Ab initio study of magnetocrystalline anisotropy, magnetostriction, and Fermi surface of L1₀ FeNi (tetrataenite)

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Abstract
We present results of ab initio calculations of several L1₀ FeNi characteristics, such as the summary of the magnetocrystalline anisotropy energies (MAEs), the full potential calculations of the anisotropy constant \( K_3 \), and the combined analysis of the Fermi surface and 3D \( k \)-resolved MAE. Other calculated parameters are the spin and orbital magnetic moments, the magnetostrictive coefficient \( \lambda_{001} \), the bulk modulus \( B_0 \), and the lattice parameters. The MAEs summary shows rather big discrepancies among the experimental MAEs from the literature and also among the calculated MAE’s. The MAEs calculated in this work with the full potential and generalized gradient approximation (GGA) are equal to 0.47 MJ m\(^{-3} \) from WIEN2k, 0.34 MJ m\(^{-3} \) from FPLO, and 0.23 MJ m\(^{-3} \) from FP-SPR-KKR code. These results suggest that the MAE in GGA is below 0.5 MJ m\(^{-3} \). It is expected that due to the limitations of the GGA, this value is underestimated. The L1₀ FeNi has further potential to improve its MAE by modifications, like e.g. tetragonal strain or alloying. The presented 3D \( k \)-resolved map of the MAE combined with the Fermi surface gives a complete picture of the MAE contributions in the Brillouin zone. The obtained, from the full potential FP-SPR-KKR method, magnetocrystalline anisotropy constants \( K_2 \) and \( K_3 \) are several orders of magnitude smaller than the MAE/\( K_1 \) and equal to \(-2.0 \) kJ m\(^{-3} \) and \(110 \) J m\(^{-3} \), respectively. The calculated spin and orbital magnetic moments of the L1₀ FeNi are equal to 2.72 and 0.054 \( \mu_B \) for Fe and 0.53 and 0.039 \( \mu_B \) for Ni atoms, respectively. The calculations of geometry optimization lead to a \( c/a \) ratio equal to 1.0036, \( B_0 \) equal to 194 GPa, and \( \lambda_{001} \) equal to \(9.4 \times 10^{-6} \).

Keywords: rare earth free permanent magnets, magnetocrystalline anisotropy energy, hard magnetic materials, full potential, L1₀ FeNi, magnetostriction, Fermi surface

(Some figures may appear in colour only in the online journal)

1. Introduction

Electric power generators, motors and transformers are just a few examples where magnetic materials find applications in modern technology. The hard magnetic materials used the most are alnicos, hexaferrites, and Nd–Fe–B alloys. The economical event from 2011 called the rare-Earth crisis [1] destabilized, among other things, the prices of neodymium, motivating efforts to find new rare-earth free permanent magnets. The ongoing search for hard magnetic materials free from rare-earth elements is summarized in several review articles [2–7]. Some of the promising candidates studied recently are e.g. Fe/Co nanowires [8, 9], Fe–Co alloys doped with B and C [10–15], (Fe/Co)ₓB [16–19], (Fe/Co)ₓXₐB₂ [20–22], MnBi
and magnetocrystalline anisotropy energy (MAE) requires fully relativistic electronic band structure calculations. The calculations are carried out by using the full-potential local-orbital minimum-basis scheme FPLO-14.0-49 [30, 42] with the generalized gradient approximation (GGA) in the Perdew–Burke–Ernzerhof (PBE) form [43]. The calculations are performed up to a 80\(^3\) k-mesh with tetrahedron method for integration, an energy convergence criterion 10\(^{-8}\)

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\[
\frac{\Delta l}{l_0} = \frac{3}{2} \lambda_{001} \left[ \sum_{i} \frac{1}{2} \alpha_i^2 \beta_i^2 - \frac{1}{3} \right] + 3 \lambda_{111} \sum_{i\neq j} \alpha_i \alpha_j \beta_i \beta_j.
\]

If the measurement is made along the [001] direction the equation simplifies to:

\[
\frac{\Delta l}{l_0} = \frac{3}{2} \lambda_{001} \left[ \frac{1}{2} \alpha_i^2 - \frac{1}{3} \right]
\]

and for a single domain system it takes the form:

\[
\lambda_{001} = \frac{1}{3} l_0(\theta = 0^\circ) - l_0(\theta = 90^\circ)
\]

