Theoretical prediction of lanthanum composition effects on structural, electronic and thermal properties of \( \text{La}_x\text{Sc}_{1-x}\text{N} \) alloys

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Abstract. In this study, we have explored the structural, electronic and thermal properties of \( \text{La}_x\text{Sc}_{1-x}\text{N} \) ternary alloys in rock-salt structure using the full-potential linearised augmented plane wave (FP-LAPW) method based on the density functional theory (DFT). To calculate the exchange-correlation energy and potential, we have used the Wu–Cohen generalised gradient approximation and the modified Becke–Johnson. We investigated the effect of composition on lattice parameters, bulk modulus and band gap. The variation of the calculated lattice constant with lanthanum composition is practically linear and shows a small deviation of the obtained results from Vegard’s law. Moreover, the effect of thermal macroscopic properties was also investigated employing the quasiharmonic Debye model, which takes into account the lattice vibrations. We have found a good accord between our results, the experimental data and previous theoretical results available in the literature for the binary compounds which can be a support for the ternary alloys in the future.

Keywords. Density functional theory; full-potential linearised augmented plane wave; electronic band structure; thermal properties; Debye model.

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1. Introduction

Different physical properties can be studied using quantum mechanical methods such as the first principle calculation which is the most used approach in the simulation methods that are framed with the density functional theory (DFT). The most used quantum mechanical approach is the DFT functional and it can be applied in a majority of atoms and molecules (finite systems) and surfaces and solids (infinite systems). This computer technique can determine different properties of solids such as crystal structure. It has diverse methods with low calculation rates and reasonable precision [1].

In recent years, attention has been given to alloys and their electronic, structural, elastic and thermodynamic properties. Thermophysical properties such as coefficient of thermal expansion, isothermal bulk modulus, Grüneisen parameter, heat capacity and Debye temperature have attracted the attention of researchers for many years, especially for alloys. Studying these properties enables us to develop these alloys to become more effective and useful in industrial and electronic fields [2–5]. Many researchers were inspired by the early transition metal, (mono) nitride compounds which crystallise in rock-salt structure (B1) at normal temperature and pressure with the possibility of a transition to the CsCl structure (B2) at high pressures and these new structures which can be metallic or semiconducting, have high hardness, mechanical strength, high superconducting transition temperatures and relatively high melting point. These properties qualify them to be used in hard coatings, magnetic storage devices and cutting tools, thanks to their excellent physical properties [6–8].

The structural and electronic properties of \( \text{ScN} \) have been determined by Takeuchi et al using first-principles calculations in the rock-salt, cesium chloride, nickel arsenide, zinc-blende and wurtzite structures [9,10]. In 2019, the structural and electronic properties of \( \text{ScN}, \text{YN}, \text{LaN} \) and \( \text{LuN} \) have been studied by Winiarski and Kowalska [11] using DFT. Ghezali et al [12] investigated the structural and electronic properties of \( \text{LaN} \) compound using two different methods: a version of the full potential linear muffin-tin orbitals method (FP-LMTO) and the full potential linear augmented plane
waves (FP-LAPW). Then, Gueddam et al [13] studied the parameters of crystal structure and elastic coefficients of LaN compound at null and increased pressure and Louhadj et al [14] studied the electronic band structures of ScN, YN, LaN and GdN compounds employing FP-LMTO.

LaN compound is one of the most important materials which attracted researchers’ attention. It is a transition metal nitride and sometimes it can be a superconductor. It is crystallised in the rock salt (B1) phase and under pressure has a probable phase change to the CsCl [12]. The ScN compound has a rock salt structure (B1) with a possible transition to the CsCl (B2) phase as a function of pressure and both possibly have a metastable wurtzite (B4) phase [15].

