The origin of perpendicular magneto-crystalline anisotropy in L10–FeNi under tetragonal distortion

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Received 30 November 2012
Published 8 February 2013
Online at stacks.iop.org/JPhysCM/25/106005

Abstract

We investigated the origin of perpendicular magneto-crystalline anisotropy (MCA) in L10-ordered FeNi alloy using first-principles density-functional calculations. We found that the perpendicular MCA of L10-FeNi arises predominantly from the constituent Fe atoms, which is consistent with recent measurements of the anisotropy of the Fe orbital magnetic moment of L10-FeNi by means of x-ray magnetic circular dichroism. Analysis of the second-order perturbation of the spin–orbit interaction indicates that spin-flip excitations between the occupied majority-spin and unoccupied minority-spin bands make a considerable contribution to the perpendicular MCA, as does the spin-conservation term for the minority-spin bands. Furthermore, the MCA energy increases as the in-plane lattice parameter decreases (increasing the axial ratio c/a). The increase in the MCA energy can be attributed to further enhancement of the spin-flip term due to modulation of the Fe d(xy) and d(x2−y2) orbital components around the Fermi level under compressive in-plane distortion.

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

1. Introduction

Ferromagnetic materials with strong magneto-crystalline anisotropy (MCA) have attracted much attention as regards application in high density magnetic recording media and nonvolatile magnetoresistive random access memory (MRAM). In order to ensure sufficient endurance against thermal fluctuations when downsizing a recording bit or memory cell, it is inevitable that ferromagnetic materials with higher MCA, such as L10-ordered FePt [1], CoPt [2], and D022-ordered Mn3−δGa, will be adopted [3]. Furthermore, perpendicularly magnetized electrodes of magnetic tunnel junctions (MTJs) lower the critical current density for current-induced magnetization switching by spin-transfer torque [4].

Recently, L10-ordered FeNi, which is free from rare-earth and/or noble metal elements, has been successfully fabricated as thin films using an alternative monatomic layer deposition technique [5–8]. The MCA energy of L10-FeNi thin films depends on the kinds of buffer layers used [7] as well as the degree of chemical order [8]. Furthermore, x-ray magnetic circular dichroism (XMCD) and magneto-optical Kerr effect measurements were carried out on alternately layered FeNi thin films grown on Ni/Cu(001) substrates [9]. By analyzing the Fe L-edge XMCD spectra using the sum rule [10, 11], the anisotropy of the orbital magnetic moment was obtained, and it was found that the Ni-sandwiched Fe layer has a positive MCA energy of 60 µeV, while the Fe-sandwiched Ni layer has a positive MCA energy of 60 µeV.

For ordered L10-type alloys TX (T = Mn, Fe, Co, Ni, X = Au, Pt, Pd, Co, Ni) the MCA has been well studied theoretically using first-principles density-functional calculations [12–20]. For L10-type FePt or CoPt, the strong perpendicular MCA was attributed to the Pt atom due to
strong spin–orbit interaction, in which Fe or Co atoms only induce the spin magnetic moment on the Pt site [12]. On the other hand, L1₀-FeNi also shows a perpendicular MCA, despite the weak spin–orbit interaction of Ni as compared with that of Pt. Therefore, the physical origin of the perpendicular MCA of L1₀-FeNi remains unclear at present. Furthermore, recent XMC measurement studies reported the anisotropy of the orbital magnetic moment [9, 22]. These reports discussed the MCA on the basis of Bruno’s relation [21], in which the MCA energy is proportional to the difference in orbital magnetic moment between two magnetization directions. However, there are additional terms related to spin-flip excitations between the exchange-splitting majority-spin and minority-spin bands in the second-order perturbation of the spin–orbit interaction. The effects of the spin-flip terms on the MCA energies can be neglected for systems with strong exchange coupling, such as a free-standing Fe or Co monolayer [23], while the spin-flip term makes a large contribution to the MCA for systems including nonmagnetic elements. In fact, previous theoretical work revealed an enhancement of the Pt orbital magnetic moment of L1₀-FePt in the in-plane magnetization direction [14, 12], while real-space analysis of the MCA energy indicated a strong perpendicular contribution from Pt atoms [12]. This means that the spin-flip term is essential for understanding the perpendicular MCA of ordered L₁₀-type alloys.

