Materials Science in Semiconductor Processing 39 (2015) 148–155

Contents lists available at ScienceDirect

Materials Science in Semiconductor Processing

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

The effect of growth conditions, point defects and hydrogen on the electronic structure and properties of p-type (Al,N) codoped ZnO: A first principles study

M.A. Lahmer [a,b,*], K. Guergouri [c]

[a] *Department of Physics, University of Boumerdes, Boumerdes 35000, Algeria*
[b] *Department of Physics, University of Constantine 1, Constantine 25000, Algeria*
[c] *Department of Physics, Larbi Ben M'Hidi University, University of Oum El Bouaghi, Oum El Bouaghi 04000, Algeria*

ARTICLE INFO

Available online 21 May 2015

Keywords:
ZnO
Hydrogen
First-principles
p-Type
Vacancy

ABSTRACT

The effects of point defects, hydrogen, and growth conditions on the electronic structure and properties of the (Al,N) codoped p-type ZnO have been investigated using the first principles method. The obtained results showed that the Al_{Zn}–N_O–V_{Zn} complex is a shallow acceptor that can play an important role in achieving the p-type conductivity in the (Al,N) codoped ZnO films. Our results showed also that the electrical conductivity type in the (Al,N) codoped ZnO films strongly depends on the donor/acceptor concentrations ratio. The codoped ZnO films prepared under both Zn-rich and O-rich growth conditions with a donors/acceptors ratio of 1:2 have a p-type conductivity, while those prepared with a ratio of 1:1 cannot be p-type unless if they are prepared under O-rich conditions. The achieved p-type quality depends also on the used nitrogen doping source. To prepare p-type ZnO film of high quality using the (Al,N) codoping method, the use of NO or NO_2 is recommended. The presence of donor defects such as oxygen vacancies and hydrogen will significantly affect the electronic properties of the (Al,N) codoped ZnO films, and if the concentration of these defects in the sample is high enough, the material can be easily converted to n-type.

1. Introduction

Zinc oxide (ZnO) is a wide-band-gap semiconductor with a band gap of 3.3 eV at room temperature. This material has recently attracted much attention because of potential applications in optoelectronic and transparent devices [1,2]. Usually, as-grown ZnO exhibits n-type conductivity. As the origin of the intrinsic n-type conductivity, native defects such as Zn interstitial (Zn_i) and O-vacancy (V_O) have been suggested. But it is difficult to explain the observed n-type conductivity using these defects, since Zn_i has a high formation energy and the donor level of V_O is too deep [3–10]. As an alternative candidate, hydrogen was suggested. It is unintentionally incorporated during crystal growth and can be responsible for the n-type conductivity [11–16].

To use ZnO in optical, optoelectronic and photonic applications and devices it is necessary to fabricate good p-type material. However, it is difficult to produce good p-type ZnO due to a large number of donor-type point defects that exist in the as grown material such as; interstitial zinc (Zn_i), oxygen vacancy (V_O) and hydrogen. Self-compensation, high ionization energy and low solubility are additional challenges for the realization of p-type ZnO using monodoping method. Several research groups have reported the fabrication of p-type ZnO using group V and group I elements such as; Li [17], N [18], P [19] and Na [20]. Others have used As [21] and Ag [22] as acceptor

---

[*] Corresponding author at: Department of Physics, University of Boumerdes, Boumerdes 35000. Algeria. Tel.: +213 24816249.
E-mail address: malahmer@yahoo.fr (M.A. Lahmer).

http://dx.doi.org/10.1016/j.mssp.2015.05.004
1369-8001/© 2015 Elsevier Ltd. All rights reserved.

dopants. Recently, the codoping method has been suggested as a solution for this problem. Yamamoto and Katayama-Yoshida [23] suggested that the use of the codoping method can solve unipolarity for the fabrication of p-type ZnO. Since then, many groups have carried out theoretical and experimental studies to realize p-type ZnO by the codoping method using different dopants such as; P–N [24], Mg–N [25], Ag–N [26–28], Li–N [29–32], B–N [33,34], Be–N [35], Al–N [36–38], Ag–S [39,40], Ga–N [41,42], As–N [43], Al–As [44] and Li–F [45]. Yamamoto et al [23] found that n-type doping with shallow donors like Al, Ga and In atoms decreases the madelung energy of ZnO. They found that the codoping method using acceptors (N) and shallow donors like Al simultaneously will decrease the madelung energy, decrease the ionization energy of the acceptors and donors, and enhance the solubility of the acceptors atoms.

In recent years, the Al–N codoping has attracted much attention and several studies have been carried out to control the structural, electrical and optical properties of Al–N codoped p-type ZnO.

In this paper, first-principles calculations have been carried out to investigate the electronic structure and properties of (Al,N) and (Al,2N) codoped ZnO as well as the effects of the growth conditions, point defects and hydrogen on these properties.

2. Computational details

All calculations are performed by using the first-principles pseudopotential method based on density functional theory (DFT) and the plane-wave method as implemented in the *Quantum Espresso* package [46]. For valence electrons, The pseudopotentials involve 12, 6, 5 and 3 valence electrons for Zn($3d^{10}4s^2$), O ($2s^22p^4$), N($2s^22p^3$) and Al($3s^23p^1$), respectively. In this work Al and N dopants are doped into ZnO in the form of substitutions. Exchange and correlation effects are treated under the generalized-gradient-approximation with Perdew–Burke–Ehrenkof (PBE) functional [47].

All calculations are performed using the $3\times3\times2$ supercell with 72 atoms (Fig. 1) and a $3\times3\times2$ grid of k-points generated by the Monkhorst-Pack scheme. The cutoff energy for plane wave basis was set to 40 Ry. The atoms positions are fully relaxed using the BFGS method.

