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ORIGINAL RESEARCH

Structural, Electronic, and Magnetic Properties of Hard Magnetic $SmNi_2Fe$ Compound: a DFT Study

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Abstract
Permanent magnets with high magnetic properties are used in many areas like motors, generators, magnetic separators, handles, electron tubes, magnetic resonance imaging systems, health, electronics, automotive, and mining. Due to their easy and inexpensive production methods, most common permanent magnets are ferrite magnets. However, the quest for finding new and alternative permanent magnets is still in progress. Thus, in this study, lattice parameters (a, c), equilibrium lattice volume (V), density (ρ), formation energy (E_f), Wyckoff positions, magnetic moments, density of states. and electronic band structure of $SmNi_2Fe$ are investigated using density functional theory (DFT) calculations. For exchange-correlation relations, PBE method within generalized gradient approximation (GGA) and (GGA + U) is used. With a 8.628 μ_B (GGA) and 9.886 μ_B (GGA + U) total magnetic moment, $SmNi_2Fe$ shows a strong permanent magnetism. Obtained negative formation enthalpy of $SmNi_2Fe$ (− 1.526 eV/f.u.) clearly shows that studied material can be synthesized experimentally. Besides, density of states and spin polarized electronic band structures indicate that $SmNi_2Fe$ is metallic. Calculated lattice parameters of $SmNi_2Fe$ are in good agreement with literature.

Keywords Permanent magnets · Density functional theory (DFT) · Magnetic moment · Formation enthalpy · Electronic properties

1 Introduction

The demeanor of magnetic nanostructures conveys not only the nanoscale properties like particle size and geometry but also the inside features of the magnetic materials. For instance, the magnetization reversal in nanodots significantly hinges on the anisotropy of the dot material. Moreover, nanostructures are frequently used in bulk forms, thereby their external features must be utilized in consideration of bulk materials. Magnetic materials can be classified as hard magnetic materials and soft magnetic materials depending on their magnetization characteristics. Hard magnetic materials known as permanent magnets, show strong demagnetization that is to say they maintain their magnetism after being magnetized [1–6]. Magnetic capability of a material is determined by its inner coercivity. For hard magnetic materials, the intrinsic coercivity value is greater than ~ 10 kAm^{-1}. Due to their extensive usage as magnets in devices such as electric motors, generators, and actuators, hard magnetic materials are intensively used in today's technology [7–13]. Since transition metal and rare earth-based compounds have utmost retentivity and coercivity and high Curie temperature, they are the best materials for permanent magnetism [14–17]. As well as these features, the large uniaxial anisotropy is also a major feature for the permanent magnet [18].

On the other hand, soft magnetic materials can be easily magnetized and demagnetized by low magnetic field. Briefly, the criteria for a material to be magnetic are high coercivity, magnetic remanence, and saturation values with high stability [19, 20]. Neodymium and samarium-cobalt-based rare-earth magnets are known to

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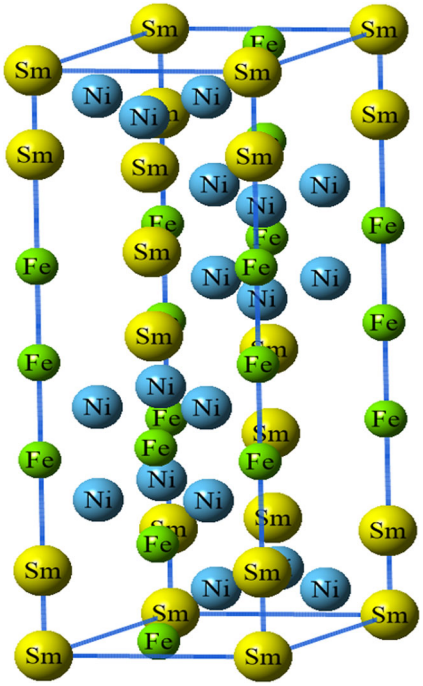

Fig. 1 (Color online) The crystal structure of the $SmNi_2Fe$ compound with $PuNi_3$ type structure

have superior magnetic properties due to their high magnetocrystalline anisotropy. However, the high prices of these rare-earth materials pave the way for the development of alternative permanent magnets composed of inexpensive materials. In addition to pure permanent magnets like Fe, Ni, and Co and their compounds, samarium-iron-containing permanent magnets are also a promising candidate for future applications due to their high resistance to demagnetization, high magnetism, and better resistance to temperature and corrosion. Thus, these magnets may be a superior choice to NdFeB magnets in the future. Contrary to intensive research about samarium-cobalt-based magnets available in literature [21–25], studies about samarium-iron-based magnets are very limited. K. Nouri et al. studied the phase diagram of Sm-Fe-Ni ternary system at 800 °C using the XRD and SEM/EDS techniques [26]. M. Matsuura et al. investigated the effect of Mn addition on magnetic properties of Sm-Fe-N core-shell by means of reduction diffusion process [27]. K. Nouri et al. investigated the magnetic properties of the bulk and nanocrystalline $SmNi_2Fe$ using mechanical milling [28].

