Molecular alignment in He nanodroplets

Patricia Vindel-Zandbergen[*1], Ji Jiang[†], Manuel Barranco[‡], Marius Lewerenz[†], Christoph Meier[*2], Martí Pi[‡], Nadine Halberstadt[*3]

[*] Laboratoire Collisions Agrégats Réactivité, Université Paul Sabatier, 118 route de Narbonne 31062 Toulouse Cedex 09, France

[†] Laboratoire de Modélisation et Simulation Multi Echelle, Equipe de Chimie Théorique, 5 Boulevard Descartes 77454 Marne-la Vallée Cedex 2, France

[‡] Departament FQA, Facultat de Física, and IN2UB, Universitat de Barcelona , Diagonal 645, 08028 Barcelona, Spain

Synopsis We present a quantum dynamical analysis of the adiabatic alignment and subsequent rotational motion of the CH_3I-He complex, excited by strong off-resonant laser pulses. In this context, the emphasis is laid on the effect of the helium atom onto the coherent time evolution of the rotor states. The results are discussed with respect to experiments of adiabatic alignment in superfluid He droplets where a strong decoherence of the rotor states was observed.

The alignment of molecular species by strong non-resonant laser fields has been widely used within the last decade and has proven to be very useful in many applications in molecular science. For a detailed review of this methodology and its applications, see e.g.[1] and references therein.

After the strong pulse interaction leading to alignment, the subsequent dynamics is then given by the time evolution of the free rotor states, which for short times typically shows a strong dephasing, leading to an almost isotropic orientational distribution. However, for an isolated system, due to the discreetness of the spectrum, and the finite number of excited rotational states involved, the rotational wave packet rephases at well-defined later times, leading to the same aligned state as at the initial time (rotational revivals). This method has the advantage of providing a field free alignment of molecules at these times.

The phenomena of revivals depends crucially on the coherent evolution of the rotational wave packet, and as such forms a sensitive tool to study the interaction with a weakly interacting environment. In this perspective, the method of adiabatic alignment within a helium droplet offers an extremely interesting aspect: to which extend can a superfluid helium environment lead to decoherence of the rotational states? This question has recently been addressed experimentally, and as surprising result, they found the rotational dynamics much slower than that of isolated molecular speciess and the absence of the rotational revivals characteristic of gas phase molecules [2, 3, 4]. In order to shine light on this process, we perform numerical simulations of the adiabatic laser alignment of CH_3I within the helium droplet.

As a first step, we analyze the CH_3I molecule with a single helium atom. To describe the intermolecular interaction, we use a potential composed of a sum over CH_3-He, I-He and He-He terms obtained by fitting to ab initio calculations. These potentials are then used for quantum dynamical propagations, including the laser pulses chosen according to the above cited experiments, to model the adiabatic alignment, and its modification due to the presence of a single helium atom.

Based on this full quantum mechanical wave packet describing the rotor dynamics together with the motion of the helium, we analyze the rotational decoherence by tracing out the degrees of freedom of the helium, and we directly model the observed alignment signal, to compare with the experiments presented in Ref. [2].

As an outlook, on the basis of this work, we discuss the possibility to model the molecular alignment within superfluid helium droplets within the framework of density functional theory, such as used by the Barcelona group [5].

References

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[1] E-mail: patricia.vindel@irsamc.ups-tlse.fr

[2] E-mail: chris@irsamc.ups-tlse.fr

[3] E-mail: Nadine.Halberstadt@irsamc.ups-tlse.fr