Half-Metallicity, Elasticity and Magnetic Moment of MgN: A First-Principles Study

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Abstract

We report on elastic, electronic and magnetic properties of MgN. This alloy is a strong candidate as a half-metallic material. Such materials are of interest in the field of spintronic applications. Our calculation is first-principles based and is devoted to the Rock salt (RS) and Zinc blend (ZB) phases of this system. Scalar and relativistic calculations, in the Local Spin Density approximation (LSDA) and Generalized Gradient approximation (GGA), are performed using the FPLO.09 code. Spin and charge maps, calculated using the wien2k code, are reported as well. Among our findings is that incorporating spin-orbit interaction impairs half-metallicity to some extent and that the energy gap in the spin-up density of states is pressure-dependent up to a specific pressure.

Introduction

Alkaline earth-nitrides in both the RS and ZB structure are of interest because of the ferromagnetic half-metallic property they exhibit. This property is exploited in the spintronic application for injecting spin-polarized electrons into semiconductors. Binary alkaline earth-nitrides with rock-salt structure e.g. CaN, SrN and BaN were studied for example by Full-potential linearized augmented plane wave FP-LAPW first principles calculation [1]. Earlier half metals e.g. half-Heusler, full-Heusler and some perovskite compounds contain transition metals [2-4]. Half-metallic ferromagnets that are transition-metal- free e.g. CaP, CaAs were first reported by Kusakabe et al [5]. Recently, a new promising material for spintronics has been reported by Droghetti et al [6] where the effects of spin-orbit interaction, compression and exchange–correlation interaction on the robustness of the half-metallicity state.
have been discussed.

In the present work, we undergo a detailed study of the electronic, magnetic and elastic properties of this material. Our first principles calculation are DFT-based [7] and were performed using mostly the electronic code FPLO.09 [8] and wien2k [9]. We have used the well-known GGA and LSDA approximations [10,11] throughout this work. For investigating the half-metallicity state, spin-orbit interaction has been taken into account in our relativistic calculation.

**Theory and Computation**

Our calculation is a first-principles study of MgN compound in both ZB and RS structures within the framework of density functional theory. The theoretical values of equilibrium lattice constant, magnetic moment, electronic structure, energy gap, bulk modulus and its pressure derivative are calculated by the full-potential nonorthogonal local-orbital minimum basis method (FPLO) [8] using both LSDA and GGA approximations [10,11]. The bulk modulus and its pressure derivative have been computed using four different forms of the Murnaghan equation of state [12]. We have used the same set of parameters in our computation to ensure unbiased comparison between the results obtained from LSDA and GGA approximations. The parameters are: the k-mesh subdivision: 24×24×24, the accuracies of the density and total energy are 10^{-6} and 10^{-8} Hartree respectively. For RS structure, the space group is ( #225 ) and the atomic positions are (0,0,0) for Mg and (1/2, 1/2,1/2) for N. For ZB structure, the space group is (#216) and the atomic positions are (0,0,0) for Mg and (1/4, 1/4,1/4) for N. We performed the calculations taking into account the spin-orbit coupling to investigate its effect on the half-metallicity state.

**Results and Discussion**

We show in Fig.1 the energy dependence on the lattice parameter for the ZB and RS structures in the GGA approximation. The RS is clearly the more stable one being less in energy and in the lattice parameter at equilibrium. A similar study we have done, using the LSDA approximation, showed the same behavior as that of the GGA approximation; however the calculated lattice parameter at equilibrium is slightly larger in the latter approximation.

In order to obtain the equation of state i.e. P(V) for MgN, we have used four different models [12] for fitting our ab initio energy vs. volume data. A detailed discussion of the Murnaghan's equation of state (EOS) is given by Fuchizaki [13]. A summary of our calculation is given in table [1]. The values of B_0 calculated using these four different models are comparable for a given structure and approximation method, however the ZB phase has bulk modulus of only about 70% of that of the RS phase. The first pressure derivative of the bulk modulus B’ is in the range 3.7 - 4.7 for the four used models. The lattice parameter at equilibrium is in excellent agreement with the work of Droghetti et al [6]. In addition, the LSDA approximation gives systematically B_0 values higher than those of the GGA approximation for both of the RS and ZB structures. We have used the B_0 and B’ values, thereafter to calculate the
P(V) data. An example, using the viento model [12], is shown in Fig. 2 for the RS and ZB structures in the GGA approximation. The corresponding data for the same two structures in the LSDA approximation is shown in Fig. 3. Using either the GGA or LSDA approximation shows that the ZB structure is relatively easier to compress, at a given pressure, as compared to the RS structure. We should mention that both of the LSDA and GGA calculation were carried out at the same computation input parameters (e.g. k-mesh, density and energy accuracy, etc.) to insure unbiased comparison.

