## PERSPECTIVES

rate radiocarbon year–calendar year calibration curve. This problem is particularly acute prior to the last glacial maximum (about 20,000 years ago). Several data sets from various natural sources have been proposed for calibration use, but no two data sets agree sufficiently to establish a consensus (13).

Scientists attempting to take advantage of the available IntCal98 calibration curve to establish subcentennial resolution chronologies must become more familiar with the calibration curve and its inherent limitations. In many circumstances, radiocarbon dates on a series of carefully chosen samples will allow considerable refinement of the derived calendar ages through constraints imposed by a priori information (such as stratigraphy) or by the pattern of the radiocarbon dates relative to calibration curve variations (an approach that is sometimes referred to as "wiggle-matching"). Even with the implementation of such methods, the establishment of reliable chronologies with centennial or better resolution will require substantial diligence and the devotion of appropriate resources to overcome the inherent limitations in the conversion of radiocarbon dates to calendar ages.

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- 12. This simple exercise uses the tree-ring portion of the IntCal98 calibration curve. This portion is constructed entirely from Northern Hemisphere trees. However, the seasonal migration of the intertropical convergence zone will yield a mixture of Northern Hemisphere and Southern Hemisphere air in the tropics (14). The Northern Hemisphere–Southern Hemisphere difference averages 41±14 years between 1850 and 950 A.D. but varies from ~8 to ~80 years for any given decade (15).
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#### Supplementary Online Material

www.sciencemag.org/cgi/content/full/307/5708/362/DC1 Table S1

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#### CHEMISTRY

# Short and Sharp—Spectroscopy with Frequency Combs

### Thomas Udem

ver since high-resolution laser spectroscopy was introduced as a way to study atoms and molecules, many experts in the field thought that only continuous ("single-mode") lasers can resolve narrow spectral features. Much work has therefore been devoted to the construction of spectrally pure single-mode lasers. These lasers work well at infrared and visible wavelengths, but they become troublesome in the near-ultraviolet and virtually impossible to realize for even shorter wavelengths, for example in the extreme ultraviolet (10 to 100 nm).

These short wavelengths can easily be accessed with pulsed lasers through the use of nonlinear interactions. The shortest wavelength to date has been achieved with a process called high harmonic generation (HHG). (Harmonics are integer multiples of a laser frequency, that is, integer fractions of its wavelength.) However, nonlinear interactions are efficient only with short laser pulses, which cause spectral broadening of the laser. The spectral width of the laser is roughly equal to the inverse pulse duration, yielding  $10^{14}$  Hz for a 10-femtosecond ( $10^{-14}$  s) pulse (a typical pulse duration for HHG). This is far too wide for precision experiments, where a resolution of ~10 Hz has been reached (1, 2).

On page 400 of this issue, Witte *et al.* (3) report an experiment that circumvents that limit. The trick is to use not just one pulse but a train of N coherent pulses. (This is similar to the interference of multiple light rays to form the spectrally narrow features of a grating.) The authors are not the first to record spectral lines that are narrower than the spectral width of a single pulse, but they are the first to use harmonics for that purpose.

The spectrum of a pulse train has an "envelope" that is given by the spectrum of a single pulse, but it is divided up into a series of fringes that are separated by the pulse repeti-

tion rate  $f_{rep}$  of the pulse train (see the first figure). The fringes are perfectly regular in frequency space if the pulses are perfectly regular in the time domain. The pulse train must be coherent, that is, the pulses must have a defined (nonrandom) phase relation to each other. This requirement is almost automatically fulfilled with a mode-locked laser. Such lasers emit pulse trains in which all pulses are copies of a single pulse. In the spectrum of a train of N > 1 pulses, the



fundamental limit of the width of a single fringe is given approximately by  $f_{rep}/N$ . For a typical mode-locked laser, this is much smaller than the spectral width of a single pulse. For N = 3 and  $f_{rep} \sim 70$  MHz, as used by Witte *et al.* (3), we expect a fringe width of 70/3 MHz = 23 MHz. Indeed, that is roughly the linewidth observed by the authors [see figure 3 of (3)]. It is still large compared to the requirements of high-resolution spectroscopy, but improvements by many orders of magnitude should be possible.

For these improvements to become a reality, one must shine more pulses on the atoms or molecules. In this case, the fringes turn into sharp spikes that can be as narrow as in a well-stabilized single-mode laser (4). Such a series of delta-shaped spikes is usually called a frequency comb and can be used to measure the frequency of any of the spikes relative to an atomic clock (a very

How to narrow the linewidth. The spectrum of a train of N pulses,  $I_N(f)$ , is shown schematically for some values of N. The single-pulse spectrum (red curve; repeated as a pink "envelope" in the subsequent spectra) is as broad as the inverse pulse duration. Multiple pulsing causes fringes (blue curves) with a linewidth of  $f_{rep}/N$  to appear. For a large number of pulses, the spectrum resembles a series of deltashaped spikes, which are the modes of the frequency comb, that is, the modes of the laser. For a laser with  $f_{rep} = 100$  MHz and a pulse duration of 10 fs, one expects 1 million modes (or fringes) to appear.