These efforts are followed by the theoretical reinvestigation of the magnetostrictive coefficient \(\lambda_{001}\) for the L1\(_0\) FeNi. The measured values of \(K_1\) of the L1\(_0\) FeNi are relatively high [34, 36] and equal up to 1.0–1.3 MJ m\(^{-3}\). Skomski and Coey [6] suggest that even such high values of \(K_1\) are insufficient to raise FeNi off the category of semi-hard magnets. However, it has been shown for the L1\(_0\) FeNi films, that their intrinsic magnetic properties can be altered e.g. by engineering larger strains [37, 38]. Furthermore, the composition and microstructure of the L1\(_0\) FeNi may be tailored as well to improve the FeNi potential for rare-earth-free permanent magnet application [34]. Skomski points out the beneficial self-organized microstructure of the L1\(_0\) FeNi being reflected in a relatively high coercivity of about 120 mT [39]. Some other characteristics indicating the L1\(_0\) FeNi as a good candidate for hard magnets are magnetization approaching that of the Nd–Fe–B and a relatively high Curie point near 550 °C [40], however preceded by the critical temperature of the ordered state of about 320 °C [27]. Considering from the application point of view, achieving and retaining the L1\(_0\) atomic order remains a serious weakness of the FeNi [38, 41].

Figure 1. The L1\(_0\) crystallographic structure. The solid lines designate a unit cell containing two formula units and the dashed lines confine a unit cell with a single formula.

In this work, we investigate theoretically the magnetocrystalline anisotropy constants \(K_1\) and \(K_3\), Fermi surface and bulk modulus of the tetrataenite. These aspects of L1\(_0\) magnetic materials can be found in a paper of Laukhin et al [29].

2. Computational details

The determination from the first principles of the magnetocrystalline anisotropy constants and the magnetostRICTic coefficient \(\lambda_{001}\) requires fully relativistic electronic band structure calculations. The calculations are carried out by using the full-potential local-orbital minimum-basis scheme FPLO-14.0-49 [30, 42] with the generalized gradient approximation (GGA) in the Perdew–Burke–Ernzerhof (PBE) form [43]. The calculations are performed up to a 80\(^3\) k-mesh with tetrahedron method for integration, an energy convergence criterion 10\(^{-8}\)
Table 1. The lattice parameters ($a$ and $c$), spin ($M_S$) and orbital ($M_L$) magnetic moments, magnetocrystalline anisotropy energies (MAE), magnetostrictive coefficients ($\lambda_{001}$) and bulk moduli ($B_0$) of the L1₀ FeNi phase.

| Quantity unit | $a$ Å | $c$ Å | $M_S$(Fe) $\mu_B$ | $M_S$(Ni) $\mu_B$ | $M_L$(Fe) $\mu_B$ | $M_L$(Ni) $\mu_B$ | MAE or $K_1$ $\mu$eV formula $^{-1}$ | MAE or $K_1$ MJ m$^{-3}$ | $\lambda_{001}$ $10^{-6}$ | $B_0$ GPa |
|---------------|-------|-------|------------------|-------------------|------------------|------------------|-----------------------------|-----------------------------|-----------------|-------|
| Experiment    | 3.57 [38] | 3.57 [38] | 2.54 ± 0.16      | 0.73 ± 0.04       | ~0.05            | 0.10 [33]        | 0.58–1.3                    | ~9 [57]                     |                 |       |
| FLAPW-GGA [31] | 3.58 | 3.58 | 2.71 | 0.052 | 0.69 | 0.038 | 32 | 0.22 | 9.7 |       |
| VASP-GGA [34] | — | — | — | — | — | — | 110 | 0.78 | — | — |
| VASP-GGA [35] | 3.56 | 3.584 | 2.65 | — | 0.61 | — | 78 | 0.56 | — | — |
| WIEN2k-GGA [35] | — | — | — | — | — | — | 69 | 0.48 | — | — |
| WIEN2k-GGA [26] | 3.56 | 3.58 | 2.69 | — | 0.67 | — | 69 | 0.48 | — | — |
| WIEN2k-GGA (this work) | — | — | 2.69 | 0.052 | 0.66 | 0.036 | 67 | 0.47 | — | — |
| ASA-SPR-KKR-GGA [26] | — | — | 2.73 | — | 0.62 | — | 110 | 0.77 | — | — |
| FP-SPR-KKR-GGA (this work) | — | — | 2.69 | 0.053 | 0.60 | 0.036 | 32 | 0.23 | — | — |
| FPLO14-GGA (this work) | 3.56 | 3.58 | 2.72 | 0.054 | 0.53 | 0.039 | 48 | 0.34 | 9.4 | 194 |

