


# A molecular probe for quantum metrology

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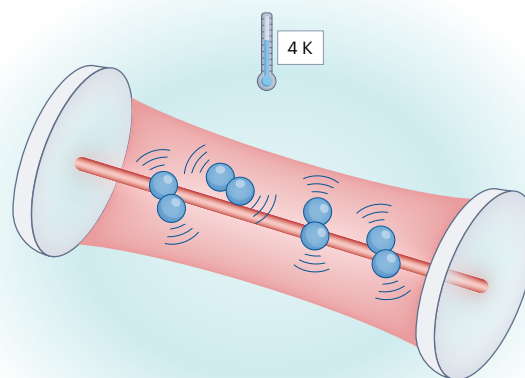
Cavity-enhanced spectroscopy has now reached temperatures as low as 4 K – colder than most of space. This removes long-standing barriers in measuring hydrogen, which is a benchmark system for testing quantum theory and relevant for metrology.

In the past, molecular spectroscopy was mainly used to analyse molecular structures. Now, it has become a far more powerful tool that is also used in studies of fundamental physics and quantum metrology. Cavity-enhanced spectroscopy offers remarkable sensitivity, but its extension to temperatures below 10 K has been challenging. Now, writing in *Nature Physics*, Kamil Stankiewicz and colleagues report a spectrometer capable of uniformly cooling both the optical cavity and the gas sample down to 4 K (ref. 1). This advance enables precise infrared spectroscopy of molecular hydrogen ( $H_2$ ), creating a versatile platform for fundamental studies at low temperatures for the examination of molecular physics theories and the realization of thermodynamic temperature metrology.

High-finesse optical cavities are characterized by low optical loss, strong light confinement and greatly enhanced sensitivity for spectroscopy. These features allow light to be reflected back and forth hundreds of thousands of times, thus amplifying light–molecule interactions. This enables the detection of weak spectral signatures, such as the absorption of hydrogen molecular vibrations. However, optical cavities are extremely sensitive to temperature fluctuations and mechanical vibrations, which has limited this technique to applications close to room temperature. Operation of such cavities at cryogenic temperatures below 20 K reduces Doppler broadening, enhances peak absorption and freezes out background contaminants, all of which are particularly attractive advantages.

Previous systems were limited to higher temperatures<sup>2</sup> or cooled only the gas sample<sup>3</sup>, not the cavity itself, thus creating detrimental thermal gradients. Stankiewicz and colleagues' design successfully cooled the entire optical assembly while effectively isolating it from thermal and vibrational noise, and integrated high-finesse optical cavities as well as precision frequency measurements (Fig. 1).

To demonstrate the capabilities of their experiment, Stankiewicz and colleagues focused on studying molecular hydrogen, the most abundant neutral molecule in the universe. Unlike other molecules, which condense into solids or liquids at low temperatures and thus become unsuitable for high-resolution absorption spectroscopy measurements, molecular hydrogen possesses the highest vapour pressure of any neutral molecules at 4 K, meaning that it remains gaseous. As the simplest neutral molecule, its energy can be determined using quantum chemical *ab initio* methods, with no requirement for the introduction of empirical or artificially adjusted parameters. Precise spectroscopy of molecular hydrogen thus allows one to test quantum theory in molecular systems<sup>3,4</sup>. Furthermore, it enables primary



**Fig. 1 | Cavity-enhanced spectroscopy of molecular hydrogen at cryogenic temperature.** The optical cavity was operated in a low-temperature environment (blue background) with temperatures as low as 4 K, while the temperature uniformity of the analysed hydrogen molecular gas sample (pair of blue dots) was preserved. Through measurements of the vibrational spectrum of molecular hydrogen, the molecular energy levels, the statistical temperature of the hydrogen molecules, and the characteristic data of hydrogen phase transitions can be precisely determined. The red shadow indicates the laser light inside the optical cavity. The laser light, as well as the molecular absorption, are substantially enhanced by the cavity.

metrology – in simple terms, a reference standard that relies on physical constants and laws rather than on physical artifacts – directly traceable through an unbroken, documented chain of calibrations or comparisons to the International System of Units (SI)<sup>2</sup>.

The spectrometer keeps the optical cavity and enclosed  $H_2$  gas at a uniform temperature of 4 K, with root-mean-square temperature fluctuations below 1 mK. A vibration-damping suspension isolates the system from cryocooler noise. Using a frequency-stabilized laser system that probes the  $H_2$  fundamental vibration band near 2.2  $\mu\text{m}$ , the team demonstrated four key applications of their platform.

First, they carried out a stringent test of quantum electrodynamics by measuring the frequency of a specific rovibrational transition in the  $H_2$  molecule. This measurement provides an important benchmark for the most advanced *ab initio* theoretical calculations. Second, they demonstrated the optical realization of the SI units for temperature (kelvin), pressure (pascal) and molecular density (mole per cubic metre) in the temperature range between 4 K and 20 K as an alternative method – and with a comparable accuracy – to helium vapour pressure thermometry. Third, they measured the  $H_2$  vapour-pressure curve with greatly improved accuracy, which provides a reference for the calibration of low-temperature sensors. Finally, they tracked the ortho-para  $H_2$  spin-isomer conversion on copper surfaces, determining

a conversion time constant of 32.4 hours in their system. Measurement of the spin-isomer conversion of molecular hydrogen allows one to elucidate the influence of various materials on nuclear spin states.

This work marks a substantial advance for cryogenic spectroscopy and quantum metrology. The choice of molecular hydrogen creates a powerful link between laboratory precision experiments, quantum mechanical calculations involving relativistic and quantum electrodynamical effects, and astrophysical models, where the molecular hydrogen spectroscopy and energy transfer rate of internal levels at cryogenic temperature are important data<sup>5</sup>. The ability to realize SI units optically at 4 K establishes a primary standard for metrology in extreme environments relevant to space science and fundamental physics.

Although currently focused on H<sub>2</sub>, the instrumental platform from Stankiewicz and colleagues is adaptable to other species that can be introduced through buffer-gas cooling or have sufficient vapour pressure at low temperatures, such as hydrogen deuteride molecules. Future developments that combine this platform with fast measurement techniques, such as rapid scanning spectroscopy or frequency comb-based methods<sup>6</sup>, could further enhance its performance. Cavity-enhanced spectroscopy at 4 K provides a unique tool to study

collision dynamics at ultralow energies and weakly bound molecular complexes, such as the formation of hydrogen molecular clusters. More broadly, it exemplifies how pushing instrumental capabilities to extreme conditions can unlock new scientific frontiers.

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## Competing interests

The authors declare no competing interests.