First-principles study of spin-wave dispersion in Sm(Fe$_{1-x}$Co$_x$)$_{12}$

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Abstract

We present spin-wave dispersion in Sm(Fe$_{1-x}$Co$_x$)$_{12}$ calculated based on first-principles. Anisotropy in the lowest branch of the spin-wave dispersion around the $\Gamma$ point is discussed. Spin-waves propagate more easily along $a^*$-axis than along $c^*$-axis, especially in SmFe$_{12}$. We also compare values of the spin-wave stiffness with those obtained from an experiment. The calculated values are in good agreement with the experimental values.

Keywords: hard-magnet compounds, first-principles calculation, ThMn$_{12}$ structure, Sm(Fe,Co)$_{12}$, spin-wave

1. Introduction

Magnetic properties at finite temperatures are important in applications of hard magnets, and ab initio modeling for spins has become one of the standard techniques today. The spin-wave dispersion can be derived from such a model. It offers intuitive description of magnetic collective modes, which is important in understanding finite temperature properties. It is also possible to compare the dispersion directly with that obtained by experiments.

Magnetic compounds with the ThMn$_{12}$ structure have regained attention since their potential as the main phase in a hard magnet was reevaluated by a first-principles study [1] and experimental works [2, 3] in these years. Hirayama et al. have recently synthesized Sm(Fe$_{1-x}$Co$_x$)$_{12}$ films for
$x = 0, 0.1, 0.2$, and shown that Sm(Fe$_{0.8}$Co$_{0.2}$)$_{12}$ has favorable magnetic properties, including the spontaneous magnetization of 1.78 T at room temperature [4].

In this paper, we present spin-wave dispersion in Sm(Fe$_{1-x}$Co$_x$)$_{12}$ calculated based on first-principles for $x = 0$ and 0.2. Because there is no experiment clarifying its spin-wave dispersion to the best of our knowledge, we compare values of the spin-wave stiffness with those obtained from the experiment by Hirayama et al.[4]. We also discuss anisotropy in the lowest branch around the Γ point: spin-waves propagate more easily along $a$-axis than along $c$-axis especially in SmFe$_{12}$.

2. Methods

We use the Korringa-Kohn-Rostoker Green function method for solving the Kohn-Sham equation of density functional theory [5, 6]. The exchange-correlation functional is approximated within the local density approximation [9]. The f-orbitals at the Sm site are treated as a trivalent open core with the spin-configuration limited by Hund’s rule, and the self-interaction correction is applied to the orbitals. The spin-orbit coupling is disregarded in the calculation except that the effect is implicitly taken into account in the spin-configuration of the f-electrons. We assume the Fe and Co atoms randomly occupy the 8f, 8i and 8j site in the ThMn$_{12}$ structure [Space group: $I4/mmm$ (#139); see also Figure 1] and their site preference is disregarded.

In the calculation of spin-wave dispersion, we treat the randomness with the virtual crystal approximation (VCA). However, for the the other part of the calculation, namely, the magnetization and the Curie temperature, we use the coherent potential approximation (CPA), which is more sophisticated than VCA concerning the randomness. To compare the CPA results with that of VCA, we calculate the magnetization and the Curie temperature also within VCA. The reason why we do not use CPA in the calculation of the spin-wave dispersion is addressed later in this section. The experimental lattice constant $a$ and $c$ given in Ref. [4] are used in those calculations. We use the calculated values of the inner parameters for SmFe$_{12}$ given in Ref. [7].

The magnetic coupling is calculated using Liechtenstein’s formula [8]. In formalism, we use those values as $J_{i,j}^{\mu,\nu}$ in the following classical Heisenberg Hamiltonian:

$$H = - \sum_{i,\mu} \sum_{j,\nu} J_{i,j}^{\mu,\nu} \vec{\sigma}_i^\mu \cdot \vec{\sigma}_j^\nu$$

(1)
Figure 1: Crystal structure of the ThMn\(_{12}\) structure. The atomic positions indicated by the Wyckoff positions (2a, 8j, 8i, 8f) are shown. Some bonds are shown to serve as eye-guides.

where \( \vec{e}_i^\mu \) is a unit vector that is in the direction of the magnetic moment at the \( \mu \)th site in the \( i \)th unit cell.

