Weakly bound clusters and tagging spectroscopy in helium nanodroplets

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Synopsis Helium nanodroplets are ideally suited habitats for weakly bound clusters and for their formation. Two examples will be discussed, where knowledge could be gained from such experiments in combination with computer simulations: (a) alkali ion enhanced H_2 storage on C_{60} and (b) He-tagging spectroscopy of $C_{60}(He)_n^+$ with recent interpretations and discussions from theory.

The rapid cooling of the superfluid helium environment allows for the formation of very weakly bound molecular clusters that are bound for example only by ion-induced dipole or van der Waals forces. In particular, it is also possible to attach He atoms or H_2 molecules to larger molecules and molecular clusters. In experiments together with DFT calculations we could get a concise picture of the effect of an alkali dopant on the hydrogen storage capability of a carbonaceous structure by studying the case of $C_{60}Cs(H_2)_n^+$ [1]. Enhanced ion abundance could be seen for the first 10 H_2 adsorbate molecules and for $32 < n < 42$ for the cesium-doped fullerene. The calculations indicated strong quantum effects that are rarely quantified and that certainly need further attention.

In a second example, the helium was employed as a messenger for spectroscopy of C_{60}^+. The results of Campbell [2] were reproduced and the shift of the electronic excitation energies was measured for tagging with up to $n \approx 100$ [3]. In addition to this shift, the line-width of the excitations vary with n, the number of He tags attached to C_{60}^+. A report shall be given on the ongoing theoretical study which is undertaken to understand this systematic effect in the broadenings and that tentatively links it to the existence of a large number of possible isomers. Calculations indicate that charge transfer can occur from the region of 6-6 bonds to centers of 5-rings (Figure 1). This charge-transfer may lead to a slightly differently balanced He-5-ring to He-6-ring interaction. Furthermore, preliminary results from path integral Monte Carlo calculations of the ground state isomers and from time-dependent density functional theory calculations will be presented.

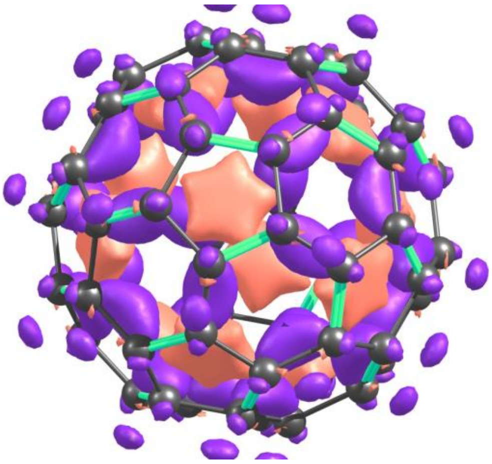

Figure 1. Charge density difference map between the ground and the excited state of C_{60}^+ as obtained by CASSCF(19,10) calculations with a small active space.

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References

[1] A. Kaiser, et al. 2017 *Int J Hydrogen Energy* 42 3078
[2] E. K. Campbell, et al. 2015 *Nature* 523 322
[3] M. Kuhn, et al. 2016 *Nat Comm* 7 13550

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[1] **Fehler! Verweisquelle konnte**

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