Electronic structure of C_{240} rings

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A tight-binding molecular dynamics program has been used to optimize the structure of 6 isomers of C_{240}. Electronic structures of the cluster and crystalline forms of these isomers have also been calculated, and their stability discussed accordingly.

1. INTRODUCTION

Motivated by previous predictions[1] and recent discoveries of larger fullerenes[2], we have considered 6 isomers of C_{240} for which we calculate the electronic structure in the cluster and crystal phase. These isomers have the simplest nontrivial topological form after the spheroidal C_{60} and the well-known nanotubes. They have a toroidal form, and can be obtained by bending a graphite sheet along both x and y directions, or in other words, by bending and connecting the two ends of an open nanotube. In order to relax the resulting stress, one has to introduce defects such as pentagons in the outer ring and heptagons in the inner ring of the torus. The details of such constructions can be found in ref.[3]. The considered structures have a 5 fold axis and contain 10 pentagons and 10 heptagons.

2. CALCULATION METHOD AND RESULTS

The force calculation for relaxations is done by using the tight-binding method using the parametrization of Xu and coworkers[4]. In general the LDA cohesive energy curves of graphite and diamond are reproduced very accurately with this orthogonal scheme, which has also predicted well the atomic coordinates and bond lengths of C_{60}.

For the electronic structure calculations however, we use the parametrization used by Saito et al.[6]. This non-orthogonal parametrization is obtained from a fit to the LDA band structure of graphite and can reproduce very well the occupied energy levels of C_{60}. We therefore adopt this formulation for the electronic

structure calculations of the cluster and crystalline phases.

The properties of the considered 6 isomers can be summarized in the following Table 1. Their geometry can be viewed in the Figure 1.

The cohesive energies of our parametrization being from a fit to LDA calculations, are larger in absolute value than the experimental values; we therefore compare the differences in the cohesive energies with respect to C_{60}. From the above table, one can notice that 1) there is noticeable change in the electronic structure after relaxation, and 2) qualitatively the results are similar to those of Itoh and Ihara, namely that the isomer a is the most stable one in cluster form with a Gap of less than 1 eV, so thermodynamically, it should be slightly less stable than C_{60}, for example. This is in direct relationship to the geometry of the pentagons and heptagons on the torus. The isomer a's height is about 6.5 Å; one could guess why it has the largest gap among the 6 considered isomers since it is the only isomer whose inner and outer tubes are both of the zigzag type. Furthermore, one can also note that for the 3 structures a,d and e where the gap and the cohesive energy are largest (most stable) the cross section of the tube has a more circular shape : smallest height (about 6.5 Å) and largest difference between the outer and inner tube radii (about 4.5 Å). For more elliptic cross sections, there is presumably a larger stress field which results in a smaller cohesive energy.

Now we turn our attention to the solid phase of these clusters. Being of circular shape, they best fit into a triangular lattice. We therefore considered periodic structures with hexagonal symmetry. The length a and

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Table 1

Electronic properties of the 6 C_{240} isomers in their cluster form. The numbers in parenthesis are from a previous calculation[3]. Our calculations however are obtained after relaxation of the original structures.

| Isomer | Gap (eV) | | Cohesive Energy -CE(C60) (eV/atom) | Inner radius(Å) | Outer radius(Å) | Height(Å) |
	Xtb	Stb				
a	0.764	0.738(0.497)	-0.04(-0.09)	3.86(4.35)	8.20(8.96)	6.55(7.46)
b	0.150	0.072(0.263)	+0.01(-0.05)	3.42(3.78)	7.60(8.03)	7.96(8.65)
c	0.077	0.020(0.017)	+0.05(-0.04)	3.88(4.42)	7.61(8.00)	8.08(8.42)
d	0.157	0.309(0.082)	-0.07(-0.10)	3.36(3.69)	8.19(8.99)	6.74(7.61)
e	0.400	0.182(0.275)	-0.03(-0.09)	3.78(4.35)	8.50(9.14)	6.57(7.52)
f	0.050	0.006(0.000)	+0.05(-0.04)	3.79(4.42)	7.62(8.00)	7.99(8.00)

the height c of the cell were first determined by minimizing the total energy (keeping the atomic coordinates constant however). Once these 2 lattice parameters were found, we used the non-orthogonal parametrization[6] to calculate the electronic energy levels of these structures at the Γ point.

The obtained results can be summarized in the following Table2:

We can see that not only the lattice constants differ from isomer to isomer, but also the shortest distance between two neighboring clusters is isomer dependent. Structure d which was insulating in the cluster form has

become a metallic crystal, whereas the structure f which had a tiny gap as a cluster, is a semiconducting solid with a gap of 0.5 eV: Open shell structures can form a semiconductor and closed-shell systems can form a metal. This is a well-known result in the case of atoms, and depends on the crystal symmetry.

