On the energy of curved carbon surfaces

A multi-scale Density Functional Theory and Monte Carlo study

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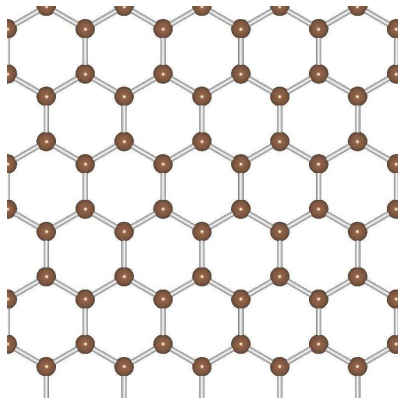
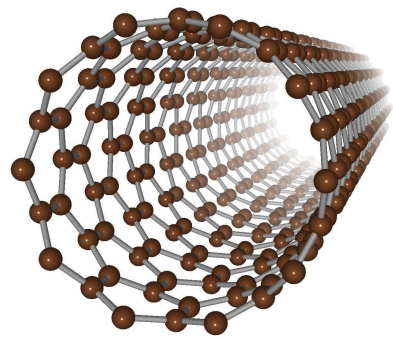
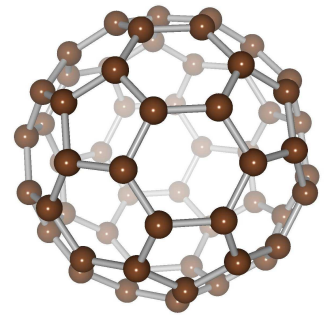
19 September 2010

Motivation – carbon nanostructures

- graphene, (single and multiwall) nanotubes, fullerenes







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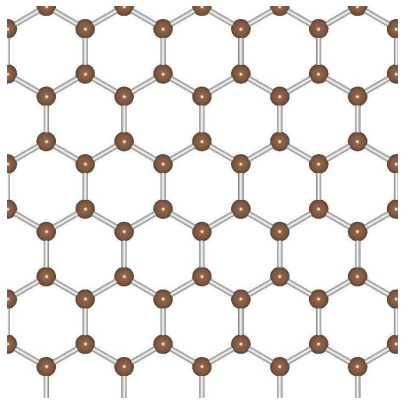

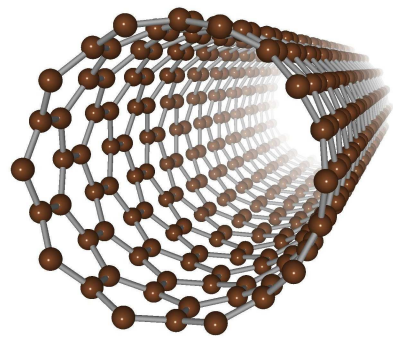

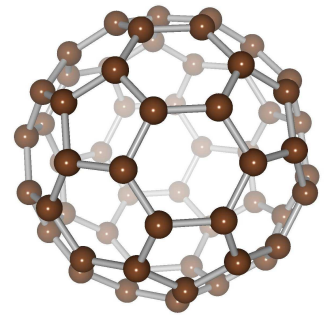

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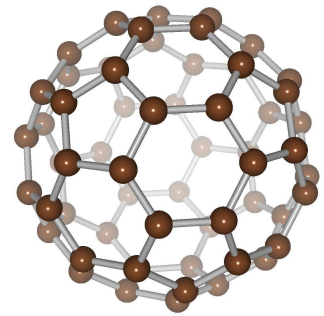

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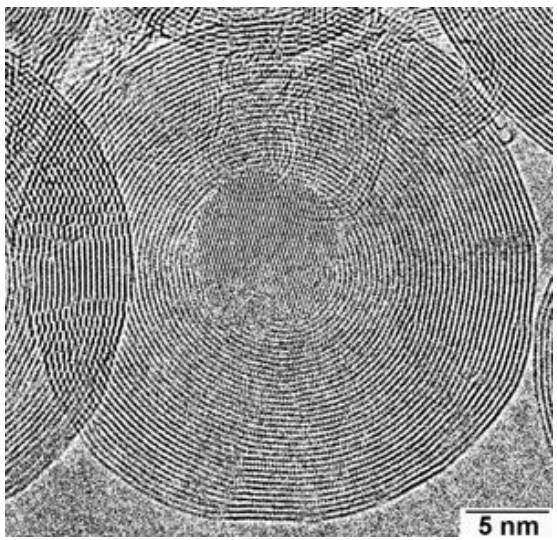

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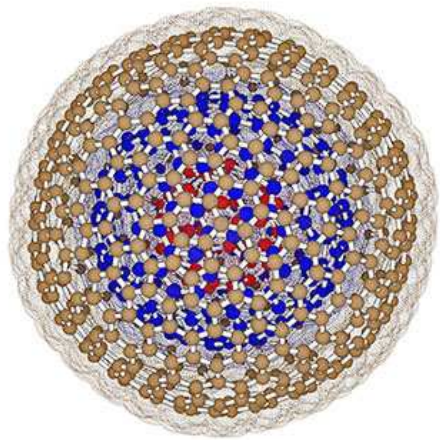

http://www.3dchem.com/

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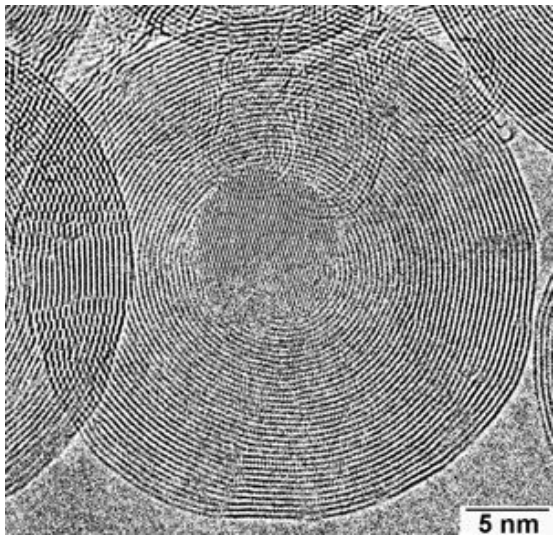
- possible use: supercapacitators, hydrogen storage



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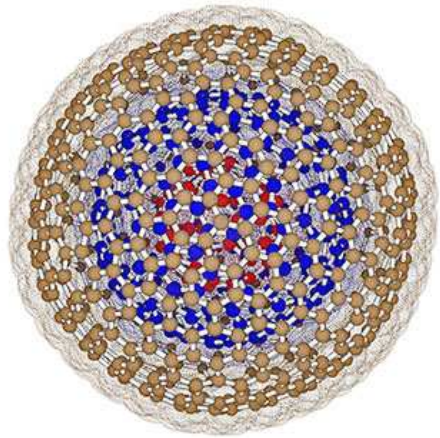