In calculation of spin-waves, we consider small fluctuation of \( \vec{e} \) from the alignment in the ground state, \( \vec{e}_{i}^{GS,\mu} \). We assume that \( \vec{e}_{i}^{GS,\mu} \) is parallel or antiparallel to the \( z \)-direction. Elementary excitations of spin-waves \( \vec{e}_i^\mu = \vec{e}_{i}^{GS,\mu} + \vec{u}_i^\mu(q) \exp(i\omega t - i\vec{q} \cdot \vec{R}_i) \) can be obtained by diagonalizing a matrix \( \mathcal{J}(\vec{q}) \) constructed from \( J_{i,j}^{\mu,\nu} \), where \( u_\mu^x(\vec{q}), u_\mu^y(\vec{q}) \ll 1 \) and \( u_\mu^z(\vec{q}) \simeq 0 \) are assumed for the \( x \)-, \( y \)- and \( z \)-component, respectively. We summarized derivation of \( \mathcal{J}(\vec{q}) \) in Appendix A without considering an equation of motion. We refer readers to Ref. [9] for its relation to the dynamics.

In Liechtenstein’s formula, spin-rotational perturbations at the \( i \) and \( j \) site are considered, and the excitation energy is interpreted as intersite magnetic interaction \( J_{i,j}^{\mu,\nu} \) [8]. The formula consists of the perturbation of the local potentials and the scattering path operator. The scattering path operator can be obtained from the Green’s function of the Kohn-Sham system, which
is usually obtained as a function in the reciprocal space. While the spin-rotational perturbation can be formally transformed into the reciprocal space (because it is Kronecker-delta-like), it is difficult to transform the scattering path operator into the real space without loss of precision. Therefore, it is advantageous to construct $\mathcal{J}(\vec{q})$ in the reciprocal space. Pajda et al has also pointed out that this type of direct calculation is possible [10].

Based on the Fourier transformed Liechtenstein’s formula, we have developed a method for obtaining the $\omega-\vec{q}$ dispersion directly in the reciprocal space that is combined with the KKR method for this study. In the present calculation, $16 \times 16 \times 16$ q-points in the Brillouin zone are considered. As a post-process, we interpolate quantities for arbitrary $\vec{q}$ vectors using the values on the $q$-mesh as follows. We first construct a Fourier series that reproduces all the values on the mesh. This includes, however, terms with high frequencies that cannot be accurately determined with the $q$-mesh. We truncate all these terms (low-pass filtering), and add a constant so that the quantity at the $\Gamma$ point becomes identical to the original one.

The reason why we use VCA instead of CPA in the calculation of spin-wave dispersion is the following. Liechtenstein’s formula (with CPA) gives $J$ that also depends on the two elements at the ends. We can use those $J$’s to construct the Heisenberg model with a random configuration of elements with the justification described in [11]. Then, we have to consider the sample average for the spin-wave dispersion. It needs too large a supercell to take the average directly when $J$ is long-ranged. Although application of CPA to the Heisenberg model was proposed in [11] to overcome this difficulty, the expected resource consumption is still too high for the systems we considered. It is because the calculation needs a fine mesh in the Brillouin zone in order to obtain spin-wave dispersion around the $\Gamma$ point, which is of particular interest here. On this ground, we abandoned using $J$ that depends on the atomic species. In order to obtain such averaged $J$, VCA would be enough.

3. Results and discussion

We first present our results for magnetization within VCA and CPA, and compare them with the experimental values in Ref. [4]. Figure 2 shows the values of the magnetization as functions of $x$ (the Co concentration). The contribution from f-electrons at Sm sites are included in those values assuming that the f-electrons have magnetic moment of $g_J\sqrt{J(J+1)} = 0.85\mu_B$, which adds approximately 0.05 T to the magnetization.
Figure 2: Values of magnetization calculated with VCA and CPA, and those from an experiment (extrapolated to 0 K) [4] as functions of concentration $x$.

