Observation of the amorphous zinc oxide recrystalline process by molecular dynamics simulation

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The detailed structural variations of amorphous zinc oxide (ZnO) as well as wurtzite (B4) and zinc blende (B3) crystal structures during the temperature elevation process were observed by molecular dynamics simulation. The amorphous ZnO structure was first predicted through the simulated-annealing basin-hopping algorithm with the criterion to search for the least stable structure. The density and X-ray diffraction profiles of amorphous ZnO of the structure were in agreement with previous reports. The local structural transformation among different local structures and the recrystalline process of amorphous ZnO at higher temperatures are observed and can explain the structural transformation and recrystalline mechanism in a corresponding experiment [Bruncko *et al.*, Thin Solid Films **520**, 866-870 (2011)]. © 2013 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4789956>]

I. INTRODUCTION

ZnO material is a typical semiconductor in the 2B-6A group, a group with a wide direct band gap¹ and higher surface-to-volume ratios for potential use in different technologies, such as gas sensors in nano-size.^{2,3} Among semiconductor materials, ZnO is one transition metal which possesses unique physical properties and is very suitable for applications in electronic and optical devices.⁴⁻⁹ Recently, indium tin oxide (ITO),^{10,11} aluminum-doped zinc oxide (AZO:F),^{12,13} and fluorine-doped indium tin oxide (ITO:F) have been broadly applied in the semiconductor industry. Two type film structures are investigated, those of polycrystal¹⁴ and non-polycrystal films. Because it is easy for polycrystal film to break through repeated crimping,¹⁴ non-polycrystalline films have been developed to ameliorate this defect.¹⁵ In experimental studies, amorphous zinc oxide films have been demonstrated to have other good characteristics; for example, in optics, they can adsorb ultraviolet light effectively.¹⁶ In addition, amorphous zinc oxide film not only avoids heteromorphism but can also be synthesized at low temperatures.¹⁷ A variety of experimental studies have focused on the production process of single-crystal ZnO, but studies of amorphous ZnO are still lacking. However, amorphous semiconductor thin film possesses some advantages which allow the amorphous solid to replace crystalline material, according the k-selection rule.¹⁸ Table I lists the advantages and disadvantages of the amorphous and crystalline ZnO structures, taken from a number of references.¹⁹⁻²⁵ Crystal ZnO materials have low hardness because the ZnO film is grown on the a-axis (0002) perpendicular to the substrate, with the interface layer absorbing defects caused by large lattice mismatch. The amorphous thin film is

cheaper to produce than the crystalline form and can be manufactured at lower temperature, even at room temperature. Experimental research has described ZnO amorphous thin film manufacturing methods such as radio-frequency magnetron sputtering,²⁶ molecular beam epitaxy (MBE),²⁷ pulse laser deposit (PLD),²⁸ sol-gel,²⁹ metal organic chemical vapor deposition (MOCVD),³⁰ and the prefiring-final annealing method,³¹ all of which can use different substrate materials and controlling temperatures, among other conditions.

A variety of crystalline internal structures have been analyzed by XRD analysis, and amorphous ZnO thin films have been grown on different material substrates by different methods. The properties of amorphous ZnO films structures depend heavily on the growth environment, especially the substrate materials and temperature. References mention different crystalline surfaces grown which are observed in the quenching process of ZnO amorphous films, and conclude that the internal structure changes depend on temperature. This study focuses on the internal molecular structures of amorphous ZnO and uses a simple and efficient method to configure the model. In many studies, wurtzite (B4)³² and zinc blende (B3)³³ structures had been constructed for simulation and analyses of phase transfer in changing temperatures and pressures.³⁴ Due to the many applications which involve high pressures or temperatures for crystal materials, any discussion of the precise pressure-volume-temperature equation that can be provided by molecular dynamics is very important.³⁵

In this study, we employ the basin-hopping method with Buckingham potential to construct the ZnO structures and find the crystalline (B3 and B4) and amorphous structures at different temperatures by MD simulation. In addition, we presented thermal elevation process of ZnO, and the average bond length, the coordinated number (CN) distribution, and the change of volume of three varieties of ZnO structures were also studied at different temperatures.