3. CONCLUSIONS

Several isomers of C_{240} were considered in this work. We can deduce from our tight-binding calculations their electronic properties and perhaps stability. To study their mechanical properties, further calculations

Table 2

Electronic properties of the 6 C_{240} isomers in their crystalline form.

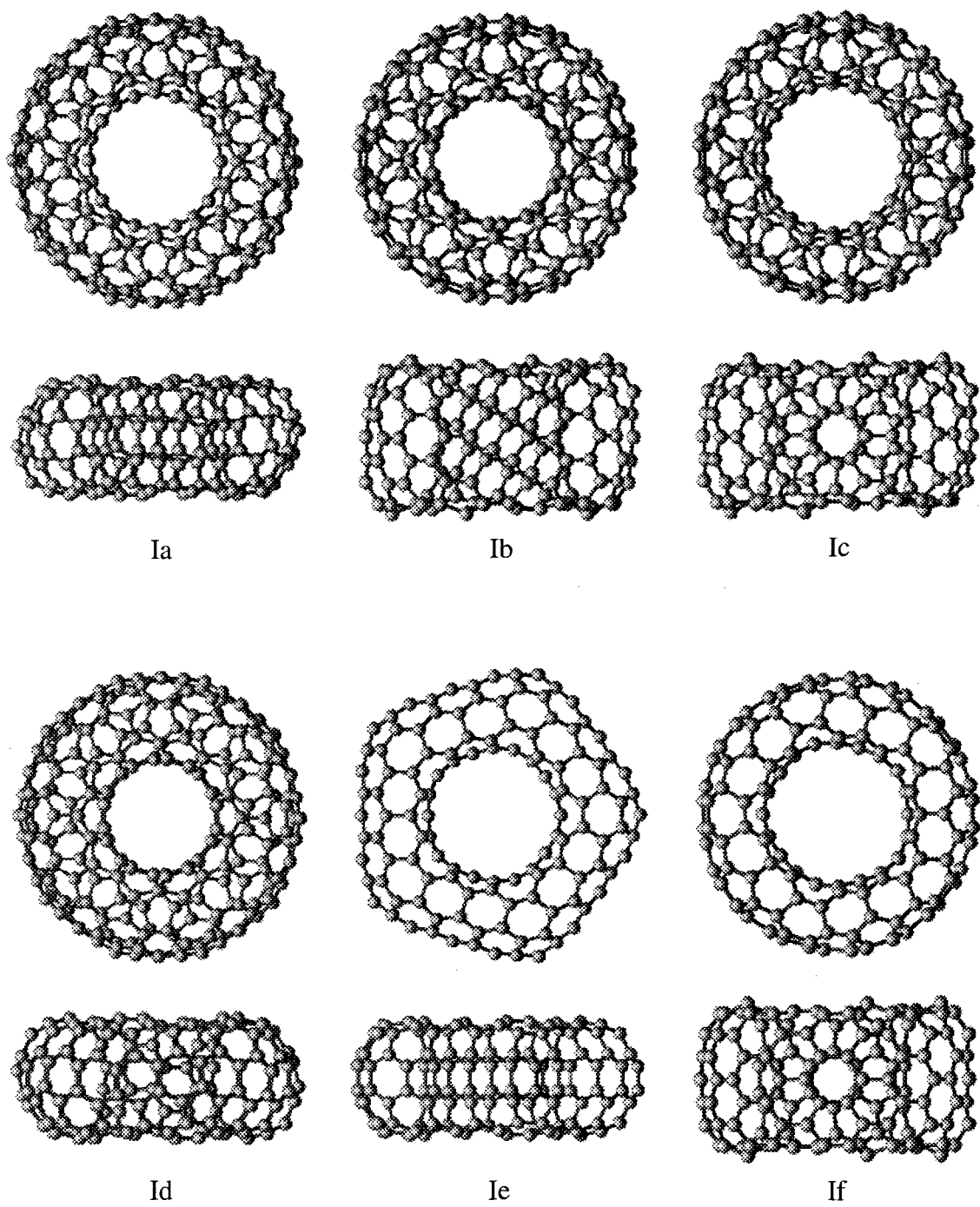| Isomer | Gap (eV) | | Cohesive Energy -CE(C60) (eV/atom) | Lattice constant a(Å) | Lattice constant c(Å) |
	Xtb	Stb			
a	0.656	0.380	-0.04	18.17	8.55
b	0.298	0.206	+0.01	16.61	9.56
c	0.164	0.041	+0.04	16.65	9.48
d	0.105	0.023	-0.08	18.16	8.54
e	0.207	0.034	-0.03	18.12	8.77
f	0.561	0.414	+0.01	16.66	9.59

Figure 1. Othographic projection of the 6 isomers, topview and sideview.

of the vibrational frequencies of these clusters are underway. In the solid phase, we could find semiconducting as well as metallic behavior in these systems. Synthesizing the solid phase would of course be very difficult; but there might be the possibility of making some of these clusters, or similar structures with the same topology, experimentally.

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