

Atomic and electronic structure of crystalline–amorphous carbon interfaces

G. Kopidakis^{a,*}, I.N. Remediakis^b, M.G. Fyta^{b,1}, P.C. Kelires^b

^a Department of Materials Science and Technology, University of Crete, P.O. Box 2208, 71003 Heraklion, Crete, Greece

^b Physics Department, University of Crete, P.O. Box 2208, 71003 Heraklion, Crete, Greece

Available online 17 July 2007

Abstract

Interfaces of diamond with amorphous carbon (a-C) are investigated using tight-binding molecular dynamics simulations. Such interfaces are found to be stable, with a-C atoms covalently bonded to the diamond surfaces. The atomic and electronic structure of the a-C region is consistent with previous results on pure a-C and does not depend critically on the diamond face exposed. However, surface properties influence the relative stability of interfaces with high density a-C. In this case, the interfacial region is small and very dense homogeneous a-C grows on diamond. At lower densities, carbon atoms nucleate on diamond surfaces and create an intermediate region between diamond and a-C. The shape of diamond crystals embedded in a-C is predicted using the Wulff construction with appropriately defined energies for surfaces with overlayer material. These predictions are verified by empirical potential simulations of nanodiamond inclusions in a-C matrix. The electronic density of states and the dielectric function, calculated for our samples containing a-C/diamond interfaces, show that optoelectronic properties of these composite materials are dominated by a-C.

© 2007 Elsevier B.V. All rights reserved.

Keywords: Diamond; Amorphous carbon; Interface; Nanocrystals

1. Introduction

Diamond–amorphous carbon heterostructures with promising mechanical and optoelectronic properties have emerged as a result of advances in carbon-based materials deposition and characterization techniques [1–4]. These materials exhibit a variety of structural features and properties that depend on growth conditions. A large number of experimental studies reveal relations to several carbon allotropes and transformations between phases, particularly at the nanoscale [5]. Some, such as nanocrystalline diamond (with thin amorphous regions at the grain boundaries), are fairly well described [1], but a more general understanding of diamond–amorphous carbon nanocomposite materials, which are predicted to have superb mechanical properties [6], is highly desirable. In order to investigate these mixed phases, it is crucial to understand the interaction between diamond surfaces and amorphous carbon (a-C).

We use Tight-Binding Molecular Dynamics (TBMD) simulations to study various a-C/diamond (a-C/D) interfaces. TBMD is a quantum mechanical method that allows for accurate calculations for cell sizes and simulation times that are much larger than those feasible with first-principles methods [7]. Our previous TBMD studies of pure a-C networks, over the whole range of possible densities, have resolved long-standing issues related to the structural, mechanical, electronic, and optical properties of these materials, directly connecting our theoretical results with experiment [8–10]. The amorphous–crystal interface in silicon has been studied with simulations based on a combination of empirical and tight-binding models [11]. In the case of carbon, where several bond hybridizations are available, these heterostructures are expected to have very interesting properties that depend both on the diamond surface and on the a-C overlayer. In the present work, TBMD simulations allow us to obtain the detailed atomic-scale picture of a-C/D interfaces and predict their structural and electronic properties. We find that for high density a-C there is no extended intermediate phase at the interface. For lower densities, higher concentrations of carbon atoms close to diamond surfaces, compared with the rest of the a-C network, are observed. In all cases, a-C atoms are covalently bonded to

* Corresponding author.

E-mail address: kopidaki@materials.uoc.gr (G. Kopidakis).

¹ Present address: Physics Department, Harvard University, 17 Oxford Str., Cambridge 02138, USA.