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TABLE I. The advantages and disadvantages of crystal and amorphous ZnO materials.

Crystal	Advantage:
	1. The visible emission in the RT PL spectra was drastically suppressed without sacrificing the band-edge emission intensity in the ultraviolet region, so they have potential applications as ultraviolet light emitters.¹⁹ 2. Good thermal stability.²⁰ 3. ZnO possesses the highest piezoelectric tensor and is comparable to GaN and AlN. It is suitable for application in a charge coupled device (CCD).²¹
Amorphous	Disadvantage:
	1. Low hardness.²² 2. It needs doped with high valence element to improve poor electric conductivity.²³
Amorphous	Advantage:
	1. Amorphous ZnO can be prepared at low temperature, and was found to be well adapted to determine the optical constant of amorphous ZnO film over a wide spectral range.¹⁷ 2. Superior thermoelectric properties for thermoelectric devices such as a hydrogen gas sensor or infrared sensor under severe conditions, but not for thermoelectric power generation.²⁴
Amorphous	Disadvantage:
	1. The bandgap is smaller than crystal, and needs to be doped with another element to produce ionic characteristics and increase band-gap for potential applications.²⁵

II. SIMULATION MODEL

Before heating the amorphous zinc oxide structure, the most important step is to obtain a reliable amorphous zinc oxide configuration. Many global optimization algorithms have been successfully developed to obtain the lowest-energy configuration, such as genetic algorithm (GA),³⁶ big-bang (BB) method,³⁷ and simulated-annealing basin-hopping (SABH) method. In the traditional BH method, a conjugate gradient method is used to reach the stable configuration with the local energy minimum.³⁸ In our BH method, the conjugate gradient method was replaced by the fast inertial relaxation engine (FIRE) method,³⁹ which can be used to simulate a system consisting of a large number of atoms. Also, using the FIRE method in the BH method is faster than the conjugate gradient method for a larger system. Furthermore, the simulated annealing (SA)⁴⁰ method was also implemented with the BH method as the SABH method, which includes a wider search within the energy space. To obtain reasonable amorphous configurations instead of crystalline ones, the searching criterion of SABH in this study reverts to the original BH method where the developing direction of a lower energy is preferable.

For calculations of the optimization theory, we must select the potential function most applicable to the element's characteristics. The potentials for ZnO that we consider in the present work take a Buckingham-type form

$$U = \frac{q_i q_j}{r_{ij}} + A \exp\left(\frac{-r_{ij}}{\rho}\right) - \frac{C_{ij}}{r^6}, \quad (1)$$

where the first term on the right-hand side of equation is the long-ranged Coulombic energy, and the second and third

terms represent the short-range repulsion and attraction, respectively. The parameters of A_{ij} , ρ_{ij} , and C_{ij} correspond to Table II.⁴¹

We confirm that the density of amorphous zinc oxide is 4.6 g/cm³, as shown in Table III^{42–47} and use it to construct different systems of atom numbers, and calculate these unit cell lengths. Then, we put these chosen systems in our SABH method to find the most unstable structure of all local minima structures. Next, we pick the most unstable structure for each individual system to revise the lattice parameter with geometric optimization by the General Utility Lattice Program (GULP) module.⁴⁸ After obtaining the theoretical density for different systems, we sort these data and compare them with experimental data. Finally, we selected a system of 2000 atoms to act as an initial structure in the heating process.

X-ray diffraction (XRD) is commonly known as the direct evidence for the periodic crystal structure by the relative diffraction intensity at different X-ray incident angles. All XRD profiles shown in the current study were conducted by the REFLEX module in Materials Studio package.⁴⁹ In REFLEX, the Bragg's law is used to get the constructive interference intensity for X-rays scattered by materials, and the formula is listed as follows:

TABLE II. Parameters of the Buckingham potential function.⁴¹

Parameter	A_{ij} (eV)	ρ_{ij} (Å)	C_{ij} (eV Å ⁶)
O ²⁻ -O ²⁻	9547.96	0.21916	32.0
Zn ²⁺ -O ²⁻	0.0	0.0	0.0
Zn ²⁺ -Zn ²⁺	529.70	0.3581	0.0

TABLE III. Theoretical and experimental density of B3, B4, and amorphous structures.