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where $\theta$ is the angle between the magnetization direction and the $c$ axis. If the $ab$ initio calculated total energies are fitted in a quadratic form:

$$E(\theta = 0^\circ) = a\theta^2 + bl + c;$$
$$E(\theta = 90^\circ) = a\theta^2 + bl + c + MAE,$$
the equation for magnetostrictive coefficient can be written as:

$$\lambda_{001} = -\frac{2}{3} \frac{\partial^2 E}{\partial \theta^2}.$$

In this work, the MAEs are evaluated based on the total energies calculated self-consistently for two perpendicular quantization axes:

$$MAE = E(\theta = 90^\circ) - E(\theta = 0^\circ).$$

In addition to the FPLO calculations, the Korringa–Kohn–Rostoker (KKR) approach as implemented in the Munich SPR-KKR package (non-public full potential version 7.6.0) is used to calculate the magneto-crystalline anisotropy constant $K_3$ [48, 49]. The advantage of using the full potential method for MAE calculations has been discussed before [16, 50]. The $ab$ initio calculations of $K_3$ are still a numerically demanding task today. But what distinguishes the SPR-KKR from the other $ab$ initio codes is a numerical accuracy on the level of about 0.1 $\mu$eV in calculating the total energy, which is the same order of magnitude as expected for the $K_3$ value of the L1$_0$ FeNi. Another argument in favor of the SPR-KKR is that it has been successfully applied before to calculate $K_3$ for the magnetic shape memory Fe–Pd alloys [51]. In order to get converged values of $K_3$ for the L1$_0$ FeNi, the FP-SPR-KKR parameters of the 10$^{-10}$ $\text{Ry}$ energy convergence criterion and up to 225 $\times$ 225 $\times$ 158 k-points (about 8 million) are necessary. For Brillouin zone integration, the special point method with a regular k-point grid is used. Khan et al have shown for L1$_0$ FePt phase [24] that the KKR calculations of total energies are also quite sensitive to the angular momentum expansion $l_{\text{max}}$ cutoff used for the multipole expansion of the Green function. Khan et al concluded that the angular momentum $l_{\text{max}} = 3$ cutoff yields to a qualitatively correct value of the MAE, however even for $l_{\text{max}} = 7$ a full convergence is still difficult to reach. Taking this conclusion into account, we choose for our calculations a maximum angular momentum value $l_{\text{max}} = 4$ ($N_L = 5$ in the KKR configuration file). Our decision is further motivated by the results of a convergence test of $K_1$ with respect to the angular momentum, performed up to $l_{\text{max}} = 6$. The test indicated that problems with the convergence occur above the $l_{\text{max}} = 4$ leading to a divergence of the $K_1$ value. This behavior may come from numerical problems in evaluating the Madelung potential and near-field corrections, pointed out by Khan et al [24]. The energy integrals are evaluated by contour integration on a circular energy path in the complex plane (GRID = 5), using 40 points of the E-mesh. The calculations within the FP-SPR-KKR are carried out with the PBE exchange-correlation potential and with the same crystallographic parameters of FeNi as used for FPLO MAE calculations. The magneto-crystalline anisotropy energy in the tetragonal crystal can be described by the following equation [52]:

$$MAE = K_1 \sin^2 \phi + K_2 \sin^4 \theta + K_3 \sin^4 \theta \cos 4\phi,$$
where \( K_i \) are the anisotropy constants, \( \theta \) is the angle between the magnetization direction and the \( c \) axis, and \( \phi \) is the angle between the magnetization and the \( a \) axis within the basal plane of a tetragonal lattice. For \( \theta = 90^\circ \), the equation (7) takes the following form:

\[
\text{MAE} - K_1 - K_2 = K_3 \cos 4\phi .
\]

