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Structural, elastic, thermodynamic and electronic properties of LuX (X = N, Bi and Sb) compounds: first principles calculations

Mohammed Ameria, Faiza Bennara, Slamani Amela, Ibrahim Amerib, Y. Al-Douri cd and Dinesh Varshneye

aLaboratoire Physico-Chimie des Matériaux Avancés (LPCMA), Université Djillali Liabès de Sidi Bel-Abbès, Sidi Bel-Abbès, Algeria; bFaculty of Exact Sciences, Department of Physics, Djillali Liabes University, Sidi Bel Abbes, Algeria; cInstitute of Nano Electronic Engineering, University Malaysia Perlis, Kangar, Malaysia; dFaculty of Science, Physics Department, University of Sidi-Bel-Abbes, Algeria; eMaterials Science Laboratory, School of Physics, Vigyan Bhavan, Devi Ahilya University, Indore, India

ABSTRACT
The structural, electronic, elastic and thermodynamic properties of LuX (X = N, Bi and Sb) based on rare earth into phases, Rocksalt (B1) and CsCl (B2) have been investigated using full-potential linearized muffin-tin orbital method (FP-LMTO) within density functional theory. Local density approximation (LDA) for exchange-correlation potential and local spin density approximation (LSDA) are employed. The structural parameters as lattice parameters $a_0$, bulk modulus $B$, its pressure derivate $B'$ and cut-off energy ($E_c$) within LDA and LSDA are presented. The elastic constants were derived from the stress—strain relation at 0 K. The thermodynamic properties for LuX using the quasi-harmonic Debye model are studied. The temperature and pressure variation of volume, bulk modulus, thermal expansion coefficient, heat capacities, Debye temperature and Gibbs free energy at different pressures (0—50 GPa) and temperatures (0—1600 K) are predicted. The calculated results are in accordance with other data.

1. Introduction
There is a growing interest in studying the electronic properties of rare earth (RE) pnictides due to their magnetic and electrical properties. Foremost the RE monopnictides and monochalcogenides have been widely studied under high pressure.[1—5] These incompletely filled f-electron shells of the lanthanide ions are forcefully relocated and, therefore, interact strongly with the lattice. Such effect has been interpreted in terms of promotion of 4f electron of RE ion to the 5d conduction band and the fusion of f states with the p states of the neighboring ion. Many theoretical works [6,7] have focused on condensed matter researches because of their diversive and unusual structural, magnetic and phonon properties. Some pnictides of lanthanide group, which crystallize in NaCl (B1) type crystal structure, have been investigated under high-pressure of X-ray diffraction technique.[8—13] The electronic structures of lighter RE mono-pnictides have been studied theoretically with great details.[14,15] Lanthanum has a large number of theoretical and experimental investigations that is reported on lanthanum compounds.[16,17] Amongst the RE mono-pnictides, Gd mono-pnictides (GdX; X = N, P, As,
Sb, Bi) are the simplest series,\cite{18,19} they are known to exhibit unusual electric and magnetic properties.\cite{20,21,22} We note that these materials have wide applications in scientific and industrial sectors as nonlinear optics, electro-optic components, glass-making, grinding alloys, composites lasers, phosphorous lasers and electronic devices. We are interested in lutetium (Lu); this is an RE of the lanthanide series. The aim of our work is to study structural, elastic and thermodynamic properties to determine the equilibrium lattice constants, bulk modulus, and first pressure derivative in B1, B2 structures in the version of local density approximation (LDA) and local spin density approximation (LSDA), of three binary LuX (X = N, Bi and Sb) using the full-potential linearized muffin-tin orbital (FP-LMTO) methods. The paper is organized as follows. Section 2 presents briefly the method of calculations. Section 3 displays the results and discussions and finally the conclusion is given in Section 4.