This study aims to calculate the structural, electronic and thermal properties of La$_{1-x}$Sc$_x$N alloys for $x$ ranging from 0, 0.25, 0.5, 0.75 to 1. The purpose of this study is also to investigate the effect of lanthanum composition on the structural, electronic and thermal properties of La$_{1-x}$Sc$_x$N alloys. The existing literature on the basic properties of La$_{1-x}$Sc$_x$N can be used as references for future experimental research on these systems. This allows the development of novel materials and improves the limitation of existing devices in electronic and optoelectronic technological applications. This study is accomplished by the Wu–Cohen generalised gradient approximations (WC-GGA) and modified Becke–Johnson (mBJ), and included the FP-LAPW method in the framework of DFT. In §2, the method of calculation is briefly outlined. The theoretical results obtained for the structural, electronic and thermal properties of La$_{1-x}$Sc$_x$N ternary alloys and discussion are presented in §3, followed by a conclusion of this work in §4.

2. Method of calculation

The total energy was calculated using the scalar non-relativistic FP-LAPW [16] method as implemented in the Wien2k computational package [17], based on DFT [18,19]. The exchange-correlation potential was treated using the WC-GGA approach [20] and the mBJ potential was used [21,22] to calculate the electronic structure. We have applied the special quasirandom structure (SQS) technique proposed by Zunger et al [23] to reproduce the randomness of the alloys for some initial shells about a given site. The SQS scheme is reasonably precise to describe the physical properties that are not affected by the errors introduced using the concept of periodicity beyond the first shells. In the FP-LAPW method, the electron wave function was expended inside the muffin-tin (MT) spheres in a mixture with a cut-off $l_{\text{max}} = 10$ as spherical harmonic functions. Also, the charge density was Fourier expended up to $G_{\text{max}} = 12$ (Ryd)$^{1/2}$. $R_{\text{MT}} K_{\text{max}}$ was determined to be 8, where $R_{\text{MT}}$ is the plane-wave radius and $K_{\text{max}}$ is the cut-off for wave function basis and we chose the energy between the core and the valence states for a cut-off of about $-6.0$ Ry. The values of muffin-tin radii ($R_{\text{MT}}$) were chosen to be 2.55, 2 and 1.7 (a.u.) for the La, Sc and N atoms respectively. An irreducible Brillouin zone mesh of 1331 special K points has been used for both binary and ternary alloys. We have studied the thermal effects employing the quasiharmonic Debye model [24] implemented in the code Gibbs2 program.

3. Result and discussion

3.1 Structural properties

Firstly, we determined the structural properties of the binary compounds LaN, ScN in rock-salt structure employing the WC-GGA approximation, and the ternary alloys La$_{1-x}$Sc$_x$N were modelled at different concentrations, as recurrent supercells for the lanthanum concentrations $x = 0.25, 0.5$ and 0.75 periodically with the described ordered structures. Thus, the structure of ternary alloys is determined by supercells of 8 atoms corresponding to $1 \times 1 \times 1$ single cells. We replaced the atoms of scandium with those of lanthanum to obtain the desired concentration. The structural optimisation was realised by minimising the total energy of cell parameters and the atomic positions. The structural optimisation was realised by minimising the total energy of cell parameters and the atomic positions to determine the optimal lattice parameter ($a$), the bulk modulus ($B$) and its pressure derivative ($B'$). We calculated the total energies at various volumes in the region of equilibrium and using the Birch–Murnaghan equation of state [25] we fitted the values. The calculated structural parameters for La$_x$Sc$_{1-x}$N at different compositions $x = 0, 0.25, 0.5, 0.75$ and 1 employing the WC-GGA approach are presented in table 1, with existing experimental measurements and theoretical results reported in the literature for comparison. Our computed structural parameters for binary compounds ScN and LaN are similar with very little variation compared to the theoretical works reported previously. Compared to the experimental results, the deviation of our results from lattice constants is less than 0.7% and 0.8% for ScN and LaN respectively, which are weakly overestimated with respect to the experiment. These results confirm the usual trend of GGA approximation to overestimate the lattice constant value [26–28], whereas the bulk modulus value of ScN is in excellent accord with our results.
Table 1. Computed lattice constants \( a_0 \) (Å), bulk modulus \( B \) (GPa) and its pressure derivative of \( \text{La}_x\text{Sc}_{1-x}\text{N}. \)

