Journal of Solid State Chemistry 241 (2016) 38–46

Contents lists available at ScienceDirect

Journal of Solid State Chemistry

journal homepage: www.elsevier.com/locate/jssc

Research report

Electronic and magnetic properties of $TbNi_4Si$: Ab initio calculations, mean field approximation and Monte Carlo simulation

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ARTICLE INFO

Article history:
Received 15 January 2016
Received in revised form
20 April 2016
Accepted 24 April 2016
Available online 26 April 2016

Keywords:
Magnetic properties
Electronic structure
Magnetocaloric effect
Ab-Initio
DFT
Mean field approximation
Monte Carlo simulation

ABSTRACT

Using the density functional theory, the electronic structure; density of states, band structure and exchange couplings of $TbNi_4Si$ compound have been investigated. Magnetic and magnetocaloric properties of this material have been studied using Monte Carlo Simulation (MCS) and Mean Field Approximation (MFA) within a three dimensional Ising model. We calculated the isothermal magnetic entropy change, adiabatic temperature change and relative cooling power (RCP) for different external magnetic field and temperature. The highest obtained isothermal magnetic entropy change is of $-14.52\ J\ kg^{-1}\ K^{-1}$ for a magnetic field of $H=4$ T. The adiabatic temperature reaches a maximum value equal to 3.7 K and the RCP maximum value is found to be $125.12\ J\ kg^{-1}$ for a field magnetic of 14 T.

1. Introduction

Cold production is an essential process; actually, it represents a very important part of the global electricity consumption. The use of conventional systems poses an environmental problem. Therefore, it is necessary to develop new cold production technique. On the one hand, eliminate refrigerant gases and secondly, to improve energy efficiency. These issues feed a lot of research axes that are oriented towards magnetic refrigeration.

Recently, the orthorhombic derivative of the $CaCu_5$-type, namely the YNi_4Si-type (space group Cmmm), RNi_4Si compounds were reported [1]. These compounds supplement the known series of the $CaCu_5$-type RNi_5 compounds and RNi_4Si solid solutions [2,3]. The orthorhombic distortion of parent $CaCu_5$-type compounds may be considered as a prospective route for optimizing their magnetic and hydrogen storage properties [4–6]. In order to test its feasibility, it requires a systematic investigation of the magnetic properties of YNi_4Si-type RNi_4Si, which could be compared with those of the well-known $CaCu_5$-type RNi_5 and RNi_4Si compounds.

The recent reports of the magnetic properties and structures of $CaCu_5$-type RNi_5, RNi_4Si [7–11] and YNi_4Si-type RNi_4Si [1,12,13] (R=Gd, Tb, Dy) conclude that in the case of {Tb, Dy} Ni_4Si the ferromagnetic ordering temperature increases from the $CaCu_5$-type RNi_5 across $CaCu_5$-type RNi_4Si to YNi_4Si-type RNi_4Si, whereas the Curie point decreases from $GdNi_5$ to the $CaCu_5$-type $GdNi_4Si$ and ($CaCu_5$-type YNi_4Si-type $GdNi_4Si$) have the same Curie temperatures. The transformation of their magnetic properties in the orthorhombic distortion of $CaCu_5$-type lattice, results from the initial ab-plane ferromagnetic-like ordering of Tb and Dy sublattices and possible c-collinear ferromagnetic ordering of Gd sublattice.

The $TbNi_4Si$ compound has a ferromagnetic order with a critical temperature close to 37 K. The experimental value of the Tb magnetic moment is of 8. 66μB, while Ni does not have a magnetic order [13]. The experimental values of the magnetization as a function of the external magnetic field in the range [−140,140] kOe, at a fixed temperature $T = 2K$, show that the saturation magnetization, M_{sat}, is equal to 7,7 μB, while, in the range [−14,14] kOe it exhibits a hysteresis loop with a remanent magnetization of M_{res}=3, 1μB and a coercive field of H_{coer}=3 kOe, the paramagnetic susceptibility for $TbNi_4Si$ follows the Curie-Weiss law $\chi=\frac{c}{T-\theta}$ with in the temperature range [30, 300] K [12,13].

