A theoretical calculation of the electronic and magnetic behaviour of C-doped zincblende AlN

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Abstract. Based on structural spin-polarized density functional theory, the electronic and magnetic properties of carbon-doped zincblende AlN were investigated. The calculations were carried out by means of the pseudopotential method employed exactly as implemented in Quantum Espresso code. For 0.625% C-doped AlN, we found a ferromagnetic and half-metallic behaviour. Per supercell, the AlC₀.₀₆₂₅N₀.₉₃₇₅ has a net magnetic moment of 1.₀µ₆. The main contribution to the magnetic moment comes from the 2p-C orbitals, with a lesser contribution of the 2p-N and 2p-Al orbitals. Our results suggest that C-doped AlN has potential applications in semiconductor spintronic devices.

1. Introduction
Aluminium nitride (AIN) is one of the compounds of group-III nitrides characterized by a wide band gap, low dielectric constant, high thermal conductivity, and large bulk modulus [1–4]. Due to these properties, currently there has been more interest in AlN, owing to its wide application in optoelectronics areas such as optical detectors, semiconductor lasers, and short-wavelength light-emitting diodes, as well as in high-temperature, high-power, and high-frequency devices [5–8]. AlN normally crystallizes in the wurtzite structure [9]. However, experimental studies of AlN epitaxially grown on most substrates yield the stable zincblende phase. This shows that AlN can also crystallize in the zincblende structure [10,11]. On the other hand, many theoretical and experimental studies have shown that AlN-doped transition metals are good candidates for use as Diluted Magnetic Semiconductors (DMS), with potentials applications in the rising field of spintronics [12–19]. Nevertheless, to make these applications a reality the compounds must exhibit ferromagnetism at room temperature. Recently, many researchers have reported high-temperature ferromagnetism in several types of Transition-Metal (TM)-doped semiconducting oxides and nitrides [20–22]. However, several investigators have proven that magnetic transition metal dopants-doped DMS segregate to form ferromagnetic clusters, secondary phases, or precipitates [23–25]. To avoid this problem, many researchers are investigating the effect of nonmagnetic ion doping on semiconductors such as C and N in order to obtain high-temperature ferromagnetic semiconductors [26–28].

In the present paper, we carry out first-principles total energy calculations in order to investigate the effects of the incorporation de C in the zincblende-AlN volume on the structural, electronic, and magnetic properties of the AlC₀.₀₆₂₅N₀.₉₃₇₅ compound.
2. Computational method
We used the pseudopotential method [29,30] within the framework of density functional theory [31] as implemented in the Quantum ESPRESSO computational code [31]. To take account of the correlation and exchange effects of the electrons, we used the generalized gradient approximation (GGA) of Perdew, Burke, and Ernzerhof (PBE) [32]. A kinetic energy cutoff of 40Ry was used for the expansion of the wave function in the plane wave. Brillouin zone integrations were performed with the special k-point method over a 6×6×4 Monkhorst-Pack mesh [33] for each unit cell. All calculations were carried out with spin polarization. To calculate the structural, electronic, and magnetic properties of pure AlN, we employed a supercell with 32 atoms, which corresponds to a 2a×2b×1c zincblende supercell (see Figure 1). For the doping of AlC$_{0.0625}$N$_{0.9375}$, only one Al atom was substituted for a C atom within the supercell. All atomic positions of pure c-AlN and AlC$_{0.0625}$N$_{0.9375}$ compounds were relaxed until all forces became smaller than 10$^{-4}$eV.

3. Results and discussions
3.1. Structural properties
The main structural parameters, such as the lattice constant ($a_0$), the equilibrium volume ($V_0$), and the bulk modulus ($B_0$) of AlN and the ternary compounds AlC$_{0.0625}$N$_{0.975}$ calculated in this paper are listed in Table 1, along with the available experimental and theoretical values. One can see that for zincblende AlN, our calculated lattice constant of 4.3815Å agrees well with values reported theoretically, 4.380Å [34] and 4.376Å [35], and experimentally, 4.370Å [36], since it differs by less than one percent. Additionally, regarding the bulk modulus of 206.52GPa calculated in this paper, this value agrees well with values reported theoretically (212.4GPa [34] and 210GPa [35]), in comparison with the experimental value of 202GPa [36]. The error in the calculations is small, with a value of 2.23%. Since the errors in the lattice constant and the bulk modulus are small, this shows that the reported results are acceptable.

![Figure 1. Unit cell of AlC$_{0.0625}$N$_{0.975}$ compound in the zincblende structure.](image)

| Compound | $a_0$ (Å) | $V_0$ (Å$^3$) | $B_0$ (GPa) |
|----------|-----------|-------------|-------------|
| AlN      | 4.3815    | 84.545      | 206.52      |
|          | 4.3800$^a$| -           | 212.4$^a$   |
|          | 4.3760$^b$| -           | 210.0$^b$   |
|          | 4.3700$^c$| -           | 202.0$^c$   |
| AlC$_{0.0625}$N$_{0.975}$ | 4.3800 | 84.528 | 202.0 |

$^{ab}$Theoretical reference [34,35]. $^c$Experimental reference [36].
On the other hand, when one N atom in the $2a \times 2b \times 1c$ zincblende supercell is replaced with a C atom, the lattice constant of the AlC$_{0.0625}$N$_{0.975}$ compound changes only slightly with respect to pure zincblende AlN. This may be because the radius of the C atom (0.914Å) is very close to the atomic radius of N (0.92 Å).

### 3.2. Electronic properties

The lattice constants calculated for pure-AlN and AlC$_{0.0625}$N$_{0.975}$ compounds in the zincblende phase shown in the Table 1 were used to calculate the spin-polarized Density Of States (DOS) along the high-symmetry paths in the first Brillouin zone.

#### Figure 2.

Total and partial density of state of (a) pure-AlN and (b) AlC$_{0.0625}$N$_{0.975}$ in the zincblende phase.

Figure 2(a) shows the total and partial density of states of pure-AlN. This compound exhibits a semiconductor behaviour with a forbidden energy gap of 3.41 eV. The magnitude of this gap calculated in this paper agrees well with values reported in other theoretical papers, for example 3.38eV [37] and 3.40eV [38]. On the other hand, the spin-polarized DOS of the ternary compound AlC$_{0.0625}$N$_{0.975}$ is shown in Figure 2(b). We note that when a carbon atom occupies the position of a nitrogen atom in the cubic AlN, it loses its semiconductor behaviour. Thus, the AlC$_{0.0625}$N$_{0.975}$ compound acquires a half-metallic character, because the up spins are semiconductors and the down spins are metallic. The ternary compound has magnetic properties, with a total magnetic moment of $1.0 \mu_B$/cell, which originates mainly from the 2p-C states and in lesser proportion from the 2p-N and 2p-Al states.

### 4. Conclusions

We carried out a study of pure-AlN and the AlC$_{0.0625}$N$_{0.975}$ compound. The structural, electronic, and magnetic properties were studied using DFT calculations. The calculated lattice constants of pure AlN and the experimental values are in good agreement. Analysis of the density of states reveals that the AlC$_{0.0625}$N$_{0.975}$ compound exhibits a half-metallic behaviour, with a total magnetic moment of $1.0 \mu_B$ per cell, which originates mainly from the 2p-C states and in lesser proportion from the 2p-N and 2p-Al states. These compounds are good candidates for potential applications in spintronics.

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