<table>
<thead>
<tr>
<th>( x )</th>
<th>This study</th>
<th>Other calculations</th>
<th>Exp.</th>
<th>This study</th>
<th>Other calculations</th>
<th>Exp.</th>
<th>This study</th>
<th>Other calculations</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>4.471</td>
<td>4.516(^e), 4.54(^d)</td>
<td>4.44(^a)</td>
<td>213.001</td>
<td>201(^d)</td>
<td></td>
<td>4.158</td>
<td>3.31(^d)</td>
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<tr>
<td></td>
<td></td>
<td>4.48(^e), 4.50(^f)</td>
<td>4.51(^b)</td>
<td>215.86(^e), 235(^f)</td>
<td></td>
<td></td>
<td></td>
<td>3.926</td>
</tr>
<tr>
<td>0.250</td>
<td>4.728</td>
<td>–</td>
<td>–</td>
<td>168.444</td>
<td>–</td>
<td></td>
<td></td>
<td>3.897</td>
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<td>0.500</td>
<td>4.938</td>
<td>–</td>
<td>–</td>
<td>148.446</td>
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</tr>
<tr>
<td>1</td>
<td>5.257</td>
<td>5.26(^e), 5.3(^g)</td>
<td>5.32(^f)</td>
<td>130.11</td>
<td>130.92(^e)</td>
<td>112(^h)</td>
<td>4.124</td>
<td>3.84(^e)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>5.28(^i)</td>
<td></td>
<td></td>
<td>4.01(^i)</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

\(^a\)Ref. [32], \(^b\)ref. [33], \(^c\)ref. [34], \(^d\)ref. [9], \(^e\)ref. [7], \(^f\)ref. [6], \(^g\)ref. [35], \(^h\)ref. [29] and \(^i\)ref. [36].

and that of 215.86 GPa calculated by Shoaib et al in [7] using the DFT-based FP-LAPW method. As for \( \text{LaN} \), our results are 13\% larger than the experimental value of 112 GPa presented in [29]. On the other hand, we noticed that the lattice parameter for \( \text{LaN} (x = 1) \) is larger than \( \text{ScN} (x = 0) \), \( a(\text{LaN}) > a(\text{ScN}) \), using the same anion atom in both compounds. These results can be clarified by taking into consideration the atomic radii of the \( \text{La} \) and \( \text{Sc} \) atoms, which are respectively given by \( R(\text{La} = 2.74 \text{ Å}) \) and \( R(\text{Sc} = 2.09 \text{ Å}) \).

The value of bulk modulus for \( \text{ScN} \) is bigger than that of \( \text{LaN} \), \( B(\text{ScN}) > B(\text{LaN}) \). The inverse relationship with \( a(\text{LaN}) > a(\text{ScN}) \) is in agreement with the well-known relation between \( B \) and the lattice constant \( B \propto V_0^{-1} \), where \( V_0 \) is the volume of the unit cell [30]. Because of the absence of experimental and theoretical works concerning lattice parameters and bulk modulus for alloy composition \( 0 < x < 1 \), our results for this composition can be considered as the predicted work and may be used as references in the future.

The variation of lattice parameters for \( \text{La}_x\text{Sc}_{1-x}\text{N} \) alloy with \( \text{La} \) concentration \( x \) is seen in figure 1. This variation is approximately linear and shows an important deviation from Vegard’s law [31] with the downward bowing parameter as \(-0.297 \text{ Å}\); this behaviour is due to the mismatches of lattice constants of \( \text{ScN} \) and \( \text{LaN} \) compounds. In figure 2, we display the variation of bulk modulus with \( x \) for \( \text{La}_x\text{Sc}_{1-x}\text{N} \). Between different values of bulk modulus of the extreme binary compounds \( \text{ScN} \) and \( \text{LaN} \), we remark a deviation of the bulk modulus with downward bowing as 96.691 GPa. This difference is generally caused by the mismatch of the bulk modulus of binary compounds. Moreover, the decrease of bulk modulus with the increase of composition from 0 to 1 indicates that the \( \text{La}_x\text{Sc}_{1-x}\text{N} \) alloys become more compressible as \( x \) increases.

