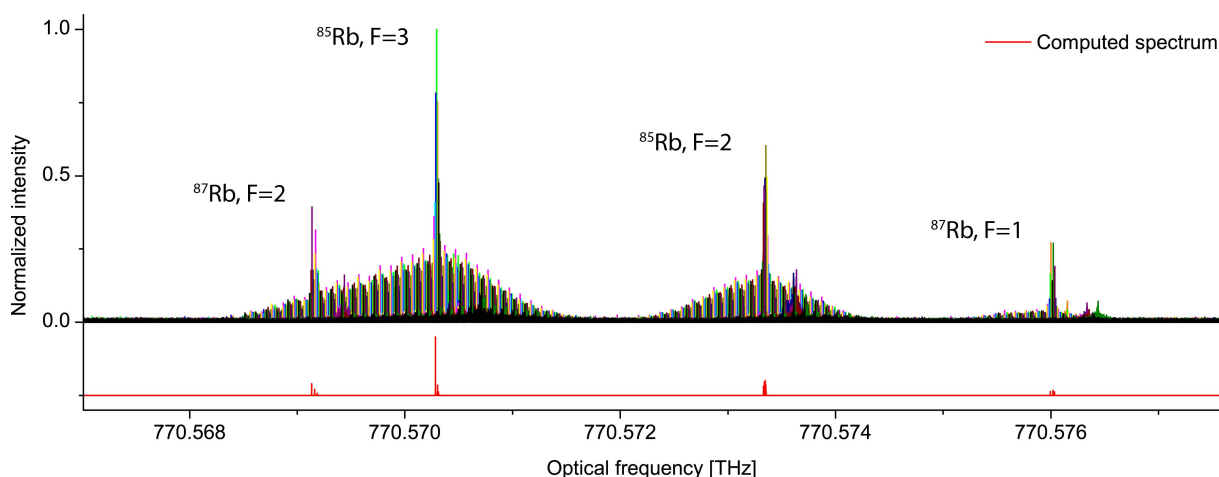

Figure 4. Two-photon excitation sub-Doppler dual-comb spectroscopy of the $5S_{1/2}$ -$5D_{5/2}$ transitions of natural rubidium. Doppler-free resonances emerging the Doppler-broadened resonances are clearly distinguishable. Thirteen individual spectra with resolved comb lines of a spacing of 100 MHz are interleaved, improving the resolution to 8 MHz. The measurement time for the full-interleaved spectrum is 52 s. F denotes the hyperfine quantum number in the ground state.

In other experiments [28,29], we harness coherent Raman effects. We have demonstrated [29] coherent anti-Stokes Raman spectroscopy and spectro-imaging of liquid samples with two laser frequency combs. All spectral elements are simultaneously measured on a single photodetector within a short time on the microsecond scale. The spectral bandwidth is determined by the span of the ultrashort pulse lasers. The resolution is only limited by the physical width of the molecular bands. With samples of liquid solvents, we demonstrate (Fig. 5) high resolution (4 cm^{-1}) Raman spectra spanning more than 1200 cm^{-1} recorded within tens of microseconds and these capabilities have been extended to hyperspectral imaging with an acquisition rate of 50 pixels/s. Real-time multiplex accessing of hyperspectral images may expand the range of applications of nonlinear microscopy.

As laser frequency comb techniques and laser technology continue to evolve, new applications come within reach. Curiosity-driven research with new instruments will lead to surprising discoveries. Let us end with a quote from Charles Townes („How the laser happened") [30]: „There is much that we don't understand; in many cases, we don't understand that we don't. And the really surprising discoveries will probably depend primarily on individuals, not teams or committees." The opportunities for individuals to make important and surprising contributions is perhaps the most important secret of success of our field of laser spectroscopy.
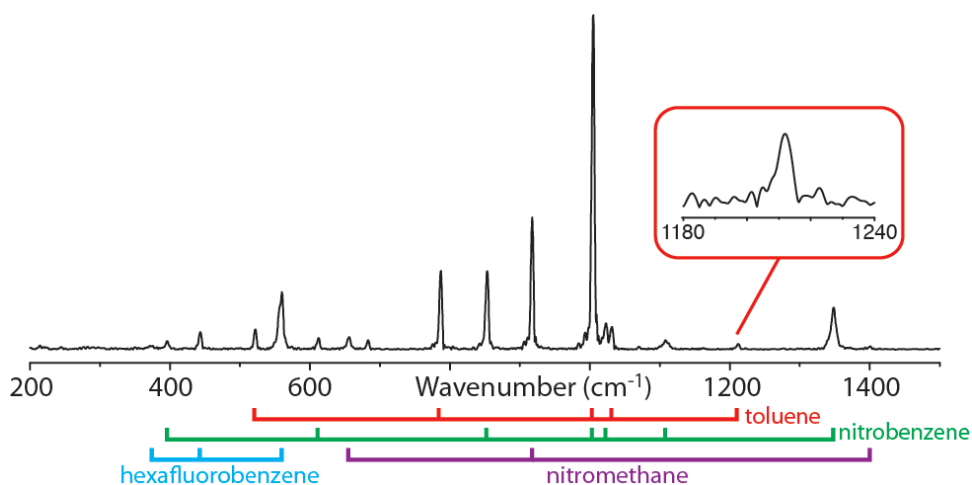
Figure 5. Dual-comb coherent anti-Stokes Raman high-resolution spectroscopy of a mixture of liquid chemicals. The spectrum measured within 295.5 μs at 4 cm^{-1} apodized resolution. The number of individual spectral elements (defined as spectral span divided by resolution) is 300.

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