Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

SUPPLEMENTARY MATERIAL: Structural, electronic and energetic properties of giant icosahedral fullerenes up to C6000. Insights from an *ab initio* hybrid DFT study

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FIG. S1: CRYSTAL geometry input required to generate the (2,2) carbon icosahedral fullerene, starting from the two-dimensional graphene sheet.

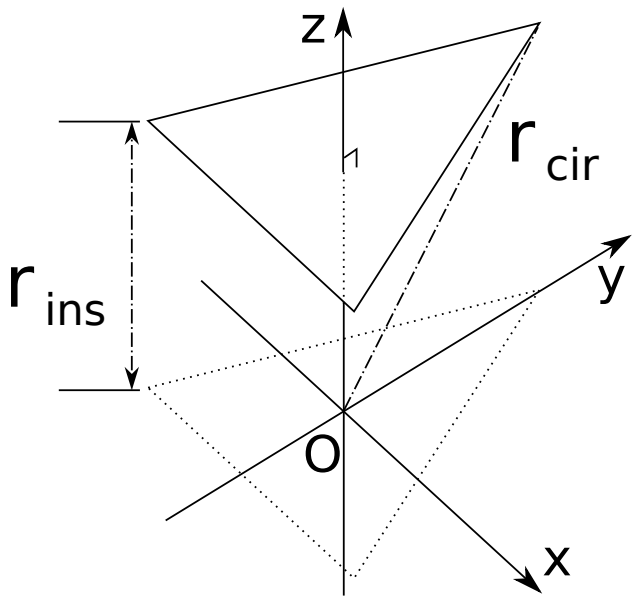

FIG. S2: Cartesian reference for the fullerene represented with respect to the triangular side of the icosahedron. O coincides with the center of the icosahedron; r_{ins} and r_{cir} are the radii of the corresponding inscribed and circumscribed spheres.

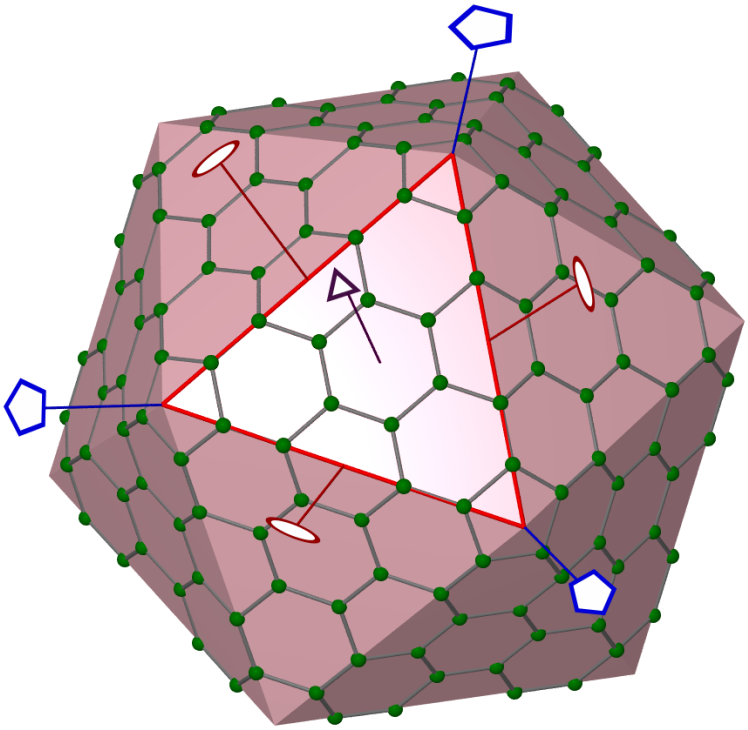

FIG. S3: Construction of the icosahedral fullerene from the monolayer. The icosahedron has been represented to emphasize the faceted configuration of the as-built, unrelaxed fullerene. Rotation axes have been reported. The (2,2) fullerene (240 atoms) is shown.