Structural optimizations for bulk ZnO in the WZ structure produced the following parameters: $a=3.284$ Å, $c=5.290$ Å, $u=0.379$ and $\Delta H_f=3.43$ eV, in good agreement with experiment ($a=3.249$ Å, $c=5.201$ Å, $u=0.382$ and $\Delta H_f=3.6$ eV).

The formation energy is a key quantity characterizing the properties of a defect or impurity in a solid. Defects with high formation energies can occur in low concentrations and by consequence will have a small or negligible impact on the host material properties; only those whose concentration exceeds a threshold will have observable effects.

The formation energy of defect D is defined as

$$E^f(D)=E_{tot}(ZnO+D)-E_{tot}(host)+\sum_i n_i\mu_i \tag{1}$$

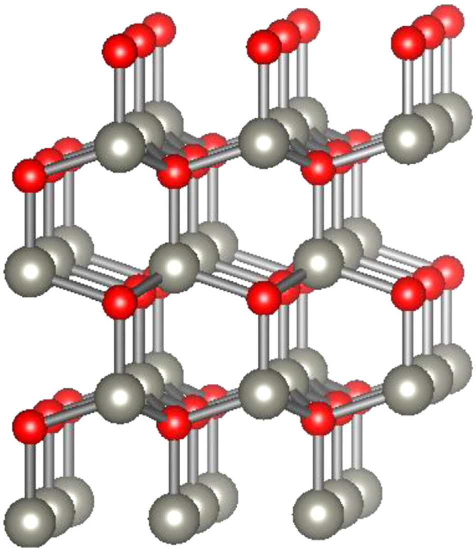

Fig. 1. The $3\times3\times3$ ZnO supercell used in this work.

where $E_{tot}(ZnO+D)$ is the total energy of a supercell containing the defect D, $E_{tot}(host)$ is the total energy of the ZnO perfect crystal in the same supercell, n_i indicates the number of atoms i (Zn, O or D) that have been added to ($n_i<0$) or removed ($n_i>0$) from the supercell, μ_i is the chemical potential of species i.

In our case, the formation energies of Al–N and Al–2N complexes are calculating according to

$$E^f(Al-N)=E_{tot}(ZnO{:}\,Al+N)-E_{tot}(ZnO)+\mu_{Zn}+\mu_O-\mu_{Al}-\mu_N \tag{2}$$

$$E^f(Al-2N)=E_{tot}(ZnO{:}\,Al+2N)-E_{tot}(ZnO)+\mu_{Zn}+2\mu_O-\mu_{Al}-2\mu_N \tag{3}$$

where $E_{tot}(ZnO{:}\,Al+N)$, $E_{tot}(ZnO{:}\,Al+2N)$ and $E_{tot}(ZnO)$ are the total energies of supercells containing Al–N, Al–2N and the pure ZnO.

The Al chemical potential μ_{Al} is set to its value in the FCC structure and is given by

$$\mu_{Al}=\mu_{Al}^{Bulk}=\frac{1}{4}E_{tot}(FCC\ Al) \tag{4}$$

The Zn chemical potential μ_{Zn} is given by

$$\mu_{Zn}=\mu_{Zn}^{Bulk}+\Delta\mu_{Zn}=\frac{1}{2}E_{tot}(hcp\ Zn)+\Delta\mu_{Zn} \tag{5}$$

where E_{tot}(Fcc Al) and E_{tot}(hcp Zn) are the total energies of the Al(FCC) and the Zn(hcp) structures respectively.

The oxygen chemical potential is restricted within the thermodynamically allowed ranges determined by the corresponding formation enthalpy of bulk ZnO (ΔH^f_{ZnO}) and gas phase of O ($\mu^{gas}_{O_2}$).

The allowed range for the chemical potential $\Delta\mu_O$ is given by

$$\Delta H^f_{ZnO}\le\Delta\mu_O\le 0 \tag{6}$$

The formation energy depends on the chemical potential of Zn and O atoms (μ_{Zn} and μ_O), and this dependence can be simplified by eliminating μ_{Zn} in favor of μ_O using the bulk thermodynamic equilibrium condition:

$$\Delta\mu_{Zn}+\Delta\mu_O=\Delta H^{f}_{ZnO} \tag{7}$$

The nitrogen chemical potential μ_N depends on the N source used in the doping of the material. Previous work of Yan et al [48] shows that the N doping efficiency in ZnO depends greatly on the dopant chemical potential. In this work we use the same N gas source as in Ref. [48]. The nitrogen chemical potential is given by

$$\mu_N=\frac{1}{2}E_{tot}(N_2)+\Delta\mu_N \tag{8}$$

where $E_{tot}(N_2)$ is the total energy of nitrogen molecule and $\Delta\mu_N$ is given by;

$$\Delta\mu_N=\begin{cases}0 & \text{for}\quad N_2\\ \Delta H_f^{No}-\Delta\mu_O & \text{for}\quad NO\\ \frac{1}{2}(\Delta H_f^{N_2O}-\Delta\mu_O) & \text{for}\quad N_2O\\ \Delta H_f^{NO_2}-2\Delta\mu_O & \text{for}\quad NO_2\end{cases} \tag{9}$$

where ΔH_f^{NO}, $\Delta H_f^{N_2O}$ and $\Delta H_f^{NO_2}$ are the formation enthalpies of the molecules; NO, N_2O and NO_2 respectively.

