Selective reflection spectroscopy of atoms and molecules: Probing microscopic layers of vapour in macroscopic vapour cells

Athanasios Laliotis[1], Joao Carlos de Aquino Carvalho[1], Junior Lukusa Mudiayi[1],Isabelle Maurin[1], Benoit Darquié[1], Sean Tokunaga[1], Martial Ducloy[1], Daniel Bloch[1]

[1] Laboratoire de Physique des Lasers, UMR7538 CNRS, Université Paris13, Sorbonne Paris-Cité, 93430, Villetaneuse, France

One does not need elaborate technologies to probe atoms next to surfaces. It suffices to measure the small resonant change of reflectivity on a window/vapour interface. Selective reflection spectroscopy is a linear sub-Doppler technique probing vapour at a typical depth $\frac{\lambda}{2\pi}$, where λ is the wavelength of excitation. This allows the use of high vapour densities in search of collisional and local-field effects. Selective reflection is also an important experimental method for measuring Casimir-Polder interactions of atomic excited states without being particularly concerned about their short lifetime. Our group is now investigating the effects of near field thermal emission (thermal excitation of polariton modes [1]), on atoms. Excited state atoms present numerous transitions in the mid-infrared, more or less resonant with surface polaritons of common dielectrics, making them ideal probes of thermal fields close to surfaces [2]. We are probing Cs($7P_{1/2}$) atoms in a high temperature sapphire-cell. The resonant coupling between the $7P_{3/2} \rightarrow 6D_{3/2}$ transition at $12.15\mu m$ with the sapphire polariton at $12.35\mu m$ allows us to measure a very strong increase of the atom-surface attraction with temperature. We are also trying to demonstrate a near field energy transfer from thermally populated polaritons to Cs($7P_{1/2}$) atoms. This will manifest as an increase of the Cs($6D_{3/2}$) population next to the surface and by a distance dependent change of the Cs($7P_{1/2}$) lifetime.

Going beyond atoms, we are now probing molecular vapours (gases) close to surfaces. Molecules have much weaker transition probabilities but they also offer significant advantages compared to atoms. For starters, Casimir-Polder interactions can be richer due to the complex molecular geometry (chirality, anisotropy) with an interest that could span to physical-chemistry. Furthermore molecules can have a series of rovibrational transitions in the near and mid-infrared window of the spectrum acting as a sort of 'frequency comb' of references. C_2H_2 and HCN transitions for example serve as frequency references in the telecommunications window. Here we present selective reflection measurements on the P(1) rovibrational transition of NH_3 at $10.6\mu m$ using a quantum cascade laser source. The frequency resolution is clearly sub-Doppler, essentially limited by the sub-MHz linewidth of the laser source. We are now exploring the possibility of making micrometric thin cells filled with molecular gases at room temperature. These will be truly compact systems for high-resolution spectroscopy, useful for frequency referencing applications as well as probing molecule-surface interactions.

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