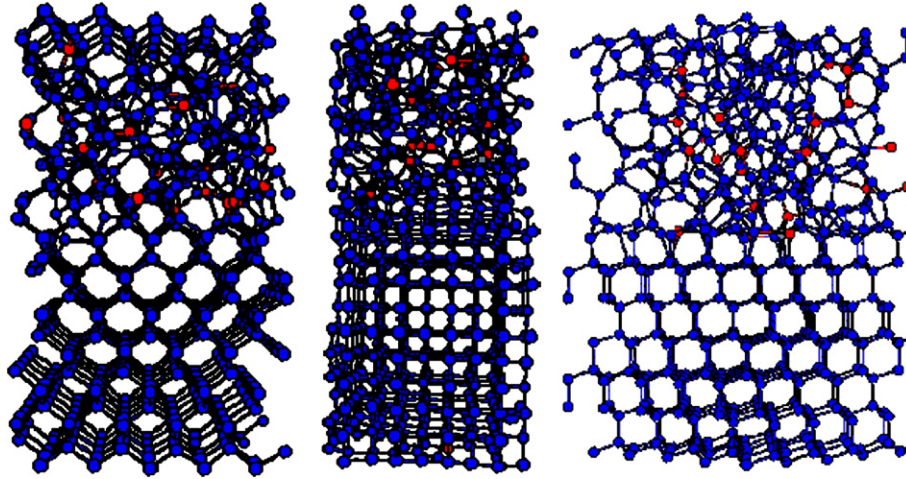


Fig. 1. Atomic structure of interfaces of high density a-C with low-index faces of diamond D(100) (left), D(110) (centre), D(111) (right). Blue atoms are 4-fold, red are 3-fold coordinated. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

diamond surfaces. Systematic calculations of interface energies between different faces of the crystal and a-C, and for several a-C densities and preparation conditions, determine the relative stability of various interfaces. We define a relevant energy per unit area for the a-C/D heterostructure and use it to predict the shape of faceted diamond crystals embedded in a-C. Our results are confirmed by empirical Monte Carlo simulations of nanocrystals in a-C matrix. The electronic properties of these same element heterostructures and possibilities to tailor them are also briefly discussed.

2. Results and discussion

The a-C/D interfaces are obtained by melting part of our initial diamond simulation cells to liquid and quenching it down to zero temperature, while keeping the rest of the atoms frozen in

their crystal positions. The as-quenched structures are then relaxed by allowing all atoms to move and reach the minimum energy configuration. Periodic boundary conditions are used in all directions, so that the supercell is a layered structure with alternating amorphous and crystalline parts. In order to study the effects of different preparation conditions, we used several quenching rates, a-C densities, amorphous and crystal sizes, and volume relaxation methods. Molecular dynamics simulations were performed using the Environment-Dependent Tight-Binding model (EDTB) of Tang et al. [12]. Typical simulation cells used in our TBMD consist of hundreds of atoms and the time step used is 1.05 fs. The results presented here are from samples obtained by quenching from temperatures of 12,000 K down to zero with quench time of 13.125 ps and 26.25 ps, corresponding to rates of about 914 K/ps (“fast”) and 457 K/ps (“slow”), respectively.

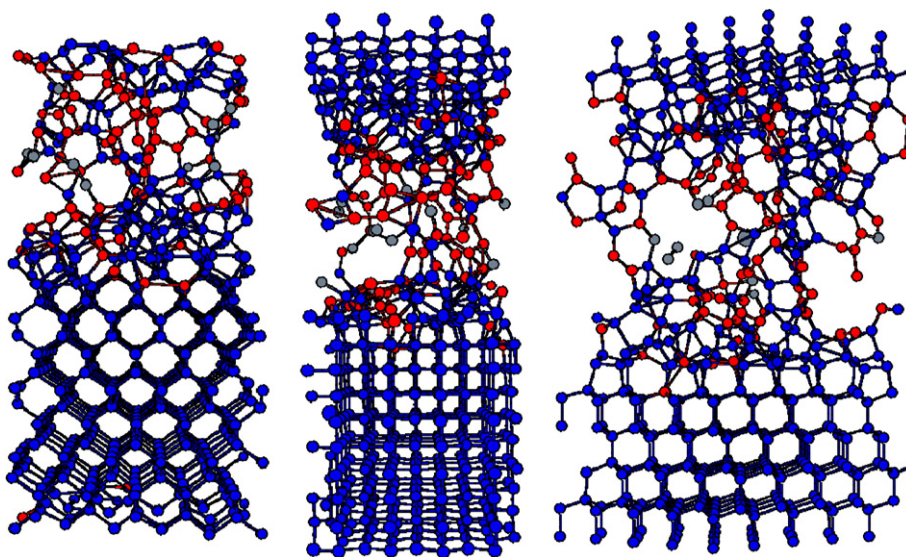


Fig. 2. Atomic structure of interfaces of low density a-C with low-index faces of diamond D(100) (left), D(110) (centre), D(111) (right). Blue atoms are 4-fold, red are 3-fold coordinated. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Download English Version:

<https://daneshyari.com/en/article/703636>

Download Persian Version:

<https://daneshyari.com/article/703636>

[Daneshyari.com](https://daneshyari.com)