The theoretical prediction reasonably agrees with the experimental values with the underestimation of 0.15 T at worst. In a previous first-principles study using a PAW-GGA method (which consider the spin-orbit coupling only in the spin-configuration at the Sm sites as in our calculation), magnetization of SmFe$_{12}$ was evaluated as 1.83 T [12], which is not much different from the present value. The deviation from the experiment is partly attributed to orbital moments. When we considered the spin-orbit coupling at the Fe sites, the gain was 0.03 T in SmFe$_{12}$.

The experimental observation that the Co introduction reduces the magnetization at low temperature is theoretically understandable. It has already been discussed that the optimal percentage of substitution of Co can significantly depend on how the host system has a room for improvement in the magnetic moment [13]. In the case of SmFe$_{12}$, the Co substitution can slightly enhance the ferromagnetism due to hybridization between Fe and Co, however, this does not overcome the expansion of the volume. This reduction is well reproduced by the CPA calculation.

As for validity of VCA, it is adequate in the sense that the deviations of the values from CPA are within a few percent. We should note, however, that the tendency as a function of $x$ is opposite. In VCA, the enhancement of the ferromagnetism with increasing $x$ is exaggerated by totally forgetting the inhomogeneity caused by the randomness.
Figure 3: Calculated values of Curie temperature within VCA and CPA as functions of concentration $x$. In both cases, the mean-field approximation is used.

Figure 3 shows values of the calculated Curie temperatures as functions of $x$ within VCA and CPA. In both cases, we use the mean-field approximation (MFA) as described in Ref. [14]. Those Curie temperatures ($T_C$) are overestimated due to the use of MFA, however, it reproduces relative change of $T_C$ with respect to $x$ in experiments (see also our previous study [14]). Figure 4 compares the numerical results with the experimental Curie temperature [4]. Results of linear regression are also shown in the figure. The relative change of $T_C$ is reproduced well in CPA, which can be seen from the value of the gradient (1.09) being close to 1. In VCA, the tendency toward ferromagnetism is again excessive, however, the values of the Curie temperature is significantly correlated with the experimental values. We therefore expect that VCA also offers informative description of Sm(Fe$_{1-x}$Co$_x$)$_{12}$.

We then present our results for the spin-wave dispersion. Figure 5 shows that in SmFe$_{12}$ (solid lines) and Sm(Fe$_{0.8}$Co$_{0.2}$)$_{12}$ (broken lines). The broken lines are aligned upward compared to the solid lines. The introduction of Co in Sm(Fe$_{0.8}$Co$_{0.2}$)$_{12}$ enhances the magnetic exchange interaction and makes the excitation energy for the collective modes higher than in SmFe$_{12}$.

In SmFe$_{12}$, the curvature of the lowest branch around the Γ point along $c^*$-axis ($D_c$) is larger than that along $a^*$-axis ($D_a$). The smallness of $D_a$ indicates that spin-waves propagates more easily along $x$-axis than $z$-axis. The value of $\omega$ at Z [= $(1 0 0)$] is conspicuously small, which characterizes the
Figure 4: Experimental values (horizontal) of Curie temperature versus theoretical values (vertical) within VCA and CPA. In both cases, the mean-field approximation is used. Results of linear regression are also shown in the figure.

smallness of $D_a$. This anisotropy originates from the tetragonal asymmetry of the system, which has two effects: (1) it elongates the Wigner-Seitz cell in the reciprocal space along $c^*$-axis in the case of $c < a$, and (2) allows the distribution of $J_{i,j}^{\mu,\nu}$ to be anisotropic. However, the elongation of the Wigner-Seitz cell makes $D_a/D_c$ larger when the values of $J_{i,j}^{\mu,\nu}$ are kept unchanged (see also Appendix C). Therefore, the smallness of $D_a$ must be attributed to the anisotropic distribution of $J_{i,j}^{\mu,\nu}$, which would be closely related to the structure of the network of the transition atoms. As for Sm(Fe$_{0.8}$Co$_{0.2}$)$_{12}$, the curvatures become more isotropic due to the enhancement of the exchange interaction.

