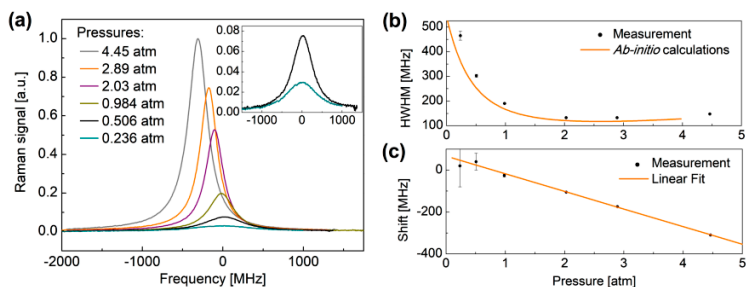


# Comb-calibrated coherent Raman spectroscopy of molecular hydrogen

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Highly accurate measurements of H<sub>2</sub> transition frequencies is fundamental for testing the quantum electrodynamics and physics beyond the standard model [1–3]. However, the retrieval of the unperturbed line positions is very challenging since it compels to work in low pressure conditions: the achievement of high signal-to-noise ratios is then hindered by the weakness of quadrupole transition moments and by the low molecular density. Alternatively, the distortion of the line profile at higher pressure could be carefully modelled in order to compensate for speed-dependent collisional effects and for the strong Dicke narrowing. High accuracy measurements of the Q(1) transition of the pure H<sub>2</sub> 1–0 band at 4155.25 cm<sup>-1</sup> have been performed from 0.2 to 5 atmosphere using stimulated Raman spectroscopy. An Er:fiber frequency comb has been used to calibrate the frequency difference between the pump and Stokes cw lasers involved in the Raman process. The pump laser emits at 737.8 nm and is kept fixed while the Stokes laser is scanned over 3 GHz around 1064 nm. The two beams are spatially superimposed and travel through a multipass cell filled with H<sub>2</sub>. Figure 1 (a) displays the line profiles measured at seven different pressures (the measurements at the two lowest pressures are displayed in the inset). As it can be noticed from panel (b) the retrieved widths are in a good agreement with *ab-initio* values based on H<sub>2</sub>–H<sub>2</sub> quantum scattering calculations. The frequency shift, plotted in panel (c), is proportional to pressure above 1 atm and the retrieved pressure coefficient agrees well with previous results [4]. The strength of the approach which provides high signal-to-noise ratio and frequency accuracy at the same time enables the use of more advanced profile models, such as the Hartmann–Tran profile, for line shape investigation.



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- [2] Phys. Rev. Lett. 2013, 110, 193601
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- [4] J. Quant. Spectrosc. Radiat. Transf. 2016, 177, 75.