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Structure and stability of threading edge and screw dislocations in bulk GaN

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ABSTRACT

Computational studies of a-type threading edge dislocations parallel to the [0001] direction in bulk GaN reveal three possible structures of their cores that are characterized as the 4-atom ring, the 8-atom ring and the 5/7-atom ring configurations. Similar calculations made on the c-type threading screw dislocation suggest the existence of a full core, although the precise structure is still a matter of debate. Most studies associate the position of the dislocation with only high-symmetry points in the lattice and do not discuss the core stability in non-equiatomic configurations. Here, we investigate how the core structures of a-type edge and c-type screw threading dislocations depend on the position in the (0001) plane at which the dislocation is inserted perpendicular to this plane. Furthermore, by successively removing the high-energy atoms from the cores of these dislocations, we track the stability of each core in non-equiatomic configurations and identify the core structures of minimum energy. The edge dislocation is shown to prefer the 5/7-atom ring structure, while the core of the screw dislocation transforms into a new Ga-rich configuration with the associated energy drop of nearly 0.4 eV per one Angström of the dislocation length. For all types of dislocation cores found in this paper, we employ isotropic linear elasticity to estimate their core radii and core energies.

1. Introduction

III-nitrides are important semiconductor materials that can be used to manufacture active optoelectronic devices emitting in the full spectral range from infrared to ultraviolet [1,2]. Under ambient conditions, they crystallize in the close-packed wurtzite structure that belongs to the space group $P6_3mc$ (No. 186). Due to the lack of cheap lattice-matched substrates, they are mostly grown on sapphire (Al_2O_3), although there has been continued interest to replace the substrates by Si wafers [3–7]. During the epitaxial growth of polar ⟨0001⟩-oriented GaN films, a large density of threading dislocations (TDs) appear with line directions perpendicular to the (0001) interface. These are the a-type $1/3[11\bar{2}0]$ edge, c-type [0001] screw, and the $(a+c)$-type $1/3[11\bar{2}3]$ mixed dislocations of which the edge dislocations constitute the largest and the screw dislocations the smallest fraction [8]. The threading edge dislocations are known to contribute only negligibly to relieving the lattice mismatch between the GaN film and the substrate [9]. At the same time, their energies scale linearly with the thickness of the film and thus one would expect that the film of sufficient thickness has to be free of threading dislocations. However, electron microscopy observations made on epitaxially grown films show the opposite. The GaN films are full of threading dislocations that propagate through the film and reduce the luminescence of quantum wells grown on the top [10,11].

It was known for a long time that dislocations are responsible for scattering electrons [12,13]. In epitaxially grown III-nitrides, the electrically active TDs act as sources of nonradiative recombination of charge carriers (excitons), i.e. electrons in the conduction band and holes in the valence band [14]. In particular, edge dislocations act as electron acceptors and thus their cores become negatively charged [15]. Similarly, screw TDs have been correlated with excess reverse-bias leakage currents in epitaxially grown GaN films [16]. In most optically active semiconductors, the density of TDs around 10^3 cm^{-2} is capable of completely quenching the luminescence of the device [10,17]. This is, however, not the case for III-nitrides which retain reasonable internal quantum efficiency up to very large densities of TDs, typically 10^7–10^{11} cm^{-2} [15,18].

The experimental efforts made in the past two decades contributed greatly to optimizing the growth of wurtzite III-nitrides on lattice-mismatched substrates, with the densities of TDs reduced down to 10^7 cm^{-2} when the nucleation layer is used [19]. On the

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theoretical side, a number of first-principles studies have been made to identify the equilibrium core structures of all types of TDs [20–23], which were reviewed in Ref. [15]. In the first principles calculations, the initial core structures are often constructed so as to match the positions of image peaks obtained from HREM measurements [24], image simulations or the core structures suggested by simplified theoretical models [25,26]. In these calculations, the limited number of atoms in the periodic block leads to the confinement of the dislocation core into a small spatial region. Various approaches have been developed to mimic the bulk environment of the dislocation, the most common of which are periodic boundary conditions in the (0001) plane and using a cluster configuration in which the dangling bonds on the surface of the cluster are passivated by hydrogens [23,27]. Large-scale computer modeling of these TDs is possible by employing empirical potentials, where the simulated block can be made effectively infinite by a careful choice of the boundary conditions in the plane perpendicular to the dislocation line. These studies were made in the past for all three basic types of TDs [28–30] using the Stillinger–Weber potential [31]. By considering only the Ga–N bonds, Béré and Serra [28] identified three equilibrium configurations of the $1/3[11\bar{2}0]$ edge dislocation in the bulk, which are designated as the 4-atom ring, 8-atom ring and 5/7-atom ring core configurations, the last of which had the lowest formation energy. Previous Z-contrast imaging of edge dislocations in free-standing foils of GaN show the existence of the 8-atom ring core configuration [24,32], whereas their recent optical sectioning in HAADF-STEM reveals also the 5/7-atom ring core configuration [33]. The core structures of [0001] threading screw dislocations were investigated by Belabbas et al. [30]. Besides the previously found full-core configuration [23], they identified a new configuration of lower energy that is obtained by inserting the dislocation at the boundary between two hexagonal lattice sites in the (0001) plane. However, in all these calculations, the dislocations were inserted only at a few special positions in the (0001) plane, which may not provide the full picture of core stability in wurtzite GaN.