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  - input: surface stress $\approx$ surface energy
- the (excess) surface energy of a fullerene (or SWNT) is an increase of its total energy with respect to (planar) graphene

Outline

Introduction

- Motivation

Methodology and Results

- Fullerenes treated with DFT
- Ring shell structure
- Monte Carlo simulations
- Description of the fullerenes
- Curvature energy

Conclusions

Fullerenes from DFT

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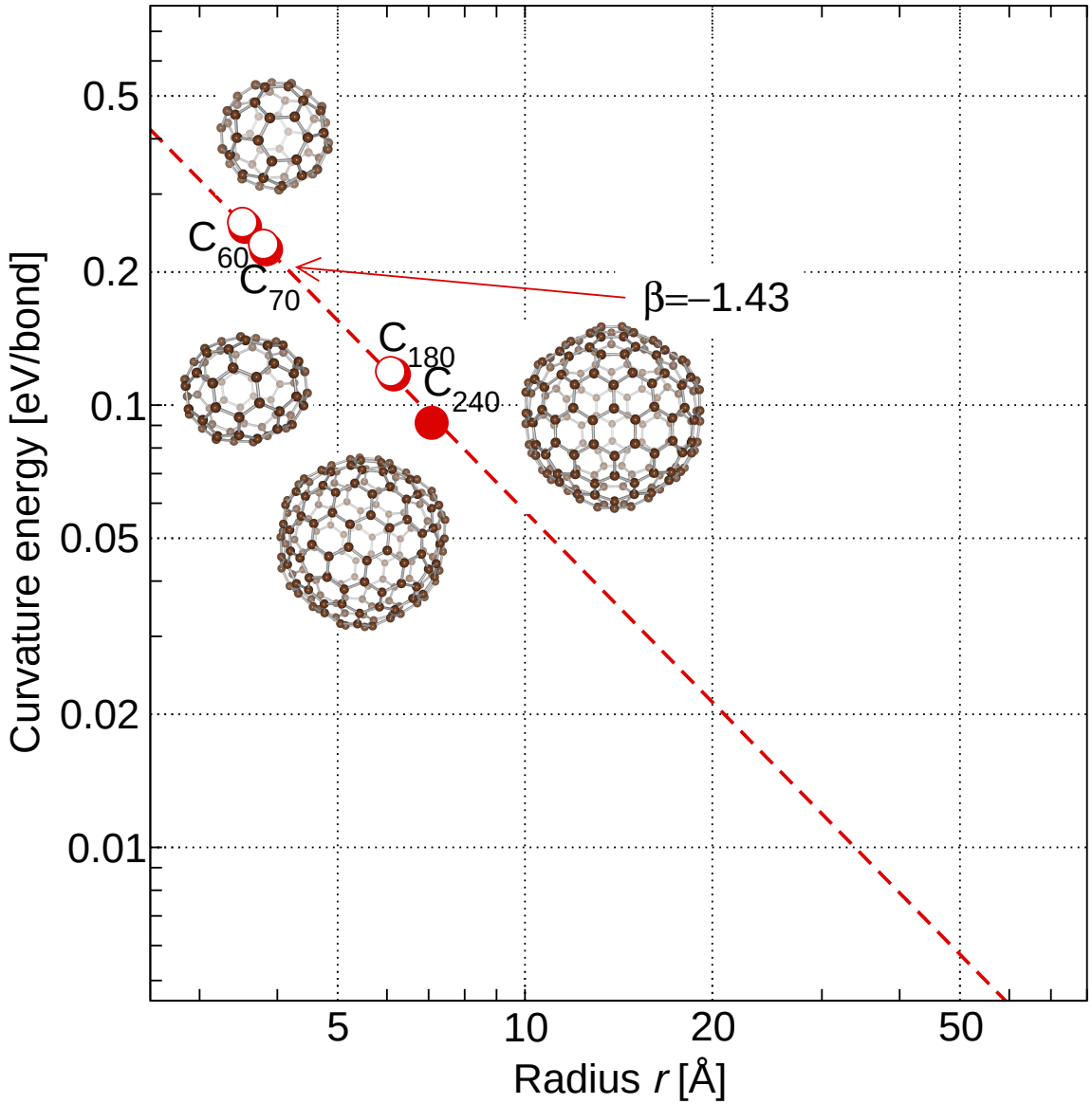
- focus on fullerenes with icosahedral symmetry (with the exception of C_{70})

Fullerenes from DFT



radius:

$$r = \langle |\mathbf{r}_i - \mathbf{R}_0| \rangle$$

power-law fit:

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C_{240}:
12 days on
16 processors

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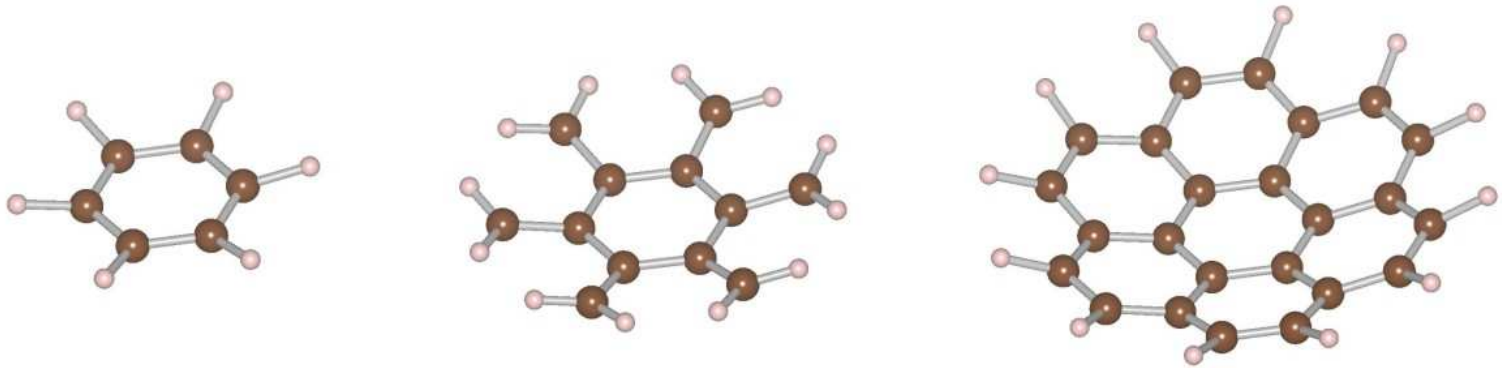
Ring shell structures