3. Results and discussions

3.1. (Al,N) and (Al,2N) codoped ZnO

Fig. 2 shows the calculated density of states (DOS) of perfect ZnO, Al–N codoped ZnO, and Al–2N codoped ZnO. The Fermi level is set to zero as usual. In the case of the perfect ZnO supercell, the Fermi level is located at mid-gap, which means a semiconductor behavior of the material. When the material is codoped with Al and N atoms, a new peak appears near the valence band maximum (VBM) and the Fermi level is shifted down in this case (Fig. 2b); however, it appears that the Al–N complex which is constituted from Al_{Zn} and N_O atoms on neighboring sites is a deep acceptor, and it cannot lead to good p-type conductivity. This is completely different from the results of Hongling et al [49] who founds by using an LDA+U method that Al–N codoping can lead to p-type conductivity in ZnO. The differences between the results of this work and the results of Ref. [49] can be attributed to differences in computational methods and supercell size used in each work. While we use GGA-PBE in our work, the authors of Ref. [49] use the LDA+U method to correct the band gap problem. The obtained gap in this case is about 2.77 eV; however, the *3d* band position is not corrected. They obtained a *3d* band position around −10 eV which is different from the experimental values (−7 eV to −8 eV).

Now, we are going to study the effect Al–2N codoping on the electronic structure and properties of ZnO. Duan et al [50] have investigated the properties of the Al–2N codoped ZnO using first principles method. They found that the N–Al–N configuration with the two N atoms in first neighboring sites of the Al_{Zn} atom is the most stable configurations. For this reason we will limit our study to this configuration only. The obtained DOS (Fig. 2c) shows that in the case of Al–2N codoped ZnO the Fermi level is shifted downward in the valence band. This means that the valence band is partially filled and a number of holes exist. The material in this case is p-type conductivity, in good agreement with previous DFT calculations [49–51].

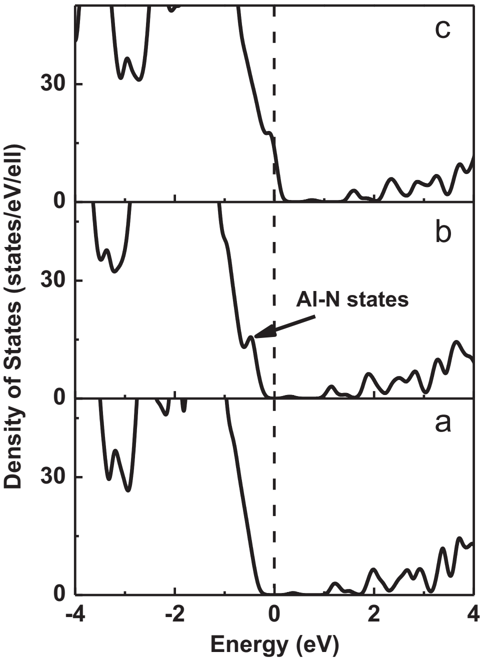

Fig. 2. Calculated density of states of ZnO: (a) pure, (b) with Al–N complex and (c) with Al–2N complex.

We turn our attention now to the effect of the growth conditions on the properties of the (Al,N) codoped ZnO. The calculated formation energies for the Al–N and N–Al–N complexes under different growth conditions are shown in Fig. 3a and b, respectively. The obtained results show that the formation energies of the Al–N and N–Al–N complexes are sensitive to the oxygen chemical potential (μ_O) and to the nature of the used N doping gas. It is very clear that among all nitrogen doping gases proposed for the realization of (Al,N) codoped ZnO, the NO gas is the most efficient N doping source under O-rich conditions because it leads to the formation of the Al–N and N–Al–N complexes with high concentrations. However, under Zn-rich conditions NO_2 gives the lowest formation energies for both Al–N and N–Al–N complexes and this means that it will be the best choice.

3.2. Role of zinc vacancy (V_{Zn}) and oxygen vacancy (V_O)

Zn vacancy (V_{Zn}) is one of the most common point defects in ZnO prepared under O-rich conditions because it has the lowest formation energy among all native defects. Fig. 4 shows the obtained density of states for Al–N and Al–2N codoped ZnO supercells with zinc vacancy. Compared to the Al–N codoped ZnO case, we can show that the presence of the zinc vacancy will create a shallow acceptor states at the top of the VBM. The Fermi level in this case, is located within the valence band which means that the valence band is partially filled and a number of holes exist in it, and this will lead to p-type conductivity. The density of acceptor shallow states in this case is not only higher

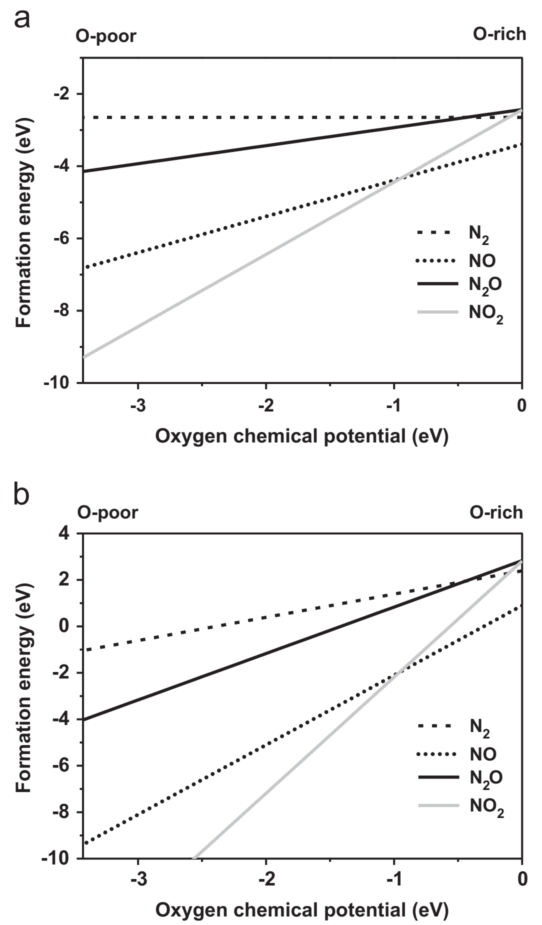

Fig. 3. Formation energy of (a) Al–N and (b) Al–2N as a function of the oxygen chemical potential. The N_2, N_2O, NO and NO_2 gases are used as a nitrogen doping source.

