Surface Science 632 (2015) 93–97

Contents lists available at ScienceDirect

Surface Science

journal homepage: www.elsevier.com/locate/susc

A DFT study on the effect of surface termination in CdTe (111)/α-Al_2O_3 (0001) heteroepitaxy

Kristoffer Meinander *, John S. Preston

Department of Engineering Physics, McMaster University, Hamilton, ON L8S 4L7, Canada

ARTICLE INFO

Article history:
Received 6 September 2014
Accepted 15 September 2014
Available online 22 September 2014

Keywords:
Density functional theory
Heteroepitaxy
Sapphire
Cadmium telluride

ABSTRACT

The heteroepitaxial growth of thin films on complex oxide substrates is highly dependent on chemical interactions in the interfacial material layers. We use first-principle density functional theory calculations to investigate the effect of alterations in the substrate termination in CdTe heteroepitaxy on c-plane sapphire, i. e., α-Al_2O_3 (0001). Comparing to previous experimental results, we rationalize the observations that an artificial alteration of the surface termination, through the addition of aluminum on the substrate surface, will affect rotational domain alignment. Our findings show that Al adatoms will occupy sites on the sapphire surface that otherwise would be beneficial for the binding of CdTe dimers, effectively altering the orientation of dimers during the initial growth process.

© 2014 Elsevier B.V. All rights reserved.

1. Introduction

The heteroepitaxial growth of thin films has become increasingly important as a step in the fabrication of many technologically important solid-state electronic and photonic devices [1]. As a deeper understanding of the intricacies of lattice matching evolves, the possibilities for substrate/thin film combinations that both have been, and potentially can be, used for the templated growth of high-quality materials increase exponentially. Recent efforts in expanding these heteroepitaxial horizons have been focused on understanding the geometrical constraints involved in the alignment of two structurally dissimilar materials, encompassing material pairs that are dissimilar in either translational [2] or rotational symmetries [3].

Where translational symmetry dissimilarities, when disparities between the surface lattice constants of the substrate and thin film occur, can be accommodated by a fairly straight-forward tilting of one material with respect to the other [2,4], differences in rotational symmetry of the materials are often accompanied by the growth of several domains within the thin films [3]. This necessarily introduces large-angle grain boundaries within the films, and can be extremely detrimental for applications where such defects affect several aspects of device performance, including carrier diffusion lengths, mobility, and lifetime. From a purely geometrical standpoint, mismatches in rotational symmetry will always result in the growth of two or more domains, if the rotational symmetry of the substrate is greater than that of the thin film material [3]. For the case of many complex substrates, including α-Al_2O_3 (0001), i.e. c-plane sapphire, and several other commonly used metal oxides, minute differences in local surface chemistry will possibly have a beneficial effect, by greatly increasing or reducing the likelihood of one domain alignment, in effect limiting the number of acceptable growth domains. A clearer understanding of the interfacial chemistry between heteroepitaxial materials will therefore be of assistance for the future design of high-quality thin film growth.

We have previously shown that the heteroepitaxial growth of CdTe thin films, through pulsed laser deposition, on α-Al_2O_3 (0001) produces high-quality thin films of a single predominant growth domain [5]. Further studies have also indicated that the specific termination of the sapphire substrate plays a huge role in the rotational alignment of the growth domain, as well as the quality of the thin film [6]. Sapphire is a complex metal oxide, for which the most stable surface termination, when truncated in the [0001] direction, has been shown to be a Al-O_3 layer [7–11]. A ball-model of this structure is shown from the side in Fig. 1a). Fig. 1b) shows a ball-model of the (0001) surface that results from this truncation, together with its rhombohedral surface unit cell in black lines. The top-most Al atoms are labeled in Fig. 1a), together with arrows indicating their positions within the unit cell.