It is obvious that samarium-iron-based magnets are still under development. Other than electronics and optics, they could become available for medical and dental applications in the future as well [29, 30]. Studies about samarium-iron based magnetic materials are very scarce, so current studies about this material are still at the starting point.

Therefore, in this paper, we have studied the structural parameters, magnetic properties, density of states, and electronic band structure of $SmNi_2Fe$ using density functional theory calculations. Paper is organized as follows: In Section 2, theoretical method is given. Section 3 is devoted to results and discussion. Finally, results are recapitulated with a conclusion part in Section 4.

2 Theoretical Method

For all calculations, we take advantage of the density-functional theory (DFT) implemented in the medeA-VASP code [31, 32]. The exchange-correlated potential is processed using the Perdew–Burke–Ernzerhof (PBE) within the GGA and GGA + U methods [33, 34]. For the plane-wave basis, an energy cutoff of 500 eV is used. A $(12 \times 12 \times 4)$ Monkhorst-Pack grid is used to sample the first Brillouin zone of $SmNi_2Fe$ with $PuNi_3$ type structure [35]. U is Coulomb parameter and J is exchange parameter, For Sm, Ni, and Fe atoms 6.0, 2.5, and 2.5 eV are used for U-J difference. And all the structures are fully relaxed with a force tolerance of 0.01 eV. Blocked Davidson algorithm is applied with a 1×10^{-5} eV convergence parameter. The Brillouin-zone integrations were performed via the Methfessel-Paxton smearing method [36] with a smearing parameter of 0.169 eV.

Table 1 Calculated lattice parameters (a, c), equilibrium lattice volume (V), density (ρ), formation energy (ΔH_f), and Wyckoff positions (z) of $SmNi_2Fe$ compound with GGA and GGA+U

Compound	Ref.	a (Å)	c (Å)	c/a	V (Å^3)	ρ (g/cm^3)	ΔH_f (eV/f.u.)	z_{Sm}	$z_{Fe/Ni}$	$z_{Fe/Ni}$
$SmNi_2Fe$	Present-GGA	5.108	24.381	4.77	550.99	8.777	− 1.526	0.1389	0.3329	0.0851
	Present-GGA + U	5.175	24.052	4.65	557.80	8.670	0.905	0.1376	0.3314	0.0845
	[22]	5.110	24.829	4.86	546.88	–	–	0.1411	0.3326	0.0794

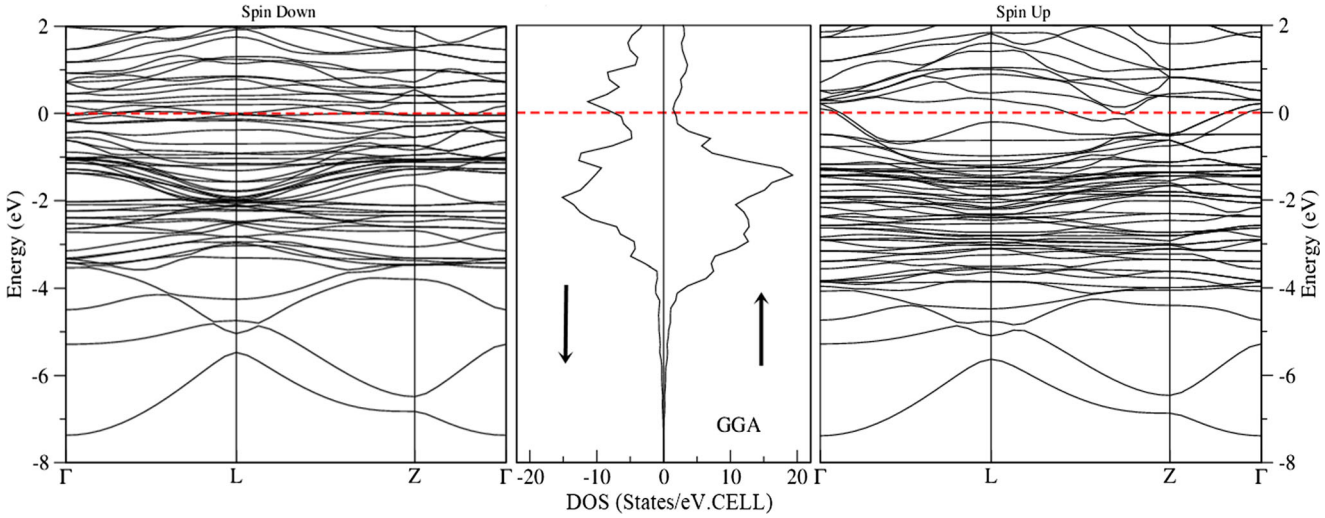

Fig. 2 (Color online) The band structure and total density of states of the $SmNi_2Fe$ compound in minority and majority spin states with GGA method

