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Electronic structure and magnetic state of $InCNi_3$

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ABSTRACT

Using the projector augmented wave method within the local density approximation and the generalized gradient approximation, we have studied the electronic structures and magnetic state of cubic antiperovskite $InCNi_3$. It is found that Ni 3d states play dominant roles near the Fermi level of this compound. The analysis of bonding nature in $InCNi_3$ implies that Ni 3d and C 2p states exhibit strong hybridization. Both the spin-polarized calculations and the fixed-spin-moment calculations indicate that cubic antiperovskite $InCNi_3$ has a stable paramagnetic (non-magnetic) state.

1. Introduction

The discovery of superconductivity at ~8 K in intermetallic compound $MgCNi_3$ [1] has stimulated considerable attention on the cubic antiperovskites $XCNi_3$ (X = Mg, Zn, Al, Ga, and In). However no superconductivity was found in $ZnCNi_3$ [2], $GaCNi_3$ [3], $AlCNi_3$ [4,5], and $InCNi_3$ [6]. For $AlCNi_3$ and $CaCNi_3$, experimental studies reported that $AlCNi_3$ is a strongly exchange-enhanced Pauli paramagnet in the very vicinity of ferromagnetic order [5] and its transition from a paramagnetic state into weak ferromagnetic one takes place at temperature near 300 K [4], while $CaCNi_3$ has a large electron–electron correlation due to the proximity of ferromagnetic order from the side of exchange-enhanced Pauli paramagnet [3]. But these magnetic properties of $AlCNi_3$ and $CaCNi_3$ reported in experiments are contradictory to the results predicted from density functional theory (DFT) calculations [7–9] that did not support $AlCNi_3$ being in a magnetic ground state and $CaCNi_3$ being in the presence of strong electron–electron correlations. DFT calculations [8] instead suggested that the tendencies toward magnetism found in experiments for $AlCNi_3$ and $CaCNi_3$ should be explained by the deviation of the Ni/C atomic ratio from the ideal stoichiometry. For the same group compound $InCNi_3$, its non-stoichiometry compound $In_{0.95}CNi_3$ has been synthesized recently [6]. From the measured results of electronic specific coefficient and Debye temperature, $In_{0.95}CNi_3$ was proposed to behave as a ferromagnetic metal below the Curie temperature (577 K) and the emergence of ferromagnetism in $In_{0.95}CNi_3$ is ascribed to the deviation of the Ni/In atomic ratio from the ideal stoichiometry [6], which is different to the findings from the DFT calculations [7,8] on $AlCNi_3$ and $CaCNi_3$. To shed more light on the understanding on the presence of ferromagnetism in $In_{0.95}CNi_3$, it is of great interest to study the physical properties of the ideal stoichiometry compound $InCNi_3$. Recently, $InCNi_3$ is proposed to be a paramagnetic nonsuperconductor from the calculated total energy, density of states at Fermi level, and electron–phonon coupling constant [9], however the detailed electronic structures of $InCNi_3$ is not presented there, especially for the bonding nature. In addition, only the generalized gradient approximation is employed in previous DFT calculations on $InCNi_3$ [9]. In order to completely understand the electronic and magnetic state of cubic antiperovskite $InCNi_3$, we conduct a comprehensive *ab initio* calculations on this compound by using the projector augmented wave method with plane-wave basis set within the local density approximation and the generalized gradient approximation. In this study, we predict the lattice parameter and bulk modulus of $InCNi_3$, discuss the bonding nature in this compound from the calculated electronic density of states and charge distribution, and also perform the fixed-spin-moment calculations to examine the stability of possible magnetic state of $InCNi_3$.

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2. Method of calculations

All calculations on $InCNi_3$ are performed by means of the projector augmented wave method [10] within the local density approximation (LDA) [11,12] and the generalized gradient approximation (GGA) [13], as implemented in VASP code [14,15]. In LDA calculations, Perdew and Zunger's parametrization [12] of Ceperley and Alder quantum Monte Carlo results [11] is used for the correlation functional. While Perdew-Wang'91 (PW91) exchange and correlation functional [13] is adopted in GGA calculations. A nickel potential with 4s and 3d electrons as valence states, an indium potential with 4d, 5s, and 5p electrons as valence states, and a carbon potential with 2s and 2p electrons as valence states have been used. Wave functions are expanded by the plane waves up to an energy cutoff of 450 eV. Brillouin-zone (BZ) integrations during the self-consistent calculations are approximated using the special k-point sampling of Monhkorst-Pack scheme [16] with a $13 \times 13 \times 13$ grid. The convergence of the total energy with respect to both k-point sampling and plane-wave energy cut-off are carefully examined, and the above calculation setup is found to provide good precision in present study. The total energies are obtained as a function of volume and then the data of total energies versus volumes are fitted to 3rd-order Birch-Murnaghan equation of state [17] to give the equilibrium lattice constant and other ground state properties of $InCNi_3$. In the calculation of density of states (DOS), a dense k-grid of $20 \times 20 \times 20$ is used and the angular-momentum-projected DOS is integrated within the spheres of radii 1.677 Å for In atom, 0.863 Å for C atom, and 1.286 Å for Ni atom, respectively.