CdTe has a strong preference toward growing as a zinc blende structure in the [111] direction, the ball-model of which is shown in Fig. 1c), together with one layer of the material in its (111) plane in Fig. 1d). The surface unit cell of CdTe, marked with black lines, has a lattice constant of 4.582 Å, which compared to the lattice constant of c-plane sapphire, 4.759 Å, gives a mismatch of approximately 3.7%. A larger difference is apparent when examining rotational symmetries, which show a threefold symmetry for CdTe as compared to a sixfold symmetry for the sapphire surface. In a purely geometrical sense, this gives a theoretical possibility of two equal and opposite domain rotations [3,12,13], which has been seen for other similar systems [14,15].

---

* Corresponding author.
E-mail address: kristoffer.meinander@gmail.com (K. Meinander).

http://dx.doi.org/10.1016/j.susc.2014.09.013
0039-6028/© 2014 Elsevier B.V. All rights reserved.

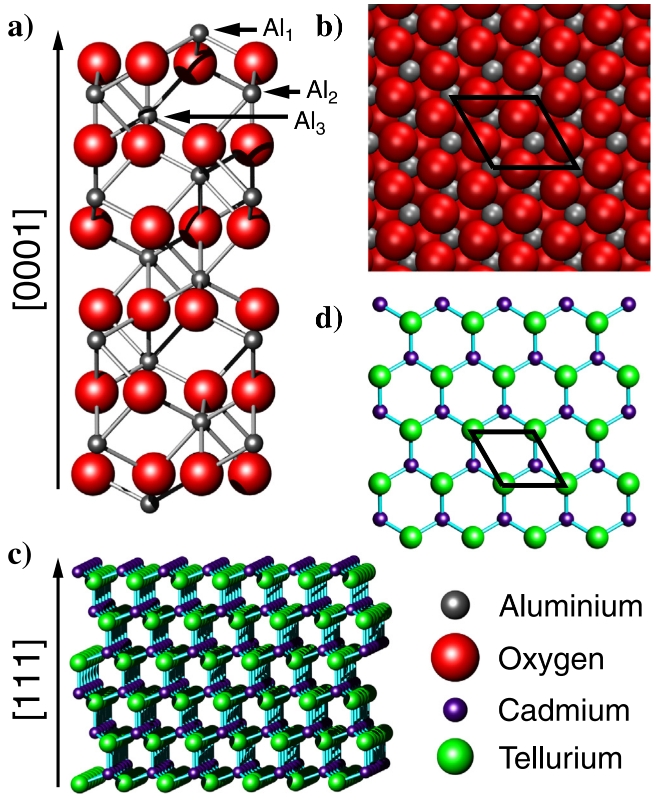

Fig. 1. a) A ball-model of the unit cell of α-Al_2O_3 shown from a side-view. The different positions of the three top-most Al atom layers are indicated with arrows. These all represent unique sites, as can be seen in b) which shows a top-view model of the (0001) surface. Al_2 occupies the corners of the unit cell, whereas Al_1 and Al_3 are in central positions within the surface unit cell. c) Side-view and d) top-view ball models of CdTe, truncated in the [111] direction are also shown, with the surface unit cell of CdTe (111) indicated in d). There is a relative mismatch of 3.7% between the two surface unit cells of sapphire and CdTe.

Because our experimental results indicate that only one domain is preferred over the other [5], local chemical interactions, both on the sapphire surface and in the interfacial layer toward the CdTe thin film, must necessarily be of great importance to the initial stages of film growth, dictating the rotational alignment of the entire thin film. Additional experiments also show that the deposition of a gradually increasing coverage of Al adatoms will effectively rotate this preferred alignment by 180°, until the coverage becomes too high and all heteroepitaxial relationship is lost. The purpose of this study is to use first-principle density functional theory (DFT) to explain these previous experimental results.