In this paper, we discuss the MCA energy of L₁₀-FeNi theoretically on the basis of first-principles density-functional calculations in order to elucidate the physical origin of the perpendicular MCA in L₁₀-FeNi. In particular, we focus our attention on the spin and atomic site dependence of the MCA energy using a second-order perturbation of the total energy due to the spin–orbit interaction. Furthermore, we discuss the dependence of the MCA energy on the in-plane lattice constant of L₁₀-FeNi, providing useful guidelines for the choice of buffer layers in L₁₀-FeNi thin films for realizing larger MCA energy.

2. The computational procedure

2.1. First-principles calculations

We carried out first-principles density-functional calculations using the Vienna ab initio simulation package (VASP) [24, 25]. For the exchange and correlation energy, we adopted the spin-polarized generalized gradient approximation (GGA) proposed by Perdew, Becke, and Ernzerhof (PBE) [26]. The nuclei and core electrons were described using the projector augmented-wave (PAW) potential [27, 28] and the spin-polarized generalized gradient approximation (GGA) [25]. For the exchange and correlation energy, we adopted the spin-polarized generalized gradient approximation (GGA) [25]. For the exchange and correlation energy, we adopted the spin-polarized generalized gradient approximation (GGA) [25].

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The wavefunctions of the valence electrons were expanded in a plane-wave basis set with a cutoff energy of 336.9 eV. The k-space integration was performed using a modified tetrahedron method with Blöchl corrections [29] with 19 800 k-points in the first Brillouin zone corresponding to the unit cell of L₁₀-FeNi. As the unit cell of L₁₀-FeNi, which we call the ‘calculation cell’ here, we adopted the tetragonal structures represented by the solid lines in figure 1. The lattice parameter c along the perpendicular (z) direction for each in-plane lattice constant a was optimized to minimize the total energy within a tolerance of 10 µeV. The spin–orbit interaction was included through the force theorem [30]. We determined the MCA energy from the difference of the sums of energy eigenvalues for magnetizations oriented along the in-plane [100] and out-of-plane [001] directions. The MCA energy was defined to be positive when the magnetic easy axis was perpendicular to the plane. We calculated the MCA energies of L₁₀-FeNi by carrying out all-electron calculations using the full-potential linearized augmented-plane-wave (FLAPW) method encoded in the WIEN2k package [31] to check our calculation results.

2.2. Second-order perturbation

In order to obtain a qualitative understanding of the origin of the MCA in L₁₀-FeNi, we consider the second-order perturbation of the total energy due to the spin–orbit interaction $H_{SO}$ [21, 32, 33], which is given by

$$E^{(2)} = - \sum_{kn} \langle \langle n \sigma | H_{SO} | kn \rangle \rangle^2,$$

where $| kn \rangle$ is an unperturbed state of energy $\epsilon_{kn}^{(0)}$ with indices for the k-point k, band n, and spin $\sigma$. In the tight-binding regime for spin–orbit coupling, $H_{SO}$ is given by the sum of the contributions from each atomic site i or j, due to the localized character of the spin–orbit coupling constant $\xi_i$, $H_{SO} = \sum_i \xi_i L \cdot S$, (2)

where S and L are the single-electron spin and angular momentum operators, respectively. By expanding $| kn \rangle$ with an orthogonal basis of atomic orbitals labeled as $\mu(\lambda)$, i.e., $| kn \rangle = \sum_{\lambda \nu} \phi_{\lambda \nu} | \mu \rangle$, we can obtain the second-order contribution of $H_{SO}$ to the total energy as a sum over terms depending on spin-transition processes, atomic orbitals, and

![Figure 1. Schematic figures of the atomic arrangement in L1₀-FeNi. The coordinate system and the unit cell used in the calculation (calculation cell) are represented by bold solid lines.](image-url)
We directly estimated the second-order perturbative contribution without the spin-orbit interaction.

