Fundamental properties of GaN(0001) films grown directly on Gd_2O_3(0001) platforms: ab initio structural simulations

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I. ABSTRACT

We present first-principles calculations to study the heterojunction between a wurtzite GaN(0001) film and a hexagonal Gd_2O_3(0001) substrate. We report that the most favorable Gd_2O_3(0001) surface is O terminated. Using the work of adhesion of isolated GaN and Gd_2O_3 slabs, our calculated interface energies suggest that the graphiticlike GaN films are fully relaxed at Gd_2O_3(0001) platforms, thereby leading to Ga-polarity in the GaN(0001) epitaxial film. Our findings agree with previously reported results. Keywords: GaN; Gd_2O_3; interface; polarity; first-principles calculations E-mail address:liupo@ms5.hinet.net or pliu@dragon.nchu.edu.tw

II. INTRODUCTION

Research on epitaxial growth of wide band gap and high gate dielectric materials based on rare earth oxides (REOs) has introduced in the Si complementary metal-oxide-semiconductor (CMOS) and GaN MOS field-effect-transistors (MOSFETs) devices[1–6]. Attempts to replace alternative dielectrics with their thermodynamic stability in these devices motivated a significant effort to develop REO thin films of the gadolinium (III) oxide (Gd_2O_3)[1,2,4–6]. Recently, the lattice parameters of a cubic bixbyite crystal gadolinium sesquioxide (α- Gd_2O_3) layer have been demonstrated to be comparable to those of its suitable template of Si(111), where the epitaxially strained α- Gd_2O_3 films impose a tensile strain (0.1%) in the in-plane direction and compressive strain (0.46%) in the out-of-plane direction with a Gd_2O_3 [$1\bar{1}0$] // Si[$\bar{1}10$] and Gd_2O_3 (111) // Si(111) epitaxial relationship, respectively[1]. Alternatives to silicon substrates, α- Gd_2O_3 layers have been grown on GaAs(100) and this represents that the epitaxially strained α- Gd_2O_3 films impose a large tensile strain (3.9%) in the in-plane direction and compressive strain (1.9%) in the out-of-plane direction with a Gd_2O_3 [$\bar{1}\bar{1}0$] // GaAs[$01\bar{1}$] and $Gd_2O_3(1\bar{1}0)$ // GaAs(100) epitaxial relationship, respectively[2]. Moreover, the 6H-SiC(0001) substrates are used to initially produce the structurally similar Gd_2O_3 islands with the hexagonal crystalline structure via molecular beam epitaxy (MBE) growths, which subsequently transform into flat films in a mixture of [111]-oriented cubic bixbyite and monoclinic structure with the superior dielectric constant of ε= 22 and leakage current of 10^{-8} $\frac{A}{cm2}$@1V[3]. Furthermore, alternate approaches involving epitaxial growth on GaN-on-sapphire substrates that can fabricate a gate dielectric for inversion-channel devices have been developed and then used to achieve the integration of MBE-grown high-temperature monoclinic Gd_2O_3 films on GaN-on-sapphire substrates at a substrate temperature of 700°C[4]. In contrast, thin films of hexagonal Gd_2O_3 (h- Gd_2O_3) films on GaN-on-sapphire substrates at lower substrate temperatures (550°C) have been shown to exhibit excellent structural quality in spite the significant mismatch (20between the two materials along the in-plane [100] direction [in-plane distances in h- Gd_2O_3 (0001) and GaN(0001) films are 3.86 and 3.189, respectively][6]. In this regard the overgrowth of GaN(0001) films shows the ability to relieve stress when grown on the stiffer h- Gd_2O_3 (0001), i.e., GaN(0001)/h-Gd_2O_3(0001)/GaN(0001)/Al_2O_3 (0001). Note that the h-Gd_2O_3 films is a high-temperature phase that exists at temperature over 2473°C and estimates the dielectric constant yielding high ε 24 reducing the capacitive effective thickness (CET) down to 0.5 nm[5]. A preliminary experimental account of the structure and unique properties in the GaN(0001)/h- Gd_2O_3 (0001) system was described in Refs. 4-6 but fundamental bonding, and electronic and energetic properties near the heterojunction of the GaN(0001)/h- Gd_2O_3 (0001) were not presented. In this paper, we report the optimized GaN(0001)/h-Gd_2O_3 (0001) heterostructure, which provides its thermodynamic and electronic properties of the interface structure and elucidates the most stable bonding configuration that enables strain relaxation.