(a) one-ring shell (b) two-ring shell (c) three-ring shell



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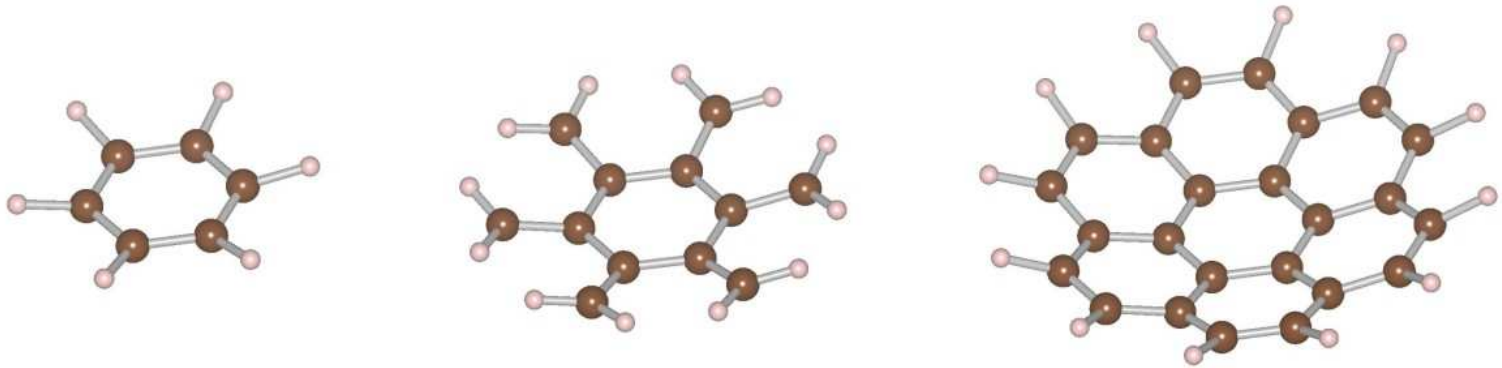

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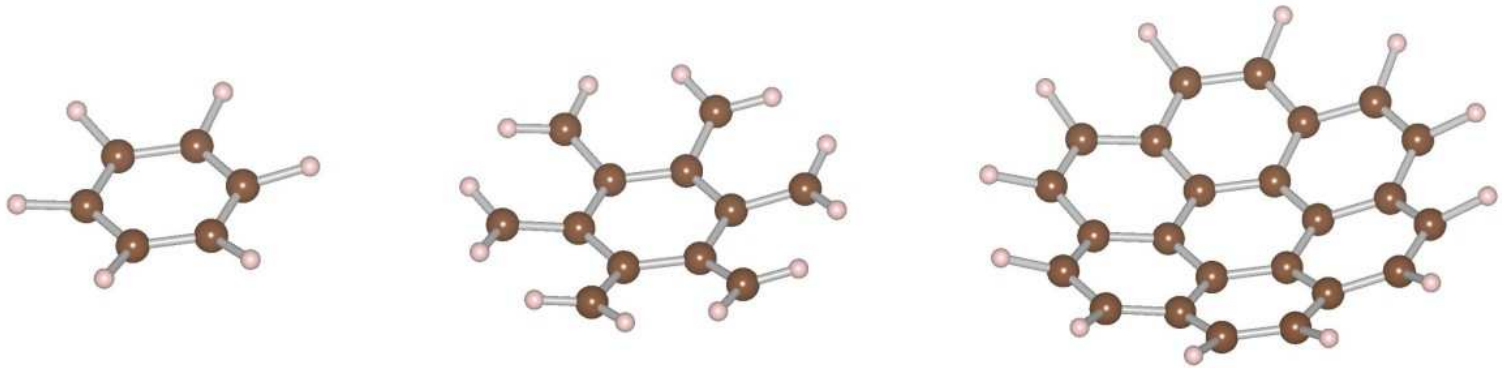

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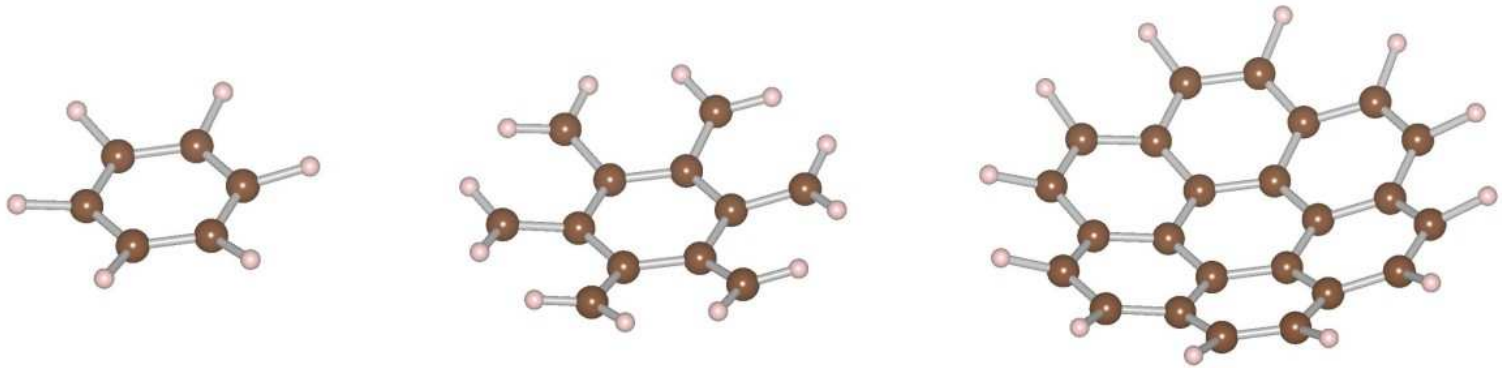

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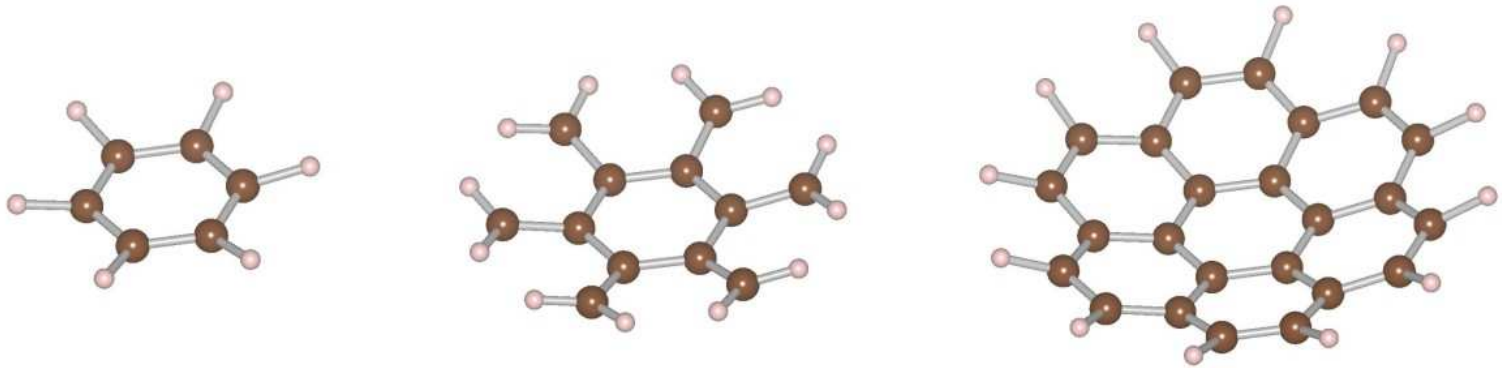