\[
E^{(2)} = - \sum_{\lambda \mu \lambda' \mu'} \langle \lambda \uparrow | L \cdot S | \lambda' \uparrow \rangle \langle \mu \uparrow | L \cdot S | \mu' \uparrow \rangle \\
\times \sum_{i,j} \xi_i^j \xi_j^i \sum_{\mu} G_{\mu \lambda}^{\mu \lambda'} (\uparrow, \uparrow; i, j) + G_{\mu \lambda}^{\mu \lambda'} (\downarrow, \downarrow; i, j) \\
- G_{\mu \lambda}^{\mu \lambda'} (\downarrow, \uparrow; i, j) - G_{\mu \lambda}^{\mu \lambda'} (\uparrow, \downarrow; i, j),
\]

(3)

where \( G_{\mu \lambda}^{\mu \lambda'} (\sigma, \sigma'; i, j) \) is an integral of the joint local density of states (LDOS) given by

\[
G_{\mu \lambda}^{\mu \lambda'} (\sigma, \sigma'; i, j) = \int_{-\infty}^{\infty} d\epsilon \int_{-\infty}^{\infty} d\epsilon' \frac{1}{\epsilon' - \epsilon} \\
\times \sum_{k} \sum_{\alpha} c_{\lambda \mu \sigma}^{kn} c_{\lambda' \sigma}^{kn} \delta (\epsilon - \epsilon_{kn\sigma}^{(0)}) \\
\times \sum_{\alpha'} c_{\lambda \mu \sigma'}^{kn} c_{\lambda' \sigma'}^{kn} \delta (\epsilon' - \epsilon_{kn\sigma'}^{(0)}),
\]

(4)

where \( E_F \) is the Fermi energy. The matrix elements of \(| \mu' \uparrow | L \cdot S | \mu \uparrow \rangle \) are given in [33] as functions of the spherical angular coordinates \( \theta \) and \( \phi \), which are defined as polar and azimuthal angles between the spin quantization axis and the crystal \( z \) axis. The MCA energy is derived as a difference between in-plane \((\theta = \pi/2, \phi = 0)\) and perpendicular \((\theta = 0, \phi = 0)\) magnetization directions, including the matrix elements \(| \mu' \uparrow | L_X | \mu \uparrow \rangle \) and \(| \mu' \uparrow | L_Z | \mu \uparrow \rangle \), respectively. We introduce a second-order contribution to the MCA energy depending on the atomic site and the spin-transition process as follows:

\[
E^{(2)}_{\text{MCA}} = \sum_i E^{i}_{\text{MCA}} \\
= \sum_i \Delta E^{i}_{\uparrow \rightarrow \uparrow} + \Delta E^{i}_{\uparrow \rightarrow \downarrow} - \Delta E^{i}_{\downarrow \rightarrow \downarrow} - \Delta E^{i}_{\downarrow \rightarrow \uparrow},
\]

(5)

and

\[
\Delta E^{i}_{\sigma \rightarrow \sigma'} = -\xi_i \sum_{\mu \lambda} \sum_{\mu'} \sum_{\lambda'} \langle \lambda \uparrow | L_X | \lambda' \uparrow \rangle \langle \mu' \uparrow | L_X | \mu \uparrow \rangle \\
- \langle \lambda \uparrow | L_Z | \lambda' \uparrow \rangle \langle \mu' \uparrow | L_Z | \mu \uparrow \rangle \\
\times \sum_{j} \xi_j G_{\mu \lambda}^{\mu' \lambda'} (\sigma, \sigma'; i, j).
\]

(6)

We directly estimated the second-order perturbative contribution to the spin and atomic site dependent MCA energy \( \Delta E^{i}_{\sigma \rightarrow \sigma'} \) by calculating \( c_{\mu \lambda, \sigma}^{kn} \) using first-principles calculations without the spin-orbit interaction. \( c_{\mu \lambda, \sigma}^{kn} \) can be obtained in the PAW formulation as a sum of the plane-wave part and the augmented part. Equation (6) includes a summation over atomic sites \( j \), indicating that not only on-site effects but also effects of hybridization with other atoms are incorporated through virtual excitations from an occupied level to an unoccupied level in the second-order perturbation. Note that \( \Delta E^{i}_{\sigma \rightarrow \sigma'} \) in equation (6) depends quantitatively on the choice of the spin-orbit coupling constant \( \xi_i \) and the atomic radius. We checked that the typical values \( \xi_F = 50 \text{ meV} \) and \( \xi_{Ni} = 100 \text{ meV} \) [21] in equations (3) and (6) give a qualitative agreement with results from first-principles calculations including the spin-orbit interaction.