III. COMPUTATIONAL METHODOLOGY

We performed a series of ab initio calculations based on the density functional theory (DFT) to elucidate the bonding, and electronic and energetic properties of various GaN(0001)/h-Gd_2O_3(0001) heterostructures. The ground state energy calculations of bulk and interface unit cells were all carried out using the Vienna Ab Initio Simulation Package (VASP)[7–9]. We employed the Vanderbilt ultrasoft pseudopotentials (USPPs) to efficiently treat ion-electron interactions. Here USPPs was derived from the projector augmented wave (PAW) method and the generalized gradient approximation (GGA) with the Perdew-Wang (PW91) exchange-correlation functional[10–12]. The electronic configurations for the valence electrons are N: $2s^22p^3$, O:$2s^22p^4$,Ga: $4s^24p^1$,and Gd: 4f $s^75d^16s^1$. An approximate formulation of the interface energy was described in Refs. 13 and 14 and is a general function of the gallium for GaN, nitrogen,

gadolinium for h-Gd_2O_3, and oxygen chemical potentials (i.e., μ_{Ga},μ_N,μ_{Gd}, andμ_O).

IV. INTERFACE STRUCTURES

Our approach for understanding the stability of the GaN/h- Gd_2O_3 heterostructures is based on the previous observation of GaN(0001) // h-Gd_2O_3 (0001) and GaN[10 0] // h-Gd2O3[10 0] [6].Accordingly, we focus on four representative structures: (i) Ga-polar GaN is grown on O-terminated Gd2O3 represented by the model 1, (ii) Ga-polar GaN is grown on Gd-terminated Gd_2O_3 represented by the model 2, (iii) N-polar GaN is grown on O-terminated Gd_2O_3 represented by the model 3, and (iv) N-polar GaN is grown on Gd-terminated Gd_2O_3 represented by the model 4. The basal dimensions of the all supercells were fixed at a = 3.726 $\mathring{A}$ and b = 6.454 $\mathring{A}$. The dimension perpendicular to the interface is approximately 70 $\mathring{A}$ and is chosen to minimize coupling between their free surfaces. The stoichiometry of all slabs was fixed at 8 f.u. of GaN and 4 f.u. of h-Gd_2O_3. Atom positions in each of the structural models are fully relaxed with a plane wave cutoff of 500 eV and a 1×6×1MonkhorstPack grid consisting of 9 irreducible k-point for reciprocal space integrations, yielding the total slab energy $E_{slab}^{GaN/Gd2O3}$ and the optimized structures shown in the four models depicted in Figure 1. The most energetically favorable interface of the model 1 leads to Ga-polar GaN(0001). We find that the interface is consistently sharp and highly epitaxial, indicating that the remarkable planarity of the GaN film is largely relaxed by the transformation into the graphiticlike structure to remove the dipole, which is close to the ideal graphitic structure. In particular, wurtzite GaN nanofilms can be readily stabilized by eliminating sp^3 tetrahedral toward sp^2 trigonal planar coordination, which are commonly observed in previous studies on the GaN(0001)/Sc_2O_3 (111) heterostructure and the stabilization of the polar surface atoms of wurtzite materials[15,16]

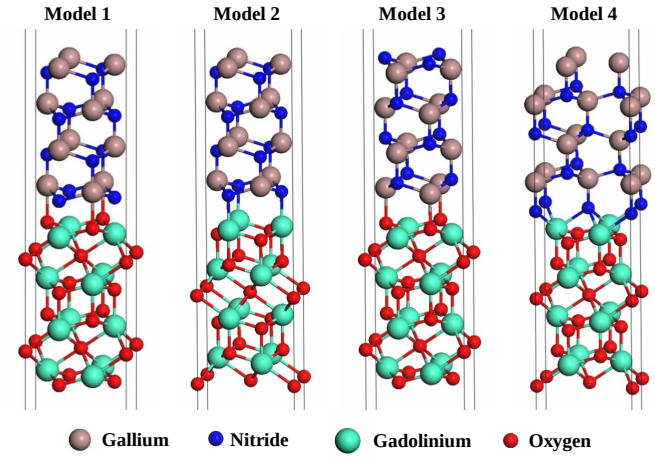

FIG. 1. Atomistic representations of the four GaN/Gd_2O_3 interface models with a 1× $\sqrt{3}$ basal dimension in the hexagonal lattice structure. Models 1 and 2 have Ga-polar GaN surfaces, while models 3 and 4 have N-polar GaN surfaces. The atoms are represented by spheres: Ga (brown), N (blue), Gd (green), and O (red). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article).

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