(8)

The \( K_3 \) is then evaluated based on the total energies \( E_{100} \) and \( E_{110} \) calculated self-consistently for \( \phi \) equal to 0° and 45° (directions [1 0 0] and [1 1 0] in the BCT unit cell) from equation:

\[
2K_3 = E_{100} - E_{110} .
\]

(9)

The computational parameters used to obtain \( K_1 \) and \( K_2 \) with FP-SPR-KKR are the same as presented above for the \( K_3 \) calculations, with an exception that a lower number of \( k \)-points, about 1.5 million, is used in the whole Brillouin zone.

Furthermore, the full-potential linearized augmented plane-wave method FP-LAPW, as implemented in the WIEN2k code [53] is used to calculate a reference value of MAE. Muffin-tin radii RMT are 2.29 \( a_0 \) for Fe and 2.29 \( a_0 \) for Ni atoms, where \( a_0 \) is the Bohr radius. The PBE exchange-correlation potential is used. The plane wave cut-off parameter \( RK_{\text{max}} \) is set to 10, which leads to above 245 basis functions. Relativistic effects are included with the second variational treatment of spin-orbit coupling. The total energy convergence criterion is set to \( 10^{-8} \) Ry. The 55 \( \times \) 55 \( \times \) 39 \( k \)-points (about 120 thousand) are used in the whole Brillouin zone.

Although the reproducibility of the results in density functional theory calculations of solids has been well established on the level of scalar relativistic estimation of the lattice parameters [54], the accurate calculations of the magnetocrystalline anisotropy energy MAE and its derivatives, like magnetostriective coefficient, remain a challenge [24]. It is even harder to calculate the values of the magnetocrystalline anisotropy constant \( K_3 \) which happens to be two orders of magnitude smaller than MAE [51]. In this work, we managed to calculate the \( K_3 \) of the L1\(_0\) FeNi with the full potential, thanks to the very accurate convergence tests.

3. Density functional theory calculations

The L1\(_0\) FeNi phase has been studied \textit{ab initio} several times before [26, 31, 35, 55]. In this work, we reinvestigate the magnetocrystalline anisotropy energy MAE and the magnetostrictive coefficient \( \lambda_{001} \) of the L1\(_0\) FeNi phase and calculate its magnetocrystalline anisotropy constant \( K_2 \), \( K_3 \), Fermi surface and bulk modulus. Our results, together with the literature data, are summarized in table 1.

3.1. Bulk modulus

We start from the calculations of the equilibrium volume (22.686 Å\(^3\)), followed by the calculations of the equilibrium \( c/a \) ratio (1.0036), see figure 2. The optimization leads to a tetragonal structure with the lattice parameters \( a = 3.56 \) Å and \( c = 3.58 \) Å. The energy-volume data, see figure 2(a), allow us to calculate the bulk modulus \( B_0 \) for the L1\(_0\) FeNi by fitting the third-order Birch–Murnaghan equation of state [58]. It leads to a \( B_0 \) equal to 194 GPa at 0 K. The previous calculations with coherent potential approximation (CPA) of the Fe\(_{0.5}\)Ni\(_{0.5}\) random alloy have given the \( B_0 \) value of about 220GPa [59]. For comparison, the experimental values of \( B_0 \) for the Fe\(_{0.5}\)Ni\(_{0.5}\) alloy measured at room temperature vary between 165 GPa and 177 GPa [60] and the experimental \( B_0 \) values for Fe and Ni are about 170 GPa and 180 GPa, respectively.