than that of the Al–N codoped ZnO but is also higher than that of the Al–2N codoped material. In the case of Al–2N codoped ZnO, the presence of zinc vacancy will enhance the p-type conductivity because it will introduce more holes in the valence band. The Fermi level in this case is shifted deep in the valence band (Fig. 4).

Under Zn-rich conditions, the oxygen vacancy is the most abundant defect in ZnO due to its low formation energy. For this reason, it can play an important role in the control of the electronic and optical properties of zinc oxide. On the other hand, oxygen vacancy is a donor defect, which means that it can compensate the Al–N acceptor states and therefore make the realization of p-type doping very difficult.

To study the effect of oxygen vacancies on the properties of Al–N and Al–2N codoped ZnO, we have calculated the DOS curves of the ZnO supercell containing the Al–N+V_O complex as well as those of the supercell with the N–Al–N+V_O complex respectively. The obtained DOS curves are shown in Fig. 5. It is very clear that the presence of oxygen vacancy affects considerably the electronic structure of the (Al,N) codoped ZnO. In the case of ZnO supercell with the Al–N–V_O complex, the corresponding Fermi level is located at mid-gap, which means that the obtained material is not p-type. In the case of the sueprcell with the N–Al–N–V_O complex, The Fermi level is shifted upward in the conduction band, which mean that a

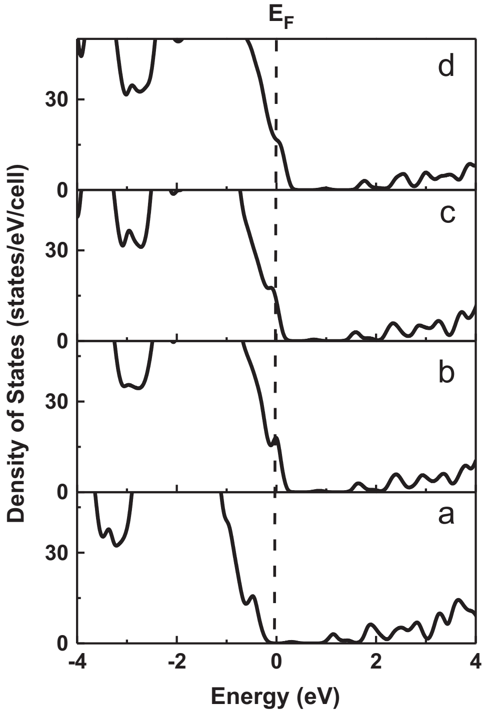

Fig. 4. Calculated density of states for ZnO supercells containing; (a) an Al–N complex, (b) Al–N+V_{Zn} complex, (c) Al–2N complex and (d) the N–Al–N+V_{Zn} complex. The vertical line shows the Fermi energy position.

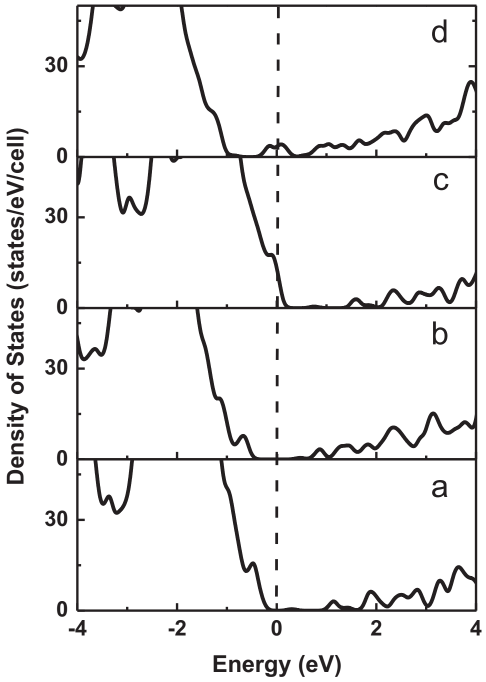

Fig. 5. Calculated density of states for ZnO supercells containing; (a) Al–N complex, (b) Al–N+V_O complex, (c) N–Al–N complex and (d) N–Al–N+V_O complex. The vertical line shows the Fermi energy position.

Table 1
Formation energies of the Al–N–V_{Zn} complex in ZnO as obtained for various nitrogen doping source. Values are given for Zn-rich and O-rich conditions at $E_F=0$ eV (valence band maximum; VBM) and $E_F=3.3$ eV (conduction band maximum; CBM).

Doping source	Charge state	Zn-rich conditions		O-rich conditions	
		VBM	CBM	VBM	CBM
N_2	0	2.50	2.50	−0.93	−0.93
	−1	−0.39	−3.69	−3.82	−7.12
N_2O	0	1.01	1.01	−0.71	−0.71
	−1	−1.89	−5.19	−3.60	−6.90
NO	0	−1.67	−1.67	−1.67	−1.67
	−1	−4.56	−7.86	−4.56	−7.86
NO_2	0	−4.15	−4.15	−0.72	−0.72
	−1	−7.04	−10.34	−3.61	−6.91

Table 2
Formation energies of zinc and oxygen vacancies in Al–N and Al–2N codoped ZnO. Values are given for Zn-rich and O-rich conditions. The N_2 gas was used as a nitrogen source.