**Table 1:** The values of $B_0$ and $B'$ calculated using four different models [12].

<table>
<thead>
<tr>
<th>EOS-models</th>
<th>Viento EOS</th>
<th>4-parameter Murnaghan EOS</th>
<th>Modified Birch-Murnaghan EOS</th>
<th>Birch-Murnaghan EOS</th>
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<tr>
<td></td>
<td>$a(A_0)$</td>
<td>$B_0$ (GPa)</td>
<td>$B'$</td>
<td>$B_0$ (GPa)</td>
</tr>
<tr>
<td>MgN-structures</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>our work</td>
<td>4.43</td>
<td>119.6</td>
<td>4.69</td>
<td>117.2</td>
</tr>
<tr>
<td>Ref.3</td>
<td>4.44</td>
<td>119.6</td>
<td>4.69</td>
<td>117.2</td>
</tr>
<tr>
<td>RS-GGA</td>
<td>4.35</td>
<td>135.2</td>
<td>3.80</td>
<td>129.6</td>
</tr>
<tr>
<td>ZB-GGA</td>
<td>4.83</td>
<td>88.94</td>
<td>4.52</td>
<td>86.30</td>
</tr>
<tr>
<td>RS-LSDA</td>
<td>4.75</td>
<td>98.88</td>
<td>3.80</td>
<td>94.00</td>
</tr>
<tr>
<td>ZB-LSDA</td>
<td>4.73</td>
<td>98.88</td>
<td>3.80</td>
<td>94.00</td>
</tr>
</tbody>
</table>

**Figure 1:** The energy dependence on the lattice parameter for the ZB and RS structures in the GGA approximation.
In order to study the half-metallicity in this system and whatever effect the spin-orbit coupling might have on the energy gap, we calculated the magnetic moment and energy gap using the LSDA and GGA approximations in the scalar and relativistic schemes. The results are shown in table [2].
Table 2: The magnetic moment, energy gap and spin polarization at E_f for both the LSDA and GGA approximations in the scalar and relativistic schemes. (*) : We have used the theoretical lattice constants from our ab initio calculations.

<table>
<thead>
<tr>
<th>Method</th>
<th>(a_{\text{theo}}(\text{A}^0)^*)</th>
<th>Total moment ((\mu_B))</th>
<th>Energy gap at E_f (eV)</th>
<th>P%</th>
</tr>
</thead>
<tbody>
<tr>
<td>RS-LSDA-scalar</td>
<td>4.35</td>
<td>0.9190</td>
<td>0.00</td>
<td>50.65</td>
</tr>
<tr>
<td>RS-LSDA-relativistic</td>
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<td>0.9390</td>
<td>0.00</td>
<td>58.26</td>
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<td>RS-GGA-scalar</td>
<td>4.43</td>
<td>1.0000</td>
<td>5.09</td>
<td>100</td>
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<tr>
<td>RS-GGA-relativistic</td>
<td>4.43</td>
<td>0.9996</td>
<td>0.00</td>
<td>95.15</td>
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<tr>
<td>ZB-LSDA-scalar</td>
<td>4.75</td>
<td>1.0000</td>
<td>4.13</td>
<td>100</td>
</tr>
<tr>
<td>ZB-LSDA-relativistic</td>
<td>4.75</td>
<td>1.0002</td>
<td>0.00</td>
<td>99.88</td>
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<tr>
<td>ZB-GGA-scalar</td>
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<td>2.90</td>
<td>100</td>
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<tr>
<td>ZB-GGA-relativistic</td>
<td>4.83</td>
<td>0.9999</td>
<td>0.00</td>
<td>99.99</td>
</tr>
</tbody>
</table>