3.2. Magnetostrictive coefficient \( \lambda_{001} \)

The total and magnetocrystalline anisotropy energies \textit{versus} the length of the lattice parameter \( c \) are presented in figure 3, as necessary to evaluate the magnetostriective coefficient \( \lambda_{001} \) from equation (5). The total energy and MAE are calculated based on the optimized crystal structure in a constant volume mode adopted for the distortion. The MAE calculated for the equilibrium L1\(_0\) FeNi structure equals 48 \( \mu \text{eV} \) formula\(^{-1} \) (0.34 MJ m\(^{-3}\)). Initial tests performed with volume relaxation for 50\(^3\) \( k \)-points have shown that the constant volume mode underestimates the \( \lambda_{001} \) of the L1\(_0\) FeNi by about 10%. For the price of this inaccuracy, in the constant volume mode, we can perform calculations up to 80\(^3\) \( k \)-points. The resultant \( \lambda_{001} \) obtained for 60\(^3\), 70\(^3\), and 80\(^3\) \( k \)-points are equal to 11.3 \( \times \) 10\(^{-6}\), 8.7 \( \times \) 10\(^{-6}\), and 9.4 \( \times \) 10\(^{-6}\), respectively. The variation of the estimated \( \lambda_{001} \) with the number of \( k \)-points comes from the numerical inaccuracy in the evaluation of a very small quantity as MAE. Nevertheless,
the calculated here $\lambda_{001} = 9.4 \times 10^{-6}$ stays in good agreement with the previous theoretical result by Wu and Freeman [31], $\lambda_{001} = 9.7 \times 10^{-6}$, and with the experimental value by Bozorth [57] $\lambda_{100} \sim 9 \times 10^{-6}$, $\lambda_{001} \sim 10 \times 10^{-6}$ for the L10 FeNi is a rather small value. It is of the same order of magnitude as for elements Fe and Ni and three orders of magnitude smaller than for the magnetostrictive material Terfenol-D.

3.3. Magnetic moments

The spin magnetic moments calculated with the FPLO for the L10 FeNi are equal to $2.72 \mu_B$ for the Fe atom and $0.53 \mu_B$ for the Ni atom, see table 1. The calculated spin magnetic moments on Fe and Ni stay in relatively good agreement with the values measured on Fe equal to $2.54 \pm 0.16 \mu_B$ and on Ni equal to $0.73 \pm 0.04 \mu_B$ for the L10 FeNi phase [56]. The calculated orbital magnetic moments are equal to $0.054 \mu_B$ for the Fe atom and $0.039 \mu_B$ for the Ni atom. They are also close to the experimental values on Fe equal to $\sim 0.05 \mu_B$ and on Ni equal to $0.10 \mu_B$ as measured for the L10 FeNi phase [33]. The calculated and measured orbital magnetic moments on Fe in the L10 FeNi are reduced in comparison to the experimental value of $0.086 \mu_B$ for the bcc iron [61]. The calculated orbital magnetic moments on Ni (0.039 $\mu_B$) in the L10 FeNi are also reduced with respect to the experimental value of $0.055 \mu_B$ for the fcc nickel [62]. The calculated total magnetic moment is equal to $3.34 \mu_B$ formula$^{-1}$ (1.67 $\mu_B$ atom$^{-1}$), which is not a particularly high value for 3d-based magnetic materials. From the perspective of hard magnetic materials, this reduction of the magnetic moment in comparison to e.g. pure bcc Fe is beneficial for the magnetic hardness but adversely affects the energy product.

3.4. Magnetocrystalline anisotropy energy

Getting consistent MAE results from different first principles codes is still a challenge. The difficulties come from such factors as a complex shape of the valence band structure or a demand of very high numerical accuracy [24]. The differences between the results from various codes may come from the application of different approximations like the atomic sphere approximation, the lack of crystal structure optimization or the insufficient number of $k$-points [16, 24, 63]. Some recent papers discuss, however, the reproducibility of the MAE between the WIEN2k and KKR methods [24] and between FPLO and WIEN2k [16].

The calculated in GGA MAEs of the L10 FeNi taken from the literature are equal to $0.22$ [31], $0.48$ [26], $0.56$ [35] and $0.78$ [34] MJ m$^{-3}$. The experimental determination of the L10 FeNi magnetocrystalline anisotropy constant $K_1$ is ambiguous as well, with a spread in the $K_1$ values from 0.58 [32], trough 0.67 [27], up to 1.3 [34] MJ m$^{-3}$. The MAEs of the L10 FeNi calculated in GGA in this work are equal to 0.23, 0.34 and 0.47 MJ m$^{-3}$ from FP-SPR-KKR, FPLO and WIEN2k, respectively.