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\+ can easily access any desired radius

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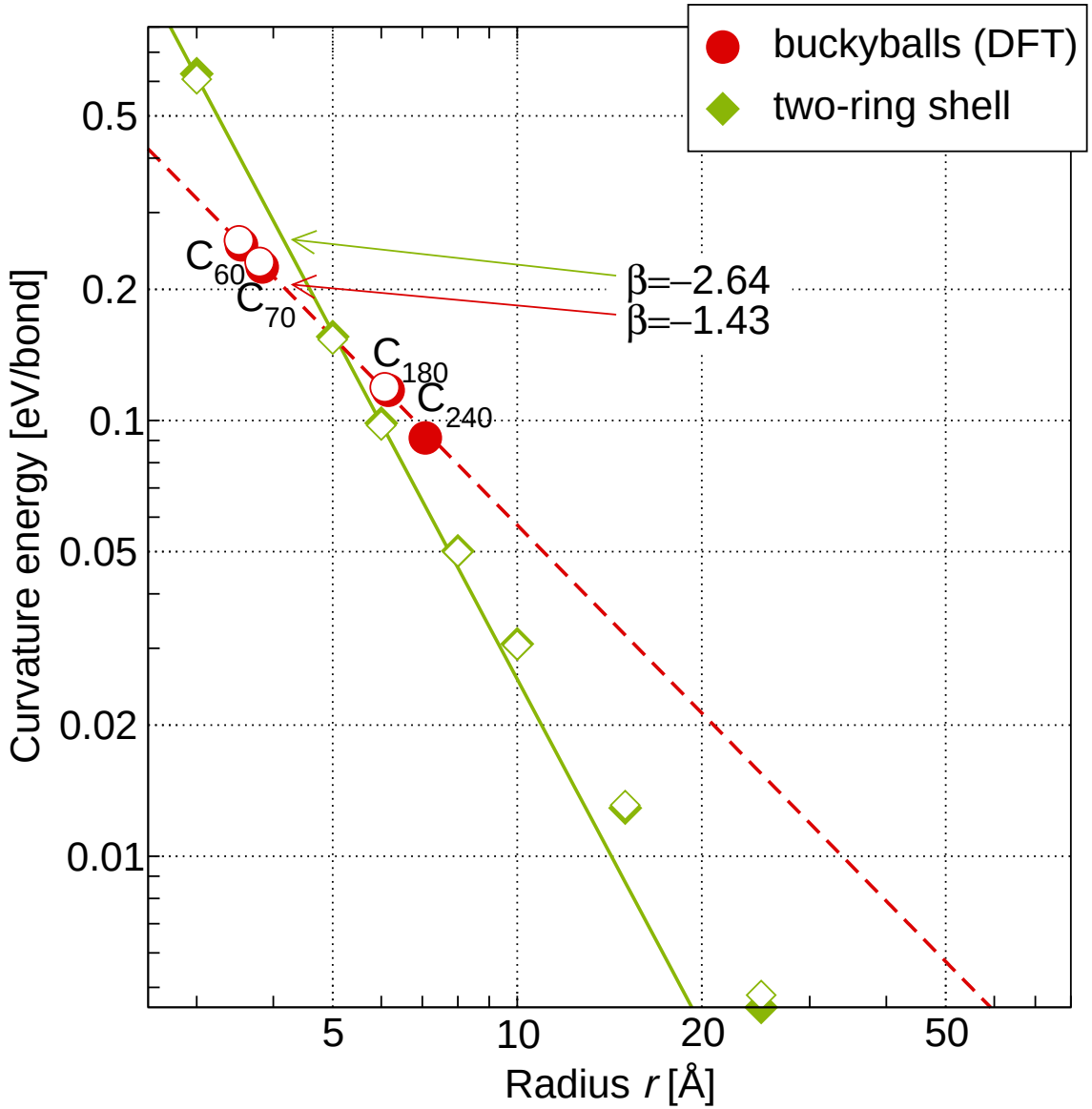

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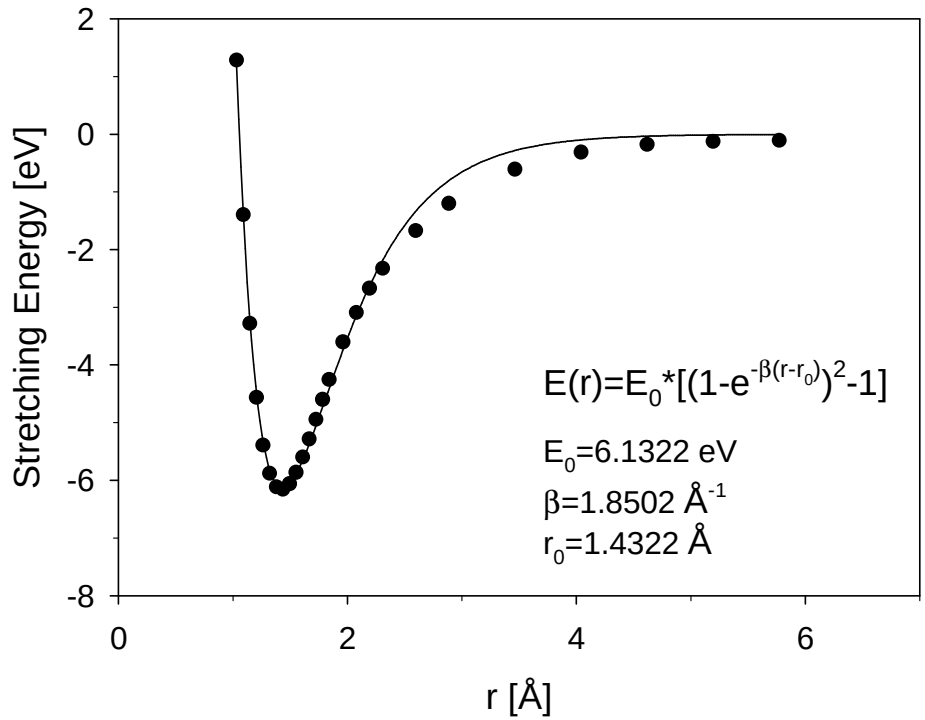
Inter-atomic potentials

construction of classical inter-atomic potentials "coherent" with the present DFT calculations