In this work, we aim to study the electronic, magnetic and magnetocaloric properties of the $TbNi_4Si$ compound, using the methods mentioned above: Density Functional Theory, implemented in Wien2k code, Monte Carlo Simulation and Mean Field Approximation.

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http://dx.doi.org/10.1016/j.jssc.2016.04.036
0022-4596/

2. Structure and model

Our compound is RNi_4Si type with R=Tb ($4f^9 6s^2$), Ni ($3d^8 4s^2$) and Si ($3s^2 3p^2$), possesses an orthorhombic structure derived from the $CaCu_5$ structure (Fig. 1). The structure belongs to the space group Cmmm with the crystal lattice parameters a=5.0626 Å, b=8.2189 Å and c=3.9516 Å. The atomic positions are: Tb (2a) [0, 0, 0], Ni1 (4i) [0, 0.3396, 0], Ni_2 (4 f) [1/4, 1/4, 1/2] and Si (2c) [0, 1/2, 1/2] (Fig. 1) [13]. The distribution of charges in *TbNi₄Si* is given by $Tb^{3+}Ni_4^{1-}Si^{1+}$. It is clear that $Ni^{1-}$and Si^{1+} ions do not have magnetic moments. As a result, the magnetic moment of this compound comes from the Tb^{3+} ion.

The Ab-initio calculations presented in this paper have been performed using Density Functional Theory (DFT) based on the Full Potential Linearized Augmented Plane-Wave (FP-LAPW) method as implemented in WIEN2K code [14]. The FP-LAPW method has proven to be one of the accurate methods [15] for the computation of the electronic structure of solids. The exchange correlation potential was treated using the Generalized Gradient Approximation (GGA) for the total energy calculations [16] and GGA+*U* which includes the on-site Coulomb interaction in the GGA Hamiltonian [17]. The atomic muffin–tin (MT) spheres, supposed not to overlap with each other, are taken as 2.5, 2.31 and 1.9 at. units (a.u) for Tb, Ni and Si, respectively. The largest reciprocal vector *G* in the charge Fourier expansion, Gmax, was equal to 12, and number of basis functions to be included, RMT × K_{max}, was equal to 7. Calculations are performed with (1000) 128 in equivalent k points in the irreducible Brillouin zone. The convergence criterion was chosen to be the total energy and set at 10^{-4} eV. In order to calculate the coupling values, we have considered a supercell of $2 \times 1 \times 1$ in real space.

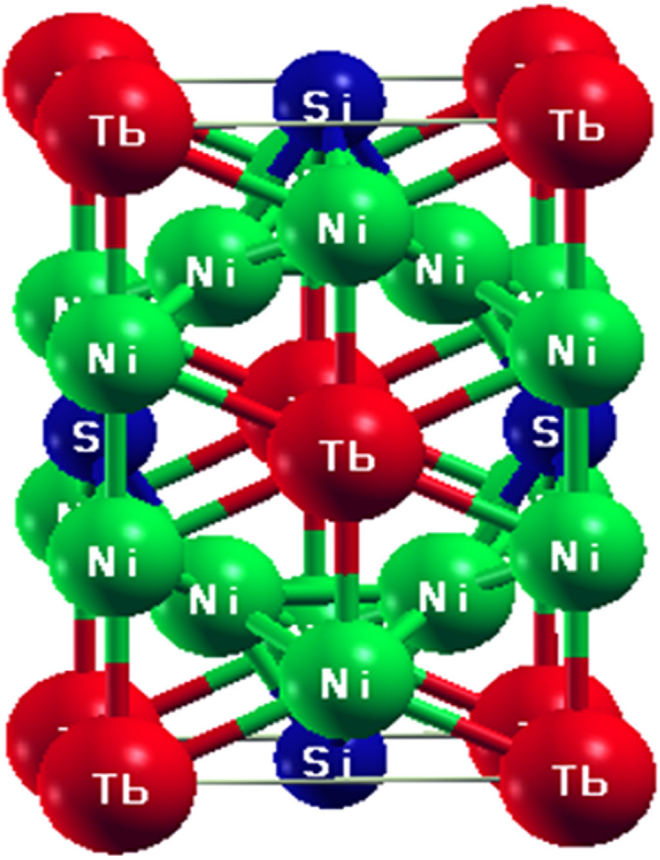

Fig. 1. The structure of $TbNi_4Si$ compound.

