Theoretical study of the dynamics of superfluid helium nanodroplets doped with potassium

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Synopsis The goal of this work is to study the photodesorption dynamics of a single atom of potassium from the surface of superfluid helium nanodroplets using the most recent and powerful theoretical simulation tools.

During the past few years, several real time dynamics experiments have been conducted on superfluid helium nanodroplets doped with alkali atoms using femtosecond pump-probe laser techniques [1]. Alkali atoms are particularly interesting as dopants because of their very weakly attractive interaction with helium, which makes them reside in a dimple at the droplet surface [2, 3]. Upon photoexcitation they usually desorb [3], except the heavy alkalis Rb and Cs excited close to the gas phase D1 transition [4, 5]. This is due to the strong repulsion between the excited state electronic orbital, which is much more diffuse and spread out than in the ground state, and the surrounding helium. The process can be rather complex, since the nanodroplet can absorb and dissipate part of the recoil energy as density waves and/or atom evaporation. In addition, the alkali atom can bring along one or a few helium atoms and desorb as an exciplex [1, 6].

From a theoretical point of view, the light mass of helium makes it a challenge to study the real time dynamics of this process because of quantum effects. The Helium density functional theory (He-DFT) approach and its time-dependent version (He-TDDFT) are very efficient semi-empirical methods which work with the helium density rather than the N-helium wave function, like quantum chemistry DFT does with electron density. They have proven to be the only way to date to simulate both the stability and the dynamics of a droplet with a size comparable to experiment [7, 8].

We study the $4p \leftarrow 4s$ and $5s \leftarrow 4s$ photo-excitation and desorption dynamics of a helium droplet doped with potassium using He-TDDFT. The method is the same as the one already used for other alkalis [9, 10]. Potassium presents the additional interest that its dynamical behavior is at the border between classical and quantum mechanical dynamics. We will present and discuss the results for a classical description of potassium photo-desorption dynamics and give a preliminary discussion of the quantum effects.

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