Table 1
Calculated lattice constants (a, in Å), bulk modulus (B, in GPa), and the first pressure derivative of B (B') for $InCNi_3$, with a comparison with available experimental values and other theoretical results

Property	Present		Previous [9]	Experimental [6]
	LDA	GGA-PW91	GGA-PBE	
a	3.7893	3.8805	–	3.7836
B	228.7	185.6	183.56	–
B'	4.70	4.74	–	–

3. Results and discussions

Recent experimental study [6] on $In_{0.9}CNi_3$ reported that this compound has the typical cubic antiperovskite structure with the space group 221($Pm\bar{3}m$) and its lattice parameter is about 3.7836 Å determined from the powder X-ray diffraction patterns. The calculated equilibrium lattice constant (a), bulk modulus (B), and first pressure derivative of bulk modulus (B') of cubic antiperovskite $InCNi_3$ are listed in Table 1. From the results in Table 1, one can see that the deviation of the predicted lattice constant of $InCNi_3$ in LDA calculation is about 0.15% and the one in GGA-PW91 calculation is about 2.5% with respect to the experimental lattice constant of $In_{0.9}CNi_3$, respectively. The bulk modulus of $InCNi_3$ is predicted to be 228.7 GPa in LDA calculation and 185.6 GPa in GGA-PW91 calculation, respectively, and especially the latter is in excellent agreement with the previous GGA-PBE calculation result (183.56 GPa) [9] from highly precise full-potential linearized augmented plane-wave (FP-LAPW) method.

In order to understand the electronic nature of this compound, its band structure and density of states (DOS) have been calculated. For $InCNi_3$ at equilibrium, the LDA and GGA-PW91 calculations give almost identical electronic band structure around the Fermi energy. Here we show only GGA-PW91 results. Fig. 1(a) shows the calculated electronic band structure of cubic $InCNi_3$ along several high-symmetry lines in the first BZ. At the same time, the total DOS of cubic $InCNi_3$ is shown in Fig. 1(b). One band around −12 eV in Fig. 1(a) is mainly contributed by the bonding states of C 2s orbital. One band around −8 eV mostly consists of In 5s orbital and Ni 4s orbital. Complex bands extending between −7 and −4 eV are composed of C 2p and Ni 3d orbital mixed lightly with In 5s orbital. Dense bands around −2 eV are dominated by Ni 3d electrons and In 5p orbital, while the latter one contributes a little to the lower part of these dense bands. Two conduction bands crossing the Fermi energy (E_F) are dominated by Ni 3d, C 2p and In 5p orbitals. The contribution of

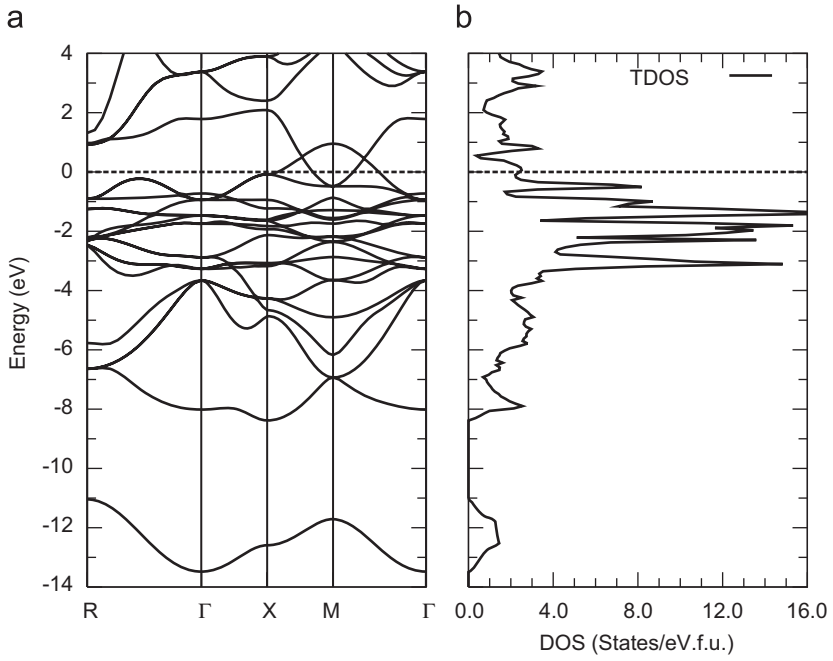

Fig. 1. Electronic band structure and total density of states (TDOS) of cubic $InCNi_3$ at equilibrium, obtained from GGA calculations.