3 Results and Discussions

3.1 Crystal Structure and Formation Enthalpy

$SmNi_2Fe$ has a $PuNi_3$ cyrstal-type structure. This material is in the form of RNi_3 series where R is a rare earth element. Atoms in $SmNi_2Fe$ compound occupy 3a (0;0;0) and 6c (0;0;z) positions, respectively, as shown in Fig. 1. They crystallize in R-3 m rhombohedral-type structure depending on the rare earth atoms. Calculated lattice parameters (a, c), equilibrium lattice volume (V), density (ρ), formation enthalpy (ΔH_f), and Wyckoff positions of $SmNi_2Fe$ are given in Table 1. It is obviously seen that calculated lattice parameter is in good agreement with literature [28]. The formation enthalpy is a significant parameter which gives deep insight about whether studied compound is structurally stable and experimentally synthesizable or not [37]. The formation enthalpy of $SmNi_2Fe$ can be obtained from the following equation;

$$\Delta H_f = E^{total}_{SmNi_2Fe} - \left[E^{bulk}_{Sm} + 2E^{bulk}_{Ni} + E^{bulk}_{Fe}\right] \quad (1)$$

where $E^{total}_{SmNi_2Fe}$ is the equilibrium total energy, E^{bulk}_{Ni}, E^{bulk}_{Sm}, and E^{bulk}_{Fe} are the energies per atom. The formation enthalpy (ΔH_f) of the $SmNi_2Fe$ compound is 1.526 eV (147.237 kJ/mol). The obtained negative formation enthalpy value shows that this compound is thermodynamically stable and may be synthesized experimentally.

3.2 Electronic Properties

Figure 2 shows electronic band structure along high symmetry directions in the first Brillouin zone in the spin up and spin down states of $SmNi_2Fe$ compound from −8 to 2 eV. The dashed horizontal red line at

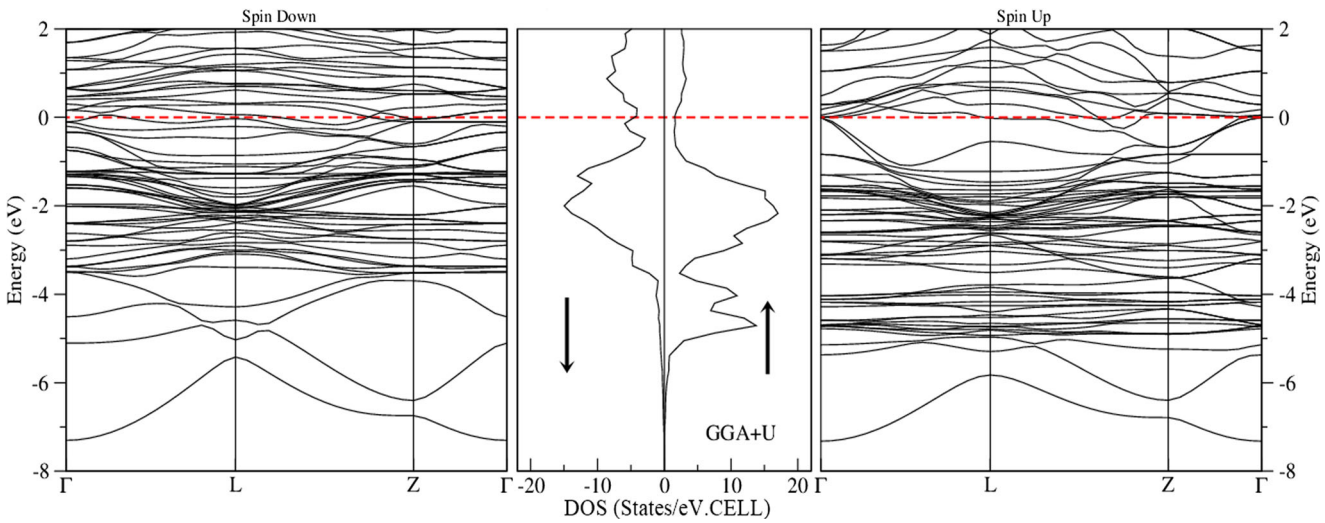

Fig. 3 (Color online) The band structure and total density of states of the $SmNi_2Fe$ compound in minority and majority spin states with GGA+U method