The calculated value of the curvatures from the dispersion are shown in Table 1. Those are determined by fitting of a quadratic function to the data points within a $0.2 \times \frac{2\pi}{a}$ radius of the $\Gamma$ point. The spin-wave stiffness $D$ corresponding to the coefficient of $T^{3/2}$ in the Bloch’s theory of the isotropic case is also determined in the following way. Consider transformation $(q_a, q_b, q_c) \rightarrow (\sqrt{D/D_a} q'_a, \sqrt{D/D_a} q'_b, \sqrt{D/D_c} q'_c)$, where $q$’s are the components of $\vec{q} \equiv (q_a, q_b, q_c)^T$. This transforms the approximate harmonic dispersion to an isotropic one: $\hbar \omega \approx D_a q'_a^2 + D_a q'_b^2 + D_c q'_c^2 \rightarrow D (q_{a'}^2 + q_{b'}^2 + q_{c'}^2)$. One should then pay attention to the change of the state density in the $q'$ space. When the Jacobian $\sqrt{D^3/(D_a^2 D_c)}$ is one the state density is kept un-
Figure 5: The spin-wave dispersion ($\hbar \omega$ as a function of $\vec{q}$) in SmFe$_{12}$ (green) and Sm(Fe$_{0.8}$Co$_{0.2}$)$_{12}$ (purple).

Table 1: Calculated values of the spin-wave stiffness for Sm(Fe$_{1-x}$Co$_x$)$_{12}$ compared with the experimental values by Hirayama et al [4]. The curvatures around the $\Gamma$ point in the lowest branch and along the $a^*$- and $c^*$-axis are also shown as $D_a$ and $D_c$. The stiffness values are in the unit of $\text{meV} \, \AA^2$.

| $x$ | $D$ (Exp.) | $D_a$ | $D_c$ |
|-----|------------|-------|-------|
| 0   | 118 (179)  | 91.6  | 194   |
| 0.1 | 252 (251)  | 220   | 332   |
| 0.2 | 324 (351)  | 290   | 407   |

changed. Therefore, $D = (D_a^2 D_c)^{1/3}$ gives the corresponding spin-stiffness.

The calculated values in Table 1 are in good agreement with the value obtained by Hirayama et al [4] for all the $x$’s. The theoretical $D$ tends to be smaller than the experimental values, which is conspicuous especially at $x = 0$. For a finite $x$, this underestimation is partly and accidentally canceled by the excessive tendency toward ferromagnetism in VCA.

The values of $D_a$ and $D_c$ can be converted to the exchange stiffness, $A$. We here consider the following macroscopic Hamiltonian for magnetization $\vec{M}$ and the magnetic coupling:

$$E = -\int_{\Omega} dx \, dy \, dz \, m^T \left( \vec{\nabla}^T \mathbf{A} \vec{\nabla} \right) m,$$  \hspace{1cm} (2)
Table 2: Calculated values of the exchange stiffness for Sm(Fe$_{1-x}$Co$_x$)$_{12}$. The stiffness values are in the unit of pJ/m (= 10$^{-12}$J/m).

| x | A  | $A_x$ | $A_z$ |
|---|----|------|------|
| 0 | 7.21 | 5.61 | 11.9 |
| 0.1 | 15.6 | 13.6 | 20.5 |
| 0.2 | 20.3 | 18.1 | 25.4 |

where $\vec{m} \equiv \vec{M}/|\vec{M}|$, $\vec{V} = (\partial_x, \partial_y, \partial_z)^T$, $\Omega$ denote the domain, and $\mathcal{A}$ is a 3 x 3 constant tensor that is real, symmetric and positive definite. We also assume that the absolute value of the magnetization, $|\vec{M}|$, is constant through the space. Our Hamiltonian is identical to that used by Belashchenko [15] except for the surface term with a partial integration.

