Supporting Information

Tkatchenko-Scheffler method

Table 1 Comparison between the Tkatchenko-Scheffler (TS) and van der Waals (optB86b-vdW) methods on one iodine molecule adsorbed on graphene layer, with 5.55 % of iodine concentration. Variation of the adsorption energy E_{ad} (eV), of the adsorption height between the molecule and the average height of the graphene layer (Å), of the minimum distance between two I_2 (Å) and the bond distance between the two iodine atoms, before and after adsorption (Å).

Physical parameters	TS method	optB86b-vdW method
E_{ad} (I_2)	-0.31	-0.38 [1]
I_2-graphene distance	3.38	3.17 [1]
I_2-I_2 distance	7.38	7.38 [1]
I-I bond (single)	2.68	2.68 [1]
I-I bond	2.69	2.70 [1]

Distortion effect of graphene layer

If we compare statistically the height difference between each carbon atoms after adsorption, see Figure 1, for non-relaxed cell a) and relaxed cell b), the distortion effect of graphene is negligible. The atom heights vary up to 0.06 Å and 0.07 Å for the system with non-relaxed and relaxed cell respectively. In addition, the median axe locates carbon atoms around a value greater than 0.02 Å and lower than 0.04 Å for the system with non-relaxed and relaxed cell respectively.

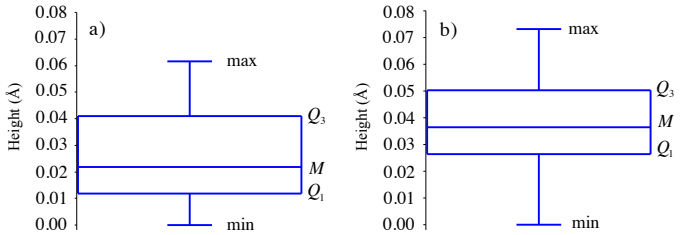
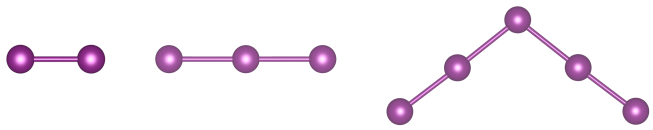


Figure 1 (Color online) Box and whisker plots of carbon atom height from agglomered iodine on graphene (15.6 % of idione concentration), with non-relaxed cell a) and relaxed cell b). The extremities of the whisker represent the minimum and maximum of all of the data, noted min and max respectively. The bottom and top of the box are the first (splits off the lowest 25 % of data from the highest 75 %) and third quartiles (splits off the highest 25 % of data from the lowest 75 %), noted Q_1 and Q_3 respectively. The band inside the box is the median (cuts data set in half), noted M.

Geometric analysis of single polyiodides

We summarized the main geometric parameters and the vibrational modes after geometry optimizations of isolated diatomic iodine, triiodide and pentaiodide ions.



(a) (b) (c)

Figure 2 (Color online) Top view of (a) diatomic iodine molecule, (b) triiodide ion and (c) pentaiodide ion.

Table 2 Variation of the bond distance between two iodine atoms (Å), of the angle between three iodine atoms (°) and of the vibrational frequencies ν (cm^{-1}) for symmetric and anti-symmetric stretching modes of single I_2, $I_3^{\delta-}$ and $I_5^{\delta-}$ in gas phase. They are compared with experimental (R = Raman, IR = Infrared) and other calculated values. Intensities are given as strong (s).

Structure	I-I	Angle	ν	
Fig.2			Sym.	A.Sym.
(a)	2.68 [1]		214	
	2.67 [2]		213 [2]	
	2.666 [3,4]		215 (R) [3]	
(b)	2.97	180	106	142
	2.93 [2]		114 [2]	147 [2]
	2.93 [5]		112 [6]	145 (IR) [7]
(c)	[2.87 ; 3.04]	[110 ; 182]	153	143
			160 (R,s) [8]	145 (R,s) [8]

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