Defect	Zn-rich		O-rich	
	Al–N	Al–2N	Al–N	Al–2N
V_{Zn}	5.16	5.01	1.72	1.58
V_O	1.76	1.09	5.20	4.52

number of electrons exist in the conduction band and the obtained material in this case is n-type conductivity.

To confirm the nature of the Al–N–V_{Zn} defect, we have calculated its formation energies in different charge states and the obtained results are listed in Table 1. Our calculations showed that the single negative charge (−1) is the stable charge of the Al–N–V_{Zn} complex over all possible values of the Fermi energy and this confirm the shallow acceptor character of this defect. The obtained results shows also that under O-rich conditions NO gives the lowest formation energy, while, under Zn-rich conditions NO_2 gives the lowest formation energy and this confirm our previous predictions concerning the choice of the nitrogen doping source. On the other hand, our calculations show that the Al–N–V_O complex is a double donor and this means that it can compensate easily the acceptor states induced by the Al–N–V_{Zn} complex.

The calculated formation energies of the zinc and oxygen vacancies in both Al–N and Al–2N codoped ZnO are listed in Table 2. The obtained results show that zinc vacancy has almost the same formation energy in Al–N and Al–2N codoped ZnO. A difference of only 0.16 eV was obtained, which means that this defect can exist with almost the same concentration in both cases. In the other hand, things are different in the case of the oxygen vacancy defect. The obtained results show that the formation energy of V_O in the Al–2N codoped ZnO is 0.7 eV lower than its value in the Al–N codoped material. This means that compared to the Al–N codoped material, the Al–2N codoped ZnO will contain more oxygen vacancies. This means also that it is very difficult to realize p-type ZnO by (Al,N) codoping under Zn-rich conditions, and that the best choice is to grow the codoped material under O-rich conditions.

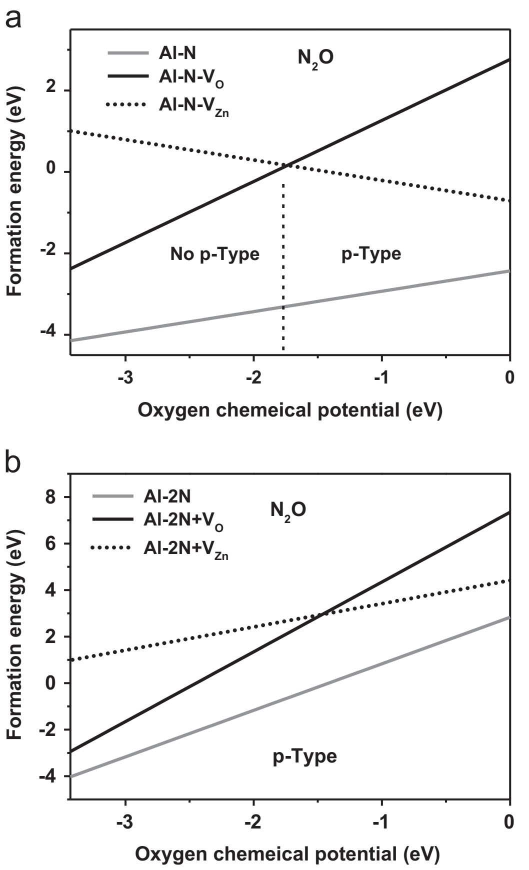

Fig. 6. Formation energy of V_O and V_{Zn} related complexes as a function of the oxygen chemical potential in; (a) The (Al,N) codoped ZnO and (b) The (Al,2N) codoped ZnO. These values are obtained using N_2O as a nitrogen doping source.

On the other hand, combining these results with the results of Section 3.1, we can suggest that it is possible to produce a p-type ZnO films by the (Al,N) codoping method under O-rich conditions through the use of N_2O or N_2 gases as a nitrogen source. To better understand this point, we have calculated the formation energy of these defects under different growth conditions (different N source and different values of μ_O) and the obtained results for N_2O are showed in Fig. 6. It is very clear that in the case of Al–2N codoped ZnO the obtained material is always p-type because the Al–2N complex has always the lowest formation energy. However, the best p-type quality can be obtained under O-rich conditions rather than the Zn-rich conditions. In the case of Al–N codoped ZnO the situation is different. The p-type doping can be achieved only under O-rich conditions and for oxygen chemical potential in the range of [−1.75 eV, 0 eV]. In the previous experimental work of Chou et al [52], the authors have showed that the conductivity type of (Al,N) codoped ZnO thin films prepared by RF magnetron sputtering method with N_2O as a nitrogen source gas depend greatly on the used $N_2O/(N_2O+O_2)$ ratio. Their results shows that the best p-type conductivity is obtained when a 30% volume ratio N_2O is used, and when the used ratio is higher than 50% the obtained material is n-type. These results appear to be in agreement with the results of this work. On the other hand, Kumar et al [53] showed that the as grown (Al,N) codoped ZnO films prepared by sputtering method with N_2/O_2 ratio in the range of 40−60% are n-type and that the p-type conductivity is obtained after thermal annealing. They explained these results in term of the formation of new V_{Zn}-related acceptor. This last result is very important because it appear to be in very good agreement with our results that show that the Al–N–V_{Zn} complex or the Al–2N–V_{Zn} complex can be the origin of the p-type conductivity in the (Al,N) codoped ZnO.