Let us consider $\mathcal{A} = \text{diag}[A_x, A_x, A_z]$ for the Sm(Fe$_{1-x}$Co$_x$)$_{12}$ systems. Following discussion in Ref. [16], one can derive the approximate relation, $\hbar \omega \approx (A_x q_a^2 + A_x q_b^2 + A_z q_c^2) 4/\rho_M$, where $\rho_M$ is the number of Bohr magnetons per unit volume. By comparing it with $\hbar \omega \approx (D_a q_a^2 + D_b q_b^2 + D_c q_c^2)$, one can see that

$$A_x = \rho_M D_a/4; \quad A_z = \rho_M D_c/4.$$  \hspace{1cm} (3)

In the same manner we derived $D = (D_a D_c)^{1/3}$, exchange-stiffness $A$ can be related to $A_x$ and $A_z$ as follows:

$$A = (A_x^2 A_z)^{1/3}.$$  \hspace{1cm} (4)

For a general $\mathcal{A}$ this relation becomes $A = (\lambda_1 \lambda_2 \lambda_3)^{1/3}$, where $\lambda$’s are the eigenvalues of $\mathcal{A}$.

The calculated values of $A$, $A_x$ and $A_z$ for Sm(Fe$_{1-x}$Co$_x$)$_{12}$ are shown in Table 2. The anisotropy in the spin-wave stiffness directly leads to anisotropy in the exchange stiffness because the relation $D_a/D_c = A_x/A_z$ holds due to eq. (3). Toga et al. [17] has recently reported the values of the exchange stiffness at finite temperatures in their Monte Carlo simulation for Nd$_2$Fe$_{14}$B, which is also an Fe-rich rare-earth compound. Though they did not present the value at zero temperature, the order of the exchange stiffness seem to agree with our results for SmFe$_{12}$.
4. Conclusion

We presented calculation of the spin-wave dispersion in SmFe\textsubscript{12} and Sm(Fe\textsubscript{0.8}Co\textsubscript{0.2})\textsubscript{12}, and discussed how the introduction of Co enhances the exchange interaction. Our calculation predicted anisotropy in the curvatures of the lowest branch around the Γ point in SmFe\textsubscript{12} and weakening of the anisotropy when Co is introduced into the system. We also calculated the spin-wave stiffness of the systems, which values are in good agreement with the experimental values by Hirayama et al [4].

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Appendix A. Heisenberg Hamiltonian for spin-waves

In this section, we consider small fluctuation of the spins from the ground state in the Hamiltonian of eq. (1). We assume the ground state to be the form of $\vec{e}^{\text{GS},\mu}_{i} = t (0, 0, \sigma_{i})$ with $\sigma_{i} = \pm 1$, which means the spins are parallel or antiparallel along the z-axis and the unit cell is large enough to describe the ground state. We assume the $x$-component of the fluctuation, $\delta x^{\mu}_{i}$, is much smaller than 1, and so is the $y$-component, $\delta y^{\mu}_{i}$. With this fluctuation, the $\vec{e}$ can be written as follows:

$$
\vec{e}_{i}^{\mu} \simeq \left( \begin{array} {c}
\delta x^{\mu}_{i} \\
\delta y^{\mu}_{i} \\
\sigma_{i} \left[ 1 - \frac{1}{2} \left\{ (\delta x_{i}^{\mu})^2 + (\delta y_{i}^{\mu})^2 \right\} \right] \end{array} \right). \quad (A.1)
$$
We here retain the second order terms of $\delta x$ and $\delta y$ because the excitation energy is the second order of the fluctuations. It can easily be seen that this vector has an absolute value of 1 within the second order approximation. Then, the Hamiltonian of eq. (1) becomes

$$H - E_{GS} = - \sum_{\alpha \in \{x,y\}} \sum_{\mu, \nu} \sum_{i, j} \delta \alpha^\mu_i \left( J^\mu,\nu_{i,j} - \delta_{\mu,\nu} \sum_{k, \ell} J^\mu,\ell_{i,k} \right) \delta \alpha^\nu_j,$$

where $E_{GS}$ is the energy of the ground state, and $\delta \alpha$ is either $\delta x$ or $\delta y$ indexed by $\alpha$ in the first summation symbol. This expression can be rewritten with the Fourier transform of $\delta \alpha$ and $J$:

$$\delta \alpha^\mu_i = \sum_\mathbf{q} \tilde{\delta} \alpha^\mu_i (\mathbf{q}) e^{-i\mathbf{q} \cdot \mathbf{R}_i},$$

and

$$J^\mu,\nu_{i,j} = \sum_\mathbf{q} \tilde{J}^\mu,\nu_i (\mathbf{q}) e^{-i\mathbf{q} \cdot (\mathbf{R}_i - \mathbf{R}_j)}$$

into the following form:

$$H - E_{GS} = - \sum_{\alpha \in \{x,y\}} \sum_{\mu, \nu} \sum_\mathbf{q} \tilde{\delta} \alpha^\mu_i (\mathbf{q}) \left( \tilde{J}^\mu,\nu (\mathbf{q}) - \delta_{\mu,\nu} \sum_\ell \tilde{J}^\mu,\ell (\mathbf{0}) \right) \tilde{\delta} \alpha^\nu_j (\mathbf{q})$$

$$\equiv - \sum_{\alpha \in \{x,y\}} \sum_{\mu, \nu} \sum_\mathbf{q} \tilde{\delta} \alpha^\mu_i (\mathbf{q}) \tilde{J}^\mu,\nu (\mathbf{q}) \tilde{\delta} \alpha^\nu_j (\mathbf{q})$$

because $\delta \alpha^\mu_i$ is real. Therefore, we can obtain elementary excitations in the Hamiltonian by diagonalizing the matrix of $\tilde{J}^\mu,\nu (\mathbf{q}) = \tilde{J}^\mu,\nu (\mathbf{q}) - \delta_{\mu,\nu} \sum_\ell \tilde{J}^\mu,\ell (\mathbf{0})$.

**Appendix B. Spin-wave stiffness in bcc Fe, hcp Co, fcc Co and fcc Ni**

In this section, we show values of the spin-wave stiffness calculated for bcc Fe, hcp Co, fcc Co and fcc Ni.

We used calculated values of the lattice constants of $a = 2.837$ Å for bcc Fe, $a = 2.494$ Å, $c = 4.042$ Å for hcp Co, $a = 3.530$ Å for fcc Co, and $a = 3.525$ Å for fcc Ni, which are obtained by using a PAW-GGA method. The spin-wave stiffness are determined by fitting of a quadratic function to the data points within a $0.2 \times \frac{2\pi}{a}$ radius (bcc, hcp) or a $0.3 \times \frac{2\pi}{a}$ radius (fcc) of the $\Gamma$ point.

Table B.3 compares our values with previous theoretical values [10] and experimental values. Our results are in good agreement with the experimental values except for Ni, for which the Heisenberg model would not be very appropriate. They are also quite similar to the previous theoretical values, however, we could not resolve the large deviation in fcc Co.
Table B.3: Calculated values of the spin-wave stiffness for bcc Fe, hcp Co, fcc Co and fcc Ni compared with theoretical values from [10] and experimental values [19, 20, 21]. The values are in the unit of meV Å².

|          | This work | Theory [10] | Exp.  |
|---------|-----------|-------------|-------|
| bcc Fe  | 259       | 250 ± 7     | a278 ± 30, b280 |
| hcp Co  | 580       | —           | b580  |
| fcc Co  | 429       | 663 ± 6     | c340  |
| fcc Ni  | 748       | 756 ± 29    | d555  |

a Ref. [19]; b Ref. [20]; c Ref. [22]; d By thin-film resonance at 295 K [21]

Appendix C. The $D_a/D_c$ ratio in bcc Fe, fcc Co and fcc Ni with tetragonal deformation

To provide more data of the anisotropy of the spin-wave stiffness in tetragonal systems we calculate the $D_a/D_c$ ratio in bcc Fe, fcc Co and fcc Ni with applying tetragonal deformation. The $c/a$ ratio of the systems is changed from -2% to +2% with their volume kept constant (see Appendix B for the systems without the deformation).