To try to go further in the understanding of critical phenomena associated with ferromagnetism from $TbNi_4Si$ compound, we choose the three-dimensional Ising model, because $TbNi_4Si$ material exhibits the b axis-collinear magnetic ordering of Tb magnetic sublattice [13], it allows describing each lattice site as a magnetic nano-dipole in which spin moments can take up-direction or down-direction. The Hamiltonian of $TbNi_4Si$ includes nearest neighbors interactions and crystal field, it is given by:

$$H = -J_1 \sum_{\langle ij \rangle} S_i S_j - J_2 \sum_{\langle ij \rangle} S_i S_j - \Delta \sum_{i=1}^{N} S_i^2 \quad (1)$$

Where ⟨ij⟩ stand for the first nearest neighbors sites i and j. Δ is the crystal field. J_1 and J2 are the exchange interactions parameters between the Tb-Tb (First nearest neighbors) and Tb-Tb (Second nearest neighbors), respectively. S_i and S_j are the spin moments at site (i) and site (j). The spin moment of Tb^{3+} ion is S=3.

3. Results and discussion

3.1. Ab initio calculations

$TbNi_4Si$ Compound has a single magnetic atom, the terbium, with three magnetic configurations; ferromagnetic, type (A) antiferromagnetic and type (B) antiferromagnetic configuration as shown in Fig. 2.

The exchange couplings J_1 and J_2, in Eq. (1), may be obtained using the following equations:

$$E(F) = -J_1 \frac{Z_1}{2} S^2 - J_2 \frac{Z_2}{2} S^2 - \Delta S^2 \quad (2)$$

$$E(AF-A) = -J_1 \frac{Z_1}{2} S^2 + J_2 \frac{Z_2}{2} S^2 - \Delta S^2 \quad (3)$$

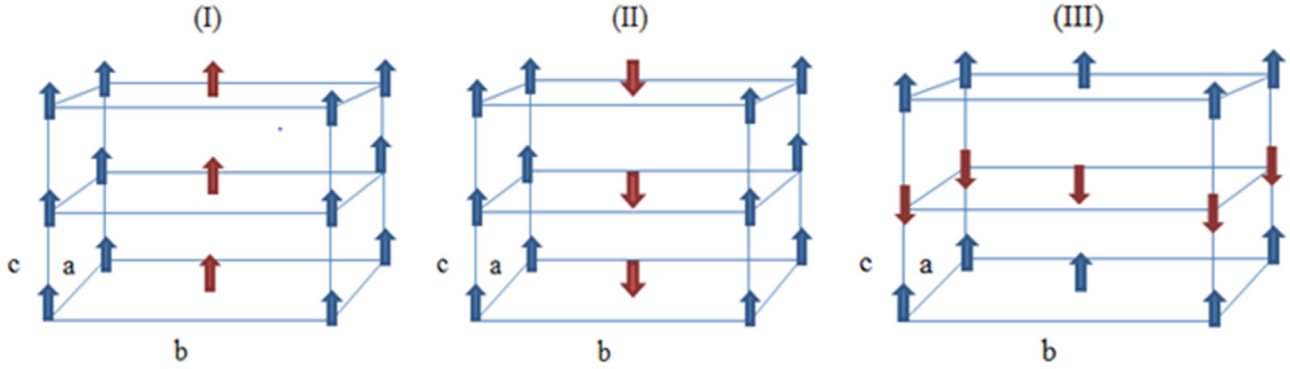

Fig. 2. The three studied configurations (I) ferromagnetic configuration (II) type (A) antiferromagnetic configuration (III) type (B) antiferromagnetic configuration.