Simulation Of The Properties Of Hydrogen Clusters And Hydrogen Adsorbed On Graphitic Surfaces

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There are several reasons that justify the present interest in hydrogen and hydrogen clusters. Hydrogen is expected to play a decisive role as a viable alternative to present fuels for the massive production of energy. In the near future hydrogen could replace gasoline in cars, and prototypes using electric motors that obtain the energy from the reaction of hydrogen with atmospheric oxygen have been already developed by most car manufacturers. The outstanding problem remaining to be solved in this area is to develop an effective way to store sufficient amount of hydrogen in the tank of a car. Intensive effort is now dedicated to find materials that could act as efficient hydrogen containers, and graphitic materials providing a large surface for the adsorption of hydrogen are one of the candidates. We have performed density functional (DFT) calculations of the adsorption of molecular hydrogen on the surface of graphene and carbon nanotubes, and also on the surface of layers and nanotubes of similar materials. The adsorption energies are not larger than 0.1 eV per hydrogen molecule, and this binding energy is not enough to reach the target of 6% of the storage system weight established by the DOE of USA. The calculations suggest two strategies to improve the storage capacity. The first one consists in doping the graphitic surfaces with adequate impurities [1], and Figure 1 shows that lithium doping increases the binding energy of the hydrogen molecule to the graphitic surface in a factor of two. The second consists in using a nanoporous material with an optimized pore size, such that the effective graphitic surface available to each hydrogen molecule becomes enhanced. As an example, the binding energy of the molecule inside a narrow carbon nanotube also doubles its value on the external surface [2].

In addition, it is expected that some day hydrogen isotopes will be the fuel in nuclear fusion reactors, once the problem of sustaining and controlling the reaction is solved. In fact, some unexpected relations to the cluster field may be established. Experiments by Zweiback et al. [3] have shown that the irradiation of a dense molecular beam of large deuterium clusters with a high intensity laser produces the strong ionization of the clusters, which leads to a violent Coulomb explosion of the positively charged skeleton. Fusion reactions induced by the collisions of flying deuterium nuclei arising from the coulomb explosion of neighbor clusters in the molecular beam have been observed in those experiments, and it has been proposed that table-top neutron sources could be constructed based on this cluster beam technique. Using the time dependent density functional formalism (TDDFT) we have performed computer simulations of the first stages of the processes occurring in those experiments, that is, of the step of the ultrafast laser irradiation of the deuterium clusters. The dynamical behavior of the clusters in

response to the laser depends on the frequency and intensity of the laser, and different processes like collective electronic excitations, slow fragmentation of the cluster, and ionization followed by Coulomb explosion, can occur.

Other properties of hydrogen clusters are also interesting. In the neutral clusters, the hydrogen atoms form strongly bonded molecules and these molecules interact weakly with each other. The shape of the molecules is not perfectly spherical; nevertheless, DFT calculations indicate that the hydrogen clusters show some magic numbers (that is, clusters that have a particularly high stability) similar to those observed in clusters of the inert gases. Comparison with the results of Raman spectroscopy experiments gives further insight on the structure of those molecular clusters.

Atomic hydrogen can be used to probe the local reactivity of metallic clusters. We have applied this idea to small planar gold clusters. Adsorption of H shows that the peripheral rim is the most reactive region of the gold clusters, and this conclusion helps understanding the reactivity of the gold clusters with oxide surfaces.

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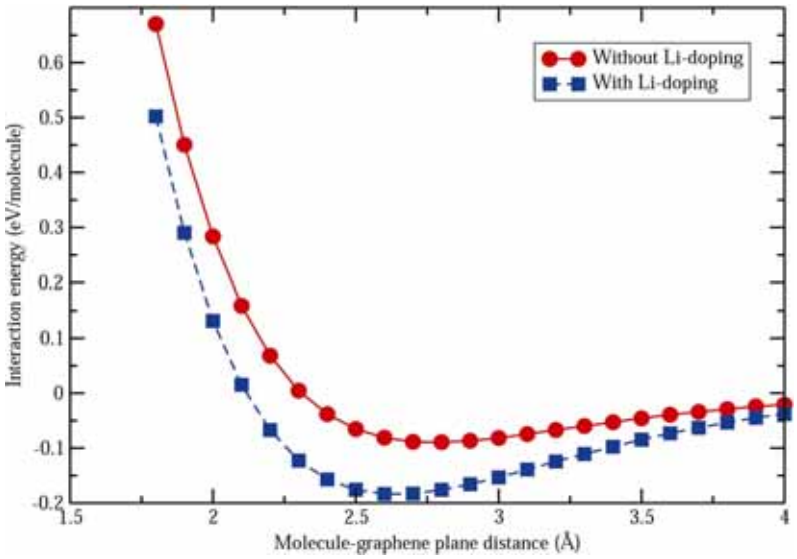

Figure1. Interaction energy of molecular hydrogen with clean and a Li doped graphene.

References

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