The objective of this paper is to identify all equilibrium core structures that the threading a-type edge and c-type screw dislocations adopt in the bulk (both parallel to the [0001] direction) and investigate their stability in configurations with Ga and/or N vacancies. The latter is accomplished by successively removing high-energy core atoms and relaxing all remaining atoms in the simulated block. It is expected that the system reduces its energy by removing a small number of these atoms but the energy will eventually rise above the initial configuration due to the cost associated with internal surfaces. This is mainly important for the screw dislocation, whose large Burgers vector may favor such a core transformation [26]. For completeness, we estimate core radii and core energies of all cores types of threading edge and screw dislocations found in this paper.

2. Details of atomistic simulations

All atomistic simulations in this paper were made using the LAMMPS code [34] with the atomic interactions described by the Tersoff–Brenner potential for GaN developed by Nord et al. [35]. Unlike the previously used Stillinger–Weber potential that characterizes the Ga–Ga and N–N bonds indirectly by fitting a data set obtained mostly from crystal defects, the Tersoff–Brenner potential in Ref. [35] is constructed by taking into account the properties of pure constituents such as α-Ga and N_2. Including these details in the parameterization of the potential leads to correct description of bonding close to extended defects, where the atomic environments are typically very different from that of the reference wurtzite lattice. It should be emphasized that this potential does not include the energies of long-range electrostatic interactions of ions. To some extent, these are accounted for in the parameterization of the reference wurtzite structure, but they are not adjusted when the atomic environments deviate from the tetrahedral coordination. Consequently, the energies of wurtzite and sphalerite (zinc-blende) structures are the same, although the former should be slightly lower ($\Delta E_{w-zb} = -9.88$ meV/atom [36]). While this difference may play role in molecular dynamics studies, it is unlikely to affect molecular statics simulations of extended defects.

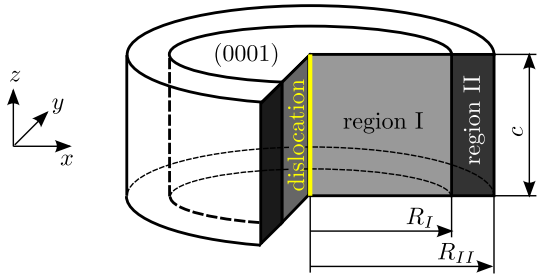

Fig. 1. A cylindrical simulation block used for the calculations in this paper. Periodic boundary conditions are applied along the z direction to study straight threading dislocations in the bulk.

All calculations to follow were made using the cylindrical simulation block shown in Fig. 1. Periodic boundary conditions were applied along the z direction to study properties of straight edge/screw dislocations. The thickness of the block in the z direction was equal to c ($= 1.633a$), where $a = 3.181$ Å is the equilibrium basal-plane lattice parameter. The x axis of the block was parallel to the $[11\bar{2}0]$ direction, the y axis parallel to the $[\bar{1}100]$ direction, and the z axis parallel to the [0001] direction. The dislocation line was parallel to the z direction and intersected the perpendicular (x, y) plane at the desired dislocation position (more on the choice of this position in the following two sections). This was accomplished by displacing all atoms by the anisotropic linear-elastic strain field of the dislocation [37,38]. The atoms in the region I of the block were then relaxed using conjugate gradients, while keeping those in the region II fixed by zeroing all components of forces acting on these atoms. The radius of the region I was $R_I = 15a$ and the radius R_{II} was adjusted so that the interaction radii of the atoms in the region I did not extend to the outer surface of the block ($R_{II} = 18a$). The atoms in the region II thus make the simulated block extend effectively to infinity in the (x, y) plane. In all calculations made in this paper, all dislocation cores were contained fully within the region I of the block and thus the boundary conditions imposed through the region II have no influence on the dislocation core structures.

3. Threading $1/3[11\bar{2}0]$ edge dislocation

When the wurtzite crystal is viewed along the [0001] direction, it is formed by atomic columns arranged in a regular hexagonal array, one of which is shown in Fig. 2(a). The interior and edges of this hexagon represent all possible positions at which the dislocation can be inserted parallel to the [0001] direction (and thus perpendicular to the plane of the figure). The corners plotted by shaded spheres in Fig. 2(a) represent the [0001] columns of atoms. In principle, inserting the dislocation at different positions within this hexagon may provide a range of dislocation cores, each corresponding to a different local minimum of energy [25]. These are identified here by inserting the dislocation successively at all positions of a regular grid within this hexagon and relaxing all atoms in the region I of the block under periodic boundary conditions applied along the z direction. During this relaxation, the atoms in