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precise clock operating in the radio frequency domain).

In principle, it should be possible to apply many pulses in series to an atom or molecule, because a typical modelocked laser emits ~70 million pulses per second. However, at room temperature, atoms or molecules tend to move out of the laser focus before they can be hit by a large number of pulses. This becomes even more of a problem if a harmonic of a laser is used. because these harmonics are usually of low power and must therefore be focused to a small spot size to obtain a reasonable intensity.

To date, spectroscopy with frequency combs has not reached the resolution of single-mode lasers. In an early experiment, Eckstein *et al.* 

reached a resolution of 4 MHz for the sodium 4s-4d transition (natural linewidth 1.6 MHz) (5). More recently, Marian *et al.* (6) and Snadden *et al.* (7) have performed comb spectroscopy on the two-photon 5s-5d transition of rubidium. The latter authors laser-cooled and trapped the atoms to keep them within the laser focus. The resulting linewidth approached the natural linewidth of 300 kHz (see the second figure). In contrast to Witte *et al.*, all these authors



**Probing the resonance frequencies with a frequency comb.** The two-photon 5s-5d transition in <sup>85</sup>Rb breaks up into several components: For the 5s state, only the F = 3 hyperfine state is used. The 5d state first breaks up into two fine structure states,  $5d_{3/2}$  and  $5d_{5/2}$ , which in turn break up into hyperfine components (red and blue curves). The black curve is the resonance fluorescence (shifted up for clarity) that is recorded as the frequency comb is scanned across the line. The two-photon excitation becomes possible whenever the frequency of two modes or twice the frequency of one mode coincides with the atomic transition. As a consequence, the two-photon absorption spectrum repeats at an interval of half the laser repetition rate (80.3 MHz in this experiment). Figure adapted from (7).

used the fundamental, not a harmonic, of a frequency comb.

Despite these advances, single-mode lasers remain more suitable than frequency combs for spectroscopy—unless one uses harmonics of frequency combs where single-mode lasers are not readily available. This is what Witte *et al.* have now demonstrated with a train of three pulses of the fourth harmonic of a Ti:sapphire modelocked laser (rather than with the femtosec-

## PERSPECTIVES

ond laser pulses themselves). Hopefully, we can expect the application of much higher harmonics with many more pulses to interesting atomic or molecular species. A whole new window for high-resolution spectroscopy thus opens. It might even become possible to use a single laser system to cover all wavelengths from the near-infrared to soft x-rays; this possibility is out of reach for single-mode lasers.

High-resolution spectroscopy in the extreme-ultraviolet regime would be very useful for investigating hydrogen-like ions. For these ions, the quantum electrodynamic contributions to the energy levels (Lamb shifts) become more important and can be determined more precisely. It might even be possible to create an optical clock that operates in the extreme ultraviolet regime. The stability of such a clock is proportional to the transition frequency in use, and would thus be very high.

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Serengeti National Park

Tanzania

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### ECOLOGY

## A Leap for Lion Populations

### Esa Ranta and Veijo Kaitala

where are substantial populations of lions in Africa and Asia. Despite being known as "the king of the jungle," the African lion is principally found on open savannas, with smaller numbers in woodland areas. Since the 1960s, the ecology of lions has been intensively studied in the Serengeti National Park of Tanzania in East Africa (1-3) (see the figure). Lions live in family groups known as prides. Both woodlands and plains prides of the Serengeti are typically composed of six related females, their cubs, and a few unrelated males who mate with the adult females. The females do most of the hunting for the pride (1), and they must hunt in groups to be successful. Most of the daily activity of males concerns maintenance of territory. Packer *et al.* (4) have completed a detailed analysis of long-term

records of lion populations in a 2000-km<sup>2</sup> area of the Serengeti National Park. As they report on page 390 of this issue, lion population size has remained remarkably stable for long periods (10 to 20 years) punctuated by sudden increases that do not seem to reflect numbers of available prey.

The population dynamics (changes in the population size) of animals obey several key rules (5, 6). Population increases are due to births of new individuals and arrival of immigrants from nearby populations, whereas any decreases are due to individuals dying or leaving their natal population. Often births and deaths are dependent on each other either directly or with an intervening time lag. Population increases also depend on the amount of resources that mature females are able to monop-

olize. However, Packer et al. now reveal an unusual feature of the population dynamics of Serengeti lions (4). They discovered that lion numbers. both in the woodlands and on the plains, undergo long periods of stability interrupted by abrupt changes. This is intriguing because no such saltatory changes in population dynamics are reported for lion prey such as wildebeest, Cape buffalo, and gazelle (4).

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