Title: ***Probing molecules next to surfaces***

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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 mostly due to the difficulty of manipulating molecules. A notable experiment was performed in the group of M. Arndt by diffraction of macromolecules on silicon nitride nanogratings [3].

Methodology

The SAI group has already performed selective reflection experiments on NH_3 and SF_6 rovibrational states, demonstrating the feasibility of probing molecules next to surfaces (probing depth of ~1.5μm) with a quantum cascade laser (QCL) source at ~10.5μm. The challenge that lies ahead is measuring molecule-surface interactions. Towards this end, we will fabricate thin cells with dielectric windows, such as ZnSe, that are transparent throughout the near and mid-infrared part of the spectrum (roughly from 1-12μm). 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.

Perspectives

We will examine the possibility of probing rovibrational transitions of acetylene at 1.5μm inside thin cells. This will allow the fabrication of compact frequency references at telecom wavelengths.

The SAI group is also developing a project that aims at probing Rydberg atoms next to surfaces in order to measure Rydberg surface interactions and demonstrate higher order effects in the Casimir-Polder interaction. Depending on the available time and the evolution of the experiments, the student may also be involved in this project.

References

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[3] C. Brand *et al.*, "A Green's function approach to molecular diffraction..," *Ann. Phys.*, 527, no. 9–10, pp. 580–591,2015.