3. Results

3.1. Magneto-crystalline anisotropy and the orbital magnetic moment

We found that the magnetization of L1\textsubscript{0}-FeNi favors the [001] direction, with an MCA energy of 0.078 meV per formula unit (f.u.) for the equilibrium lattice constants \( a_0 = 3.556 \text{ Å} \) and \( c_0 = 3.584 \text{ Å} \), leading to \( K_a = 0.56 \text{ MJ m}^{-3} \). \( K_a \) is the uniaxial MCA constant and is defined by \( K_a = E_{\text{MCA}}/V \), where \( V \) is the volume of the calculation cell. This value is comparable with experimental observations; i.e., 0.63 and 0.70 MJ m\textsuperscript{-3} [5, 7]. Note, however, that the long-range chemical order parameters have been 0.5 or less for samples fabricated so far [8]. On the other hand, for completely L1\textsubscript{0}-ordered FeNi, the calculated order parameter value is 1. Experimental observations have indicated that the \( K_a \) of L1\textsubscript{0}-FeNi is roughly proportional to the order parameter [8]. For L1\textsubscript{0}-ordered FePt, it was observed experimentally that \( K_a \) increases as a quadratic function of the order parameter [34]. The same behavior was confirmed in previous theoretical studies based on the coherent-potential approximation (CPA) [17, 15, 20]. The spin magnetic moments of the constituent atoms in L1\textsubscript{0}-FeNi are independent of the magnetization direction, and were evaluated as 2.65 \( \mu_B \) and 0.61 \( \mu_B \) for Fe and Ni atoms, respectively. On the other hand, the orbital magnetic moment of each atom shows a characteristic dependence on the magnetization direction. The anisotropy of the orbital magnetic moment defined by \( \Delta \mu_{\text{orb}} = \mu_{\text{orb}}^{[001]} - \mu_{\text{orb}}^{[100]} \) takes a positive value, 0.006 \( \mu_B \), for Fe, whereas it is negative, \(-0.002 \mu_B \), for Ni in L1\textsubscript{0}-FeNi.

Recently, Kotsugi and co-workers performed XMC measurements of alternately layered FeNi thin films grown on Cu/Au buffer layers [22]. They found that the angular
distribution in the Fe orbital magnetic moment shows a clear and strong angular dependence, while the Ni orbital magnetic moment displays no clear angular distribution. This indicates that the anisotropy of the Fe orbital magnetic moment is a main contributing factor for the perpendicular MCA of L1₀-FeNi, which is reasonably consistent with the present theoretical results.

3.2. The origin of the perpendicular magneto-crystalline anisotropy

According to Bruno’s relation [21], the MCA energy is proportional to the anisotropy of the orbital magnetic moment. To confirm the validity of Bruno’s relation in L1₀-FeNi, we evaluate the second-order perturbative contribution to the MCA energy $\Delta E_{\sigma \rightarrow \sigma'}^{\text{Fe}}$ given in equation (6). Figure 2 shows a bar graph of $\Delta E_{\sigma \rightarrow \sigma'}^{\text{Fe}}$ for L1₀-FeNi in the second-order perturbation. We found that the Fe atom makes a positive contribution to the perpendicular MCA, while the Ni atom makes a negative contribution. These results qualitatively agree with the behavior of the orbital magnetic moment. However, the spin-flip term $\Delta E_{\uparrow \rightarrow \downarrow}^{\text{Fe}}$ makes a considerable contribution to the perpendicular MCA as does the spin-conservation term $\Delta E_{\uparrow \rightarrow \uparrow}^{\text{Fe}}$. This means that the evaluation of MCA from the Fe orbital magnetic moment using Bruno’s relation [21] will underestimate the MCA energy of L1₀-FeNi because of the absence of the spin-flip term in the orbital magnetic moment. The large spin-flip term $\Delta E_{\uparrow \rightarrow \downarrow}^{\text{Fe}}$ can be attributed to the large majority-spin LDOS of Fe just below the Fermi level (see figure 3(a)), which is due to the hybridization of Fe d orbitals with Ni d orbitals having weak exchange splitting compared with Fe. The small contributions of $\Delta E_{\uparrow \rightarrow \uparrow}^{\text{Fe}}$ and $\Delta E_{\downarrow \rightarrow \downarrow}^{\text{Fe}}$ to the MCA energy can be attributed to the small LDOS of the unoccupied majority-spin states, which is typical of the properties of alloys including more than half transition metal elements.