2. Computational method

Computations depicted here, were performed using the FP-LMTO method within. The density functional theory (DFT) and implemented in the Lmtart computer code in this method, the space is divided in two parts: an interstitial regions (IR) and non-overlapping (MT) spheres centered on the atomic sites. In the IR regions, the Fourier series represent the basic function. In side on MT spheres, the basic set is treated as a linear combination of radial functions times spherical harmonics. The charge density and the potential are represented inside the muffin-tin sphere radius by spherical harmonics up to \(l_{\text{max}} = 6\). The exchange-correlation (XC) effects are treated with the LDA using the scheme developed by Perdew and Wang and the LSDA. The \(k\)-integrations over the Brillouin zone (BZ) is carried out up to a (6,6,6) grid in the irreducible Brillouin zone, using the tetrahedron method the values of the sphere radii, energy cut-off and the number of PWs used in our calculations are listed in Table 1.

![Table 1. The plane wave number PW, cut-off energy (Ry) and the muffin-tin radius (RMT) (a.u.) used in calculation for LuN, LuBi and LuSb in B1, B2 structures in LDA and LSDA.](image)

3. Results and discussion

3.1. Structural properties

It is useful to calculate the structural properties of LuX (X = N, Bi and Sb) compounds to determine the equilibrium lattice constants, bulk modulus and first pressure derivate of bulk modulus that are computed by minimizing the total energies for different values of lattice constant by means of Murnaghan’s equation of state (EOS) [23] in B1 and B2 phases using LDA and LSDA. The values of lattice parameters, bulk modulus and the first derivate of bulk modulus are summarized in the Table 2 for each structure of LuX (X = N, Bi and Sb) in the two approximations (LDA and LSDA). Figure 1 illustrates the energy based on volume of structures (NaCl-type B1 and B2-type CsCl), which clearly shows that the minimum energy ration is marked for B1 structure (NaCl type) for LuX (X = Sb and Bi) using LSDA, while for the LuN crystallizes in the same structure utilizing LDA. For the present values of lattice parameters in LDA, for B1 structure of LuN, LuBi and LuSb compounds are 4.826, 6.105 and 5.903 Å, respectively. The lattice parameters in LSDA for B1 of LuN, LuBi and LuSb are 4.827, 6.106 and 5.921 Å, respectively, as given in Table 2. The present values of bulk modulus in LDA are found to be 391.102, 70.721 and 97.775 GPa for B1 structure of LuN, LuBi and LuSb compounds, respectively. In LSDA, the values of bulk modulus are 385.809, 69.692 and 91.894 GPa for LuN, LuBi and LuSb, respectively. The same variation is recorded for the pressure derivate of bulk modulus for B1 structure of LuN, LuBi and LuSb compounds as obtained in Table 2 are 3.72, 3.59 and 3.47, respectively. But in LSDA, the recorded values are 5.83, 4.98 and 5.10. Our results are in accordance with other available data.[24–26]

3.2. Electronic properties

We study the electronic properties using the method FP-LMTO, performing the calculations of the band structure for the two compounds LuBi and LuSb using LSDA, and for LuN using LDA. In the most stable phase B1, the calculated band structures are defined in Figure 2 for LuBi, LuSb and LuN. Figure 2 shows LuBi and LuSb are metallic. The electron configuration

Table 2. The values of equilibrium lattice constant (a₀), bulk modulus (B) and the pressure derivate of bulk modulus (B’) of LuN, LuBi and LuSb in B1, B2 structures in LDA and LSDA.