The $D_a/D_c$ ratio can be approximated by $(c/a)^{-2}$ when the change of $J_{i,j}^{\mu,\nu}$ caused by the deformation can be disregarded. Consider a linear regular transform $T$ for the lattice vectors, $\vec{R} \rightarrow \vec{R}' = T\vec{R}$. The tetragonal deformation we are considering here can be expressed by $T = \text{diag} \left[ \sqrt[3]{a/c}, \sqrt[3]{a/c}, \sqrt{(c/a)^2} \right]$. This transforms a vector $\vec{q}$ in the reciprocal space to $\vec{q}' = T^{-1} \vec{q}$, which keeps $\vec{q} \cdot \vec{R}$ invariant. With the fixed values of $\tilde{J}_{i,j}^{\mu,\nu}$ we can obtain $\tilde{J}^{\mu,\nu}(\vec{q}') = \tilde{J}^{\mu,\nu}(\vec{q})$ by the inverse Fourier transform. This leads to the approximation

$$\tilde{J}^{i,\mu,\nu}(\vec{q}') \approx \tilde{J}^{i,\mu,\nu}(\vec{q})$$

(C.1)

Note that the Fourier transform with respect to the lattice after the operation of $T$ (for which we put a prime) must be distinguished from that with respect to the original lattice.

Therefore, the approximate relation in the cubic system $\hbar\omega \approx D(q_a^2 + q_b^2 + q_c^2)$ is transformed into the form of $\hbar\omega \approx D'_a q_a'^2 + D'_b q_b'^2 + D'_c q_c'^2$ with $D'_a = D'_b = (c/a)^{-2/3}D$ and $D'_c = (c/a)^{4/3}D$. Finally, we obtain the approximate relation $D'_a/D'_c = (c/a)^{-2}$. 12
Figure C.6: The $D_a/D_c$ ratio for bcc Fe, fcc Co, and fcc Ni with tetragonal distortion as functions of the $c/a$ ratio. The volumes are fixed to those of the cubic systems ($c/a = 1$) in the calculations. The dotted line shows the function of $(c/a)^{-2}$.

Figure C.6 shows the $D_a/D_c$ ratio for bcc Fe, fcc Co, and fcc Ni with the tetragonal deformation. The $D_a/D_c$ ratio in Fe and Ni tend to have a negative correlation with $c/a$, which is opposite to the cases of SmFe$_{12}$ (this work) and Nd$_2$Fe$_{14}$B [17]. In contrast, fcc Co has a positive correlation to $c/a$. These can be attributed to a difference in dependency of $J_{i,j}^{\mu,\nu}$ on $c/a$, which leads to the deviation from the $(c/a)^{-2}$ function.

[1] Takashi Miyake, Kiyoyuki Terakura, Yosuke Harashima, Hiori Kino, and Shoji Ishibashi. First-principles study of magnetocrystalline anisotropy and magnetization in NdFe$_{12}$, NdFe$_{11}$Ti, and NdFe$_{11}$TiN. *Journal of the Physical Society of Japan*, 83(4):043702, 2014.

[2] Y Hirayama, Y K Takahashi, S Hirosawa, and K Hono. NdFe$_{12}$N$_x$ hard-magnetic compound with high magnetization and anisotropy field. *Scripta Materialia*, 95:70–72, 2015.

[3] Yusuke Hirayama, Takashi Miyake, and Kazuhiro Hono. Rare-earth lean hard magnet compound NdFe$_{12}$N. *JOM*, 67(6):1344–1349, 2015.

[4] Y Hirayama, Y K Takahashi, S Hirosawa, and K Hono. Intrinsic hard magnetic properties of Sm(Fe$_{1-x}$Co$_x$)$_{12}$ compound with the ThMn$_{12}$ structure. *Scripta Materialia*, 138:62–65, 2017.
[5] Pierre Hohenberg and Walter Kohn. Inhomogeneous electron gas. *Physical Review*, 136(3B):B864, 1964.

[6] Walter Kohn and Lu Jeu Sham. Self-consistent equations including exchange and correlation effects. *Physical Review*, 140(4A):A1133, 1965.

