The Cs_x-C_{60} reaction in superfluid helium nanodroplets: An example for a spatially quenched electron transfer

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Synopsis An environment of superfluid helium effectively quenches the electron transfer which is intrinsic to the reaction of an alkali-metal atom with a fullerene.

Recently, the interaction between heliophilic C_{60} molecules and heliophobic Cs atoms on helium nanodroplets has been studied experimentally.[2] The experiments indicate that the subsequential doping of a C_{60}-doped He-droplet with a single Cs atom does not to lead to a reaction between the two dopants, while a reaction between the heliophilic and the heliophobic dopants does take place in cases where Cs_2 is formed on the droplet beforehand.

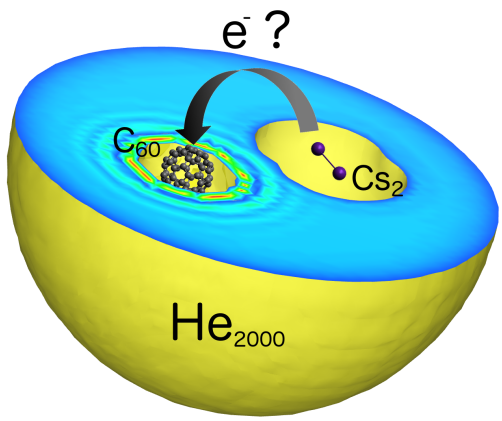

Figure 1. Cut through a helium droplet consisting of 2000 He atoms, doped with a fullerene and a Cs_2 molecule.

Here we describe this phenomenon observed by our colleagues via a combination of electronic structure calculations and orbital-free, bosonic helium DFT. One-dimensional potential energy scans are generated as approximations to the reaction pathway which describes the interaction between a single Cs atom or an alkali metal dimer and a C_{60} fullerene. These free-gas curves are then corrected for the interaction with the surrounding helium. Our studies show that the experimental findings can be interpreted in the light of a quenched electron-transfer reaction between the fullerene and the alkali dopant, which is additionally hindered by a reaction barrier stemming from the necessary extrusion of helium upon approach of the two reactants.[1]

References

[1] A. W. Hauser and M. P. de Lara-Castells. Spatial quenching of a molecular charge-transfer process in a quantum fluid: the Cs_x-C_{60} reaction in superfluid helium nanodroplets. *Phys. Chem. Chem. Phys.*, 19:1342–1351, 2017.

[2] M. Renzler, M. Daxner, L. Kranabetter, A. Kaiser, A. W. Hauser, W. E. Ernst, A. Lindinger, R. Zillich, P. Scheier, and A. M. Ellis. Communication: Dopant-induced solvation of alkalis in liquid helium nanodroplets. *J. Chem. Phys.*, 145(18):181101, Nov. 2016.

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