https://doi.org/10.1038/s41567-026-03278-4

Laser-focused on precision

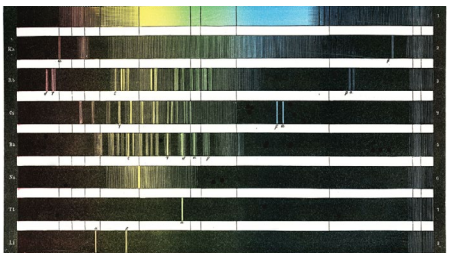
The advent of the laser transformed spectroscopy into a tool for precision measurements across scales, from nuclei to stars. In this Editorial we reflect on its far-reaching influence.

The vast majority of what we know about the inner workings of atoms and molecules comes from analysing the absorption or emission spectra produced when light interacts with matter — otherwise known as spectroscopic studies. Although spectroscopy has been practiced for centuries, beginning with early observations of sunlight dispersed by a prism, technological advances since the invention of the laser have evolved the field into laser-based precision spectroscopy.

In its most basic form, atomic spectroscopy involves measuring the wavelengths or frequencies of spectral lines — sharp lines in absorption and emission spectra — to determine the energy levels of atomic and molecular systems. However, the ability to resolve spectral lines using conventional light sources is limited by various factors, such as Doppler broadening caused by thermal motion. The introduction of the laser in 1960 provided a new light source for spectroscopy — coherent, narrow-bandwidth, intense and highly tunable — offering improvements in spectral resolution and sensitivity. Increasingly stable laser systems further advanced techniques for measuring transitions between energy levels.

One major breakthrough was in Doppler-free spectroscopy, where saturation effects in the absorption of laser light were used to eliminate Doppler broadening[1]. By revealing sharp, unbroadened spectral features, this technique enabled precise measurements of simple atomic systems, such as hydrogen, improving determinations of the Rydberg constant with a higher degree of precision[2]. In addition, laser spectroscopy has been used to probe fundamental physics, including measurements of the proton–electron mass ratio, variations of fundamental constants over time and testing quantum electrodynamics in simple atoms and ions.

Many of the optical transitions related to these advances, however, were difficult to access as their frequencies were too high to be



measured accurately and directly[3]. Previously, high-precision frequency measurements were mainly restricted to radio and microwave frequencies that oscillate slowly enough to be measured electronically. Extending spectroscopy to the optical regime necessitated large-scale frequency chains — to bridge the gap between known microwave frequencies and unknown frequencies of optical waves — which were extremely complex and costly.

The invention of the optical frequency comb offered an indirect means of measuring optical frequencies. The basic concept is that of a measuring stick: an unknown frequency is compared to a series of frequency spikes known as a frequency comb. To generate the comb, a mode-locked femtosecond laser produces a series of very short pulses of light whose phase-locked modes form a spectrum of evenly spaced frequencies[3]. The unknown optical frequency is then determined by measuring the frequency difference between the unknown light and the closest tooth on the comb — this is a radiofrequency beat which is easy to measure electronically.

Optical frequency combs were originally developed for optical atomic clocks. Before their introduction, microwave transitions in caesium had been adopted as the basis of the SI second, defined as 9,192,631,770 oscillations between two levels of caesium-133 (ref. 4). Although such microwave atomic clocks offer exceptional stability, optical clocks based on certain transitions in, for example, strontium operate at frequencies that are more than 10,000 times higher — and therefore lie in the optical range — and promise even greater precision. The optical frequency comb made precision measurements of optical-range transitions possible and enabled the development of state-of-the-art optical atomic clocks that now outperform microwave caesium clocks by two orders of magnitude[5]. Given this improved performance, there is now growing interest in redefining the SI second based on an optical transition.

As well as interrogating atomic transitions, laser spectroscopy is a common tool employed in nuclear physics. Indeed, specific transitions within certain nuclei may also serve as suitable candidates for clock transitions. Furthermore, perturbations in the electronic shell induced from the nucleus manifest as hyperfine structure and isotopic shifts in optical transitions[6]. Laser spectroscopy can resolve these features, allowing access to the charge radius of the nucleus, as well as its shape, spin and electromagnetic moments.

As well as lessons in nuclear studies, laser spectroscopy offers lessons in chemistry. Ultra-fast laser spectroscopy is a technique used to probe dynamic processes such as excited state relaxation, chemical reaction dynamics and energy transfer in photoactive materials by tracking rapid changes in their absorption spectra. Laser-based spectroscopy even finds applications at astronomical scales. Exoplanets induce tiny Doppler shifts in the emission spectra of their parent star that can be detected using laser frequency combs.

Laser-based spectroscopy has taken us from early demonstrations of Doppler-free precision measurements in simple atoms to frequency comb techniques that now influence studies from nuclear to astronomical scales. Its far-reaching impact continues to drive new developments in metrology — as evidenced by the enhanced sensitivity of low-temperature optical cavity spectrometers reported in an Article by Stankiewicz et al. in this issue of *Nature Physics* — with potential implications for global time keeping, fundamental physics and our ability to measure ultra-fast processes with ever greater accuracy.

Published online: 16 April 2026

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