![Figure 1. Lattice parameter as a function of La concentration (filled squares) compared with Vegard’s law (dashed line).](image1)

![Figure 2. Bulk modulus as a function of La concentration (filled squares) compared with the linear composition dependence prediction (dashed line).](image2)
3.2 Electronic properties

We have computed the electronic band structures of ScN and LaN binary compounds and their ternary alloys La$_x$Sc$_{1-x}$N in rock-salt structure for $x = 0.25$, 0.5 and 0.75 using the optimised value of lattice parameter along the high symmetry directions in the first Brillouin zone within both WC-GGA and mBJ approaches.

The band structures of ScN and LaN compounds obtained by mBJ approximation are displayed in figure 3. In figure 3a, we can see that the indirect band gap of ScN, $E_g(\Gamma - X) = 0.918$ eV. Our results are in accord with the previous LDA (mBJ) 0.9–1.0 eV [21,37] and 1.0 eV [38] works. Interestingly, the optical spectroscopy measurements for ScN suggested a higher experimental band gap of 1.3 eV [39]. On the other hand, the electronic band structure of LaN is also represented in figure 3b. We noticed that the highest valence band (VB) is located at the X high symmetry point and the smallest conduction band (CB) is situated at the X point in the Brillouin zone. This is in agreement with the fact that LaN is a direct (X-X) band-gap semiconductor with a value of 0.694 eV, which is close to the experimental value of band gap $E_{g\text{exp}} = 0.75$ eV [6] and is in good accord with the theoretical value of 0.73 eV [7] and 0.59 eV [11] results.

Otherwise, the binary compounds have metallic characters using WC-GGA approximation. We can note from the calculated values of the electronic band gap, that the variance compared to the experimental value is probably due to the GGA potential of exchange and correlation included in the density functional theory which underestimates the value of the energy gap in semiconductors, because of the self-interaction error and the absence of derivative discontinuity in the exchange and correlation potential. For this reason, the energy band gap can be underestimated up to 50% [40–42].

The electronic band structure of ternary alloys La$_x$Sc$_{1-x}$N with La concentrations $x (0 < x < 1)$ within the TB-mBJ approximation are displayed in figure 4. We can see that the direct band gaps of La$_{0.5}$Sc$_{0.5}$N and La$_{0.75}$Sc$_{0.25}$N at $\Gamma$ point are equal to 0.02 eV and 0.38 eV respectively, while the La$_{0.25}$Sc$_{0.75}$N alloy indicates the metallic character with a negative gap of $-0.004$ eV. The calculated band structure using WC-GGA approximation indicates the metallic character for La compositions $x (0 < x < 1)$ in La$_x$Sc$_{1-x}$N. Our calculated band-gap energies for La$_x$Sc$_{1-x}$N ternary alloys at different concentrations using the mBJ approach and a comparison of these values with the experimental and the previously published theoretical results are presented in table 2.
Figure 4. Band structure of La$_x$Sc$_{1-x}$N ternary alloys ($x = 0.25, 0.5$ and $0.75$) calculated using the mBJ approximation.
Table 2. The calculated band-gap energy (in eV) at high symmetry points for La$_x$Sc$_{1-x}$N ternary alloys at different values of $x$ computed using mBJ.

<table>
<thead>
<tr>
<th>$x$</th>
<th>This study</th>
<th>Exp</th>
<th>Other</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.918 ($\Gamma$ – X)</td>
<td>1.3$^a$</td>
<td>0.9$^b$, 1.0$^c$, 1.0$^d$</td>
</tr>
<tr>
<td>0.25</td>
<td>–0.004 ($\Gamma$ – $\Gamma$)</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>0.5</td>
<td>0.02 ($\Gamma$ – $\Gamma$)</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>0.75</td>
<td>0.38 ($\Gamma$ – $\Gamma$)</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>1</td>
<td>0.694 (X–X)</td>
<td>0.75$^e$</td>
<td>0.73$^f$, 0.59$^g$</td>
</tr>
</tbody>
</table>

$^a$Ref. [39], $^b$ref. [37], $^c$ref. [21], $^d$ref. [38], $^e$ref. [6], $^f$ref. [7] and $^g$ref. [11].