For the ZB structure, the total magnetic moment, energy gap and spin polarization calculated in the scalar LSDA approximation are 1.0 \(\mu_B\), 4.13 eV and 100\% respectively. Including the SO interaction removes the gap, almost preserves the integer moment and reduces the spin polarization. Similar behavior is found for RS in the GGA case. For example, a moment of 1.0 \(\mu_B\), an energy gap of 5.09 eV and a 100\% polarization are in excellent agreement with Droghetti et al [6]. That is to say including SO interaction may impair half metallicity in both phases. We may mention that LSDA approximation does not predict half metallicity at all in the RS phase.

Examples of the density of states for both RS and ZB phases, in the GGA approximation are shown in Figs.4 and 5 respectively. The zoomed-in insert in Fig.5 shows a small, but non-vanishing, DOS counts at E_f in the spin-up channel. The corresponding band structures for the ZB phase in both scalar and relativistic cases are shown in Figs.6 (a,b). The band structure of the spin-up channel alone using wien2k is displayed in Fig.7. A band gap is obvious in the spin-up channel in Figs.6a and 7 however, no such a gap is found in the relativistic case.

The dependence of the total magnetic moment on the lattice parameter is shown for the RS and ZB structures in the GGA approximation in Fig.8. The major features of this figure are: two plateaus, one in the lattice parameter range 8-10 A^0, where the magnetic moment reaches the constant value of 3 \(\mu_B\) and another one between 4.4 and 6 A^0 where moment saturates to 1 \(\mu_B\). However, the drop to zero moment takes place at relatively smaller lattice parameters in the case of the ZB phase. The fixed-spin moment calculation done at a = 4.72 A^0 for ZB structure in the LSDA approximation is shown in Fig.9. Minimum energy is located at 1.0 \(\mu_B\).

We have studied the effect of applying a pressure on the partial and total magnetic moments of the ZB and RS phases and also its effect on the energy gap. For example the GGA calculation for the ZB structure is shown in Fig.10. At ambient pressure (P=0 GPa) the gap is ~4.13 eV. As the pressure increases the gap increases up to ~12 GPa then it tends to saturate at higher pressures. This behavior is similar to that found in other half-metallic systems, e.g. CdSe and CdZSe [14].

Charge and spin density maps in the (110) plane are shown in Figs.11(a,b)
respectively for the ZB phase using the spin-polarized calculation in the GGA approximation as implemented in wein2k. The plane contains 6 Mg and 2 N atoms. The N atoms only carry a spin as shown in part b of the figure. Future studies are planned to calculate and analyze more quantitatively the spin and charge maps at smaller lattice parameters and also to include SO interaction in our calculation.

**Figure 4:** The partial and total DOS’s of MgN in the GGA approximation.
Figure 5: The density of states for the ZB structure using the GGA.

Figure 6a: The band structure for the ZB phase in scalar case.

Figure 6b: The band structure for the ZB phase in relativistic case.
Figure 7: The spin-up band structure for the ZB phase in scalar case using wien2k.

Figure 8: The dependence of the total magnetic moment on the lattice parameter for RS and ZB structure.

Figure 9: The fixed-spin moment calculation for the ZB structure in the LSDA approximation.
Figure 10: Pressure vs. energy gap for the ZB structure using the GGA approximation.

Figure 11a: Charge maps in the (110) plane for the ZB phase using the spin-polarized calculation in the GGA approximation.

Figure 11b: Spin density maps in the (110) plane for the ZB phase using the spin-polarized calculation in the GGA approximation.

Conclusions
The RS phase is more stable than the ZB phase of MgN. For both the GGA and LSDA approximations, the RS is systematically harder to compress. The bulk
The modulus of the RS phase is higher than that of the ZB phase as calculated by four different EOSs. Scalar calculations showed evidence of half-metallicity in both phases. However, switching the spin-orbit interaction on in the calculation has led to a relative impairment of this electronic property. Reducing the cell size has the effect of changing the magnetic state from half-metallic, integer magnetic moment ferromagnet to non-integer ferromagnet. Further reduction in the lattice parameter led to complete disappearance of the magnetic moment in both RS and ZB phase.

References