- stretching: Morse potential

$$E^S(r_{ij}) = E_0 \left[(1 - \exp(\beta(r_{ij} - r_0)))^2 - 1 \right]$$



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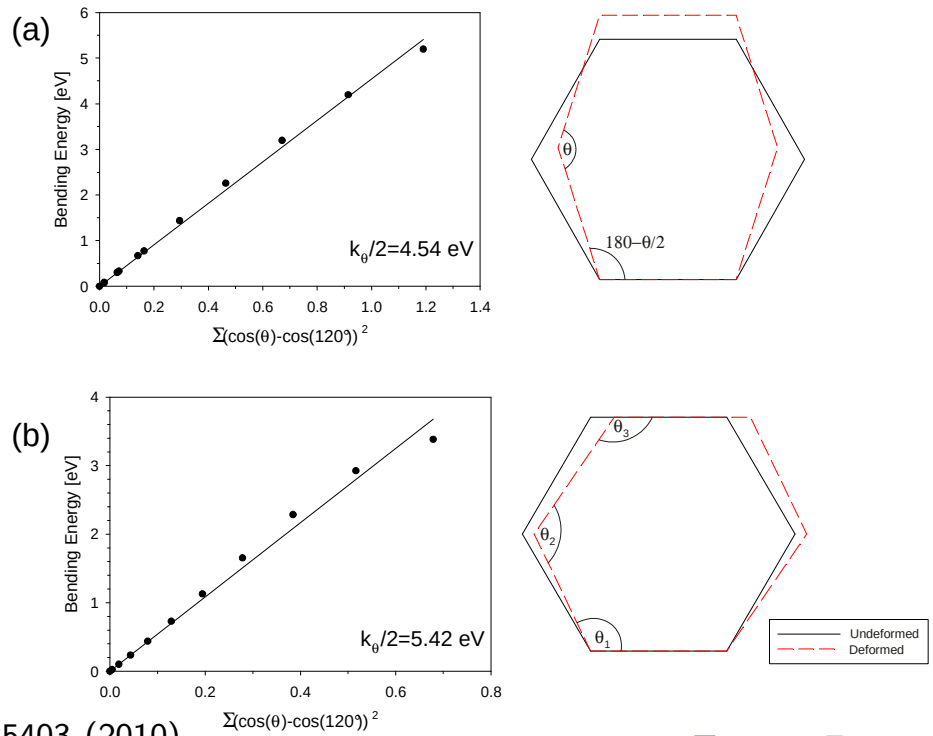

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Inter-atomic potentials

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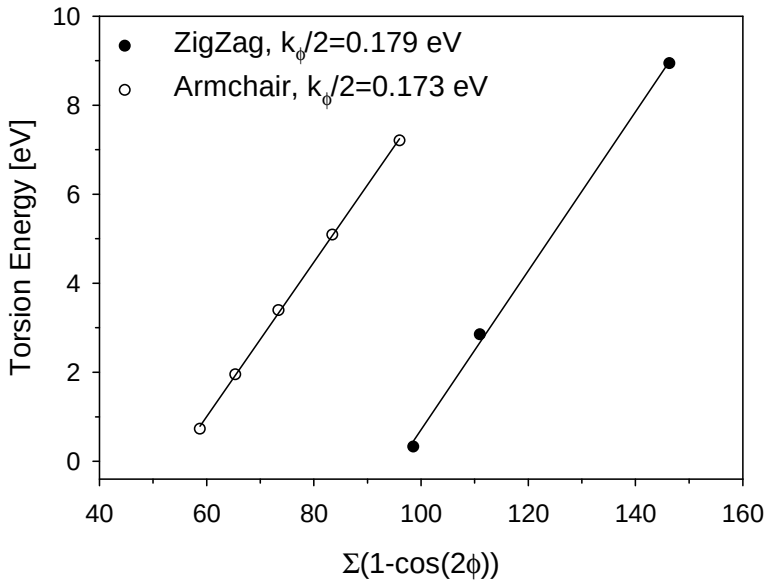
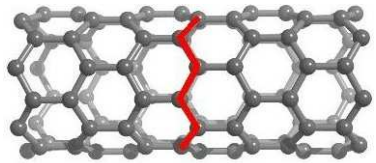
- stretching: Morse potential
- bending: harmonic potential
- torsion

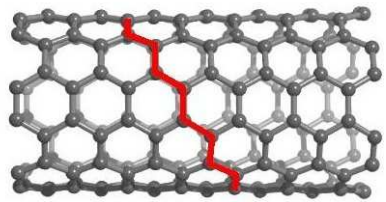
$$E^T(\phi_{ijkl}) = \frac{1}{2} k_\phi \left(1 - \cos 2\phi_{ijkl}\right)$$



Zig Zag



Armchair



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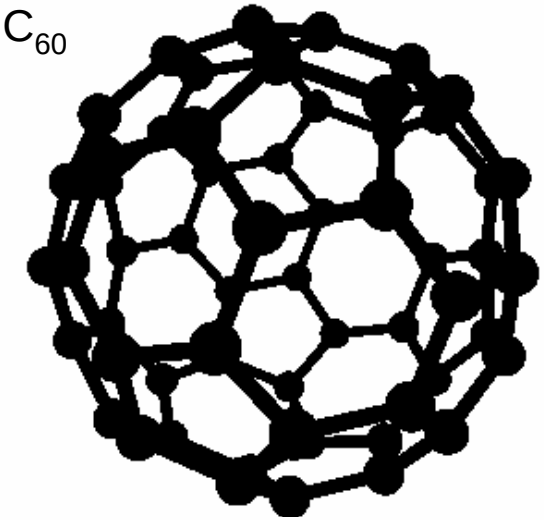
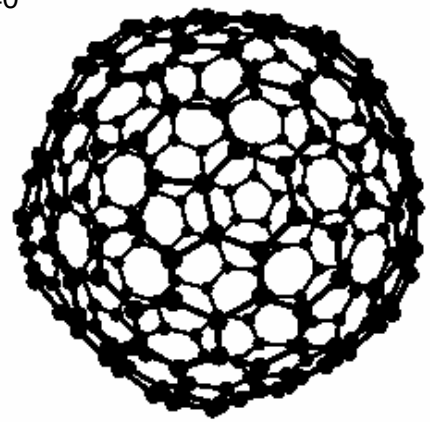
Shape of the fullerenes