$x = 0.25$, 0.5 and 0.75 in La$_x$Sc$_{1-x}$N ternary alloys, our results are only predictions.

To clarify the character of the band structures of La$_x$Sc$_{1-x}$N ternary alloys, we have computed the partial density of states at different La concentrations using the mBJ approach, and our results are presented in figure 5. Three separate regions for the partial density of states of La atoms and p, d states of Sc atoms. We observed that the metallic character of the La$_x$Sc$_{1-x}$N alloy is principally determined by the d states of La atoms and p, d states of Sc atoms.

For La$_{0.25}$Sc$_{0.75}$N, the lowest valence band in the energy range around $-16.5$ eV up to $14.5$ eV and around $-14$ eV to $10.5$ eV, are due to Sc-p states, while the s states of N are localised around $-14.5$ eV to $-14$ eV. The highest valence band near the Fermi level around $-5$ eV to $0$ eV is mainly derived from N/La-p states and the weak contribution of La-d states and Sc-p/d states. The conduction band is formed by Sc-d states and a small contribution of La-p/d states and N-s/p states. We observed that the metallic character of the La$_{0.25}$Sc$_{0.75}$N alloy is principally determined by the d states of La atoms and p, d states of Sc atoms.

For La$_{0.5}$Sc$_{0.5}$N, the lowest valence band extending between $-16.5$ and $-14.5$ eV and between $-14$ and $-10.5$ eV is due to the La/Sc/N-s states, around $-14$ to $-14$ eV is derived by a small contribution of Sc-s/p/d states. The highest valence band extending from $-5$ to $0$ eV is mainly formed by La/Sc/N-p states and a contribution of La/Sc-d states. The conduction band is formed by mixed La/Sc-p states and N-s states.

For La$_{0.75}$Sc$_{0.25}$N the contribution of La/Sc/N-s states is observed in the lowest valence band between $-16.5$ and $-14$ eV and between $-13$ and $-11$ eV, but the highest valence band near the Fermi level between $-4$ and $0$ eV is derived from La/Sc/N-p states and a weak contribution of La/Sc-d states. The conduction band is characterised by the mixed La/Sc-p and d states and N-p states.

3.3 Thermal properties

It is necessary to study the thermal properties of materials to determine their specific behaviour when they are submitted to constraints such as high temperature and high pressure. In this section, we use the quasi-harmonic Debye model of crystals [24] implemented in the Gibbs2 program and combined with the FP-LAPW method, to determine the thermal properties of La$_x$Sc$_{1-x}$N alloys. The energy volume data are calculated from the Britch–Murnaghan equation of states using WC-GGA approximation, and these data are the input data to the Gibbs2 program to calculate the thermal properties of the La$_x$Sc$_{1-x}$N alloys as a function of temperature, in the range of 0–2000 K. The present study of the thermal properties is the first theoretical prediction in our case of the studied alloys.

Figures 6 and 7 present the variation of the volume and bulk modulus of La$_x$Sc$_{1-x}$N alloys at 0 GPa, respectively. We notice that the effect of temperature $T$ on volume at every concentration is almost stable with the temperature rise. However, the bulk modulus varies linearly in a very moderate way for La concentrations at $x$ ($0 < x < 1$) in La$_x$Sc$_{1-x}$N but for the binary compounds, it is nearly constant from 0 to 200 K, and diminishes when $T > 200$ K. Therefore, the binary compounds ScN and LaN become more compressible with increasing temperature.

In figure 8, we present the effect of temperature $T$ on the Debye temperature $\theta_D$ for the La$_x$Sc$_{1-x}$N alloys. It can be noticed clearly from this figure that the value of Debye temperature is almost constant for $x = 0.25, 0.5$ and 0.75, but for binary compounds at $T > 200$ K, $\theta_D$ decreases linearly with increasing temperature. From figure 9, we can notice that for a given temperature $T$, $\theta_D$ increases linearly with increasing pressure. Our results show that ScN has a higher $\theta_D$ than LaN, and we can predict that the value of bulk modulus is higher for ScN than for LaN, suggesting that ScN is harder than LaN.