To obtain further understanding of the origin of the perpendicular MCA, we show in figure 4 the second-order perturbative contribution to the total energy of nonvanishing angular momentum matrix elements connecting d states, i.e., $\pm|\langle \mu'|L_x|\mu \rangle|^2 G_{\mu\mu'}^{\mu\mu'}(\sigma, \downarrow; i, i)$ and $\pm|\langle \mu'|L_z|\mu \rangle|^2 G_{\mu\mu'}^{\mu\mu'}(\sigma, \downarrow; i, i)$ in equation (3). We found large matrix elements of the spin-conservation term connecting Fe d(xy) and Fe d(x²−y²), which is the main contributing factor for the perpendicular MCA of L1₀-FeNi. Furthermore, a spin-flip term, from the majority-spin Fe d(3z²−r²) to the minority-spin Fe d(yz), also makes a significant positive contribution to the perpendicular MCA. These results can be confirmed in the Fe LDOS of L1₀-FeNi at a/a₀ = 1.00 in figure 3(a), where there are large LDOS of Fe d(x²−y²), d(3z²−r²), and d(yz) in the vicinity of $E_F$. 

Figure 3. Local density of states (LDOS) of the d orbitals of Fe and Ni in L1₀-FeNi for (a) a/a₀ = 1.00 and (b) a/a₀ = 0.904 without spin–orbit interaction as a function of energy relative to the Fermi energy. The equilibrium lattice constant a₀ is 3.556 Å.

Figure 4. Bar graph of the second-order perturbative contribution to the MCA energy of nonvanishing angular momentum matrix elements connecting d states $\pm|\langle \mu'|L_x|\mu \rangle|^2 G_{\mu\mu'}^{\mu\mu'}(\sigma, \downarrow; i, i)$ and $\pm|\langle \mu'|L_z|\mu \rangle|^2 G_{\mu\mu'}^{\mu\mu'}(\sigma, \downarrow; i, i)$ of L1₀-FeNi for an equilibrium lattice constant a = a₀ = 3.556 Å. ‘↓’ and ‘↑’ indicate spin-conservation and spin-flip terms, respectively.
3.3. The effect of tetragonal distortion

We examined the dependence of $K_u$ on the tetragonal distortion of L$_{10}$-FeNi. Here, ‘tetragonal distortion’ indicates a ‘distortion’ with tetragonal lattice deformation compared with the equilibrium lattice parameter of L$_{10}$-FeNi. We optimized a perpendicular lattice parameter $c$ for each in-plane lattice constant $a$. The change in the axial ratio $c/a$ and the total energy are shown in figure 5(a). As shown in figure 5(b), $K_u$ increases with decreasing in-plane lattice constant (with increasing perpendicular lattice parameter $c$), reaching a maximum value of 1.6 MJ m$^{-3}$ at $a = 3.182$ Å ($c/a = 1.242$). The present result suggests that the compressive in-plane stress utilizing the lattice mismatch between L$_{10}$-FeNi and the buffer layers is effective in achieving a higher MCA energy. Ta, Nb, W, and Mo of bcc type having the lattice constant 3.1–3.3 Å can be buffer layer candidates, giving a compressive distortion of over 5% to L$_{10}$-FeNi. Indeed, previous experiments have indicated that $K_u$ increases monotonically with increasing axial ratio $c/a$ of L$_{10}$-FeNi [7]. According to previous first-principles calculations [19], the MCA energy of L$_{10}$-FeNi reaches a maximum value of 0.90 MJ m$^{-3}$ at $c/a = 0.95$. The discrepancy between this result and that obtained in the present study may partly be attributed to the computational method adopted in the previous study, i.e., the linear muffin-tin orbital method within the atomic sphere approximation, which assumes a spherically symmetric potential in each atomic sphere.

