

Electrostatic models for fullerene-fullerene interactions

Michael Gatchell^{*1}, Stefan E. Huber^{†,‡}, Fredrik Lindén^{*}, Henrik Cederquist^{*}, Andreas Mauracher[†], Henning Zettergren^{*}

^{*} Department of Physics, Stockholm University, Stockholm, Sweden

[†] Institut für Ionenphysik und Angewandte Physik, Leopold-Franzens-Universität Innsbruck, Innsbruck, Austria

[‡] Institut für Grundlagen der Technischen Wissenschaften, Leopold-Franzens-Universität Innsbruck, Innsbruck, Austria

Synopsis We have used simple electrostatic models to investigate the stability limits of multiply charged fullerene clusters and to model charge transfer between fullerene ions.

Experimental studies of charged fullerene clusters $[C_{60}]_n^q$ have shown that the appearance size is $n = 5$ for both dications and dianions despite their different chemical natures [1, 2]. For higher charge states the appearance sizes are $n = 10$ for $q = +3$, $n = 21$ for $q = +4$, and $n \approx 33$ for $q = +5$ [1]. The ultimate stability limit of such systems is dictated by the balance between cohesive dispersion and polarization forces, and repulsive Coulomb forces.

Here we present an electrostatic model to study the stability of multiply charged C_{60} clusters where we describe each fullerene molecule as a metal sphere with the excess charge distributed over the cluster [3]. To accurately describe the interactions between the fullerene molecules, this model includes dispersion forces and an accurate description of mutual polarization effects [3, 4]. The geometries of a fullerene hexamer dication ($n = 6$, $q = +2$) from a single-point DFT calculation with the neutral structure and in the metal sphere model are shown in Figure 1. We find that the simple metal sphere model reproduces the DFT potential energy surfaces for doubly charged clusters in this size range. Doubly charged clusters consisting of six or more molecules are thermodynamically stable [3], in agreement with the experimental findings [1, 2]. This suggests that the simple model may be successfully used to study the kinetics also of multiply charged ($q > 2$) clusters.

We have implemented the metal sphere model for use in a classical molecular dynamics code in order to calculate dissociation rates as a function of cluster size, charge state, and internal energy. We will present results from these simulations at the meeting.

We have also derived the exact analytical solution for the potential energy for a point charge located between two charged dielectric spheres and shown that it can be accurately approximated by a much sim-

pler analytical expression [4]. The latter can for instance be used for fast estimates of mutual neutralization rates in C_{60}^+ and C_{60}^- collisions. Such experiments are planned to be performed at the Double ElectroStatic Ion Ring Experiment (DESIREE) at Stockholm University [5, 6]. This model may also find use in describing the evolution, growth, and dynamics of nanometer sized dielectric objects such as molecular clusters or dust grains in different environments, e.g. in astrophysical ones [4].

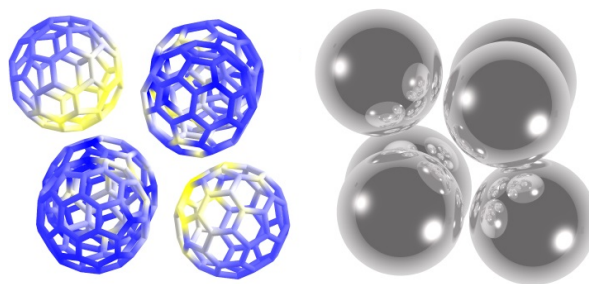


Figure 1. Left: Mulliken charge distribution of $[C_{60}]_6^{2+}$ from DFT calculations. Right: Metal sphere description of the same system.

References

- [1] B. Manil *et al.* 2003 *Phys. Rev. Lett.* **91** 215504
- [2] A. Mauracher *et al.* 2014 *Angew. Chem. Int. Ed.* **53** 13794
- [3] S. E. Huber, M. Gatchell, H. Zettergren, and A. Mauracher 2016 *Carbon* **109** 843
- [4] F. Lindén, H. Cederquist, and H. Zettergren 2016 *J. Chem. Phys.* **145** 194307
- [5] R. D. Thomas *et al.* 2011 *Rev. Sci. Instrum.* **82** 065112
- [6] H. T. Schmidt *et al.* 2013 *Rev. Sci. Instrum.* **84** 055115

¹E-mail: gatchell@fysik.su.se