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Young's moduli of carbon materials investigated by various classical molecular dynamics schemes

Florian Gayk, Julian Ehrens, Tjark Heitmann, Patrick Vorndamme, Andreas Mrugalla, Jürgen Schnack [*]

Dept. of Physics, Bielefeld University, P.O. Box 100131, D-33501 Bielefeld, Germany

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ABSTRACT

For many applications classical carbon potentials together with classical molecular dynamics are employed to calculate structures and physical properties of such carbon-based materials where quantum mechanical methods fail either due to the excessive size, irregular structure or long-time dynamics. Although such potentials, as for instance implemented in LAMMPS, yield reasonably accurate bond lengths and angles for several carbon materials such as graphene, it is not clear how accurate they are in terms of mechanical properties such as for instance Young's moduli. We performed large-scale classical molecular dynamics investigations of three carbon-based materials using the various potentials implemented in LAMMPS as well as the EDIP potential of Marks. We show how the Young's moduli vary with classical potentials and compare to experimental results. Since classical descriptions of carbon are bound to be approximations it is not astonishing that different realizations yield differing results. One should therefore carefully check for which observables a certain potential is suited. Our aim is to contribute to such a clarification.

1. Introduction

Many carbon-based materials cannot be simulated by quantum mechanical means, not even by Density Functional Theory (DFT), since they are either too extended or not regular. The latter is for instance the case for nanometer thin carbon membranes of macroscopic lateral size, which are produced from molecular precursors [1–5]. Although the precursors are well-characterized, not much is known about the internal structure of such nanomembranes [6]. Very likely the material is disordered. Mechanical properties on the other hand, such as Young's moduli, can be determined [7]. Similar statements hold true for many other carbon materials, for instance carbon nanotubes under tension [8] or amorphous carbon in general [9].

Advanced classical carbon potentials together with classical molecular dynamics are employed in order to simulate structure and dynamics of challenging carbon materials. Often the mechanical properties of the investigated materials are of special interest [6,8,10,11]. But since the classical calculations suffer from their approximate nature, we propose in this article to quantify the accuracy of various carbon potentials in view of the resulting Young's moduli for known carbon materials, before evaluating moduli for unknown systems. A very valuable comparison along these lines, in which the graphitization of amorphous carbon was studied, has been published recently [12]. As expected, none of the classical potentials works perfectly for a complex process such as graphitization, and some of the investigated potentials perform poorly.

Since the quality of a classical description might very much depend on the investigated observable, we are continuing the efforts of [9,12–14] by investigating the Young's moduli of three well-known carbon materials in large scale calculations. As materials we choose graphene, a carbon nanotube, and diamond. For the simulations we used various carbon interatomic potentials as included in LAMMPS [15] as well as the modified EDIP potential of Marks [13,16,17]. Our investigations, as well as those by other authors, might guide future developments and the proposition of new carbon potentials [18], but primarily we want to provide valuable information to users (of LAMMPS) on which potential to choose for certain investigations.

A final word might be in order regarding possible modifications of the investigated potentials. One could of course tweak every potential by multiplying it with an appropriate factor to achieve the correct experimental modulus (at least for one substance). Such a procedure would leave the ground state configurations largely the same. But this would cure only one observable and modify the properties of the

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* Corresponding author.
E-mail address: jschnack@uni-bielefeld.de (J. Schnack).

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potentials with respect to other observables in an uncontrolled way as was e.g. discussed in Ref. [19]. Our goal is to contribute to the knowledge about well-defined and often-used carbon potentials and their predictive power, especially in view of the recent upsurge of carbon research.

The article is organized as follows. In the next section we shortly repeat the essentials of classical molecular dynamics calculations. The main section is devoted to the simulations of the three carbon materials. The article closes with a discussion and conclusions.

2. Classical carbon-carbon interaction

A realistic classical carbon-carbon interaction must be able to account for the various sp^n–binding modes. The program package LAMMPS [15] offers several of such potentials, among them those developed by Tersoff and Brenner in various versions [20–22] as well as new extensions built on the original potentials.

In addition to the implemented potentials we are going to use the improved EDIP potential by Marks [16] which so far is not included in standard versions of LAMMPS. Taking this potential as an example, we want to qualitatively explain how such potentials work. These potentials comprise density-dependent two- and three-body potentials, U_2 and U_3 in this example respectively,

$$U\left(\vec{R}_1,\ldots,\vec{R}_N\right) = \sum_{i=1}^{N}\left(\sum_{\substack{j=1\\ j\neq i}}^{N} U_2(R_{ij},Z(i)) + \sum_{\substack{j=1\\ j\neq i}}^{N}\sum_{\substack{k=j+1\\ k\neq i}}^{N} U_3(R_{ij},R_{ik},\theta(i,j,k),Z(i))\right) \tag{1}$$

which account for the various binding modes. This is achieved by an advanced parameterization in terms of a smooth coordination variable $Z(i)$ as well as by appropriate angle dependencies $\theta(i,j,k)$. The EDIP potential employs a cutoff of 3.2 Å and a dihedral penalty.

