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Femtosecond Comb Technique Vastly Simplifies Optical Frequency Measurements

A recent measurement of the transition frequency between the 1S and 2S states of hydrogen has now reached a precision of 1.8 parts in 10^{14}. As the precision of optical spectroscopy has developed, further improvements appear to be getting easier instead of harder. So says Theodor Hänsch, of the Max Planck Institute of Quantum Optics in Garching, Germany (near Munich), who has been a leader in the field since the 1970s. At that time, hydrogen spectroscopy could be done at the level of 1 part in 10^7 or 10^8. But Hänsch dreamt that eventually one would be able to reach a level of 1 part in 10^{18} by splitting the natural linewidth for the 1S–2S resonance (which is 5 parts in 10^{16}) to about 1%. Now he's within four orders of magnitude of his dream. The new hydrogen measurement was done jointly by Hänsch and his collaborators in Munich, and by Christoph Salomon and his collaborators at the Paris Observatory.[1]

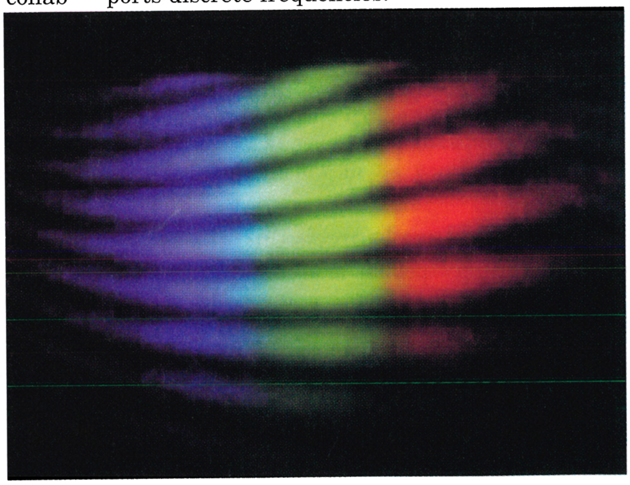
In the past, measuring optical frequencies with high precision has required heroic efforts. Gigahertz microwave frequencies can be measured by electronically counting oscillations in time. But when you reach the terahertz (10^{12} hertz) and petahertz (10^{15} hertz) frequencies in the infrared, visible, and ultraviolet, you can't count fast enough electronically. Instead, experimenters have built elaborate chains of harmonic generators and frequency-locked lasers that begin with a cesium atomic clock operating in the microwave range and then generate higher and higher harmonics in nonlinear diode mixers, crystals, and other nonlinear devices.

First make a comb

In 1978, Hänsch and his colleagues demonstrated that the periodic pulse train of a mode-locked CW laser behaves in the frequency domain like a precise set of equally spaced modes, resembling the teeth of a comb. The space between the modes (the teeth) is just the pulse repetition rate. With such a frequency comb, they believed one could eventually eliminate the frequency multiplying chain.[2] Later, Veniamin Chebotayev (Institute of Laser Physics in Novosibirsk, Russia) discussed similar ideas.

> A laser frequency comb lets you do away with a factory full of generators and lasers.

At first glance, says Hänsch, making precise measurements with short pulses doesn't make sense because a short pulse has a broad spectrum. In a laser, however, a pulse is caught between two mirrors. The pulse inside such a resonator can be described as a superposition of the resonator's modes, and the resonator only supports discrete frequencies.



INTERFERENCE FRINGES show a high degree of mutual phase coherence between two white-light continuum pulses, which are produced by focusing the light from a femtosecond laser at two separate spots inside a nonlinear optical medium. A similar phase coherence is observed between successive pulses, when the output of a mode-locked laser is spectrally broadened in a microstructured optical fiber.[3,4] The resulting broad frequency comb provides a precise ruler to measure the frequency of light throughout the visible spectrum. (Photo from ref. 6.)

Two decades ago, only picosecond lasers were available. Because the spectral width of these short-pulse lasers varies inversely with the pulse length, only the advent of femtosecond lasers made it possible to directly reach terahertz frequencies.

