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Crystalline structures and defects in liquid GaN during rapid cooling processes

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ABSTRACT

The formation of crystalline structures and defects in GaN at three different cooling rates were investigated based on molecular dynamics simulations by using the Stillinger–Weber potential. The structural properties of GaN during the solidification processes were analyzed by several structural analysis methods. The system formed crystals at 200 K when cooled at rates of 10^{11} K/s and 10^{12} K/s, whereas the amorphous state that formed at the cooling rate of 10^{13} K/s included some crystal clusters. The ⟨4 0 0 0⟩ and ⟨2 3 0 0⟩ Voronoi polyhedrons were dominant in GaN during the rapid quenching processes. The ⟨4 0 0 0⟩ polyhedrons can aggregate to form cubic and hexagonal phases in GaN at lower temperatures. After solidification, cubic and hexagonal structures were coexistent in the system with a hybrid disordered region and crystal defects. When the temperature was decreased at a rate of 10^{11} K/s, coherent twin boundaries, having excellent structural stability and configural continuity, were easily formed between the cubic and hexagonal crystal structures.

1. Introduction

Research and application of GaN materials are popular topics at the frontier of present global semiconductor research. GaN is an important semiconductor material for developing microelectronic and optoelectronic devices. GaN has a wide direct band-gap, strong bonding features, high thermal conductivity, good chemical stability, and strong anti-irradiation ability, and is thus widely applied in optoelectronics [1], high-temperature and high-power devices [2], and high-frequency microwave devices [3,4], and has shown considerable potential for electroluminescence applications [5].

Researchers have elucidated many details concerning the physical and chemical properties of solid GaN [6]. Computer simulation is an efficient tool that has been applied to the study of microstructures to promote a deeper understanding of the physical and chemical properties of GaN. Several potential models for describing interatomic interactions in GaN have been presented [7–10]. By employing suitable potentials, computer simulations can be used to predict the structural and morphological properties of GaN clusters at the atomic level [11,12]. In contrast, the experimental methods generally describe the structures beyond the short-range order. The nanostructures and crystallinity of GaN can be probed by several characterization methods such as X-ray diffraction and transmission electron microscopy [13].

The thermal conductivity of GaN, which is known to depend on the crystal quality, has been investigated by molecular dynamics simulation [14] and by employing the Monte Carlo approach [15]. However, crystal defects generally reduce the thermal conductivity [16], influence the mechanical behavior [17] and plasticity [18], and also act as recombination centers in multicrystalline materials [19]. Therefore, research on the structural properties of point defects [20] and dislocation cores [21] in such systems is important to understand the mechanisms of defect formations to improve the crystal quality.

However, quantitative analysis of the local ordered structures in GaN is still limited, and structural information on point defects and dislocations is required to develop high-quality materials in the future. In this study, molecular dynamics simulation is performed to evaluate the crystal structures and defects of GaN during rapid solidification at three cooling rates using the Stillinger–Weber (SW) potential [22]. This is a suitable potential for simulating the liquid and solid-state structures in tetrahedral semiconductors such as Si, Ge, and GaN. The generation of crystalline structures in the system is investigated in detail based on the radial distribution function (RDF) *g(r)*. Quantitative analysis is performed to investigate the local ordered units that form the crystal structures by employing the Voronoi polyhedron index [23,24]. Visualization technology is used herein to trace the evolution of crystal defects.

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2. Simulation conditions and methods

In this study, the formation processes of crystalline structures in GaN were simulated via molecular dynamics techniques using the S-W potential. Molecular dynamics (MD) simulation was carried out in a cubic box containing 8000 atoms (4000 Ga atoms and 4000 N atoms) under periodic boundary conditions. The simulations were performed in the NPT ensemble with zero pressure and the time step was 1.0 fs. The initial configuration was equilibrated at 300 K with 200,000 time steps, after which several successive equilibration calculations were carried out by increasing the temperature at intervals of 200 K; this increment was decreased to 50 K when the temperature approached the melting temperature. Bulk GaN melts at 4150 K in the simulation. The system was melted completely at 4600 K and the simulation was run for 100 ps to guarantee an equilibrium liquid state, as judged by the total energy and the microstructures. Subsequently, the temperature was gradually decreased to 200 K at cooling rates of 10^{11} K/s, 10^{12} K/s, and 10^{13} K/s. The atomic configurations were recorded at intervals of 50 K during the quenching process. Finally, the RDF, Voronoi polyhedron number, and the visualization technology were applied to indicate the crystal structures and the defects in GaN.

3. Results and discussion

3.1. Radial distribution function (RDF)

The RDF describes the distribution of the distance between two atoms in a system, and is widely used to reveal the structural characteristics of liquid and amorphous structures. The peak value of the *g(r)* indicates the probability of finding a particle at a distance *r*, away from a given reference particle. To consider the general trends in the microstructural changes during the solidification of liquid GaN in the overall process, the *g(r)*s at various temperatures are presented in Fig. 1. As shown in Fig. 1, the intensity of the first peak gradually increased and the peak becomes sharper with a decrease in the temperature. The position of the first peak shifted to a lower value between 4600 K and 2500 K; at 2500 K, the first peak approached 1.97 Å, and the position remained unchanged below 2500 K. The trough of *g(r)*, which is situated between the first and the second peak, became resonant with decreasing temperature.

