High-resolution spectroscopy of sub-wavelength confined molecular gases

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The quantum physics of sub-wavelength confined atomic vapors has been studied with precision spectroscopy [1,2]. The main techniques used are selective reflection spectroscopy in macroscopic vapour cells, probing atoms at distances comparable to the reduced wavelength $\approx \lambda/2\pi$ and thin cell spectroscopy, where atoms are confined within the windows of a cell, of thickness as small as a few tens of nanometers [2]. Performing similar experiments with molecular gases is an attractive prospect, of interest for fundamental Casimir-Polder measurements of the molecule-surface interaction and for applications such as compact frequency references. However, this has proved elusive due to low molecular transition probabilities but also due to the lack of appropriate laser sources. Here, we discuss linear rovibrational spectroscopy of NH_3 and SF_6 molecules confined in sub-wavelength dimensions with sub-Doppler frequency resolution. We use a quantum cascade laser emitting at $\lambda = 10.6$ μm rendered compatible with high-resolution spectroscopy by various technical advances on our set-up. We describe selective reflection experiments performed in a macroscopic cell, as well as thin-cell transmission spectroscopy. The cell used in our experiments is of $\lambda/2$ thickness to allow the observation of Dicke narrowing [3]. It was fabricated in our laboratory by sandwiching an annular spacer between two ZnSe windows. In both set-ups we perform linear spectroscopy of molecular rovibrations with a resolution of 0.6 MHz, limited by laser linewidth. These newly developed techniques, allow us to get novel spectroscopic information on greenhouse gases, such as SF_6, that could be used to enrich molecular databases (see Fig.1). We are currently exploring acetylene-filled thin cells as compact frequency references for telecommunication wavelengths. We are also fabricating cells of nanometric thickness for measuring Casimir-Polder interactions with molecules.

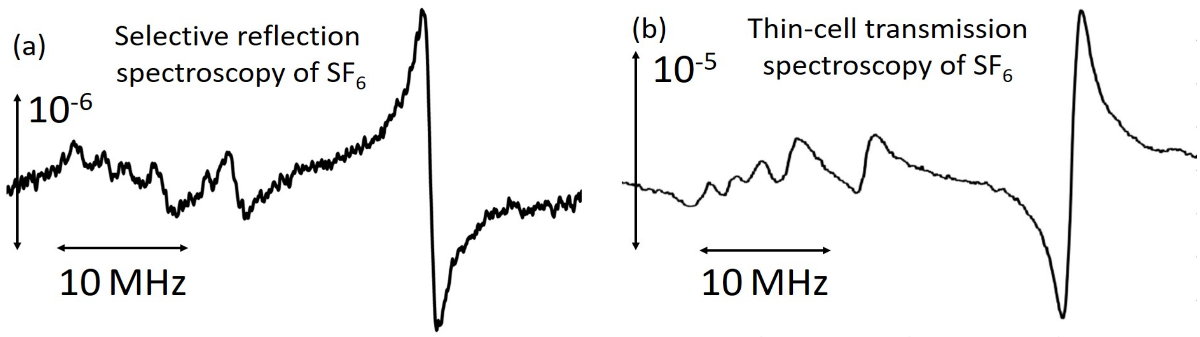

Fig. 1 Selective reflection (a) and thin cell (b) spectra of SF_6. Additional experiments, for a wider frequency range of ≈300 MHz, feature transitions not recorded in molecular databases.

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[2] H. Kubler et al., Nature Photonics 4, 112 (2010).
[3] R. H. Romer and R. H. Dicke, Phys. Rev., 99, (1955); G. Dutier et al., EPL, 63, (2003).