Quantum Molecular Dynamics and Ab initio Studies of The Crystal Structure of Hydrogen and Deuterium

Carlo Guerrero[1)], Jose Manuel Perlado [1)], Santiago Cuesta-Lopez [2)]
[1)] Universidad Politécnica de Madrid, DENIM-IFN, E-28006 Madrid (SPAIN)
[2)] University of Burgos, I+D+I building,Plaza Misael Bañuelos, s/n 09001, BURGOS (SPAIN)

Abstract: Hydrogen isotopes play a critical role both in inertial and magnetic confinemen Nuclear Fusion. Since the preferent fuel needed for this technology is a mixture of deuterium and tritium. The study of these isotopes particularly at very low temperatures carries a technological interest in other applications. The present line promotes a deep study on the structural configuration that hydrogen and deuterium adopt at cryogenic temperatures and at high pressures. Typical conditions occurring in present Inertial Fusion target designs. Our approach is aims to determine the crystal structure characteristics, phase transitions and other parameters strongly correlated to variations of temperature and pressure.

1. Introduction

Target pellets used in inertial confinement fusion, based on Deuterium-Tritium fuel, follow a complex manufacturing process of permeation at temperatures near the triple point [1]. Details of such process seem to induce the nucleation of anisotropic crystal structures organized in layers with pseudo nanocrystalline properties. This spatial order posses a serious problem since may affect the efficiency of Inertial Fusion implosion [2].

We report theoretical studies on the hydrogen isotopes and their mixtures, looking for metastable structures and their characteristics, as reported in previous experimental work[3]. Working in conditions close to the triple point is a difficult experimental task. On the contrary, our methodology, based in a quantum molecular dynamics approach, is able to provide a good description of the properties and the spatial arrangement exhibited by the hydrogen isotopes under extreme conditions.

2. Methodology

The targets used in inertial confinement fusion, consists of small spheres of a certain material, filled with a mixture of deuterium-tritium (either solid or liquid, depending on the case), at cryogenic temperatures. For our design and model we consider temperatures below the triple point of hydrogen, about 14 K (and atmospheric pressure) [4]. In this process, the spheres have a diameter of 0.4 to 1.8 mm, and are processed at a pressure of about 25 to 1000 atm (~2.5 to 100 MPa).

Our simulation procedure starts by making a solid structure in an HCP configuration either in different size scales for the simulation of a cluster, or by placing periodic boundary conditions. Since hydrogen molecules can change their orientation in each of the points of the lattice where they are placed, this fact define an initial orientation for all the solid.

Though it really has devised a code that gives the random orientation of the molecules. We define similar initial conditions for each of our simulations. The orientation is a parameter to observe that varies with temperature and pressure. Moreover, according to Toledano et al, solid molecular hydrogen show three phases, which are related to the freedom that the molecule has to change orientation in variation with temperature and pressure[5]. Therefore, orientation is a parameter critical to observe.

The lattice constant changes according to the size or number of atoms in the solid (molar volume) [6], the first step including our first study was to find the optimum lattice parameters for each of the settings shown (see Figure 1A).

Taking then this initial structure we apply an *ab initio* methodology, to relax the structure and the different configurations obtained looking for energy minimum. After that, we launch a study of structural stability involving different temperatures by means of a thermostat type *Nose-Hoover*, coupled to quantum molecular dynamics. We analyse changes in the orientation of the molecular axis, or possible changes in the solid phase at different pressures (for this last step is expected to have finally implemented a *Parrinello-Rahman* barostat). Using nearest neighbour analysis, and the radial correlation function, we are able to obtain information about phase changes occurred during the thermalization of the structure, which has a great relevance since experimental fingings point to. Experimentally it has been found the existence of meta-stable states in particular areas of DT ice targets [3] that could affect the homogeneity of the samples, and therefore the efficiency of implosion. All simulations were optimized with the help of the parallelization of the codes used.

3. Results

Our results obtained so far are aimed at studying the parameters and structures needed to develop establish the basis of a solid methodology aimed to overcome the simulation of the deuterium-tritium mixtures of wide interest in Fusion Science.

Our results reproduce the structural order showed by molecular hydrogen at any temperature and pressure (see Figure 1B), in particular, at pressure and temperatures of relevance in the manufacture process of DT-ice targets.