Based on the above theoretical results, we expect that the accurate GGA value of the MAE for the L10 FeNi is below 0.5 MJ m$^{-3}$. Such a value could be an argument for removing the L10 FeNi from the list of candidates for rare-earth free permanent magnets. Some authors suggest, however, that bare GGA is insufficient for describing the L10 FeNi and a
consideration of orbital polarization corrections is necessary [35, 55]. It has been shown that the inclusion of the orbital polarization causes a significant increase of the MAE of the L10 FeNi (from ~0.55 to ~1.23 MJ m⁻³) [55] and (from 0.48 to 0.84 MJ m⁻³) [35]. Unfortunately, even the MAE of about 1 MJ m⁻³ may be insufficient to raise the L10 FeNi from the category of semi-hard magnets [6]. To reach that goal, further efforts on increasing the magnetocrystalline anisotropy have to be made.

3.5. Magnetocrystalline anisotropy constant \(K_3\)

The magnetocrystalline anisotropy constant \(K_3\) of the L10 FeNi is particularly interesting from a perspective of spintronic applications of the material, e.g. for the magnetic tunnel junction [35]. In a tetragonal crystal, the \(K_3\) can be defined by equation (7). Unfortunately, we cannot calculate the \(K_3\) with the FPLO code, which was applied to obtain previous results. To get the \(K_3\), we then use the FP-SPR-KKR package, which produces the value of \(K_3\) equal to 110 J m⁻³. It is four orders of magnitude smaller from the value of MAE or \(K_1\) for the L10 FeNi. For comparison, the values of \(K_3\) calculated in ASA-SPR-KKR for magnetic shape memory alloys Fe–Pd [51] have a similar order of magnitude (~10³ J m⁻³) as the one presented above. The details of the calculation method and the motivations for using the FP-SPR-KKR package for evaluating \(K_3\) are presented in the introductory section. As the \(K_3\) is \(k\)-mesh sensitive, the convergence test is made, see figure 4. It can be noticed that a large number of \(k\)-points is necessary to get a satisfactory convergence of \(K_3\). To complete the FP-SPR-KKR analysis, the \(K_1\) and \(K_3\) magnetocrystalline anisotropy constants are also calculated. In figure 5, we present the energy dependence as a function of the polar angle \(\theta\) between the magnetization direction and the \(c\) axis. Parameters from a fit to \(K_1 \sin^2 \theta + K_2 \sin^4 \theta\) are \(K_1 = 0.23\) MJ m⁻³ and \(K_2 = -2.0\) kJ m⁻³, see equation (8). The lowest energy in figure 5 corresponds to the [001] quantization axis and the highest energy to the [100] axis.

3.6. Fermi surface

Figure 6 presents the Fermi surface of the L10 FeNi. The tetragonal crystal structure of the L10 FeNi is reflected in the uniaxial anisotropy and a four-fold symmetry of Fermi surface sheets. The seven sheets are divided into two groups of closed hole pockets and open sheets. The nested hole pockets (a)–(c) are centered at the high symmetry point \(M\). One other hole pocket is centered at point \(\Gamma\), see panel (e). When the hole pockets permit for only closed orbits, the mainly open sheets (d)–(f) allow for both the opened and closed orbits.

We have calculated the Fermi surface of the L10 FeNi both as the basic characteristic of the material and as the basis for a \(k\)-resolved MAE analysis, which will be presented in the next paragraph. Basic knowledge on the Fermi surface of the crystal lets us think on manipulating its properties using the emerging method called Fermi surface engineering. The method makes relations between the shape of the Fermi surface and external factors like doping or strain. The Fermi surface engineering technique has been successfully applied e.g. to transparent conductors [64] and superconductors [65].