		Density (g/cm ³)	
		Experiment	Theory
Crystal	B3	5.48145 ⁴²	5.42139 ⁴⁵
			5.43579 ⁴⁶
	B4	5.67536 ⁴³ 5.67868 ⁴⁴	5.674961 ^a
			5.70389 ⁴⁵ 5.43972 ⁴⁶
5.676363 ^a			
Amorphous		4.6 ⁴⁷	4.47 ^a

^aThis work.

$$d = \frac{n\lambda}{2 \sin \theta}, \quad (2)$$

where θ is a certain angle of incidence when the cleavage faces of crystals appear to reflect X-ray beams. d is the distance between atomic layers in a crystal, and λ is the wavelength of the incident X-ray beam. When n is an integer, the diffraction is constructive with a higher intensity. While n is an half integer, the diffraction is destructive and the intensity approaches zero.

Figure 1(a) shows the optimized structure of amorphous ZnO by GULP, with the corresponding XRD profile for wurtzite (B4) and zinc blende (B3) shown in Figure 1(b) for comparison. The characteristic XRD peaks for B4 and B3 ZnO phases are consistent with those reported in previous studies,⁵⁰ and the XRD profile of amorphous ZnO has proven that the structure obtained from SABH is amorphous.

During the simulation process, the Nosé–Hoover method⁵¹ is adopted to ensure a constant system temperature during the simulation process. The Verlet algorithm is also employed to calculate the trajectories of the positional and angular correlation, and a time step of 1 fs is set for the time integration. Isothermal-isobaric ensemble (NPT) was applied to the current system with initial temperature set at 300 K, so the averaged pressure calculated over a time interval will be a constant value, this system does not affect the comparison between the experimental and the MD simulation results. Due to the 3-dimensional periodic boundary conditions are employed during the thermal elevating process, and there is no surface for oxygen atom desorption, the stoichiometry of Zn and O maintains 1 during the heating process.⁵² Furthermore, in the simulation module, the annealing method is used in our simulation to obtain the ultimate atom position, velocity, and force; therefore, the temperature in the system is increased from 300 K to 3800 K with a rate of increase of 25 K per step, and then the temperature is maintained at every step for 3 ps. Finally, the MD simulation was performed to equilibrate the simulation system. After letting our system achieve the correct structural configurations in the vicinity of the temperature setting, we produce some molecular structures which are used for statistics and analysis.

Finally, we calculated the coordination number, volume, density, and average bond lengths between Zn–O bonding of the structural configurations at every temperature point

FIG. 1. The configuration of ZnO amorphous structure (a) and XRD spectra of B3, B4 and amorphous structure at 300 K (b).

during the heating process. In this study, we compare the simulation results of the three varieties of structures: the amorphous, and two ZnO crystals configuration (B3 and B4) systems.

III. RESULT AND DISCUSSION

To determine whether the system size has an influence on the amorphous ZnO local structures, systems with 2000,

2500, 3000, 3500, and 4000 atoms with a Zn/O ratio of 1 were used. Figure 2(a) shows the densities obtained by the SABH for systems with different atom numbers, and it seems that the densities of all cases are very close to the average value of 4.47, which is a slight 2.8% lower than the experimental density (4.6 g/cm^3).⁴⁷ This indicates that the potential in the current study is very reliable in modeling the amorphous structures. To check the local structures in the amorphous ZnO, the CN of Zn was calculated, with CN defined as the average first neighbor Zn atom number around a reference Zn atom. Figure 2(b) shows the fractions of CN5, CN4, CN3, and CN2 for systems with different atom numbers, with the integer after “CN” representing the value of CN. As one can see, CN4 is the dominant local structure with an occupation of about 0.6 and the second most dominant local structure is CN3 with a fraction of about 0.32. CN5 occupies about 0.07 of the total CN, and CN2 is close to 0. The fractions of different CN numbers vary insignificantly with atom