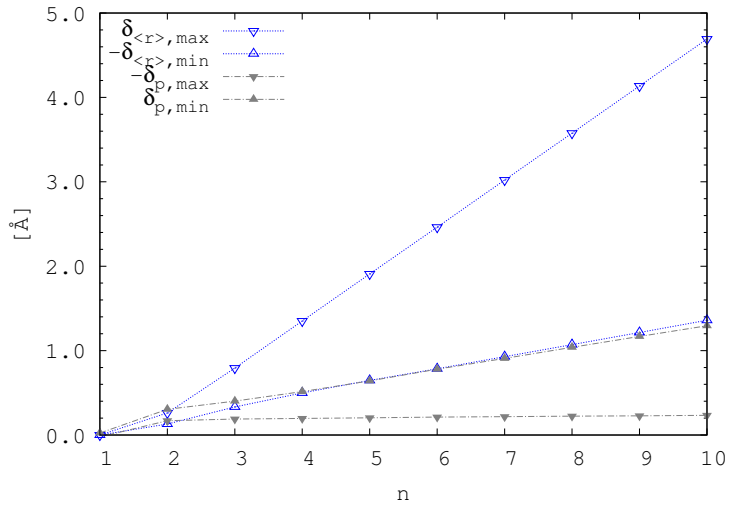

FIG. S4: Differences among minimum and maximum radii of (n, n) fullerenes and other radii: minimum ($\delta_{p,min}$) and maximum ($\delta_{p,max}$) radius of the corresponding polyhedral fullerenes; average radius of the fullerene ($\delta_{<r>,min}$ and $\delta_{<r>,max}$). All values in Å. See Figure 2 in the manuscript as a reference.

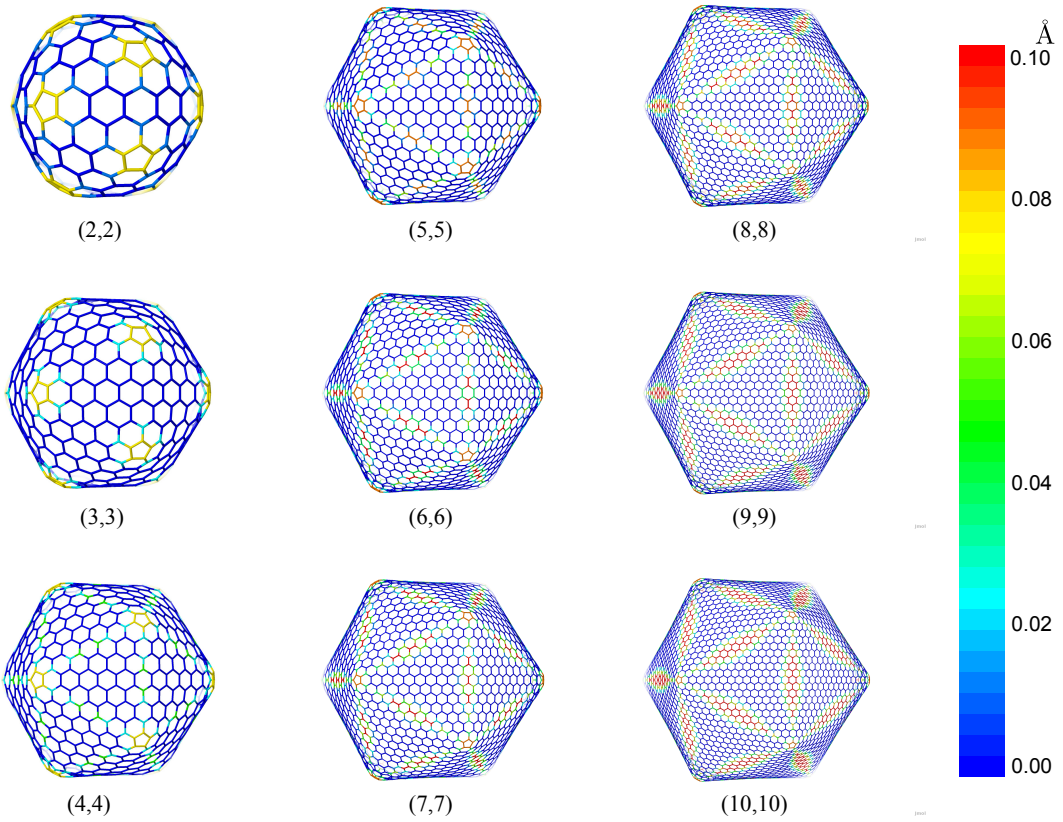

FIG. S5: Three-dimensional structures of (n, n) fullerenes ($n = 1$ not shown). For each face, atomic positions have been compared to the case of an ideally flat graphene triangle; the xy components of the relative displacements ("in-face" coordinates) are rendered by means of a color scale (values in Å). The graphene triangle is taken so that its z coordinate corresponds to the radius of the inscribed sphere to the ideal polyhedral fullerene, and its central hexagon is in-plane aligned with the central hexagon of the fullerene face.