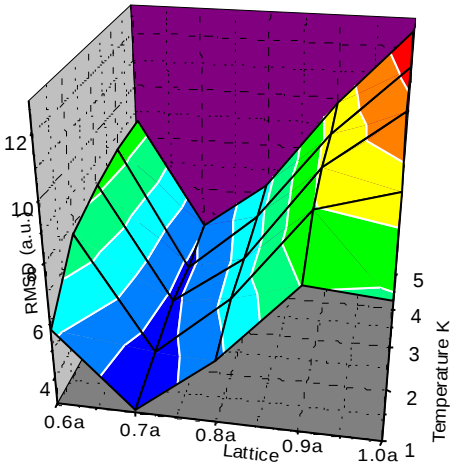

Fig. 1. Lattice constant and root-mean-square deviation (RMSD), difference between initial and final structural configuration. Taking the different lattice constants as ***0.6a, 0.7a, 0.8a, 0.9a*** y ***1.0a*** (where ***a=3.783*** *Å* [7]). Comparison is made between the structures through RMDS, for the structural difference between the initial configuration (ideal) and the final configuration after 2000 fs relaxation of the structure. The study was carried out also five temperatures from 1K to 5K, studying the mobility of hydrogen atoms with temperature.

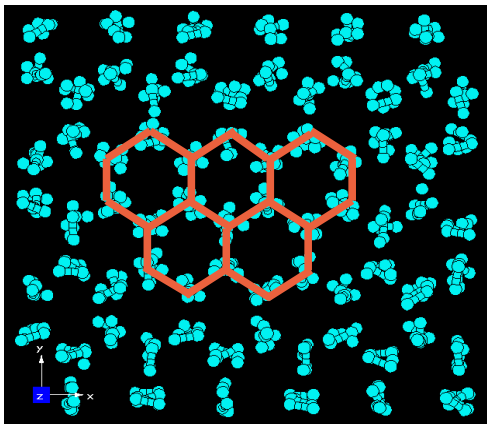

Fig. 2 Hexagonal structure of the solid molecular hydrogen to 15K and 1 GPa. Shows the free rotor which is in accordance with phase I [5]

Moreover the validity of our methodology has been tested after showing the reproduction of the pressure induced Phase I - Phase II transition for a molecular structure made of *para*-hydrogen at cryogenic temperatures, and very high pressures [8]. This transition has been predicted both theoretically and experimentally [5], and it is very characteristic because the hydrogen jumps from a configuration with molecules as free rotors (the atoms can describe a sphere), to an order where such rotations are restricted.

All results presented are for a solid molecular hydrogen, with the same orientation, and different sizes.

4. Conclusions

We have developed a method of multiscale simulation methodology, by which able to describe the behaviour of hydrogen and its isotopes, varying under high pressure conditions and at temperatures, observing the structural changes below the triple point. Our results are of crucial interest in achieving a full understanding of the DT-fuel behaviour and efficiency of implosion in Inertial Fusion Technology.

In addition such a methodology opens the possibility to study small hydrogen clusters and where the effects of size represents a wealth of information about the way in which hydrogen interacts or can be captured by other materials.

References

[1] I. V Aleksandrova, S V Bazdenkov, V I Chtcherbakov, A I Gromov, E R Koresheva, E A Koshelev, I E Osipov and L S Yaguzinskiy. “An efficient method of fuel ice formation in moving free-standing ICF/IFE targets”. Journal of Physics D: Applied Physics, **37** 1163–1178 (2004)

[2] R. Harding, L. M. Elasky, S. Verbridge, et al., “Formation of deuterium-ice layers in OMEGA targets,” LLE Review, Quarterly Rept., 99, 160 (2004).

[3] B. J. Kozioziemski, A. A. Chernov, E. R. Mapoles, and J. D. Sater. “Metastable polymorphs of hydrogen isotopes solidified near the triple point”. Physical Review B **82**, 012104 (2010)

[4] I.V Alekxandrova, E. R. Koresheva, I. E. Osipov, S. V. Bazdenkov,A. A. Belolipetskiy, V. I. Chtcherbakov, T. P. Timasheva, A. A. Tonshin, L. S. Yaguzinskiy, V. M. Dorogotovtsev, and A. A. Akunets. “Cryogenic fuel targets for inertial fusion: optimization of fabrication and delivery conditions”. Journal of Russian Laser Research, **Volume 28**, Number 3 (2007).

[5] P. Toledano, Hannelore Katzke, Alexander F. Goncharov, and Russell J. Hemley. “Symmetry Breaking in Dense Solid Hydrogen: Mechanisms for the Transitions to Phase II and Phase III”. PRL **103**, 105301(2009)

[6] P.C. Souers. “Hydrogen Properties for Fusion Energy”. University of California. (1986)

[7] N.N. Galtsov A.I. Prokhvatilov, G.N. Shcherbakov, and M.A. Strzhemechny. “Properties of solid hydrogen doped by heavy atomic and molecular impurities . Fizika Nizkikh Temperatur”. v. 29, Nos.9/10, p. 1036–1040 (2003).

[8] C.Guerrero, S. Cuesta-López,J.M.Perlado. “Structural propieties of hydrogen isotopes in solid phase in the context of inertial fusion target manufacturing”. Eur. Phys. Journal Web of conferences. Proceedings of the Seventh Conference on Inertial Fusion Sciences and Applications (IFSA 2011). In press 2012.