HIGH-RESOLUTION LINEAR SPECTROSCOPY ON A MICROMETRIC LAYER OF MOLECULAR VAPOR

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Molecular rovibrational transitions can potentially provide high-resolution frequency references for the visible to the mid infrared including the telecommunications window at 1.5μm (C_2H_2 or HCN). Going towards compact miniaturized molecular spectroscopy is an extreme challenge as molecular transition probabilities are weak and long propagation length is required. Compact systems based on hollow core fibers filled with acetylene gas (C_2H_2) at high pressures have been presented[a] but the propagation length remains macroscopic. Additionally, collisional broadening (high pressures) or the Doppler effect hinder the available resolution.

Here we present selective reflection (SR) spectroscopy measurements on a molecular gas of NH_3 and SF_6 molecules at 10.6μm. Frequency modulated SR is a high-resolution (sub-Doppler) technique, linear in laser power that is essentially only sensitive to a layer of molecules whose depth is defined by the wavelength of optical excitation ($\sim \lambda/2\pi$), even when the cell remains macroscopic. Initial measurements, were performed with a CO_2 laser, subsequently changed for a more user friendly QCL laser rendered compatible with high-resolution spectroscopy. The core of our experiments is performed on the P(1) line of ammonia at 948.23cm^{-1} with best resolution limited by the laser linewidth (~ 0.5MHz). For pressures below 50mTorr (collisional broadening ~ 1.4MHz) the hyperfine structure of ammonia can be clearly resolved.

Our experiments pave the way towards miniaturized molecular frequency references such as nanocells[b,c], and allow us to envisage the first precision measurements of the Casimir-Polder molecule-surface interaction.

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[a] F. Benabid, *Compact, stable and efficient all-fibre gas cells using hollow-core photonic crystal fibres*, Nature **434**, 488 (2005).

[b] G. Dutier et al., *Collapse and revival of a Dicke-type coherent narrowing in a sub-micron thick vapor cell*, Europhys. Lett. **63**, 35 (2003)

[c] J.-M. Hartmann et al., *Infrared look at the spectral effects of submicron confinements of CO_2 gas*, Phys. Rev. A **93**, 012516 (2016).