Zhu et al [36] have reported the realization of p-type ZnO using the (Al,N) codoping method and a high temperature homo-buffer layer. They found that increasing the buffer layer deposition time will lead to a decrease in the quality of the p-type ZnO thin films. They explained this result in term of the formation of more Al–N complexes instead of N–Al–N complexes and this appear to be consistent with our results suggesting that Al–N complex do not lead to p-type in ZnO. Another important point is related to the nitrogen source used in this study. The NH_3 gas was used as a nitrogen doping source in this case. Recent DFT calculations of Bang et al [54] show that the $(NH_3)_{Zn}$ defect is more stable under O-rich conditions than N_O, $(N_2)_{Zn}$ and $(NO)_{Zn}$. This study shows also that $(NH_3)_{Zn}$ is a neutral complex; however, it can be transformed to acceptor by capturing H donor. On the other hand, Lu et al [37] have reported the fabrication of (Al, N) codoped p-type ZnO using N_2O gas as a nitrogen and oxygen source. They have reported that the best p-type quality can be obtained for growth temperatures in the range of 450–500 °C and that the use of lower or higher growth temperatures will decrease the p-type quality of the ZnO films. The use of N_2O gas as a source of nitrogen and oxygen means that the ZnO films are prepared under O-poor conditions. Based on the results of Fig. 6 we can see that the formation energies of Al–N and N–Al–N complexes under Zn-rich conditions are very close to each other. The formation energy of the Al–2N–V_O complex in this case is slightly higher than that of the Al–2N acceptor complex. This means that it is very difficult to obtain high p-type quality in this case. However, by combining the results of Figs. 6 and 8 we believe that it is possible to understand the results of Ref. [37]. We suggest that the formation of Al–2N–H_i or Al–2N–H_O complexes is responsible of the low p-type quality observed in ZnO films prepared at 400 °C. By increasing the growth temperature to 450–500 °C, the H_O and N–H complexes become thermally unstable and the hydrogen will diffuse out the sample [54,55] and this lead to the formation of Al–2N complex. Previous work [56] showed that H plays an important role in N-doped p-type ZnO. At an annealing temperature of 500 °C, the H shows an important reduction in concentration in the ZnO film, and at the same time the N concentration remains constant and this leads to the obtaining of p-type material. For high temperatures, nitrogen become thermally unstable and will diffuse out and consequently the p-type quality decrease or vanish completely.

3.3. Effect of hydrogen

Interstitial hydrogen plays an important role in the control of the electronic properties of ZnO, because it has a shallow donor character [11], it has a low diffusion energy [57,58] and it is present in most of the ZnO growth methods. For this reason it is very important to investigate the effect of hydrogen on the electronic structure and properties of (Al,N) codoped ZnO. The obtained results in this work show that interstitial hydrogen atom prefer bonding with the N atom to form the N–H complex. The formation energy of this complex in its neutral charge is 0.09 eV.

Due to their low formation energies and their donor character in ZnO, both interstitial and substitutional H can be regarded as a serious impedance for the realization of (Al,N) codoped p-type ZnO. The calculated DOS curves for H_i and H_O defects in Al–N codoped ZnO are shown in Fig. 7. The obtained Fermi level in this case is located inside the

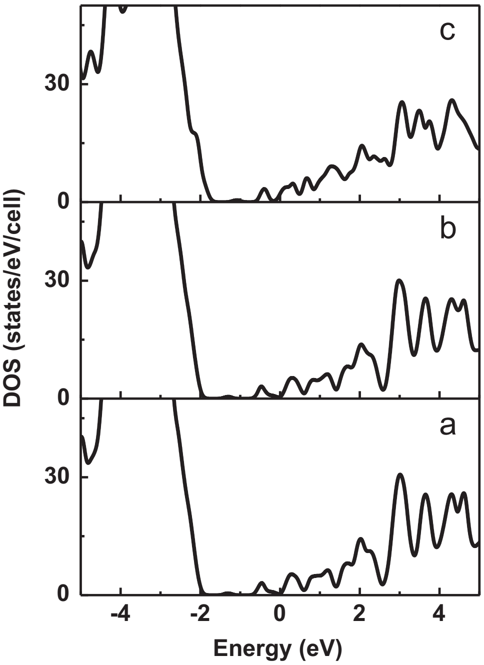

Fig. 7. Calculated density of states for ZnO supercells containing different H-related defects; (a) The Al–N–H_i complex, (b) The H_i–Al–N complex and (c) The Al–N+H_O complex.

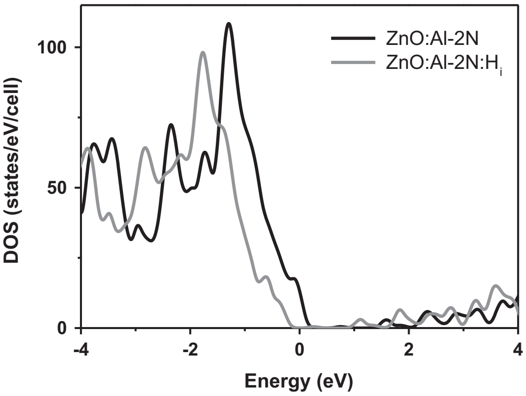

Fig. 8. Calculated density of states for ZnO supercells containing N–Al–N and N–Al–N–H_i complexes.

conduction band, which means that the obtained material is n-type conductivity. In the case of the Al–2N codoped ZnO, the situation is somewhat different. The calculated DOS shows that the Fermi level for the supercell containing Al–2N with the H_i bonded to a nitrogen atom is located at mid gap (Fig. 8). This means that the Al–2N related acceptor state is passivated by H atom and if a high amount of H_i is introduced in the sample ($c_H > c_N$) the material will be n-type conductivity.