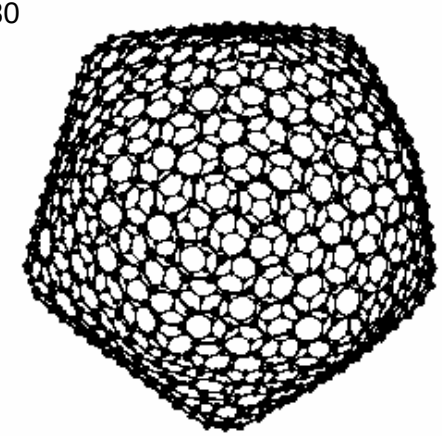
C_{60}



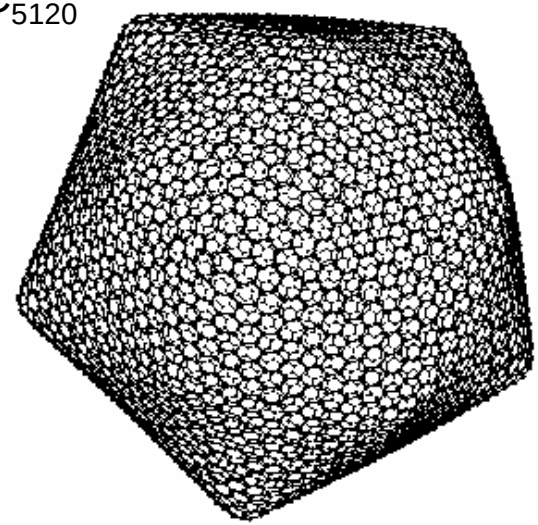
C_{240}



C_{980}



C_{5120}



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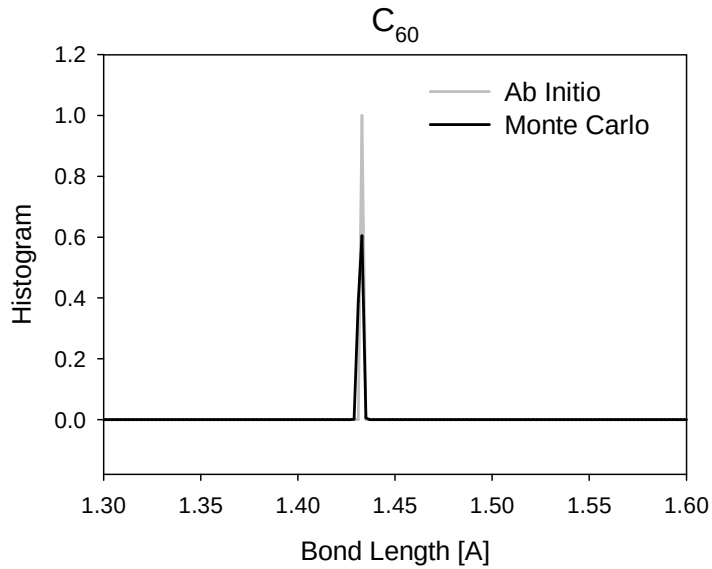
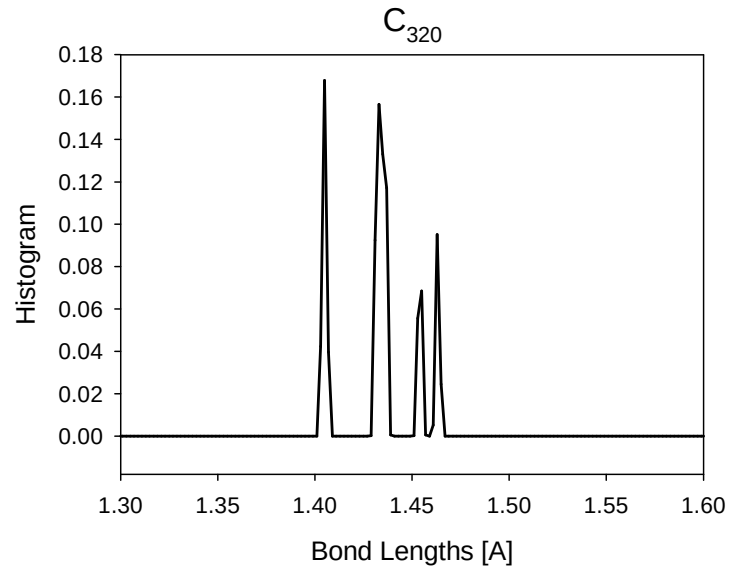
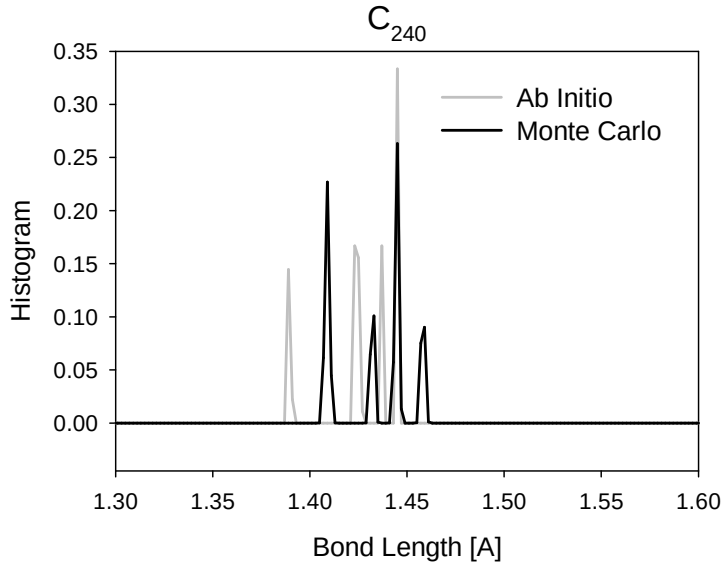
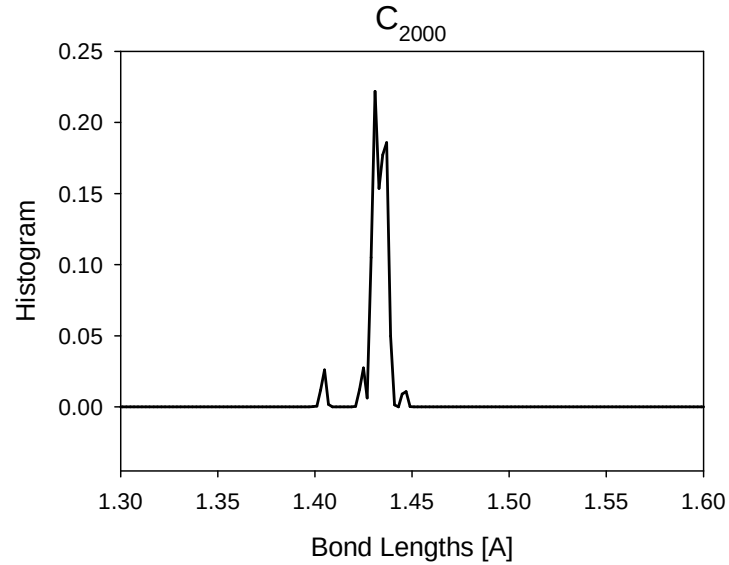
Bond length distribution









