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First Principles Calculations of the Optical Properties of Hydrogen Terminated Carbon Nanoparticles ANDREW WILLIAMSON, GIULIA GALLI, Lawrence Livermore National Laboratory, NEIL DRUMMOND, RICHARD NEEDS, TCM Group, Cavendish Laboratory, Cambridge, UK — First-principles density-functional (DFT) and quantum Monte Carlo (QMC) calculations of the single-particle and optical gaps of hydrogen-terminated carbon nanoparticles are presented. Both diamondoid structures constructed from adamantane cages and spherical diamond structure particles are studied. The DFT calculations confirm the previous predictions of Raty et al.[1] that for carbon nanoparticles larger than 1nm quantum confinement effects disappear and the gaps drop below those of the bulk material. The QMC calculations follow the DFT trends for the size dependence of the gap, but predict optical gaps 1-2 eV larger. We illustrate that the LUMO orbital of hydrogen-terminated carbon nanoparticles is a delocalized surface state, in contrast to silicon and germanium nanoparticles, where the LUMO is core-confined. This delocalized nature of the LUMO results in a small exciton binding, a negative electron affinity, and optical gaps of larger clusters that are below the bulk gap. This work was performed under the auspices of the U.S. Department of Energy by the University of California, Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48. [1] J.-Y. Raty, G. Galli, C. Bostedt, T.W. van Buuren, and L.J. Terminello, Phys. Rev. Lett. 90, 037401 (2003)

Andrew Williamson
Lawrence Livermore National Laboratory

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