<table>
<thead>
<tr>
<th>Material</th>
<th>Structure</th>
<th>Reference</th>
<th>a (Å)</th>
<th>B (GPa)</th>
<th>B’</th>
</tr>
</thead>
<tbody>
<tr>
<td>LuN</td>
<td>B1</td>
<td>Present (LDA)</td>
<td>4.826</td>
<td>391.102</td>
<td>3.72</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Present (LSDA)</td>
<td>4.827</td>
<td>385.809</td>
<td>5.83</td>
</tr>
<tr>
<td></td>
<td>B2</td>
<td>Present (LDA)</td>
<td>2.963</td>
<td>345.963</td>
<td>4.14</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Present (LSDA)</td>
<td>2.975</td>
<td>422.273</td>
<td>3.87</td>
</tr>
<tr>
<td>LuBi</td>
<td>B1</td>
<td>Present (LDA)</td>
<td>6.105</td>
<td>70.721</td>
<td>3.59</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Present (LSDA)</td>
<td>6.106</td>
<td>69.692</td>
<td>4.98</td>
</tr>
<tr>
<td></td>
<td>B2</td>
<td>Present (LDA)</td>
<td>3.772</td>
<td>72.683</td>
<td>5.16</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Present (LSDA)</td>
<td>3.769</td>
<td>74.103</td>
<td>5.59</td>
</tr>
<tr>
<td>LuSb</td>
<td>B1</td>
<td>Present (LDA)</td>
<td>5.903</td>
<td>97.775</td>
<td>3.47</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Present (LSDA)</td>
<td>5.921</td>
<td>91.894</td>
<td>5.10</td>
</tr>
<tr>
<td></td>
<td>Exp.</td>
<td>6.040ᵃ</td>
<td>53 ± 4ᵇ</td>
<td>6 ± 8ᵇ</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Theo.</td>
<td>6.113ᶜ</td>
<td>58.0ᶜ</td>
<td>3.75ᶜ</td>
<td></td>
</tr>
<tr>
<td></td>
<td>B2</td>
<td>Present (LDA)</td>
<td>3.653</td>
<td>111.596</td>
<td>4.48</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Present (LSDA)</td>
<td>3.645</td>
<td>108.361</td>
<td>5.73</td>
</tr>
<tr>
<td></td>
<td>Exp.</td>
<td>3.719ᵇ</td>
<td>66.40ᵇ</td>
<td>4.11ᵇ</td>
<td></td>
</tr>
</tbody>
</table>

of the atoms marks the state d for the chemical elements Sb and Bi and the state f and d for the chemical element Lu, duplication of spin up and spin down with respect to the Fermi energy is noticed and seen the poverty of the N atom in electron. The topography of the LuN band structure clearly shows a gap which means these materials are semiconductors. As N is poor of electrons, LuN is a semiconductor indirect gap near x-\(\gamma\) with a value of 0.0568. Figure 3 displays the density of states for LuBi, LuSb and LuN compounds. The three compounds revealed indirect characteristics of electronic properties and support the gap nature of the band structures as shown in Figure 2, followed by structural behavior of LuN, LuBi and LuSb compounds.

### 3.3. Elastic properties

The elastic constants provide very important information about the mechanical and dynamical properties such as inter atomic potentials, EOS and phonon spectra. Elastic properties contribute in
thermodynamic properties such as specific heat, thermal expansion, Debye temperature, melting point and Grüneisen parameter.\cite{27–29} Materials with cubic symmetry have only three independent elastic constants $C_{11}$, $C_{12}$ and $C_{44}$ which should be calculated. The elastic constants $C_{ij}$ are obtained by calculating the total energy as a function of volume-conserving strains using Mehl method.\cite{30,31} To calculate $C_{11}$ and $C_{12}$, a volume-conserving orthorhombic strain tensor is applied by the following expression:

$$
\vec{\epsilon} = \begin{bmatrix}
\delta & 0 & 0 \\
0 & \delta & 0 \\
0 & 0 & \frac{1}{(1-\delta)^2} - 1
\end{bmatrix}
$$

The strain application influences the total energy and expressed as

$$
E(\delta) = E(-\delta) = E(0) + 6(C_{11} - C_{12})V_0\delta^2 + O[\delta^4]
$$
The bulk modulus is

\[ B = \frac{(C_{11} + 2C_{12})}{3} \]

(3)

For identification of \( C_{44} \), we used a monoclinic strain conserving volume defined as

\[ \bar{\varepsilon} = \begin{bmatrix} 1 & \frac{\delta}{2} & 0 \\ \frac{\delta}{2} & 1 & 0 \\ 0 & 0 & \frac{4}{(4 - \delta^2)} \end{bmatrix} \]

(4)

The total energy is thus

\[ E(\delta) = E(-\delta) = E(0) + \frac{1}{2} C_{44} V_0 \delta^2 + O[\delta^4] \]