Recently, Hänsch and his collaborators demonstrated the use of a mode-locked femtosecond laser as a comb generator[3] to measure absolute optical frequencies. They have applied for a patent on the comb technique—for controlling not only the spacing of the teeth, but also the absolute position of the frequency comb. The output of a mode-locked femtosecond laser turns out to be a ruler in frequency space, says Hänsch. "It gives us a comb of frequencies, and the spacing of the teeth is precisely given by the repetition rate of the laser, which we can control with a microwave cesium atomic clock. To measure the differences between two frequencies, for each of these two frequencies, we observe a beat note with a given tooth of the comb and then count the number of teeth in between."

To measure an unknown absolute frequency, first the experimenters have to produce a frequency gap that they can bridge with the comb. One way to do this is to produce two harmonics or subharmonics of the laser frequency to be measured. For example, the ruler could be used to measure the frequency gap between the laser frequency, f, and its second harmonic, $2f$, which is of course just equal to the absolute frequency, f. The experimenters also tested the uniformity of the modes, finding that the mode separation agrees with the pulse repetition rate to the astonishingly precise value of three parts in 10^{17} over a range of 20 THz (PHYSICS TODAY, August 1999, page 9).

Initially, notes Hänsch, one might suspect that many factors could degrade the quality of the frequency comb; for example, dispersion in the laser material and the mirrors. Femtosecond lasers are designed to compensate for dispersion, but one would expect, at least in the outermost regions of the comb spectrum, says Hänsch, "where this compensation doesn't work well, that the modes can do what they want, that they won't be precisely equally spaced." However, the intensity-dependent refractive index of the laser crystal produces a so-called Kerr lens that focuses the light pulse so that a laser can operate without losses. The resulting nonlinear coupling between the modes produces a precise equal spacing. In the frequency domain, that spacing means the phases of the modes and, of course, the frequencies also have to be in lockstep to create such a pulse, says Hänsch, "a sort of collective dictate. We were pleasantly surprised when we found experimentally that the pulses are enormously reproducible, and there's a very high degree of phase coherence between successive pulses."

Hydrogen precision record

In the Munich–Paris work, the experimenters made a phase-coherent comparison of the hydrogen 1S–2S transition frequency with an atomic cesium fountain clock.[1] In an atomic fountain clock, the linewidth is narrowed by lengthening the observation time. Cooled cesium atoms are propelled upwards and then made to pass through a microwave cavity twice—first on their way up and then on their way down. The hydrogen transition frequency and the fountain clock frequency are linked with a wide frequency comb emitted by a 70-fs Ti:sapphire mode-locked laser. The pulse repetition rate of the laser is electronically phase locked to the atomic fountain clock, fixing the frequency scale of the comb. Then the experimenters employ the comb to measure the difference in frequency between two submultiples of the dye laser's optical frequency.

The new 1S–2S result is an order of magnitude better than recent measurements done by the Munich group using a conventional cesium atomic clock.[3] The Munich–Paris experimenters note that their accuracy of 1.8 parts in 10^{14} establishes the 1S–2S resonance in hydrogen as the best frequency standard in the optical region. If ultracold hydrogen were used, the accuracy of that frequency standard could be improved further. At present, the cesium atomic fountain clocks at the Paris Observatory and at the National Institute of Standards and Technology serve as primary time standards. They are believed to be accurate to better than 2 parts in 10^{15}.

Says MIT's Daniel Kleppner, "The definition of the meter is based on the measurement of the frequency of light, which means that an ultra-precise length measurement requires measuring an optical frequency. Up to now, that required a factory full of lasers. Ted has transformed the defined meter into an honest workaday practical unit."

The all-Munich experiment, just published,[3] shows for the first time the use of a laser frequency chain that measures absolute optical frequencies with the help of a femtosecond pulse train. JILA's John Hall says, "Ted's group was the first to make an absolute measurement from microwaves to the optical region. With the comb, they could bridge an insuperable barrier. So they got on the scoreboard big time." In their paper, the experimenters say, "These new tools will ultimately replace the large, delicate, complex and highly specialized harmonic laser frequency chains of the past with compact, reliable, and versatile solid-state systems for the measurement and synthesis of optical frequencies." Already, they say, they have demonstrated a potential accuracy of a few parts in 10^{18} for this new type of frequency chain, which they believe could lead to atomic clocks based on sharp optical resonances in atoms, ions, or molecules. But Hänsch admits a lot of work remains to be done.