To elucidate these results, we show in figure 5(c) the anisotropy of the orbital magnetic moments $\Delta \mu_{\text{orb}}^{\text{Fe}}$ and $\Delta \mu_{\text{orb}}^{\text{Ni}}$ between cases in which the magnetization is along the [001] or [100] direction as a function of in-plane lattice constant. $\Delta \mu_{\text{orb}}^{\text{Fe}}$ decreases with decreasing in-plane lattice constant from $a/a_0 = 1.00$, then slightly increases with increasing $a/a_0$. On the other hand, $\Delta \mu_{\text{orb}}^{\text{Ni}}$ decreases with decreasing in-plane lattice constant from $a/a_0 = 1.00$. These behaviors are inconsistent with the dependence of the MCA energy. To understand this point, we show in figure 6 the spin-resolved MCA energy $\Delta E_{\downarrow \Rightarrow \downarrow}^{\text{Fe}}$ and $\Delta E_{\uparrow \Rightarrow \downarrow}^{\text{Ni}}$ as a function of the in-plane lattice constant. First, the spin-conservation terms $\Delta E_{\downarrow \Rightarrow \downarrow}^{\text{Fe}}$ and $\Delta E_{\uparrow \Rightarrow \downarrow}^{\text{Ni}}$ qualitatively reproduce the in-plane lattice dependence of the difference.
in orbital magnetic moments $\Delta \mu_{\text{Fe}}^{\text{orb}}$ and $\Delta \mu_{\text{Ni}}^{\text{orb}}$ shown in figure 5(c), because the orbital magnetic moment originates only from the spin-conservation term. The abrupt change at around $a = 3.4$ Å of the spin-conservation term and the anisotropy of the Fe orbital moment in figure 5(c) can be attributed to the reduction of minority-spin LDOS of Fe $d(x^2 - y^2)$ in the vicinity of the Fermi level due to the compressive distortion. By comparing the LDOS of Fe between figures 3(a) and (b), we can find that the LDOS of occupied Fe $d(x^2 - y^2)$ and unoccupied Fe $d(z\gamma, z\kappa)$ reduce around the Fermi level in the minority-spin states due to the compressive distortion. These changes of the electronic structures near the Fermi level decrease the negative contribution to the MCA energies from the matrix elements of $L_x$ connecting the minority-spin $d(x^2 - y^2)$ and the minority-spin $d(z\gamma)$, leading to the increase of the MCA energies.

Furthermore, the spin-flip terms $\Delta E_{\text{Fe}}^{\uparrow \Rightarrow \downarrow}$ and $\Delta E_{\text{Ni}}^{\uparrow \Rightarrow \downarrow}$ increase with decreasing in-plane lattice constant. This means that the large perpendicular MCA energy of L1$_0$-FeNi under compressive in-plane distortion can be attributed to an increase in the spin-flip terms of Fe and Ni ($\Delta E_{\text{Fe}}^{\uparrow \Rightarrow \downarrow}$ and $\Delta E_{\text{Ni}}^{\uparrow \Rightarrow \downarrow}$). In figure 6, we show the sum of each of the terms $E_{\text{MCA}}^{(2)}$ given by equation (5) and the MCA energy from the first-principles calculations including the spin–orbit interaction $E_{\text{MCA}}$ (broken line). We can see that the compressive distortion enhances $E_{\text{MCA}}^{(2)}$ to around $a = 3.2$ Å, then it reduces $E_{\text{MCA}}^{(2)}$. This feature qualitatively agrees with that for $E_{\text{MCA}}$. The difference between $E_{\text{MCA}}^{(2)}$ and $E_{\text{MCA}}$ can be attributed to the $c_{\text{FeNi}}$ in equation (4), which can be obtained by the projection of the Bloch wavefunction to the atomic orbital around the given atomic radius. Here, we use the covalent radii of Fe (1.32 Å) and Ni (1.24 Å) [35]. It is very difficult to obtain a quantitative agreement between $E_{\text{MCA}}$ and $E_{\text{MCA}}^{(2)}$, because $E_{\text{MCA}}^{(2)}$ does not include the contribution from the interstitial region.

![Figure 6](image_url)

**Figure 6.** The second-order perturbative contribution to the MCA energy $E_{\text{MCA}}^{(2)}$ for L1$_0$-FeNi. The sum of each term $E_{\text{MCA}}^{(2)}$ (solid line) given by equation (5) and the MCA energy from the first-principles calculations including the spin–orbit interaction $E_{\text{MCA}}$ (broken line) are also shown. The equilibrium lattice constant $a_0$ is 3.556 Å.

