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First laser spectroscopy of a rovibrational transition in the molecular hydrogen ion H_2^+

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The molecular hydrogen ion H_2^+ is the simplest molecule. This iconic system has been the subject of innumerous theoretical studies, from the earliest days of quantum mechanics [1] until today, culminating in highly precise predictions of its level energies [2]. Comparisons of these predictions and measured vibrational transition frequencies would offer excellent opportunities in fundamental physics that go beyond the results achieved with the related molecule [3, 4]: a direct determination of the proton-electron mass ratio and the proton's charge radius. Furthermore, achieving precision spectroscopy of H_2^+ is an essential prerequisite for a future CPT test that compares H_2^+ with its antimatter counterpart [5, 6].

In this work we report the first vibrational laser spectroscopy of H_2^+, between low-lying rovibrational levels of para-H_2^+ [7]. We employed sympathetically laser-cooled and trapped H_2^+ ensembles. A first-overtone electric-quadrupole (E2) transition [8, 9] was driven by a unique 10^{-13}-level optical frequency metrology system reliably delivering Watt-level laser power at 2.4μm. Both hyperfine components were measured. We determined the spin-averaged rovibrational transition frequency with 3×10^{-8} fractional uncertainty, finding agreement with the predicted value. By using HD^+ as a test molecule, we also show that E2 spectroscopy is possible with 1×10^{-12} uncertainty. This demonstrates that E2 transitions are suitable for precision spectroscopy of molecular ions and that determining m_p/m_e spectroscopically with accuracy competitive with mass spectroscopy is a realistic prospect.

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