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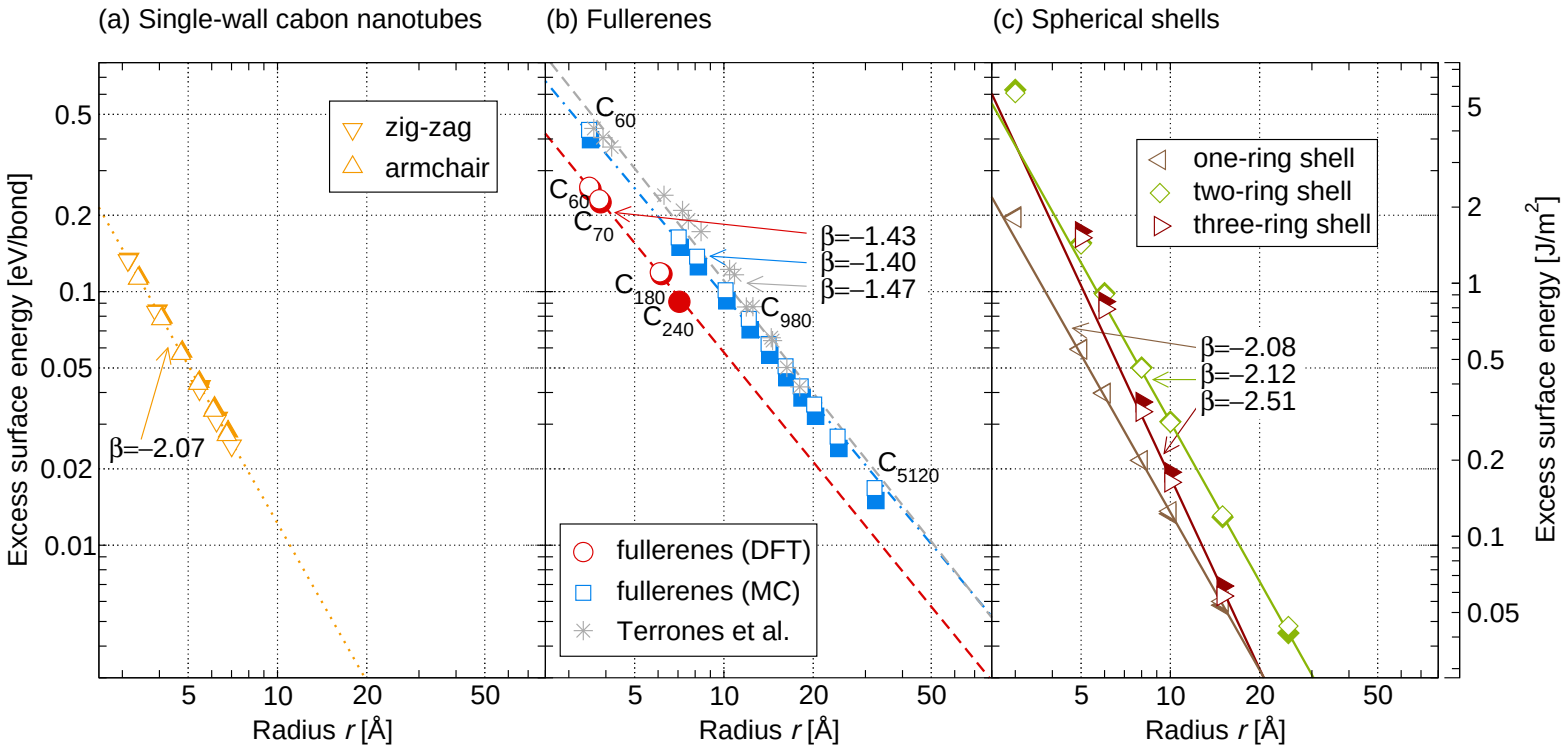
Curvature energy



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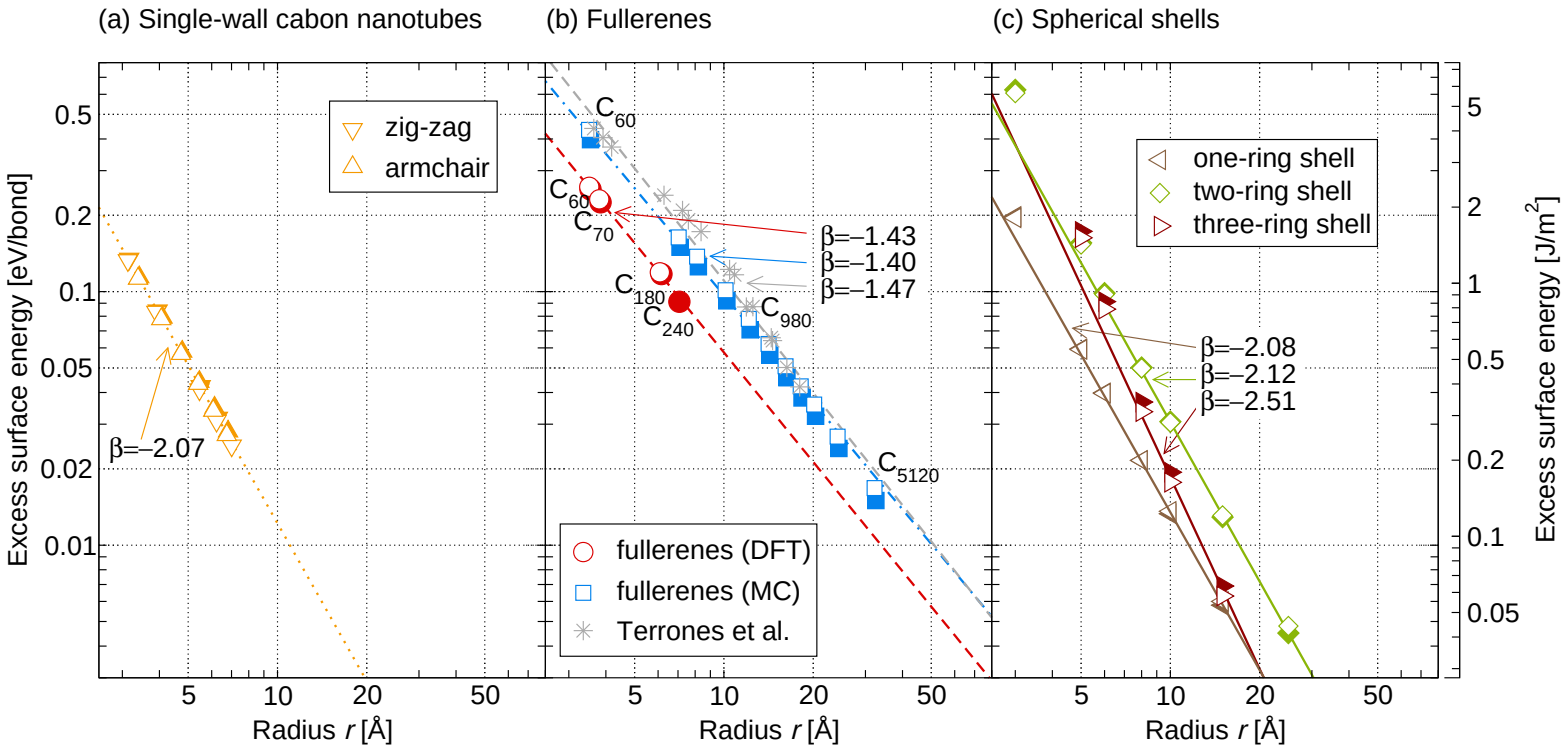