FIG. 2. The density distribution (a) and (b) CN ratio of systems with different numbers of atoms.

number, indicating that the fractions of CN values are independent on the system size when the atom number is higher than 2000. Consequently, to save simulation time while studying the temperature elevation process for amorphous ZnO, the system with 2000 atoms was used. The densities of B3 and B4 ZnO crystal structures are also listed in Table II, and the predicted values are in good agreement with previous experimental and theoretical results, indicating that the potential function is able to simulate both crystal and amorphous ZnO materials.

To compare the local structural variations with those in amorphous ZnO, the B4 and B3 crystal structures are also used during the temperature elevation process. Figure 3(a) shows the unit cell of B4 ZnO crystal structure, and Figures 3(b) and 3(c), respectively, illustrate the variations of CN fractions and the average Zn–O bond length with density at different temperatures. The corresponding snapshots at

FIG. 3. (a) The unit cell, (b) coordination number, and (c) average bond length of B4 structure versus temperature.

FIG. 4. (a) The unit cell, (b) coordination number, and (c) average bond length of B3 structure versus temperature.

different temperatures are also shown in Figure 3(b). At temperatures lower than 900 K, the fraction of CN4 is 1 and the ZnO structure is under thermal expansion, where average Zn–O bond length and system volume increase with temper-

ature. Within this temperature range (0–900 K), both Zn and O atoms vibrate at their equilibrium positions. As temperature increases to between 900 and 1800 K, the fraction of CN3 gradually increases from 0 to 0.14 and the CN4 fraction

decreases continuously from 1 to 0.84. The fractions of CN2 and CN5 are close to 0, and this indicates that a small amount of atoms at this temperature range (900–1800 K) vibrate with larger amplitudes and that some first neighbor Zn atoms at lower temperatures leave their referenced Zn atoms, resulting in the decrease of the first neighbor Zn–Zn pairs and the increase in the fraction of CN3. The increase of average Zn–O bond length shown in Figure 3(c) can be attributed to both the thermal expansion and the decrease in Zn–Zn first neighbor pairs. As the temperature rises to between 1800 and 2050 K, a small fraction of CN5 appears, and significant decreases of CN4 and increases of CN3 can be seen, as shown in Figure 3(b). However, the average bond length does not change significantly within this temperature range although the system volume increases with the temperature, implying that some local amorphous structures have been generated. When the temperature is continuously elevated to higher than 2050 K, the fraction of CN2 begins to appear and increases with temperature. The fraction of CN4 displays a considerably decrease with temperature while CN3 increases. Even though there is significant local structural transformation between CN2, CN3, CN4, and CN5, the average bond length at temperatures higher than 2050 K does not change significantly. This reveals that the fraction of local amorphous structures increases with temperature. When temperature is higher than 3100 K, the system volume and the CN fractions vary very considerably, whereas average bond length maintains a relatively constant value.

For the B3 ZnO crystal structure, Figure 4(b) shows the CN and system volume variations, and Figure 4(c) displays temperature and average Zn–O bond length with density for different temperatures. Lower than 1000 K, the fraction of CN4 remains 1, but system volume and average bond length increase with temperature. There is no local structural transformation within this temperature range and the system is under thermal expansion. Once the temperature rises to higher than 1000 K, the fraction of CN3 begins to increase from 0 while CN2 and CN5 remain at 0 until the temperature reaches 2300 K. Consequently, between 1000 and 2300 K, the decrease of first neighbor Zn–Zn pairs results in the increase of CN3 fraction and decrease in CN4 fraction, leading to an increase in system volume and average Zn–O bond length. As temperature is elevated to higher than 2300 K, the transformation between CN4 and CN3 becomes more significant because the fluctuations of CN4 and CN3 fractions are larger. The increasing system volume with slightly decreasing average bond length indicates the occurrence of active local structural transformation and the increase in the number of local amorphous structures.