[7] Taro Fukazawa, Hisazumi Akai, Yosuke Harashima, and Takashi Miyake. First-principles study of intersite magnetic couplings in NdFe$_{12}$ and NdFe$_{12}$X (X= B, C, N, O, F). *Journal of Applied Physics*, 122(5):053901, 2017.

[8] A I Liechtenstein, M I Katsnelson, V P Antropov, and V A Gubanov. Local spin density functional approach to the theory of exchange interactions in ferromagnetic metals and alloys. *Journal of Magnetism and Magnetic Materials*, 67(1):65–74, 1987.

[9] S V Halilov, H Eschrig, A Y Perlov, and P M Oppeneer. Adiabatic spin dynamics from spin-density-functional theory: Application to Fe, Co, and Ni. *Physical Review B*, 58(1):293, 1998.

[10] Marek Pajda, J Kudrnovský, I Turek, Vaclav Drchal, and Patrick Bruno. Ab initio calculations of exchange interactions, spin-wave stiffness constants, and Curie temperatures of Fe, Co, and Ni. *Physical Review B*, 64(17):174402, 2001.

[11] Paweł Buczek, Leonid M Sandratskii, Nadine Buczek, Stefan Thomas, Giovanni Vignale, and Arthur Ernst. Magnons in disordered nonstoichiometric low-dimensional magnets. *Physical Review B*, 94(5):054407, 2016.

[12] Yosuke Harashima, Kiyoyuki Terakura, Hiori Kino, Shoji Ishibashi, and Takashi Miyake. First-principles study of structural and magnetic properties of R(Fe,Ti)$_{12}$ and R(Fe,Ti)$_{12}$N (R= Nd, Sm, Y). In *Proceedings of Computational Science Workshop 2014 (CSW2014)*, volume 5 of *JPS Conference Proceedings*, page 1021, 2015.

[13] Yosuke Harashima, Kiyoyuki Terakura, Hiori Kino, Shoji Ishibashi, and Takashi Miyake. First-principles study on stability and magnetism of NdFe$_{11}M$ and NdFe$_{11}MN$ for M= Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn. *Journal of Applied Physics*, 120(20):203904, 2016.
[14] Taro Fukazawa, Hisazumi Akai, Yosuke Harashima, and Takashi Miyake. 
First-principles study of intersite magnetic couplings and Curie temperature in 
RFe$_{12-x}$Cr$_x$ (R = Y, Nd, Sm). *Journal of Physical Society of Japan*, 87(4):044706, 2018.

[15] K D Belashchenko. Anisotropy of exchange stiffness and its effect on the 
properties of magnets. *Journal of magnetism and magnetic materials*, 
270(3):413–424, 2004.

[16] Conyers Herring and Charles Kittel. On the theory of spin waves in 
ferromagnetic media. *Physical Review*, 81(5):869, 1951.

[17] Yuta Toga, Masamichi Nishino, Seiji Miyashita, Takashi Miyake, and 
Akimasa Sakuma. Anisotropy of exchange stiffness based on atomic-
scale magnetic properties in rare-earth permanent magnet Nd$_2$Fe$_{14}$B. 
*arXiv preprint arXiv:1804.05824*, 2018.

[18] QMAS—Quantum MAterials Simulator Official Site. http://qmas.jp.

[19] R Hasegawa and Ranjan Ray. Low-temperature magnetization study of 
crystalline and glassy Fe-B alloys. *Physical Review B*, 20(1):211, 1979.

[20] R Pauthenet. Experimental verification of spin-wave theory in high 
fields. *Journal of Applied Physics*, 53(11):8187–8192, 1982.

[21] G Shirane, VJ Minkiewicz, and R Nathans. Spin waves in 3d metals. 
*Journal of Applied Physics*, 39(2):383–390, 1968.

[22] H. A. Mook, J. W. Lynn, and R. M. Nicklow. Temperature dependence 
of the magnetic excitations in nickel. *Phys. Rev. Lett.*, 30:556–559, Mar 
1973.