Ground states are then found by the method of steepest descent, by conjugate gradients or damped dynamics (frictional cooling). The Young's modulus E in the ground state, i.e. at temperature $T = 0$ K, can be evaluated from the curvature of U at the ground state configuration (the kinetic energy is zero) [23].

$$E_V = \frac{1}{V_0}\left(\frac{\partial^2 U}{\partial\alpha^2}\right)_{\alpha=1} \quad , \tag{2}$$

where α is the factor by which all positions are scaled along the direction of the dimensionless unit vector $\vec{e}_\alpha$, i.e.

$$\vec{x}_i \to \vec{x}_i + (\alpha-1)\,\vec{e}_\alpha\cdot\vec{x}_i \quad \vec{e}_\alpha \quad . \tag{3}$$

V_0 denotes the cuboidic volume of the sample in equilibrium.

For two-dimensional systems such as graphene, which do not have a volume in classical molecular dynamics, (2) can be replaced by

$$E_S = \frac{1}{S_0}\left(\frac{\partial^2 U}{\partial\alpha^2}\right)_{\alpha=1} \quad , \tag{4}$$

where S_0 is the area of the stretched material in equilibrium [23]. Several authors introduced an artificial thickness h_0 in order to stay with definition (2). This thickness is often taken either as the graphite interlayer distance $h_0 = 3.35$ Å or the carbon-carbon distance of graphene, i.e. $h_0 = 1.42$ Å. In this article we choose $h_0 = 3.35$ Å.

3. Theoretical investigations

We included the following carbon potentials in our investigations: Tersoff in various versions [20,24–26], REBO-II [22] and AIREBO as well as ABOP [27]. The AIREBO potential [28,29] is investigated with its flavors: "naked" AIREBO, AIREBO with additional long range Lennard-Jones potential (AIREBO+LJ), AIREBO with additional torsion term (AIREBO+t), and AIREBO with both terms (AIREBO+LJ+t). If not otherwise stated, the cutoff of the Lennard-Jones potential has been chosen as 10.2 Å. All of these potentials are discussed in great depth in Ref. [12]. In addition we performed simulations with the EDIP potential of Marks [16]. For all potentials the respective ground states are determined, which do not need to be the same. Then the moduli are evaluated according to Eqs. (2) and (3) for $T = 0$ K. For the latter purpose we employed the script "ELASTIC" that is part of the LAMMPS folder, but without relaxation in the stretched state and a self written script in case of Mark's EDIP potential.

3.1. Graphene

Our theoretical investigations consist in the generation of initial arrangements of approximately square graphene sheets of N atoms with open boundary conditions. N was chosen as 28, 66, 120, 190, 276, 378, 496, 630, 780. As we let N grow to large numbers, finite size as well as boundary effects decrease.

The experimental value for the Young's modulus of graphene is about 1 TPa [30], which is also reproduced as 1.05 TPa by DFT calculations for this regular structure [31]. Fig. 1 shows the results obtained with the various potentials on the l.h.s., whereas the r.h.s. displays the moduli obtained for several versions of the Tersoff potential. The modulus turns out to be isotropic in accordance with Refs. [32,33]. The majority of potentials converges with N against values for the modulus in the range of 1.1 to 1.3 TPa. The various investigated AIREBO potentials yield identical results. The EDIP potential comes closest to 1 TPa, practically on top of REBO-II, whereas the ABOP modulus falls below 0.8 TPa.

The chosen Tersoff potentials, displayed on the r.h.s. of Fig. 1, exhibit a similar spread of results. Earlier parameterizations of 1989 and 1994 deviate by about 0.3 TPa from the value of 1 TPa, whereas the more recent parameterizations of 2005 and 2012 yield values of 1.1 TPa similar to the EDIP or REBO-II potentials. It should be noted that the Tersoff potential of 1990 [25] does not reproduce the correct graphene structure in our simulations.

For C—C— bond distances compare Table 1.

3.2. Carbon nanotubes

The investigated carbon nanotube (CNT) is a (20,20) tube with armchair geometry. In the investigation we varied the number of carbon atoms $N = 120,\ 200,\ 360,\ 680,\ 1040,\ 1360,\ 1720,\ 2040,\ 2400,\ 2720$; N is thus also a measure of length.

Since CNTs share the sp^2 structure with graphene, one would expect that Young's moduli of single walled CNTs are very similar to that of graphene, which is indeed the case at least for large enough radii [34,35]. For our calculations this similarity also holds. Again, the majority of potentials converges with N against values in the range of now 1.0 to 1.3 TPa, see l.h.s. of Fig. 2. The various investigated AIREBO potentials once more yield identical results. The EDIP potential comes closest to 1 TPa, again together with REBO-II, whereas the modulus calculated with ABOP again stays below 0.8 TPa.

Also for the Tersoff potentials we obtain results similar to those for graphene, compare r.h.s. of Fig. 2. The large deviation for the Tersoff potentials of 1990 and 1994 correlates again with deficiencies to reproduce the structure. Using the version of 1994 the transverse section of the CNT is not a circle but more a rounded square in our simulations, whereas we could not obtain a reasonable structure with the 1990 version at all.

For C—C— bond distances compare Table 1.

3.3. Diamond

The studied diamond structures have cuboidic shape and contain $N = 280,\ 621,\ 1166,\ 1963,\ 3060,\ 4505,\ 6346,\ 8631$ atoms. The largest

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