Who needs precision?

"Many people feel some fascination with precision," Hänsch says. "It's another frontier, where you are likely to discover something unexpected. Unlike the high-energy frontier, for high-precision measurements, our apparatus gets smaller and cheaper as we make progress. High precision is not just a sport, like mountain climbing." Precision measurements of the behavior of hydrogen, the simplest of the atoms, can test quantum electrodynamics to a level previously unobtainable. At CERN, experiments are now being prepared to produce antihydrogen, so its properties can be compared with hydrogen. "Because, on a superficial level, for sure, matter and antimatter will behave the same, the experiments are only interesting if the measurements can be done at the highest precision."

Dirac, among others, speculated that fundamental constants might actually vary with time. Last year, John Webb and his collaborators found a hint (PHYSICS TODAY, March 1999, page 9) that the fine-structure constant might be changing as the universe evolves. At the current precision achieved by the Munich–Paris team, if the measurement were to be repeated ten years from now, a change at the level of 10^{-15} per year could perhaps be observed.

Hänsch believes the improvements in measuring optical frequencies are useful not only for fundamental physics, and for precision time and frequency metrology, but "the techniques might also be useful for telecommunications," says Hänsch, "because now we can provide a grid of precisely known reference frequencies in the visible and infrared spectrum, including the communications band for fiber-optical communication."

Development of the femtosecond laser comb has brought additional surprises. It turns out to be possible to broaden the spectrum of such a femtosecond laser to span a whole rainbow of colors, from the visible to the near infrared, and to still maintain a precise frequency comb, says Hänsch. The Munich group has teamed up with Hall and his collaborators at JILA and NIST in Boulder, Colorado, and at Bell Labs, Lucent Technologies, in Murray Hill, New Jersey. The team used air–silica microstructure optical fibers recently developed at Bell Labs and also at Bath University in the UK, which broaden the laser frequency comb to span an entire optical octave. The air-filled holes in the fibers serve to concentrate the light and create the enormous nonlinear effects that yield the spectral broadening (see the figure on page 19). With this broadened spectrum, the team set up a frequency chain that let them directly measure optical frequencies in terms of the cesium microwave standard.[4] At the American Physical Society meeting in Long Beach, California in May, Hänsch reported comparing the frequency of the air–silica fiber chain with the original Munich chain and found they agreed to 6 parts in 10^{16}, thus verifying the precision of both chains.

Hall and his colleagues at JILA, NIST, and Bell Labs[5] have reported controlling the phase of very short pulses. Rather than measuring the frequency gap between a CW laser and its second harmonic, they use a comb to measure the frequency gap between modes of the comb itself and the harmonic frequencies. So, a CW laser is no longer needed. Subsequently, the Munich group produced a similar one-laser chain.

The Munich group,[3] the Boulder–Bell Labs group,[5] and a collaboration between Munich and the Technical University of Vienna have used a femtosecond frequency comb to achieve control over the absolute phase of light pulses relative to the envelope. Ferenc Krausz reported the Munich–Vienna results in a postdeadline paper at the Conference on Laser and Electro-Optics 2000 in San Francisco in mid-May. The group used a short piece of ordinary fiber but achieved pulses that last only a few cycles, operating below 6 fs.

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Titanium Carbide Nanocrystals Shed Light on Late Phase of Stellar Lifecycle

An infrared emission feature from individual nanocrystals has been measured in the lab and matched to a similar feature in the spectra of old giant stars.