3.7. MAE analysis in reciprocal space

Most often, the anisotropic magnetic properties of the materials are analyzed in the real space. The spin and orbital moments and magnetocrystalline anisotropy constants are presented as functions of the atomic position or angle [23, 35, 66]. The magnetocrystalline anisotropy energy (MAE) is one of the most important intrinsic properties of the hard magnetic crystals, which can be analyzed in reciprocal space as a \(k\)-resolved quantity. The MAE can be determined with the magnetic force theorem [31, 67, 68] from a formula:

\[
\text{MAE} = E(\theta = 90^\circ) - E(\theta = 0^\circ) = \sum_{\text{occ}} \epsilon_i(\theta = 90^\circ) - \sum_{\text{occ'}} \epsilon_i(\theta = 0^\circ) + O(\delta \rho^s),
\]

(10)

where \(\epsilon_i\) is the band energy of the \(i\)th state, \(\theta\) is the angle between the magnetization direction and the \(c\) axis and \(O(\delta \rho^s)\) is the correction term. This modern computer technique allows us to visualize the MAE(\(k\)) structure in a three-dimensional (3D) Brillouin zone. The 3D maps of magnetocrystalline anisotropy have been recently calculated for (Fe/Co)₃B alloys [18]—another candidate for rare-earth free permanent magnets. However, usually the MAE(\(k\)) dependences are presented along the 1D \(k\)-path [16, 31, 47]. Although various authors have attempted to interpret the MAE(\(k\)) without...
a 3D-resolution, in our opinion, the \( \mathbf{k} \)-path or single \( \mathbf{k} \)-point analyses do not properly cover the complexity of magnetocrystalline anisotropy. We think that in order to get a full picture of it, the entire Brillouin zone should be considered [16]. For a hard magnetic material, the calculated 3D-MAE(\( \mathbf{k} \)) plot is a unique characteristic as for example the Fermi surface is for a metal. What is interesting is that the connections between the 3D-MAE(\( \mathbf{k} \)) and the Fermi surface go beyond the above comparison and a fact is a close relation between these two. Both the 3D-MAE(\( \mathbf{k} \)) and Fermi surface have reciprocal space electronic structure characteristics. Furthermore, as the Fermi level is an upper integration boundary of the MAE, the sheets of the Fermi surface indicate the borders between the distinct regions of the MAE. Because of that, the Fermi surface is a link between the calculated 3D-MAE(\( \mathbf{k} \)) and the experiment. The 3D-MAE(\( \mathbf{k} \)) analysis is also relevant for hard magnetic materials because it can determine directions to improve the MAE. In relation to the Fermi surface engineering, an improvement technique based on the MAE(\( \mathbf{k} \)) analysis could be called magnetocrystalline anisotropy engineering.

In figure 7, we present the calculated 3D-MAE(\( \mathbf{k} \)) plots of the L1\(_0\) FeNi. We can see that the whole Brillouin zone is filled by the positive (red) and negative (blue) contributions. The magnitude of the \( \mathbf{k} \)-resolved contributions is in the order of the \( 10^{-3} \) eV per \( \mathbf{k} \)-point, where the value of the total MAE is three orders of magnitude smaller (48 \( \mu \)eV formula\(^{-1} \)). A comparison between the MAE contributions and total value indicates a fine compensation of the relatively large negative and positive components. From figure 7, it is easy to notice that the overall shape of the MAE(\( \mathbf{k} \)) is dictated by the Fermi surface. The Fermi surface sheets divide the Brillouin zone into several mainly positive or negative regions. The two most prominent positive contributions come from (1) the vicinity of the R-Z line along the [100] quantization axis and (2) from the spherical region around the M point. Actually, around the M point, we observe a sequence of negative, positive and again negative regions filling the volumes between the corresponding nested hole pockets of the Fermi surface. A similar alternating ordering of the \( \mathbf{k} \)-resolved contributions to the anisotropy constant \( K \) has been presented for the (Fe/Co)\(_2\)B alloys [18].

