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Surface Science 585 (2005) 17–24
SURFACE SCIENCE
www.elsevier.com/locate/susc

Surface energy, stress and structure of well-relaxed amorphous silicon: A combination approach of ab initio and classical molecular dynamics

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Received 27 July 2004; accepted for publication 31 March 2005
Available online 18 April 2005

Abstract

In this study, a combined method of a classical molecular dynamics calculation with the Tersoff potential and an ab initio calculation based on density functional theory has been performed. This combination method can provide quantitative evaluation of the surface energy and surface stress of well-relaxed amorphous silicon in addition to its structure. Using this method, a surface energy of 1.05 ± 0.14 J/m^2 and a surface stress of 1.5 ± 1.2 N/m are obtained. This calculation also leads to a new discovery of the microscopic characteristic of a-Si surfaces, which is not revealed through the use of an empirical potential. It is shown that there are two types of threefold coordinated atoms at the surface region; one with p^3-like bonding and the other with sp^2-like bonding. In addition, the investigation indicates that the microstructures of these defects are different from those of a threefold coordinated atom observed in the bulk structure.
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Keywords: Molecular dynamics; Density functional calculations; Amorphous surfaces; Silicon; Surface energy; Surface stress; Surface relaxation and reconstruction

1. Introduction

The surface properties of amorphous silicon (a-Si) have attracted much attention in recent years. Particularly, the structural properties of surfaces at an atomistic level are of great interest in gaining a better understanding of the growth mechanism during the deposition process of thin films [1]. Despite a-Si being one of the most fundamental thin film materials, little is known about the microscopic details of a-Si surfaces.

Other properties of particular interest are physical properties, such as surface energy and surface

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doi:10.1016/j.susc.2005.03.061

stress. More recently, Floro et al. [2] have pointed out that these two quantities play a key role in the theoretical prediction of film stresses. However, the present authors note that there is still a lack of experimental data regarding the surface energy and surface stress of a-Si.

At present, the most sophisticated technique available to evaluate these properties is an ab initio approach, which is a parameter-free quantum–mechanical calculation. This approach has successfully reproduced a (1 1 1) 7×7 dimer-adatom-stacking-fault (DAS) structure [3], a (1 1 1) 2×1 π-bonded structure [4] and a (1 0 0) 2×1 asymmetric dimer structure [5] confirmed by experimental observation [3,6]. However, creating an a-Si surface model that provides results comparable to experimental results is still a matter of debate. For example, using an ab initio molecular dynamics approach, Stich et al. [7] directly created the bulk a-Si through the rapid quenching of liquid silicon, followed by a thermal annealing within the range of a few picoseconds. The resulting structure involved a high bond-angle deviation, compared with the experimental value. The origin of this disagreement arises from the insufficient thermal annealing for structural relaxation due to its heavy computational burden. One possible way to resolve this time-scale problem is to employ a classical molecular dynamics (CMD) approach. As demonstrated in previous works, a CMD method can realize nano-second simulation, which is sufficient to completely relax the amorphous bulk [8,9] and surface [10] structure.

The approach taken here is to use a combination method of a CMD calculation and an ab initio calculation. The main role of the CMD method is the full relaxation of a-Si. Subsequently the results of the CMD calculations are used as starting points for ab-initio calculations based on density functional theory (DFT) [11]. In the present work, this combination method gives quantitative values of the surface energy and surface stress of well-relaxed a-Si. In addition, the structural analyses reveal the remarkable difference in surface microstructure between the model obtained from the classical simulations and that from the simulations based on quantum mechanics.

2. Computational methods

2.1. Combination method

The first step is to prepare well-relaxed amorphous structures by a CMD method. The Tersoff potential [12] was chosen to compute the interatomic forces among silicon atoms. In general, the Tersoff potential can reproduce the structural and physical properties regarding bulk crystal phase, bulk amorphous phase and the surfaces of both phases [8,13,14].

In the next step, DFT calculations are carried out to relax the atomic geometries obtained from CMD simulations. The present calculations are performed using Vienna ab initio simulation package (VASP) [15,16] based on pseudopotential and plane-wave techniques. The ultra-soft Vanderbilt pseudopotential [17] was employed to describe the interaction between ions and electrons. The approximation associated with the exchange-correlation energy was the generalized-gradient approximation (GGA), suggested by Perdew and Wang [18]. For the Brillouin-zone (BZ) integrations, various k-grids of Monkhorst–Pack special points [19] were used together with the Methfessel–Paxton technique [20]. In order to find the minimum-energy structure, the entire system is first allowed to dynamically relax at a finite temperature for several picoseconds and is then cooled down to 0 K. In this dynamical scheme, only the Γ-point for BZ integration was used and the plane-wave cut-off was set to 150 eV. After quenching, we perform the structural optimization using the DFT calculation involving the more k-points and larger value of cut-off energy than those in the dynamical calculations. These detailed conditions would be mentioned in the next section.

2.2. Calculation of surface energy and surface stress

The surface energy and surface stress are defined in the same manner suggested in Ref. [21]. It should be noted that, because the size of the systems that can be handled with the ab initio scheme is considerably limited, the spatial-related scattering of these two quantities cannot be neglected. Therefore, in the present work, we performed a to-

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