From Quantum Nuclear Motion of ^{4}He and N_2 Clusters in Carbon Nanotubes to the Soft, ^{4}He Droplet-Mediated, Deposition and Aggregation of Metallic Nanoparticles

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High-surface areas and precisely tuned pores of carbon nanotubes make them relevant materials for applications such as in gas adsorption, selective separation of light isotopes, and nanoreactors for quasi one-dimensional confinement of metal nanoparticles. Understanding the role of quantum nuclear effects and intramolecular interactions in the motion of molecules in carbon nanotubes is deeply fundamental. Very recent experimental measurements at low temperatures (2−5 K) of Ohba [1] revealed that much more molecules of nitrogen than helium atoms absorb in small diameter (below 0.7 nm) carbon nanopores, despite of the larger kinetic diameter of the former. Using the helium density-functional formulation for a large ^{4}He droplet containing a carbon nanotubes inside, we first show that the experiment can be understood by considering very large zero-point effects in the helium motion, which includes the formation of cavities with zero helium densities [2]. Second, we present an *ad-hoc* developed nuclear wave-function treatment to provide a detailed insight into the effects of quantum confinement for both N_2 and ^{4}He clusters in carbon nanotubes as a function of the tube diameter [3]. Third, we introduce a novel pairwise potential model [3] describing the gas adsorption to carbon materials which relies on DFT-based symmetry-adapted perturbation theory [4]. Finally, we propose an embedding approach combining nuclear density functional and wave-function treatments [3].

As a second topic, we discuss the upscaling problem to the nanoscale of the ^{4}He droplet-mediated soft-landing of metallic atoms [5]. Particularly, it will be shown how recent experimental measurements of Vilesov's group on the soft-deposition of few-nm sized metallic particles onto room temperature surfaces can be understood through our *ab-initio*-driven atomistic simulations [6].

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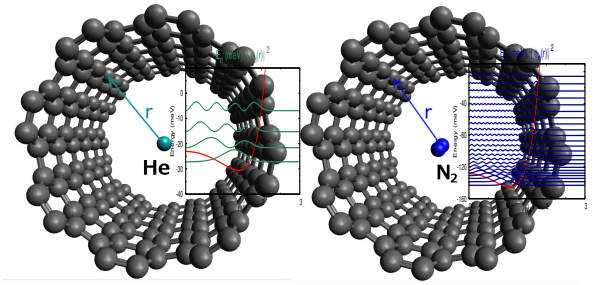

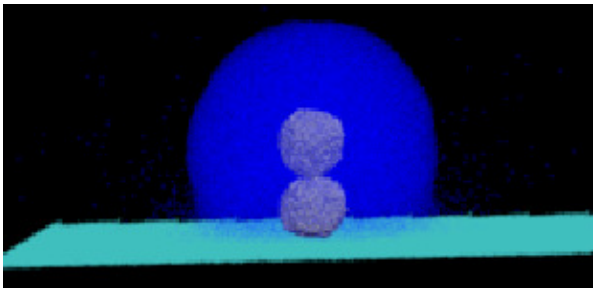

References

[1] Ohba 2016 *Sci. Rep.* **6** 28992

[2] A. W. Hauser & M. P. de Lara-Castells 2016 *J. Phys. Chem. Lett.* **7** 4929

[3] A. W. Hauser, A. O. Mitrushchenkov, and M. P. de Lara-Castells 2017 *J. Phys. Chem. C* **121** 3807

[4] M. P. de Lara-Castells *et al.* 2016 *J. Chem. Phys.* **144** 244707

[5] M. P. de Lara-Castells *et al.* 2015 *J. Chem. Phys.* **142** 131101

[6] R. Fernández-Perea, L. F. Gómez, M. Pi, C. Cabrillo, A. F. Vilesov, and M. P. de Lara-Castells, in progress.

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