In figure 8, the cross-sections of the MAE(\( \mathbf{k} \)) for planes \( \Gamma-X-M \) and R-Z-A are shown together with the corresponding cross-sections of the Fermi surface. With these plots, it is even easier to notice how the Fermi surface splits the regions with differing MAE(\( \mathbf{k} \)) components. The MAE cross-sections also confirm that the first hole pocket around the M point contains mainly negative \( \mathbf{k} \)-resolved contributions and the regions between the next two hole pockets are respectively positive and negative. Although the MAE(\( \mathbf{k} \)) structure reflects the uniaxial anisotropy of the crystal, it is easy to notice how the four-fold symmetry is broken. A reason for this is that the [100] direction has been distinguished as a quantization axis. The above-described MAE(\( \mathbf{k} \)) analysis reveals details of the structure of magnetocrystalline anisotropy of the L1\(_0\) FeNi, which are the distribution of positive and negative components, the magnitude of these shares, or the detailed relationship between the MAE(\( \mathbf{k} \)) and the Fermi surface. Unfortunately, an extra fine compensation of the large MAE(\( \mathbf{k} \)) contributions makes it difficult to predict the roads to increase the MAE of the L1\(_0\) FeNi. Because of the compensation effect, even large changes of the MAE(\( \mathbf{k} \)) structure will finish as hard to predict small changes to the MAE. Based on the combined 3D-MAE and Fermi surface analysis, we are able, however, to point out the sheets of Fermi surface which should be extended or reduced to enlarge the regions of positive MAE contributions. Unfortunately, this simple line of reasoning is difficult to realize in practice. Taking into account the innovative nature of the combined 3D-MAE and Fermi surface analysis, we do not exclude that this approach will be more fruitful for another hard magnetic material.

4. Summary and conclusions

The magnetocrystalline anisotropy energy MAE of the L1\(_0\) FeNi calculated in this work within GGA goes below 0.5 MJ m\(^{-3} \). It has been shown in the literature that the more reliable model including orbital polarization corrections gives about twice this value, whereas the experimental values of the anisotropy constant \( K_1 \) from the literature oscillate around 1.0 MJ m\(^{-3} \). Regarding the application of the L1\(_0\) FeNi as rare-earth free permanent magnets, the above numbers of MAE/\( K_1 \) are rather discouraging. Other known limitations of tetrataenite are the practical difficulties with synthesis of the ordered phase and the relatively low critical temperature of the ordered state. However, the L1\(_0\) FeNi still has potential to improve its MAE by modifications, like e.g. a tetragonal strain or an interstitial or substitutional alloying. In favor of the L1\(_0\), FeNi speaks also to the high saturation magnetization and Curie temperature. The L1\(_0\) FeNi with well defined anisotropic parameters may also find applications in modern electronics. From the point of view of computations, it can be also used as a reference standard for advanced \textit{ab initio} calculations, due to a relatively simple crystal structure.

In the reciprocal space analysis of the hard magnetic materials, it is common to present the MAE(\( \mathbf{k} \)) contributions together with the bandstructure along the \( \mathbf{k} \)-path. In this work, we have shown the 3D \( \mathbf{k} \)-resolved analysis of the MAE (3D-MAE), which gives a complete picture of the MAE contributions in the Brillouin zone. Unfortunately, in case of the L1\(_0\) FeNi, we cannot use the \( \mathbf{k} \)-resolved analysis to give specific suggestions. In return, we show the close interconnection between the 3D-MAE and the Fermi surface. We expect that analysis of the effect of strain or alloying on the \( \mathbf{k} \)-resolved MAE may allow us to improve the properties of hard magnetic materials.

The calculated spin and orbital magnetic moments and magnetostrictive coefficient \( \lambda_{001} \) stay in acceptable agreement with the previous theoretical results and measurements. The calculated bulk modulus \( B_0 \), magnetocrystalline anisotropy constants \( K_2 \) and \( K_3 \) and Fermi surface require experimental confirmation.

The conducted full relativistic calculations can be also used to formulate several general conclusions regarding the computational parameters. The fact that that spin-orbit coupling is...
the origin of the orbital magnetic moment, magnetocrystalline anisotropy and magnetostriction is well known. In order to accurately calculate the magnetic parameters, it is also crucial to consider the full potential. The significant discrepancies between the calculated MAEs even from various full potential GGA implementations show how sensitive the calculations are. To improve the reliability of the results, we suggest choosing the computational parameters very carefully and using a second code for cross-checking. For better description of the magnetic moments in the considered system, the orbital polarization corrections and dynamical mean field theory can be used. In this work, we have also shown that it is technically possible to calculate the anisotropy constant $K_3$ with full potential. The calculated value of $K_3$ is four orders of magnitude smaller than the MAE and a huge number of $k$-points is indispensable to get a consistent value.

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