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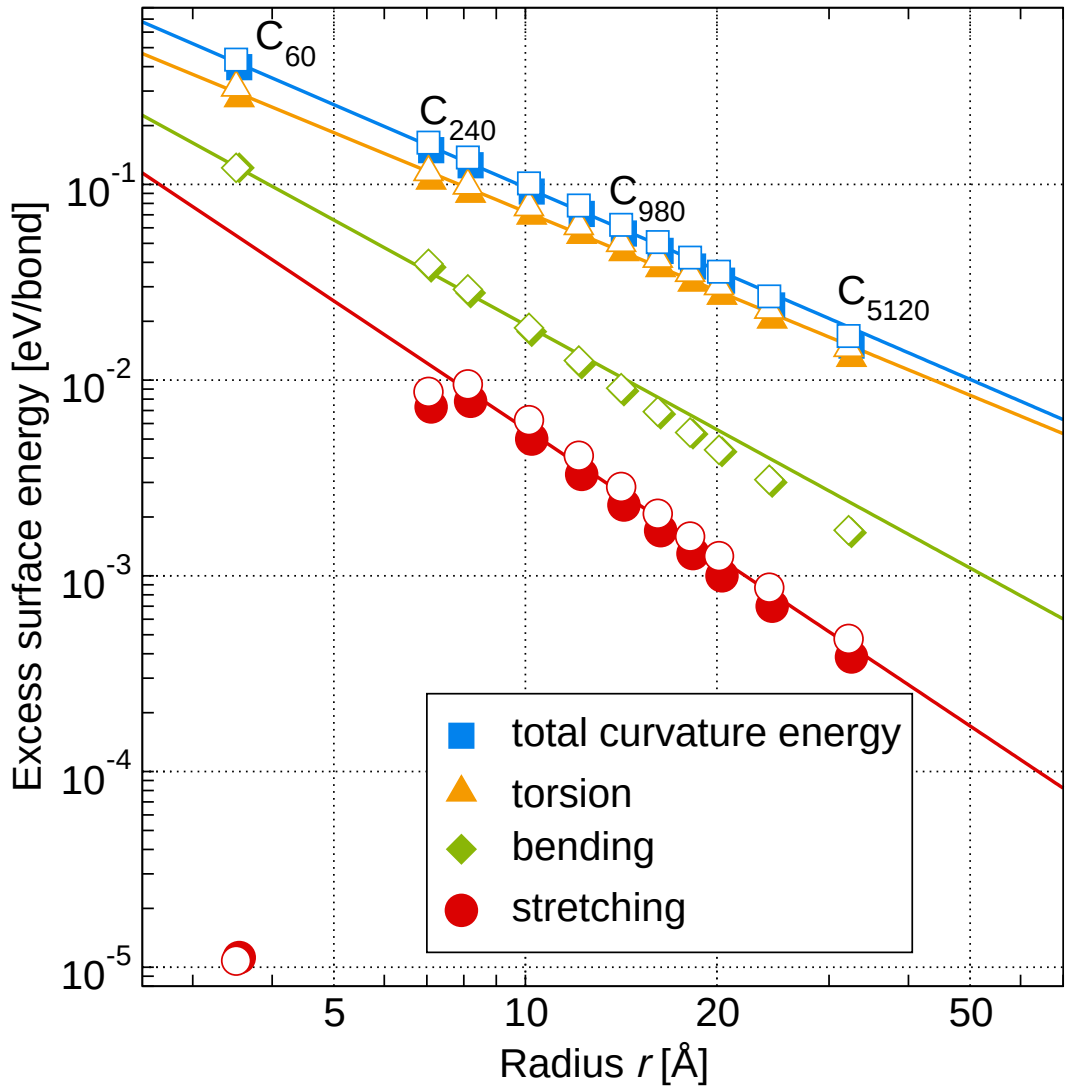
Contributions to the curvature energy



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- perfect spherical structures using approximative shell models yielded $\beta \approx -2.5$
- only stretching (two-particle) and bending (three-particle) interactons are not able to reproduce the correct fullerene geometries (i.e. bond-length and angle distributions leading to extended flattened areas), for which at least the four-particle interaction (i.e. torsion) has to be included.

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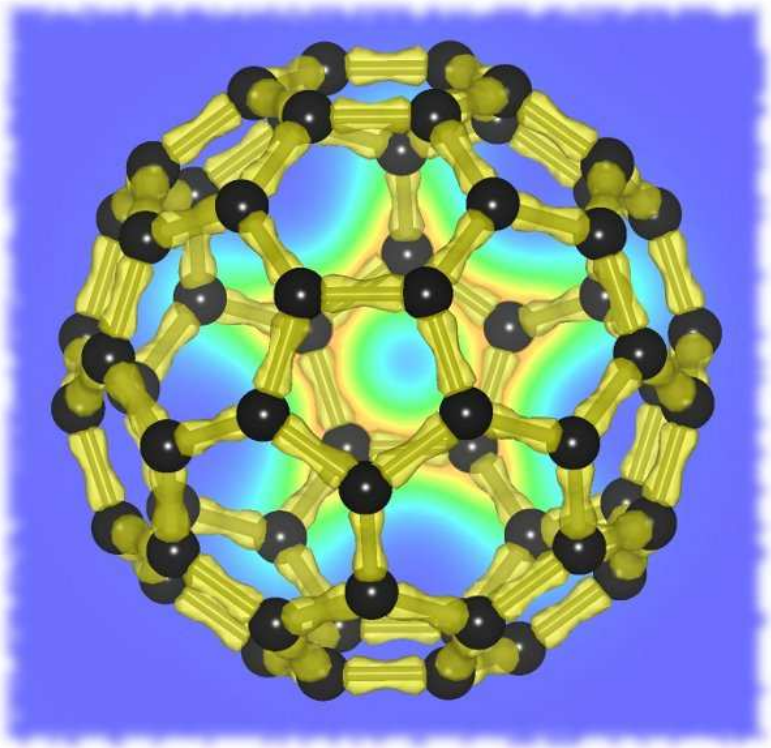
Acknowledgements



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