![Figure 7](image_url)

**Figure 7.** The 3d orbital contribution to the MCA energies of L1$_0$-FeNi as a function of the in-plane lattice constant $a$ for Fe ((a)–(d)) and Ni ((e)–(g)). Here, we use abbreviated notation for the MCA contribution of the nonvanishing angular momentum matrix elements connecting d states, i.e., $\pm |\langle \mu' \downarrow | L_2 | \mu \sigma \rangle|^2$ ($\uparrow\downarrow$ corresponds to $\sigma = \uparrow$ and $\downarrow\uparrow$ to $\sigma = \downarrow$) for $\pm |\langle \mu' \downarrow | L_2 | \mu \sigma \rangle|^2$ $G_{\mu' \mu}^{\uparrow \downarrow} (\sigma, \downarrow; i, i)$, and $\pm |\langle \mu' \downarrow | L_2 | \mu \sigma \rangle|^2$ ($\uparrow\downarrow$ corresponds to $\sigma = \uparrow$ and $\downarrow\uparrow$ to $\sigma = \downarrow$) for $\pm |\langle \mu' \downarrow | L_2 | \mu \sigma \rangle|^2$ $G_{\mu' \mu}^{\uparrow \downarrow}$ (\sigma, \downarrow; i, i). The equilibrium lattice constant $a_0$ is 3.556 Å.
Figures 7(a)–(h) show the MCA energy of nonvanishing angular momentum matrix elements connecting d states $\pm \langle \mu | L_x | \sigma \rangle$ and $\pm \langle \mu | L_y | \sigma \rangle$ in equation (3) as a function of the in-plane lattice constant. We conclude that the significant reductions of the negative contribution related to the matrix elements of the spin-flip terms are remarkably modulated by compressive in-plane distortion compared with the LDOS of other orbital components. As can be seen in figures 3(a) and (b), the LDOS of the majority-spin $d(x^2 - y^2)$ and $d(xy)$ orbitals resulting from strong bonding between the in-plane d orbitals under the compressive in-plane distortion.

4. Discussion

4.1. The effect of orbital polarization

It is well known that the orbital polarization energy related to the second Hund’s law, which is not considered in the present calculation, enhances the MCA energies of most metals and alloys. According to Ravindran et al [14], the MCA energy of L10-FeNi is enhanced from 0.077 to 0.172 meV/f.u. due to the orbital polarization energy. To clarify the effect of the orbital polarization (OP) on the MCA energy in the tetragonal distortion of L10-FeNi, we performed calculations of the MCA energy including the orbital polarization energy using the FLAPW method with the WIEN2k code [31] using the GGA-PBE function [26]. Figure 8 plots the MCA energy and the anisotropy of the orbital magnetic moment with and without OP as a function of the in-plane lattice constant.

We can confirm that the MCA energy dependence on the in-plane lattice constants obtained by the VASP-PAW code was almost reproduced by the WIEN2k code. The difference might be due to the sensitivity of MCA energies to the calculation parameters, such as the cutoff radius of the PAW potential and muffin-tin radius in the FLAPW method. We found that at the equilibrium lattice constant ($a/a_0 = 1.00$), the MCA energy with OP is roughly two times that without OP, which is consistent with previous calculation results [14]. The origin of the enhancement was well discussed in [14], i.e., the orbital polarization energy effectively enhances the spin–orbit coupling parameter $\xi_i = \xi_i + BL$, where $B$ is a Racah parameter [36] expressed in terms of Slater integrals of the single-particle wavefunctions for the d orbitals. Furthermore, we found that the MCA energy with OP increases with decreasing in-plane lattice constant, which is similar to the results without OP. However, the enhancement of the MCA energy under the compressive in-plane distortion is suppressed compared with the MCA energy without OP. The MCA energies with OP around $a/a_0 = 0.90$ are almost identical to the MCA energies without OP, despite the increase in the anisotropy of the orbital magnetic moment caused by the orbital polarization energy, as shown in figure 8(b). This can be better understood by considering the different origin of the perpendicular MCA of L10-FeNi under tetragonal distortion. As was discussed in the previous section and shown in figure 6, the perpendicular MCA around the equilibrium lattice constant ($a/a_0 = 1.00$) is mainly caused by the spin-conservation term of Fe ($\Delta E_{\text{Fe}}^{\parallel}$), while the spin-flip
terms of Fe ($\Delta E_{\text{Fe}}^{\uparrow\downarrow}$) and Ni ($\Delta E_{\text{Ni}}^{\uparrow\downarrow}$) provide the largest contribution under highly compressive in-plane distortion ($a/a_0 < 0.95$). The orbital polarization energy increases the orbital magnetic moment and its anisotropy, which is equivalent to an enhancement of the spin-conservation term, but it does not enhance the spin-flip term because of the absence of a spin operator. Therefore, the effect of the orbital polarization energy is remarkable in the MCA energy arising from the spin-conservation term compared with that from the spin-flip term.