Significant changes in the second peak occurred between 3050 K and 3000 K when the system was cooled at a rate of 10^{11} K/s, as seen in Fig. 1(a), and between 2950 K and 2900 K for a cooling rate of 10^{12} K/s (Fig. 1(b)). Finally, the second peak split entirely into two small peaks at 200 K. The *g(r)* trends indicate that crystalline structures were formed at 200 K at cooling rates of 10^{11} K/s and 10^{12} K/s. However, there was no fundamental change in the second peak and an amorphous state was formed at 200 K at a cooling rate of 10^{13} K/s, as seen in Fig. 1(c).

3.2. Voronoi polyhedron

The Voronoi polyhedron can clearly describe the structural relationship between a target atom and its neighboring atoms in amorphous structures, which is one of the methods most commonly used for the analysis of amorphous structures [25]. To obtain information on the arrangements of the atoms in GaN, the atomic configurations were analyzed using the Voronoi polyhedron with the nearest-neighbor atoms. The Voronoi polyhedrons in amorphous GaN usually produce thousands of results according to the vertical mid-planes between the central atom and all its neighboring atoms. The approximation approach that only considers the impact of the nearest-neighbor atoms significantly reduces the computation complexity and increases the observability.

The Voronoi polyhedrons consist of a series of facets that are bisection of lines between the object atoms and their neighboring atoms. This model utilizes four indices ⟨n3, n4, n5, n6⟩ to denote different types of structures, where n3, n4, n5, and n6 denote the trilateral, quadrilateral, pentagonal, and hexagonal numbers, respectively. For example, the simple cubic lattice and tetrahedral crystal can be expressed as ⟨0 6 0 0⟩ and ⟨4 0 0 0⟩, respectively. The common polyhedrons are presented in Fig. 2 (insets).

The Voronoi polyhedrons in GaN that dominate during the rapid quenching processes are ⟨4 0 0 0⟩ and ⟨2 3 0 0⟩. In contrast, the number of ⟨2 2 2 0⟩ and ⟨0 6 0 0⟩ polyhedrons constitutes only a small portion at higher temperatures and actually none at lower temperatures. As shown in Fig. 2, the evolution of the polyhedrons during the rapid quenching processes can easily be divided into two stages. From the comparison of different cooling rates, the same tendency was found for the number of ⟨4 0 0 0⟩ and ⟨2 3 0 0⟩ polyhedrons at higher temperatures. Moderate growth of these polyhedrons was maintained with decreasing temperature until an inflection point in the temperature emerged.

At a cooling rate of 10^{12} K/s, a significant change occurred between 3050 K and 3000 K, where the number of ⟨4 0 0 0⟩ polyhedron increased dramatically from 3764 to 7373, in contrast to the sharp drop in the number of ⟨2 3 0 0⟩ polyhedron. A similar change appeared between 2950 K and 2900 K at a cooling rate of 10^{13} K/s. With the continuous increase in the number of ⟨4 0 0 0⟩ polyhedron, the number of ⟨2 3 0 0⟩ polyhedron decreased to some extent and a few members were maintained to prevent crystallization. Based on the above results, it is clear that the cooling rate has an important impact on the number of ⟨4 0 0 0⟩ and ⟨2 3 0 0⟩ polyhedrons.

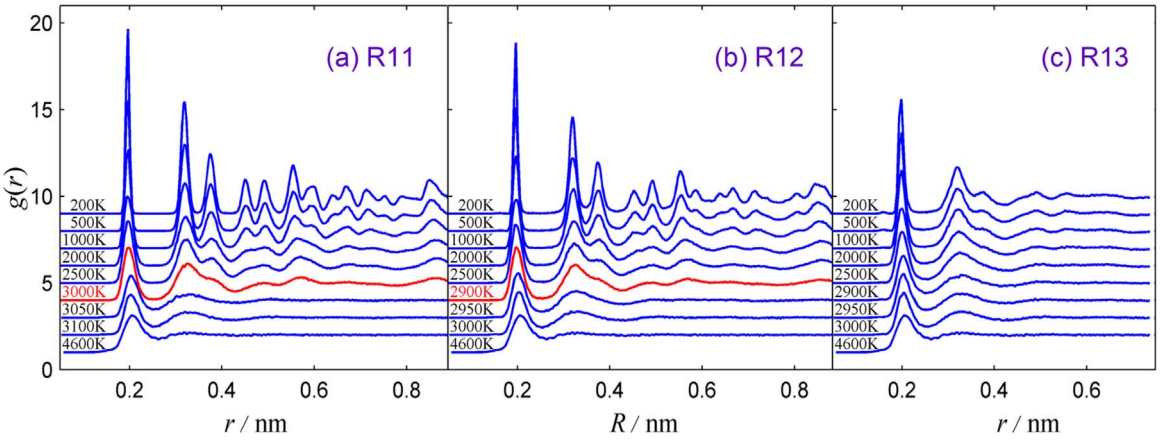

Fig. 1. Evolution of RDF at cooling rates of (a) 10^{11} K/s, (b) 10^{12} K/s, and (c) 10^{13} K/s.