2. Computational methods

In order to understand how the rotational domains of CdTe are affected by surface termination, we calculated ground state potential energies for the adatoms of Cd, Te, and Al at different lateral positions over the (0001) surface of α-Al_2O_3. Using the `ABINIT` package [16,17], which employs a plane-wave basis set and periodic boundary conditions, total energies and forces where calculated within the generalized gradient approximation (GGA) of Perdew, Burke, and Ernzerhof (PBE) [18]. To describe the ionic cores, both ultrasoft pseudopotentials and projector-augmented-wave (PAW) potentials were tested, both of which gave the same structural results and similar ratios between relative energies. The plane-wave cutoff for all calculations was 400 eV, which resulted in a convergence of the total energy to within 10 meV. Initial relaxations of the sapphire substrate structure were done according to similar previous calculations [11]. Systematic k-point convergence was checked for smaller initial bulk and surface calculations, but for the large surface used in the thin film calculations the Γ-point was sufficient. For substrate surface calculations, a 2 × 2 surface unit cell with the thickness of one unit cell was used together with a vacuum layer of 20 Å, which was tested to be sufficient for a total isolation of the sapphire slab from its mirror images. This is a typical means of studying the adsorption of atoms on similar surfaces [19].

The main results of the DFT calculations were surface potential energy maps, showing the total potential energy of the system when the adatom of choice was fixed to specific lateral positions over the surface. Energies were calculated for every point on a uniform grid of 11 × 11 positions over the surface. Intermediate points were generated by interpolation from the calculated points of the surface matrix. Only the lateral coordinates of the adatom were fixed, while its position normal to the surface, as well as all other coordinates of other atoms, were relaxed until all force components for all atoms were below 0.03 eV/Å.

3. Results and discussion

A ball-model of the surface unit cell of α-Al_2O_3 (0001), together with the surface potential energy map of a single Al adatom in different positions over the surface, as calculated by DFT, can be seen in Fig. 2a) and b), respectively. The orientation of the potential energy map corresponds to that of the ball-model, from which it can be seen that an Al adatom preferentially binds to the Al_3 site. The color scheme in Fig. 2b) is such that darker blue colors represent lower total energy,

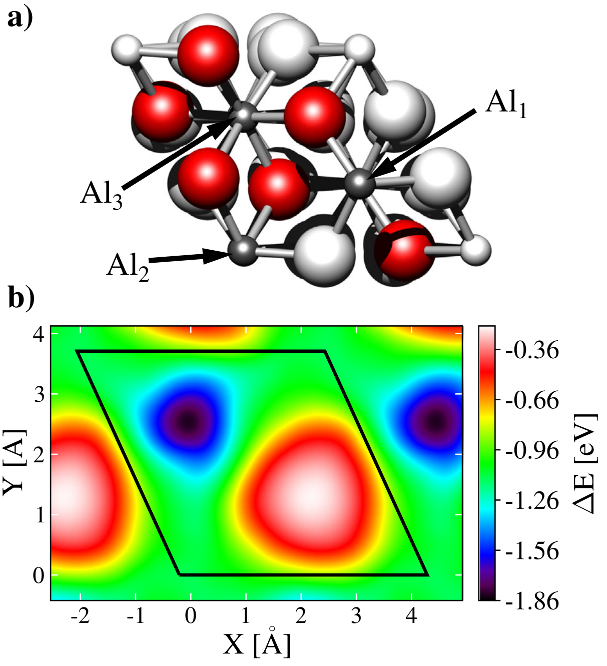

Fig. 2. a) A ball-model of the surface unit cell of α-Al_2O_3 (0001), where the positions of the top-most cations have been labeled and indicated with arrows. Only unique atomic sites are colored, leaving duplicate atoms as white. In b) the binding energy of a single Al adatom is shown as a function of lateral position over the sapphire surface unit cell, where the black rhombus represents the boundaries of the unit cell in a). The color scheme is such that the dark blue colors represent a high binding, with lower total potential energy of the system, and the lighter red colors represent a weaker binding. From this surface potential energy map it can be seen that Al adatoms preferentially bond to the Al_3 site on the sapphire surface, corresponding to a natural continuance of the sapphire crystal (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Download English Version:

https://daneshyari.com/en/article/5421993

Download Persian Version:

https://daneshyari.com/article/5421993

Daneshyari.com