Figure 5(a) shows the temperature elevation process of the amorphous ZnO material, with morphologies at several temperatures displayed in Figure 6. At temperatures lower than 500 K, variations of CN fractions, system volume, and bond length are not apparent. From 500 to 1200 K in Figure 5(a), it is clear that the fraction of CN4 increases whereas the fraction of CN3 decreases with temperature. The system volume decreases with temperature when it rises from 500 to 1200 K because some portions of the amorphous ZnO have gradually transformed into local crystal structures, which can

FIG. 5. (a) Coordination number, and (b) average bond length of amorphous structure versus temperature.

be seen in Figure 6(a). As the temperature continuously increases to 1200 to 2200 K, the system volume does not change significantly with temperature although there are local structural transformations among CN5, CN4, and CN3. Within this temperature range, the local crystal structures distributed within the amorphous ZnO adjust their orientations for crystal growth. At 2200 K, an abrupt drop of system volume in Figure 5(a) or an abrupt jump of density in Figure 5(b) can be seen, and the corresponding morphology as well as its XRD profile can be found in Figure 6(b). The distinct characteristic XRD peaks at 31° , 34° , and 35° indicated by arrows in Figure 6(b) indicate that the majority of the ZnO material is in the wurzite (B4) arrangement. As temperature increases from 2200 K, the variation of local structure is very similar to that of B4. Annealing experiments reported by Bruncko *et al.*⁵² found that the recrystallization of amorphous ZnO thin film begins to occur when annealing temperature becomes higher than about 473 K. The amorphous structure will transfer into a polycrystalline wurtzite phase

FIG. 6. The configuration and XRD spectra of ZnO amorphous structures at (a) 1000 K, (b) 2200 K, and (c) 3250 K.

with randomly oriented local crystal structures, proved by the corresponding XRD measurements. In our simulation, the recrystallization begins once the local crystal structures appear at temperatures higher than 500 K. The fraction of crystal structure gradually increases with temperature; however, the polycrystalline phase with grain boundaries is not found within the wurtzite structure obtained at 2200 K because the expensive computational cost for MD simulation limits our simulation system size to 2000 atoms. Moreover, temperatures much higher than the experimental ones (473, 673, 873, and 1073 K) are used to enhance the occurrence of local structural transformation because of the large amount of calculation time for the integration of Newton's second law. We believe that despite the artificial enforcements, the simulation can still reflect the recrystalline process during the annealing experiment.

IV. CONCLUSION

We have combined SABH method and MD simulation to investigate B3, B4, and the ZnO amorphous structural variations during the temperature elevation process. CN4 is the dominant local structure with the occupation of about 0.6, and the second most dominant local structure is CN3 with the fraction of about 0.32. The CN5 occupies about 0.07 of the total CN, and CN2 is close to 0. The changes of CN, volume, and average bond lengths are very dependent on temperature. In B3 and B4 structures, the fraction of CN4 falls while CN3 increases. System volume and temperature are shown to have a linear relationship. The volume of B4 structure can quickly expand with temperature, but the volume of B3 slowly increases with temperature to 3000 K. The B3 and B4 deconstruct at 3450 K and 3200 K, respectively.

However, the ZnO amorphous structure has the opposite tendency in CN transfer. At about 500 K, the recrystallization begins once the local crystal structures appear. The crystal structure fraction gradually increases with temperature, unlike B3 and B4. At 2200 K, the polycrystalline phase with grain boundaries is not found within the wurtzite structure because the expensive computational cost for MD simulation limits our simulation system size to 2000 atoms because of the large amount of calculation time for the integration of Newton's second law. Furthermore, temperatures much higher than experimental (473, 673, 873, and 1073 K) are used to enhance the occurrence of local structural transformation. Finally XRD analysis calculated by REFLEX module shows results in good agreement with previous experimental results, indicating that our MD simulation is able to reflect the experimental phenomenon of both crystal and amorphous ZnO materials in the temperature elevation process.

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