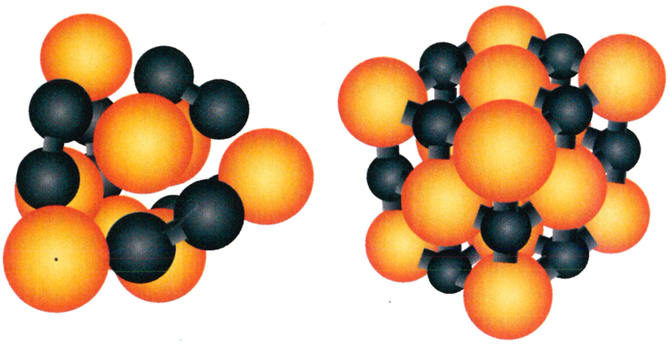
By January 2006, NASA's Stardust spacecraft will have orbited the Sun three times and spent almost seven years trawling for interplanetary dust. On the sixteenth of that month, if all goes according to plan, a washbasin-sized capsule containing the netted dust will separate from the spacecraft, make its way toward Earth, and land softly in the Utah desert.

Impressive though this feat of engineering may be, Stardust's Sample Reentry Capsule will not be the first space traveler to bring interplanetary dust to Earth. For aeons, meteorites bearing tiny grains of stardust have sporadically pummeled Earth. By extracting the grains in the lab and analyzing their isotopic abundances, scientists discovered that a tiny fraction of the grains originated in the atmospheres of giant stars that existed before the Solar System. (See Thomas Bernatowicz and Robert Walker's article "Ancient Stardust in the Laboratory," Physics Today, December 1997, page 27.) For one kind of grain in particular, titanium carbide, Bernatowicz and coworkers deduced four years ago that the grains formed in an unusually dense phase of the stellar atmosphere.[1]

Now, a multidisciplinary team based in the Netherlands has taken this work one step further.[2] By synthesizing TiC grains in the lab, measuring their infrared spectra, and comparing their spectra to those of various kinds of giant star, Gert von Helden and his colleagues have pinpointed where and when the TiC grains form.

The poster in the corridor

It was luck that gave the Dutch TiC project its start. Last October, von Helden invited Groningen-based astronomer Xander Tielens to visit the Foundation for Fundamental Research on Matter's Institute for Plasma Physics in Nieuwegein and discuss their mutual interest in polycyclic aromatic hydrocarbons (PAHs). Tielens believed that certain unidentified features in infrared spectra collected by the Infrared Astronomical Satellite (IRAS) in the 1980s could be explained by emission from PAHs, whose spectral properties von Helden's group investigates in their lab. Before they got down to business, Tielens was shown around the lab. "During the tour," recounts Tielens, "we happened to walk down a corridor, and there was a poster on the wall that described their results on TiC clusters." The Nieuwegein-based researchers study nanoclusters of TiC, but were quite unaware of the astrophysical significance of their work.



Titanium and carbon combine in various proportions to form clusters. Here, Ti_8C_{12} and $Ti_{14}C_{13}$ are shown. (Courtesy of Gert von Helden.)

Tielens told them that Bernatowicz had found TiC in meteoritic graphite grains. He also told them about a set of spectra in the IRAS archive whose emission lines at 21 μm had not been identified. "When we showed Xander some of our spectral data," von Helden recalls, "he was absolutely excited, and so were we." Indeed, after directly comparing the lab and astronomical spectra, the team found a one-to-one match.

To obtain the TiC data, von Helden had designed and carried out an experimental program, together with Deniz van Heijnsbergen and lab director Gerard Meijer (who, like von Helden, are based at the Institute of Plasma Physics and the University of Nijmegen) and Michael Duncan (University of Georgia). The clusters, of 27 to 125 atoms, were made with the standard technique of laser ablation. Once formed, the TiC grains were wafted *in vacuo* across the firing line of Felix, the institute's whimsically named free electron laser for infrared experiments. Thanks to its unique combination of a wide, tunable passband (4.5 μm < λ < 200 μm; 0.004 < $\Delta\lambda/\lambda$ < 0.07) and high power (100 MW), Felix can measure the infrared spectrum of the TiC nanoclusters by bombarding them with photons of incrementally varying wavelength. When the laser wavelength resonates with a TiC emission feature, the laser's photon fusillade is powerful enough to cause the cluster to emit a thermal electron. The resulting yield of ionized clusters, which are easily trapped, is proportional to the number of infrared photons that a warm cluster would emit if left alone.

The Felix experiments revealed not only the precise wavelength of the TiC feature—20.1 μm—but also that