Title: ***Spectroscopic measurements of the molecule-surface interaction***

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The SAI group of Paris13 has developed the selective reflection technique for probing Casimir-Polder interactions between an excited state atom and a macroscopic surface at distances comparable to ~$\lambda/2\pi$ where λ is the excitation wavelength. The group has also pioneered atomic thin cell spectroscopy, demonstrating the Dicke narrowing effect at optical wavelengths [1] and performing the first atom-surface interaction measurements in thin cells of nanometric thickness (down to 50nm) [2].

Probing of molecular gases by selective reflection or thin cell spectroscopy is a very attractive prospect that allows envisaging molecular frequency references in a compact and miniaturised apparatus. Additionally, it paves the way for spectroscopic probing of the Casimir-Polder interaction with molecules. Molecule-surface interactions are of fundamental interest due to the complex molecular geometry (anisotropy or chirality). However, experimental measurements remain so far scarce and difficult to interpret mostly due to the difficulty of manipulating molecules [3].

Methodology

The SAI group has already probed NH_3 and SF_6 molecules at micrometric distances away from a surface with selective reflection spectroscopy at ~10.5μm using a quantum cascade laser (QCL) source. The challenge that lies ahead is measuring molecule-surface interactions. Towards this end, we will fabricate thin cells with dielectric windows, transparent throughout the near and mid-infrared part of the spectrum. This will allow us to probe molecules confined in the nanometer range.

1) Theory of molecule-surface interactions

We will estimate the rotational, vibrational end electronic dipole moment matrix elements for simple molecules, in gaseous form, that are relevant for our experiments, such as NH_3, SF_6, CO_2, CO and OCS. This will allow us to calculate the atom-surface interaction potentials of these molecules. We will collaborate with with Benoit Darquié (LPL, MMTF group) and the group of S. Scheel in U. Rostock.

2) Thin cell fabrication

We plan to fabricate cells ranging from 5μm down to 50nm. The basic principles of molecular thin cell fabrication are the following: An annular spacer will be placed between two thick (~7mm) windows. A hole will be drilled on one of the windows and then connected to a vacuum flange via a metallic tube glued onto the window. The cell can be outgazed and filled with molecules using a system of valves.

3) Thin cell spectroscopy of NH_3, SF_6

We will probe rovibrational transitions of NH_3 and SF_6 molecules by thin cell spectroscopy using a QCL laser (λ=10.6μm) that is already in use by the SAI group. Thin cell experiments will start with a L=5μm thick cell (L being the cell thickness) corresponding to a thickness of ~$\lambda/2$. At this thickness we expect to have sub-Doppler resolution signals due to the Dicke effect [1]. We will then try to probe molecules in cells of decreasing thickness, going down, if possible, to about L=50nm thickness. This will increase the molecular confinement and therefore the effects of the molecule-surface interaction.

4) Anisotropy of molecule surface interactions

The Casimir-Polder interaction between molecules and surfaces depends on the molecular orientation relative to the flat surface. This anisotropic effect is stronger for molecules with non-symmetric geometry, such as linear molecules that present strong anisotropic polarizability. An excellent candidate for investigating the anisotropy of molecule-surface interactions is the OCS molecule that has strong rovibrations around 4.85μm. One of the goals of this project is to probe OCS molecular gas inside a thin cell. Using the angular momentum of the probing light beam we can probe the difference of molecule-surface interaction potentials between two quantum molecular states with different orientation compared to the surface.

References

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