(5)
Figure 4 shows the evolution of the total energy as a function of $\delta^2$. We determine the elastic constants, which are very important characteristics parameters. They are linked to the bulk modulus $B$, shear modulus $G$, Young’s modulus $E$ and Poisson’s ratio $\nu$, that are frequently measured for approaches line materials when investigating their hardness. To study the stability of our compounds in B1 structure, we have calculated the elastic constants $C_{ij}$ at the equilibrium lattice constant as listed in Table 3 for LuN, LuBi and LuSb compounds. The requirement of the mechanical stability of the cubic structure results in the following restrictions on the elastic constants must
check the Born—Huang criteria stability [32]:

\[ C_{11} - C_{12} > 0, C_{11} > 0, C_{44} > 0 \text{ and } (C_{11} + 2C_{12}) > 0 \]  

We have found that in the structure B1, the Born—Huang criteria are satisfied and approach is mechanically stable. The bulk modulus $B$, shear modulus $G$, Young’s modulus $E$ and Poisson’s ratio $\nu$ are the most interesting elastic properties for approach line materials when investigating their hardness. With the knowledge of bulk modulus $B$, shear modulus $G$, Young’s modulus $E$ and Poisson’s ratio $\nu$ are also calculated in terms of the computed data using the following relations as given in Table 3.

The Zener anisotropy factor is a measure of the degree of anisotropy in solid. It takes the value 1 for a completely isotropic material. For the $A$ values smaller or greater than unity, it is measured degree of elastic anisotropy. The calculated Zener anisotropy factor ($A$) from the present calculated values of the elastic constants is defined by the given relation [33]: $A = 2 \frac{C_{44}}{(C_{11}-C_{12})}$. The calculated anisotropy factor is given in Table 3. The Zener anisotropy calculated factors show that $A$ is smaller than 1 for LuN (0.796) and greater than 1 for LuBi (1.570) and
LuSb (1.468) as given in Table 3. The shear modulus $G$, Young’s modulus $E$ and Poisson’s ratio $\nu$ are often measured for approach line materials, and calculated in terms of the computed elastic constants $C_{ij}$ using the relations (1)–(3). The shear modulus $G$ is a measure of resistance to reversible deformations upon shear stress, $G$ is the ratio of shear stress to the shear strain:

$$G = \frac{C_{11} - C_{12} + 3C_{44}}{5}$$

Table 3. Calculated elastic constant $C_{11}, C_{12}, C_{44}$ (in GPa), bulk modulus (in GPa), Young $E$ and shear modulus $G$ (in GPa), Poisson’s ratio $\nu$, and the anisotropic $A$ for LuX ($X = N, Bi$ and Sb).

<table>
<thead>
<tr>
<th>Material</th>
<th>Reference</th>
<th>$C_{11}$</th>
<th>$C_{12}$</th>
<th>$C_{44}$</th>
<th>$C_{11} - C_{12}$</th>
<th>$B$</th>
<th>$G$</th>
<th>$E$</th>
<th>$A$</th>
<th>$\nu$</th>
<th>$B/G$</th>
</tr>
</thead>
<tbody>
<tr>
<td>LuN</td>
<td>LDA</td>
<td>802.92</td>
<td>185.193</td>
<td>221.996</td>
<td>555.58</td>
<td>391.102</td>
<td>256.383</td>
<td>631.219</td>
<td>0.796</td>
<td>0.231</td>
<td>1.525</td>
</tr>
<tr>
<td>LuBi</td>
<td>LDA</td>
<td>220.792</td>
<td>-8.626</td>
<td>186.968</td>
<td>238.046</td>
<td>70.721</td>
<td>158.064</td>
<td>271.741</td>
<td>1.570</td>
<td>-0.140</td>
<td>0.4474</td>
</tr>
<tr>
<td>LuSb</td>
<td>LDA</td>
<td>317.976</td>
<td>-12.32</td>
<td>242.536</td>
<td>330.296</td>
<td>97.775</td>
<td>211.5808</td>
<td>368.753</td>
<td>1.468</td>
<td>-0.128</td>
<td>0.4621</td>
</tr>
<tr>
<td>Other theory</td>
<td>122.77$^a$</td>
<td>23.14$^a$</td>
<td>23.34$^a$</td>
<td>-</td>
<td>-</td>
<td>122.36$^b$</td>
<td>25.89$^b$</td>
<td>21.37$^b$</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