4. Conclusions

In this work, the effects of growth conditions, point defects and hydrogen on the electronic structure and properties of the (Al,N) codoped p-type ZnO have been investigated using the first-principles method. The obtained results shows that the conductivity type of the (Al,N) codoped ZnO depend greatly on the (Al,N) doping level, the used nitrogen gas source, the point defects concentrations and the growth conditions.

Our results showed that the Al–N–V_{Zn} complex is a shallow acceptor that can play an important role in achieving the p-type conductivity in the (Al,N) codoped ZnO films. This complex can be the source of the p-type conductivity in the Al–N codoped ZnO films prepared under O-rich conditions. The obtained results showed also that the electrical conductivity type in the (Al,N) codoped ZnO films strongly depend on the donor/acceptor concentrations ratio. While the use of a donor/acceptor dopants ratio of 1:2 will produce p-type material under both O-rich and Zn-rich growth conditions, the (Al,N) codoped ZnO films prepared with a dopants ratio of 1:1 cannot be p-type unless if they are prepared under O-rich conditions. On the other hand, these results suggest that the p-type quality of the codoped material depends greatly on the nature of the used nitrogen doping source. To prepare p-type ZnO film of high quality using the (Al,N) codoping method, the use of NO or NO_2 is recommended.

Finally, the effect of the oxygen vacancies and hydrogen on the properties of the (Al,N) codoped ZnO was also investigated. The obtained results show that these defects may play an important role in the control of the electronic structure and properties of the (Al,N) codoped ZnO. The presence of the hydrogen defect or the oxygen vacancies in the sample with a high concentration will convert the p-type conductivity of the (Al,N) codoped ZnO material to the n-type.

Acknowledgments

M.A. Lahmer would to acknowledge the *nanohub* (www.nanohub.org) help in computational resources.