4.2. Properties of the transport through the MgO barrier

Finally, we discuss the potential of L1$_0$-FeNi as a ferromagnetic electrode for magnetic tunnel junctions with an MgO barrier. Figure 9 shows the electronic band structures along the [001] direction of bulk L1$_0$-FeNi. The totally symmetric band $\Delta_1$ in FeNi crosses the Fermi level in both the majority-spin and minority-spin states. The minority-spin $\Delta_1$ band is predominantly composed of Fe and Ni $d$ orbitals at $E_F$, while the majority-spin band is mainly constructed from Fe and Ni $p(z)$ orbitals. Similar results have been obtained for the band dispersion of L1$_0$-FePt [37]. Since the $\Delta_1$ band electrons predominantly transmit the MgO barrier [38, 39], huge TMR ratios cannot be expected in L1$_0$-FeNi/MgO/L1$_0$-FeNi (001) MTJs from the viewpoint of symmetry compatibility between the electronic band structures in bulk FeNi and MgO. In fact, we obtained a very small tunneling magnetoresistance ratio of 51% for the Fe-terminated L1$_0$-FeNi/MgO/L1$_0$-FeNi (001) MTJ (11% for the Ni-terminated MTJ) using Landauer-type ballistic conductance calculations. For details regarding these conductance calculations, see [37] and the references therein. Thus, to achieve higher TMR ratios, it is necessary to insert appropriate ferromagnetic layers, e.g. Fe or CoFeB, between the L1$_0$-FeNi electrodes and the MgO barrier [40].

5. Summary

We evaluated the MCA energy of L1$_0$-FeNi using first-principles density-functional calculations. The perpendicular MCA found in L1$_0$-FeNi can be attributed predominantly to the constituent Fe atoms. The MCA energy of L1$_0$-FeNi increases with decreasing in-plane lattice constant. The perpendicular MCA under tetragonal distortion was elucidated as follows. The perpendicular MCA at the equilibrium lattice constant is mainly caused by the spin-conservation term in the second-order perturbation of the spin–orbit interaction. On the other hand, the perpendicular MCA under highly compressive in-plane distortion can be attributed to the spin-flip terms of Fe and Ni. The orbital polarization energy increases the MCA energy with a small tetragonal distortion due to enhancement of the spin-conservation term, while the effect of the orbital polarization energy is suppressed under highly compressive in-plane strains. We concluded that the modulation of the in-plane lattice parameter of an L1$_0$-FeNi thin film by choosing appropriate buffer layers is effective in obtaining large MCA energies corresponding to 1.0 MJ m$^{-3}$, and is worth further investigation.

Acknowledgments

The authors are grateful to M Mizuguchi, M Kotsugi, T Kojima, and K Takanashi for sharing their experimental data prior to publication. Thanks are also due to Y Koda, C Mitsumata, and A Sakuma for valuable discussion on MCA in disordered systems. This work was partly supported by Collaborative Research based on Industrial Demand ‘High Performance Magnets: Towards Innovative Development of Next Generation Magnets’ from JST, a Grant-in-Aid for Scientific Research (Grants Nos 22246087, 22360014 and 22760003) from JSPS/MEXT, and the FIRST program from JSPS/CSTP. YM and KA gratefully acknowledge support from Mayekawa Houonkai Foundation.

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