Our shear modulus values for LuN, LuBi and LuSb can be seen in Table 3. The Young’s modulus $E$ is very important for industrial application; it is defined as the ratio of tensile stress and tensile strain, used to provide a measure of approach of the solid.[38] We calculated Young’s modulus $E$ using the following expression:

$$E = \frac{9BG}{3B + G}$$  \hspace{1cm} (8)

It is calculated that LuN (631.21) is higher than LuBi (271.74) and LuSb (368.75). The values of $E$ are mentioned in Table 3. Poisson ratio ($\nu$) is associated with the volume change during uniaxial deformation, it provides more information about the characteristics of the bonding forces in solids [39] and is expressed as

$$\nu = \frac{3B - E}{6B}$$  \hspace{1cm} (9)

The obtained values are LuN (0.231), LuBi (−0.140) and LuSb (−0.128), it is concluded that the interatomic force is noncentral forces as seen in Table 3. The material is brittle if the $B/G$ ratio is less than 1.75 and a high or $B/G$ value is associated with ductility.[39–41]
The critical value which separates ductile and brittle materials is about 1.75. Table 3 displays LuN (1.525), LuBi (0.447) and LuSb (0.462) which means that these materials are brittle.

### 3.4. Thermodynamic properties

To study the thermal properties of the compounds LuX (N, Bi and Sb) in B1 phase, we applied the quasi-harmonic Debye approximation. The thermal properties are determined in the temperature range 0°C to 1600 K (1 K = −457.87°F), where the quasi-harmonic model remains fully valid. The pressure effect is studied in the 0–50 GPa range. The quasi-harmonic Debye Model as implemented in the Gibbs program [42] is applied to predict the thermal properties. The quasi-harmonic Debye model allows us to obtain all thermodynamic quantities from the calculated energy–volume points, in which the non-equilibrium Gibbs function $G^*(V; P; T)$ is expressed as follows:

$$G^* (V; P; T) = E(V) + PV + A_{\text{vib}}[\theta_D(V); T]$$  \hspace{1cm} (10)

where $E(V)$ is the total energy per unit cell, $PV$ corresponds to the constant hydrostatic pressure condition, $\theta_D(V)$ is the Debye temperature and $A_{\text{vib}}$ is the vibration Helmholtz free energy.
According to the quasi-harmonic Debye model of phonon density of states, one writes $A_{\text{vib}}$ as

$$A_{\text{vib}}(u_D, T) = \frac{n K_B T}{8} \left[ \frac{9 \theta_D}{8 T} + 3 \ln \left( 1 - e^{-\theta_D/T} \right) - D\left( \frac{\theta_D}{T} \right) \right]$$

(11)

where $n$ is the number of atoms per formula unit, $K_B$ is Boltzmann’s constant, $D(\theta_D/T)$ represents the Debye integral. For an isotropic solid, $\theta_D$ is expressed as

$$\theta_D = \frac{\hbar}{K_B \left( \frac{6 \pi^2 n V^{1/2}}{2} \right)^{1/3} f(\sigma) \sqrt{\frac{B_S}{M}}}$$

(12)

where $M$ is the molecular mass per unit cell and $B_S$ is the adiabatic bulk modulus measuring the compressibility of crystal, which is approximated by static compressibility as

$$B_S \approx B(V) = V \frac{d^2 E(V)}{dV^2}$$

(13)

where $f(\sigma)$ and $B_S$ are given in Refs [47–49]. Therefore, the non-equilibrium Gibbs function $G^*$ is
a function of \((V; P; T)\) and is minimized with respect to volume \(V\):

\[
\left[ \frac{\partial G(V,P,T)}{\partial V} \right]_{P,T} = 0
\]  

(14)