References

[1] A. Kołodziejczak-Radzimska, T. Jesionowski, Materials 7 (2014) 2833–2881.
[2] A. Janotti, C.G. Van de Walle, Rep. Prog. Phys. 72 (2009) 126501.
[3] A. Janotti, C.G. Van de Walle, Phys. Rev. B 76 (2007) 165202.
[4] A. Janotti, C.G. Van de Walle, J. Cryst. Growth 287 (2006) 58–65.
[5] F. Oba, A. Togo, I. Tanaka, J. Paier, G. Kresse, Phys. Rev. B. 77 (2008) 245202.
[6] M.S. Kim, Yong-Sung. Kim, C.H. Park, Curr. Appl. Phys. 11 (2011) S288–S291.
[7] F. Oba, M. Choi, A. Togo, A. Seko, I. Tanaka, J. Phys.: Condens. Matter 22 (2010) 384211.
[8] R. Vidya, P. Ravindran, H. Fjellvag, B.G. Svensson, E. Monakhov, M. Ganchenkova, R.M. Nieminen, Phys. Rev. B 83 (2011) 045206.
[9] F. Oba, M. Choi, A. Togo, I. Tanaka, Sci. Technol. Adv. Mater. 12 (2011) 034302.
[10] A.F. Kohan, G. Ceder, D. Morgan, C.G. Van de Walle, Phys. Rev. B. 61 (2000) 15019.
[11] C.G. Van de Walle, Phys. Rev. Lett. 85 (2000) 1012.
[12] C.G. Van de Walle, Phys. Status Solidi B 235 (2003) 89–95.
[13] K. Shimomura, K. Nishiyama, R. Kadono, Hyperfine Interact. 136/137 (2001) 659–662.
[14] Vl. Kolkovsky, L. Dobaczewski, K. BondeNielsen, V. Kolkovsky, A. Nylandsted Larsen, J. Weber, Physica B 404 (2009) 5080–5084.
[15] E.V. Lavrov, F. Börrnert, J. Weber, Physica B 376–377 (2006) 694–698.
[16] E.V. Lavrov, Physica B 404 (2009) 5075–5079.
[17] Y.J. Zeng, Z.Z. Ye, W.Z. Xu, D.Y. Li, J.G. Lu, L.P. Zhu, B.H. Zhao, Appl. Phys. Lett. 88 (2006) 062107.
[18] Ming-Lung Tu, Yan-Kuin Su, Chun-Yang Ma, J. Appl. Phys. 100 (2006) 053705.
[19] A. Allenic, W. Guo, Y. Chen, M.B. Katz, G. Zhao, Y. Che, Z. Hu, B. Liu S.B. Zhang, X. Pan, Adv. Mater. 19 (2007) 3333–3337.
[20] S.S. Lin, J.G. Lu, Z.Z. Ye, H.P. He, X.Q. Gu, L.X. Chen, J.Y. Huang B.H. Zhao, Solid State Commun. 148 (2008) 25–28.
[21] S.P. Wang, C.X. Shan, B.H. Li, J.Y. Zhang, B. Yao, D.Z. Shen, X.W. Fan J. Cryst. Growth 311 (2009) 3577–3580.
[22] M.A. Myers, J.H. Lee, Z. Bi, H. Wang, J. Phys.: Condens. Matter 24 (2012) 145802.
[23] T. Yamamoto, H. Katayama-Yoshida, Jpn. J. Appl. Phys. 38 (1999) L166–L169.
[24] Y.R. Sui, B. Yao, L. Xiao, L.L. Yang, J. Cao, X.F. Li, G.Z. Xing, J.H. Lang X.Y. Li, S.Q. Lv, X.W. Meng, X.Y. Liu, J.H. Yang, Appl. Surf. Sci. 287 (2013) 484–489.
[25] L. Chen, Z. Xiong, Q. Wan, D. Li, Opt. Mater. 32 (2010) 1216–1222.
[26] L. Duan, W. Zhang, X. Yu, Z. Jiang, L. Luan, Y. Chen, D. Li, Appl. Surf. Sci. 258 (2012) 10064–10067.
[27] R. Swapna, M.C. Santhosh Kumar, Ceram. Int. 39 (2013) 1799–1806.
[28] W. Li, C. Kong, G. Qin, H. Ruan, L. Fang, J. Alloy. Compd. 609 (2014) 173–177.
[29] Y.Z. Zhang, J.G. Lu, Z.Z. Ye, H.P. He, L.P. Zhu, B.H. Zhao, L. Wang, Appl. Surf. Sci. 254 (2008) 1993–1996.
[30] Q. Hou, J. Li, C.W. Zhao, C. Ying, Y. Zhang, Physica B 406 (2011) 1956–1960.
[31] T. Prasada Rao, M.C. Santhosh Kumar, J. Alloy. Compd. 509 (2011) 8676–8682.
[32] Y. Gai, G Tang, J. Li, J. Phys. Chem. Solids 72 (2011) 725–729.
[33] Y.R. Sui, B. Yao, Z Hua, G.Z. Xing, X.M. Huang, T. Yang, L.L. Gao T.T. Zhao, H.L. Pan, H. Zhu, W.W. Liu, T. Wu, J. Phys. D: Appl. Phys. 42 (2009) 065101.
[34] Z. Xiong, L. Chen, C. Zheng, L. Luo, Q. Wan, Scr. Mater. 63 (2010) 1069–1072.
[35] X. Tang, Y. Deng, D. Wagner, L. Yu, H. Lü, Solid State Commun. 152 (2012) 1–4.
[36] Q.Y. Zhu, Z.Z. Ye, G.D. Yuan, J.Y. Huang, L.P. Zhu, B.H. Zhao, J.G. Lu, Appl. Surf. Sci. 253 (2006) 1903–1906.
[37] J.G. Lu, L.P. Zhu, Z.Z. Ye, F. Zhuge, Y.J. Zeng, B.H. Zhao, D.W. Ma, Appl. Surf. Sci. 245 (2005) 109–113.
[38] C. Zhang, X. Li, J. Bian, W. Yu, X. Gao, Surf. Coat. Technol. 198 (2005) 253–256.
[39] T.N. Xu, X. Li, Z. Lu, Y.Y. Chen, C.H. Sui, H.Z. Wu, Appl. Surf. Sci. 316 (2014) 62–65.
[40] Y. Xu, T. Yang, B. Yao, Y.F. Li, Z.H. Ding, J.C. Li, H.Z. Wang, Z.Z. Zhang L.G. Zhangc, H.F. Zhao, D.Z. Shen, Ceram. Int. 40 (2014) 2161–2167.
[41] H. Wang, H.P. Ho, K.C. Lo, K.W. Cheah, J. Phys. D: Appl. Phys. 40 (2007) 4682–4685.
[42] P. Li, Sh.H. Deng, L. Zhang, Y.B. Li, X.Y. Zhang, J.R. Xu, Comput. Mater. Sci. 50 (2010) 153–157.
[43] E. Przeździecka, E. Kamińska, K.P. Korona, E. Dynowska, W. Dobrowolski, R. Jakieła, Ł. Kłopotowski, J. Kossut, Semicond. Sci. Technol. 22 (2007) 10–14.
[44] L. Balakrishnan, S. Gowrishankar, J. Elanchezhiyan, N. Gopalakrishnan, Physica B 406 (2011) 4447–4452.
[45] L. Cao, L. Zhu, Y. Li, M. Yang, Z. Ye, Mater. Lett. 86 (2012) 34–37.
[46] Quantum-ESPRESSO is a community project for high-quality quantum-simulation software, based on density-functional theory, and coordinated by paolo giannozzi, see, 〈http://www.quantum-espresso.org〉 and 〈http://www.pwscf.org〉.

[47] J.P. Perdew, K. Burke, M. Ernzerhof, Phys. Rev. Lett. 77 (1996) 3865–3868.

[48] Y. Yan, S.B. Zhang, S.T. Panelides, Phys. Rev. Lett. 86 (2001) 5723–5726.

[49] L. Honglin, L. Yingbo, L. Jinzhu, Y. Ke, Mater. Sci. Semicond. Process. 27 (2014) 599–604.

[50] X.M. Duan, C. Stampfl, M.M.M. Bilek, D.R. McKenzie, Phys. Rev. B 79 (2009) 235208.

[51] P. Li, S.H. Deng, Y.B. Li, J. Huang, G.H. Liu, L. Zhang, Physica B 406 (2011) 3125–3129.

[52] S.-M. Chou, M.-H. Hon, I.-C. Leu, Appl. Surf. Sci. 255 (2008) 2958–2962.

[53] A. Kumar, M. Kumar, B.P. Singh, Opt. Commun. 283 (2010) 3994–3997.

[54] J. Bang, Yi-Yang Sun, D. West, B.K. Meyerb, S. Zhang, J. Mater. Chem. C 3 (2015) 339–344.

[55] J. Bang, K.J. Chang, Appl. Phys. Lett. 92 (2008) 132109.

[56] J.G. Lu, S. Fujita, T. Kawaharamura, H. Nishinaka, Chem. Phys. Lett. 441 (2007) 68–71.

[57] M.G. Wardle, J.P. Goss, P.R. Briddon, Phys. Rev. Lett. 96 (2006) 205504.

[58] N.H. Nickel, Phys. Rev. B 73 (2006) 195204.