The computed heat capacity ($C_V$) for the La$_x$Sc$_{1-x}$N alloys at 0 GPa is presented in figure 10. The evolution of $C_V$ of La$_x$Sc$_{1-x}$N for $x = 0, 0.25, 0.5$ and 0.75 is similar. Also at $T = 0$ K, $C_V$ tends to zero, and increases with $T^3$ at $T < 1600$ K whereas LaN increases with $T^3$ at $T < 1400$ K. The heat capacity ($C_V$) results depend on temperature, due to the use of harmonic approximation within the Debye model which is used in our calculation. Otherwise, at higher temperatures, the harmonic effect on heat capacity vanishes, and also tends to a constant value called Dulong–Petit limit, which is similar for different materials at higher temperatures. Figure 11 determines the results for $C_V$ as a function of pressure, indicating that, at different concentrations,
Figure 5. Partial density of states of La$_x$Sc$_{1-x}$N ternary alloys ($x = 0.25, 0.5$ and $0.75$) calculated using the mBJ approximation.

Figure 6. Variation of volume with temperature for La$_x$Sc$_{1-x}$N alloys.

Figure 7. Variation of bulk modulus with temperature for La$_x$Sc$_{1-x}$N alloys.

The temperature decreases approximately with increasing applied pressure.

The variations of heat capacity ($C_p$) of La$_x$Sc$_{1-x}$N alloys at different concentrations with temperature and pressure are presented in figures 12 and 13 respectively. From figure 12, we observe that the variation of $C_p$ at low temperature is similar to that heat capacity $C_V$. However, at high temperatures, $C_p$ exhibits a different characteristic, and the value of heat capacity does not converge to a constant value. It can be noticed from figure 13 that for a given temperature, $C_p$ decreases approximately linearly with increasing pressure.

The variation of entropy ($S$) with temperature is shown in figure 14. We can observe that $S$ increases when the temperature increases for all compositions because the increase in temperature increases the vibrational contribution to the entropy.

4. Conclusion

In this paper, we have studied the structural, electronic and thermal properties of La$_x$Sc$_{1-x}$N ternary alloys by employing the FP-LAPW method within the WC-GGA and mBJ approximations. The calculated equilibrium lattice parameters and bulk modulus for the ScN and
Figure 8. Variation of Debye temperature with temperature for La$_x$Sc$_{1-x}$N alloys.

Figure 9. Variation of Debye temperature with pressure for La$_x$Sc$_{1-x}$N alloys.

Figure 10. Variation of heat capacity ($C_V$) with temperature for La$_x$Sc$_{1-x}$N alloys.

Figure 11. Variation of heat capacity ($C_V$) with pressure for La$_x$Sc$_{1-x}$N alloys.

Figure 12. Variation of heat capacity ($C_p$) with temperature for La$_x$Sc$_{1-x}$N alloys.

Figure 13. Variation of heat capacity ($C_p$) with pressure for La$_x$Sc$_{1-x}$N alloys.
LaN were found to be in good agreement with the published theoretical and experimental results. The variation of the lattice parameter with La concentration has a small deviation from Vegard’s law. The values of bulk modulus increase with alloy composition $x$. The use of the mBJ approximation considerably improved the electronic band-gap energy in comparison with experimental results. We have confirmed that both ScN and LaN are semiconductors with an indirect gap ($\Gamma$–X) and a direct gap (X–X) respectively. The band gap is found to be direct ($\Gamma$–$\Gamma$) as the concentration increases from 0.5 and 0.75 but the alloy composition with $x = 0.25$ is metallic with a negative gap. The character of the band structure for different values of $x$ was discussed from the standpoint of the partial density of states. The quasiharmonic Debye model was successfully used to determine thermal properties at different temperatures from 0 to 2000 K and pressures from 0 to 20 GPa. The results presented in this manuscript are predictions and can be served as references for future experimental studies on these systems. In future works, we look to explore other properties for the same compounds.

References

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