By solving Equation (14), we obtain the thermal EOS. Heat capacity at constant volume \(C_V\) and thermal expansion coefficient \(\alpha\) are given by [50]:

\[
C_V = 3nK_B \left[ 4D \left( \frac{\theta_D}{T} \right) - \frac{(3\pi^2)}{T} e^{\frac{\theta_D}{T}} \right] - 1
\]

(15)

\[
\alpha = \frac{\gamma C_V}{B_T T}
\]

(16)
The Grüneisen parameter $\gamma$ is defined by

$$\gamma = -\frac{d\ln \left( \frac{\Theta_D}{D(V)} \right)}{d\ln V} \tag{17}$$

The relation between normalized volume and pressure at different temperature is shown in Figure 5 for LuX (N, Bi and Sb); it can be seen that when temperature increases the normalized volume $V$ increases linearly.

The variation of bulk modulus with temperature is shown in Figure 6 for LuN, LuBi and LuSb. We can clearly see that for temperatures lower than 100 K, the $B$ remains almost constant because of a small change in the volume of the primitive cell, but for temperatures higher than 100 K and up to 1600 K, we see that $B$ decreases with increasing of temperature. At zero temperature and zero pressure, the value of the bulk modulus is found to be equalled to 390.57 GPa for LuN, 70.61 GPa for LuBi and 96.80 GPa for LuSb. The variations of the thermal expansion coefficient $\alpha$ with temperature at approach pressure 0, 10, 20, 30, 40 and 50 GPa are shown in Figure 7. The temperature dependence of the heat capacity at constant volume $C_V$ is presented at 0, 10, 20, 30, 40 and 50 GPa.
as shown in Figure 8 for LuN, LuBi and LuSb, respectively. We can note that with increasing temperature, \( C_V \) rapidly increases at low temperature, then increases slowly at high temperature due to harmonic approximations of Debye model as seen \( T < 500 \, \text{K} \), where \( C_V \) increases with \( T^3 \),\(^\text{[41]}\) while at temperature higher than \( 1000 \, \text{K} \) the \( C_V \) approaches the Dulong–Petit limit.\(^\text{[42]}\) At highest temperature, the \( C_V \) tends to 49.48 J K\(^{-1}\) mol\(^{-1}\) for LuN, 49.5 J K\(^{-1}\) mol\(^{-1}\) for LuBi, 49.31 J K\(^{-1}\) mol\(^{-1}\) and 49.7 J K\(^{-1}\) mol\(^{-1}\) for LuSb. Debye temperature \( \theta_D \) is an important physical parameter to relate successfully with pressures 0, 10, 20, 30, 40 and 50 GPa for LuN, LuBi and LuSb as shown in Figure 9. The calculated values of Debye temperature (J K\(^{-1}\) mol\(^{-1}\)) at zero pressure and zero temperature is found to be equalled to 625.52, 209.90 and 276.35 K for LuN, LuBi and LuSb, respectively. The variation of Gibbs free energy \( (G) \) as a function of temperature at pressures ranging from 0 to 50 GPa for LuN, LuBi and LuSb is shown in Figure 10.

4. Conclusion

The FP-LMTO plane-wave method within DFT was used. The LDA for the XC potential and the LSDA were employed to conduct the structural properties of equilibrium lattice constants, bulk modulus and first pressure derivate of bulk modulus for LuX (X = N, Bi and Sb) compounds in B1, B2 phases within LDA and LSDA. The results are found in accordance with those found experimentally and theoretically. Our calculated structural and elastic properties
showed accordance with other data. In addition, we studied the thermodynamic properties for LuX using the quasi-harmonic Debye model. The heat capacity, thermal expansion, Debye temperature and Gibbs free energy reveal temperature dependence at different values of pressure. The three compounds prove their gap characterisitcs. Finally, the calculated electronic band structure and density of state (DOS) in B1 phase of LuBi and LuSb compounds are metallic and LuN is a semiconductor.

**Disclosure statement**

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**ORCID**

Y. Al-Douri [http://orcid.org/0000-0002-5175-6372]

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