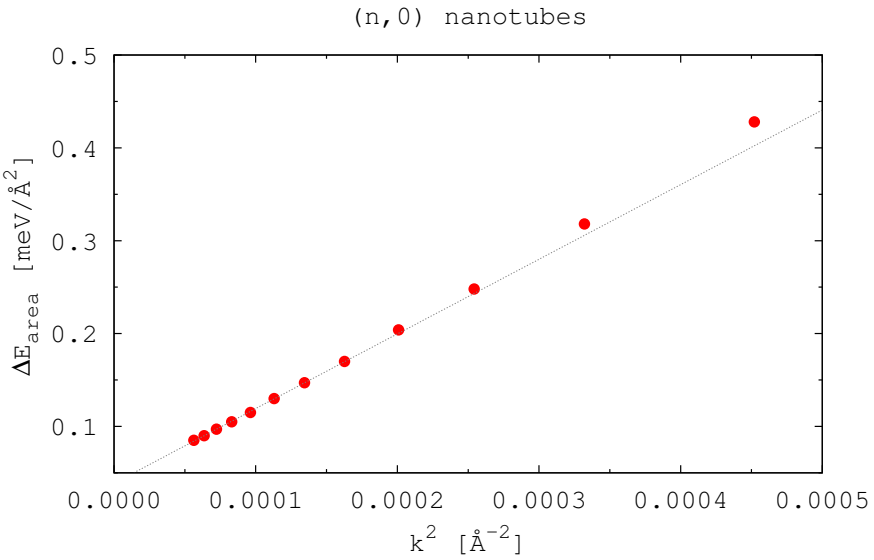

FIG. S6: Excess energy per unit area ΔE_{area} (or bending energy) of $(n, 0)$ nanotubes, in meV/Å^2, as a function of the square of the tube curvature $k^2 = 1/r^2$, in Å^{-2}. Points correspond to values obtained with the *ab initio* calculations. The curve is obtained through a linear best-fit. The fit has been performed on the last eight nanotubes of Table S1 ($n = 200 \div 340$; first eight points in the Figure starting from the left).

$(n,0)$	N_{at}	r	k	ΔE_{area}
(8,0)	32	3.184	0.3141	83.484
(16,0)	64	6.292	0.1589	21.276
(20,0)	80	7.858	0.1273	13.863
(24,0)	96	9.423	0.1061	9.832
(32,0)	128	12.554	0.0797	5.566
(40,0)	160	15.687	0.0637	3.600
(60,0)	240	23.522	0.0425	1.639
(80,0)	320	31.362	0.0319	0.925
(100,0)	400	39.199	0.0255	0.601
(120,0)	480	47.037	0.0213	0.428
(140,0)	560	54.877	0.0182	0.318
(160,0)	640	62.716	0.0159	0.248
(180,0)	720	70.553	0.0142	0.204
(200,0)	800	78.393	0.0128	0.170
(220,0)	880	86.231	0.0116	0.147
(240,0)	960	94.069	0.0106	0.130
(260,0)	1040	101.91	0.0098	0.115
(280,0)	1120	109.748	0.0091	0.105
(300,0)	1200	117.587	0.0085	0.097
(320,0)	1280	125.426	0.0080	0.090
(340,0)	1360	133.266	0.0075	0.085

TABLE S1: Properties of a set of $(n,0)$ nanotubes. N_{at} is the number of atoms, r [Å] and $k = 1/r$ [$Å^{-1}$] are the tube radius and curvature, respectively, ΔE_{area} [meV/$Å^2$] is the excess energy per unit area (or bending energy) with respect to the graphene sheet.