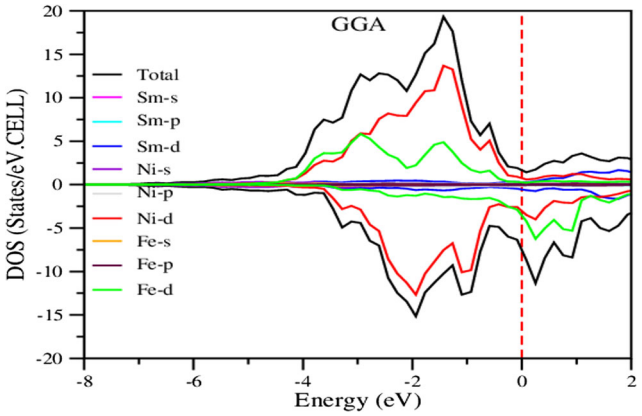

Fig. 4 (Color online) The spin-polarized total and partial DOS for $SmNi_2Fe$ compound with GGA method

0 eV shows the Fermi energy (E_F). From Fig. 1, it is observed that $SmNi_2Fe$ has no band gap along the fermi level. Therefore, we can say that studied material is metallic in nature for spin up and spin down states. We have also obtained the spin-polarized total and partial density of states (PDOS) of $SmNi_2Fe$ compound which is given in Figs. 2, 3 and 4. In Fig. 3, we have shown total and partial density of states of $SmNi_2Fe$ with GGA + U. From Fig. 4, we make an inference that between − 8 and − 5 eV contribution of the orbitals of atoms are negligibly small in both spin states. Between − 5 and − 3 eV, most of the contribution to spin up density of states (PDOS) comes from the d orbitals of Fe atoms. For the spin down state, most of the contribution comes from the d orbitals of Ni atom. Besides, between − 3 and 0 eV for spin up and spin down states, most of the contribution to PDOS significantly comes from the d orbitals of Ni atom. Between 0 and 1 eV for spin up state, contributions of the d orbitals of Ni atom and of Sm atoms are almost same. In the spin down state, most notable contribution comes from the d orbitals of the Fe atoms. Lastly, between 1 and 2 eV for spin up state, majority of the contribution comes from the d orbitals of Sm atoms. In the spin down state, contributions of the d orbitals of Ni, Fe, and Sm atoms are very close to each other.

3.3 Magnetic Properties

Obtained total and atomic magnetic moments of $SmNi_2Fe$ compound are given in Table 2. Total magnetic moment of the studied compounds for GGA and GGA + U methods are 8.628 μ_B and 9.886 μ_B, respectively. It can be clearly seen from Table 2, that the majority of the total magnetic moment comes from Fe atoms. This is an expected outcome because Fe shows strong permanent magnetism. There is also a small positive contribution from Ni atom. However, Sm atom has a small negative contribution to magnetic moment as shown in Table 2. Obtained results show that $SmNi_2Fe$ is a hard magnetic material. There is no study in the literature concerning both magnetic and electronic properties of the $SmNi_2Fe$ compound. This is the first study in which magnetic and electronic properties of $SmNi_2Fe$ are calculated.

4 Conclusion

A detailed computational study on the structural, electronic, and magnetic properties of the hard magnetic $SmNi_2Fe$ compound has been carried out using first-principle methods. The calculated lattice parameters, equilibrium lattice volume, and Wyckoff positions of $SmNi_2Fe$ are in good agreement with available experimental result. Moreover, the obtained negative formation enthalpy indicates that $SmNi_2Fe$ is thermodynamically stable and synthesizable for technological applications. The spin-polarized electronic band structures and total and partial density of states of this material have been obtained. Band structure calculations revealed that $SmNi_2Fe$ is metallic since there is no gap between the spin up and spin down states along the fermi level. The total magnetic moment of this compound is found to be 8.628 μ_B (GGA) and 9.886 μ_B (GGA + U) per formula unit, indicating that the studied material is hard magnetic material. Since studies about samarium-based magnets are limited and current neodymium-based magnets are expensive, results obtained in this paper are of special importance. High total magnetic moment showed that this material can be used as a good alternative to neodymium and samarium-cobalt-based rare-earth magnets in future technological applications. As far as we know, there is only one experimental study about $SmNi_2Fe$. However, this study is the first theoretical study in which we used the density functional theory calculations. Since the need for alternative cheap magnetic

Table 2 The calculated total and atomic magnetic moments of $SmNi_2Fe$ compound with GGA and GGA+U

Compound	Ref.	M_t (μ_B)	$M_{Sm}{}^{-1}$ (μ_B)	$M_{Sm\text{-}2}$ (μ_B)	M_{Ni} (μ_B)	$M_{Fe}{}^{-1}$ (μ_B)	$M_{Fe\text{-}2}$ (μ_B)
$SmNi_2Fe$	Present-GGA	8.628	− 0.183	− 0.134	0.370	2.560	2.351
	Present-GGA + U	9.886	− 0.196	− 0.197	0.369	3.031	2.985

materials is inevitable, the results obtained in this study will fill an important gap.

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