The Forced Magnetostrictions and Magnetic Properties of Ni$_2$MnX (X = In, Sn) Ferromagnetic Heusler Alloys

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Abstract: Experimental studies into the forced magnetostriction, magnetization, and temperature dependence of permeability in Ni$_2$MnIn and Ni$_2$MnSn ferromagnetic Heusler alloys were performed according to the spin fluctuation theory of itinerant ferromagnetism proposed by Takahashi. We investigated the magnetic field ($H$) dependence of magnetization ($M$) at the Curie temperature ($T_C$), and at $T = 4.2$ K, which concerns the ground state of the ferromagnetic state. The $M$-$H$ result at $T_C$ was analyzed by means of the $H$ versus $M^5$ dependence. At $4.2$ K, it was investigated by means of an Arrott plot ($H/M$ vs. $M^2$) according to Takahashi’s theory. As for Ni$_2$MnIn and Ni$_2$MnSn, the spin fluctuation parameters in $k$-space (momentum space, $T_A$) and that in energy space (frequency space, $T_0$) obtained at $T_C$ and $4.2$ K were almost the same. The average values obtained at $T_C$ and $4.2$ K were $T_A = 342$ K, $T_0 = 276$ K for Ni$_2$MnIn and $T_A = 447$ K, $T_0 = 279$ K for Ni$_2$MnSn, respectively. The forced magnetostriction at $T_C$ was also investigated. The forced linear magnetostriction ($\Delta L/L$) and the forced volume magnetostriction ($\Delta V/V$) were proportional to $M^4$, which followed Takahashi’s theory. We compared the forced volume magnetostriction $\Delta V/V$ and mechanical parameter, bulk modulus $K$. $\Delta V/V$ is inversely proportional to $K$. We also discuss the spin polarization of Ni$_2$MnIn and other magnetic Heusler alloys. The $p_C/p_S$ of Ni$_2$MnIn was 0.860. This is comparable with that of Co$_2$MnGa, which is a famous half-metallic alloy.

Keywords: ferromagnetic Heusler alloy; magnetostriction; magnetization; itinerant ferromagnetism; spin polarization

1. Introduction

Spin fluctuation theories have been proposed to explain the physical properties and the principles of itinerant electron systems [1–7]. Recently, the spin fluctuation theory of itinerant magnetism, known as Takahashi’s theory, was proposed by Takahashi [1–4]. The self-consistent renormalization (SCR) theory was first proposed by Moriya and Kawabata, taking into account the non-linear mode–mode coupling between spin fluctuation modes [5–7]. Concerned about the magnetic field dependence of
magnetization ($M$–$H$), the effect of non-linear mode–mode couplings is associated with the second lowest expansion of free energy in regard to magnetization $M$. In this theory, the spin fluctuations of the higher order coefficient are neglected. Takahashi’s theory is the SCR theory according to zero-point spin fluctuations, considering the transverse and longitudinal components of the fluctuations. In this theory, the spin fluctuations of the higher order coefficient are considered, and the relationship between the magnetic fields $H$ and magnetization $M$ at $T_C$ is obtained theoretically by Equation (1):

$$\left( \frac{M}{M_S} \right)^4 = 1.20 \times 10^6 \times \left( \frac{T_C^2}{w_A T_A^3 p_S^4} \right) \times \left( \frac{H}{M} \right).$$

(1)

where $M_S$ is spontaneous magnetization in the ground state, $p_S$ is the magnetic moment in the ground state ($T = 0$ K), $T_A$ is the spin fluctuation parameter in $k$-space (momentum space) in units of Kelvin, $w_A$ is the molecular weight in units of g, and $H$ is the magnetic field in units of kOe. Takahashi transcribed the spin fluctuation parameter in $k$-space at temperature $T_A$ (K) [2]. The dynamical spin susceptibility, as shown in Equation (3.1) in reference [2], is demonstrated by the double-Lorentzian function of the $k$-space (parameter: $q$) and the energy space (frequency $\omega$-space). The Lorentzian function of the $k$-space is proportional to $\chi(q = 0, \omega = 0)$. The half-width of this function, $\Delta q$, which indicates a spin fluctuation in $k$-space, is proportional to the inverse of $\chi(q = 0, \omega = 0)$. The unit of $1/\chi(q = 0, \omega = 0)$ is a dimension of the energy. Finally, $\Delta q$ is shown in a dimension of the energy. Therefore, $\Delta q$ is proportional to $k_B T_A$, where $k_B$ is the Boltzmann function and $T_A$ is the spin fluctuation parameter, as mentioned above. $T_A$ is expressed in the form of $T_A = \overline{A} q_B^2$, where $q_B$ indicates the effective zone boundary wave vector, and $\overline{A}$ indicates the non-dimensional parameter, as shown in Equation (3.6) in reference [2]. Another parameter, $T_0$, is a spectral distribution $\Gamma_{QB}$ in the frequency space, which was defined by $\Gamma_{QB} = 2 \pi k_B T_0$. In this way, the spin fluctuation parameters in $k$-space (momentum space), $T_A$, and that in energy space (frequency space), $T_0$, were defined. From the spontaneous magnetic moment $M_S$ and magnetization at $T_C$, we obtained $T_A$. Investigations into the itinerant magnetism of 3$d$ and 5$f$ electron systems were carried out by means of Equation (1) [1,8–13]. Moreover, this theory has been applied to the ferromagnetic Heusler alloys [11,14–17]. The spin fluctuation parameter in energy space $T_0$ is derived from Equation (3.16) in reference [1]:

$$p_S^2 = \frac{20 T_0}{T_A} \times C_{4/3} \times \left( \frac{T_C}{T_0} \right)^{4/3}, C_{4/3} = 1.006089 \ldots$$

(2)

From Equations (1) and (2), $T_A$ and $T_0$ are obtained.

The other method to derive the parameters $T_A$ and $T_0$ is determination from magnetic field dependence of the magnetization in the ground state ($T << T_C$) [1,13,15].

The magnetization in the ground state is expressed by the following equation:

$$H = \frac{F_1}{N_0^3 (2\mu_B)^4} \times (-M_0^2 + M^2) M,$$

(3)

where $g$ indicates the Landé $g$-factor, $N_0$ indicates Avogadro’s number, and $F_1$ indicates the mode–mode coupling term of the spin fluctuations written as:

$$F_1 = \frac{2 T_A^2}{15 c T_0}.$$

(4)

In Equation (4), $c$ is equal to 1/2 and $M_0$ is the spontaneous magnetization. Further, $F_1$ is derived from the slope of the Arrott plot ($H/M$ versus $M^2$ plot) at low temperatures by Equation (5):

$$F_1 = \frac{N_0^3 (2\mu_B)^4}{k_B \zeta}.$$

(5)
where \( k_B \) indicates the Boltzmann factor, and \( \zeta \) indicates the slope of the Arrott plot (\( M^2 \) versus \( H/M \)). Then, \( T_0 \) and \( T_A \) are provided by the following equations, respectively:

\[
\left( \frac{T_C}{T_0} \right)^{5/6} = \frac{P_S^2}{5g^2C_{4/3}} \times \left( \frac{15cF_1}{2T_C} \right)^{1/2},
\]

\[
\left( \frac{T_C}{T_A} \right)^{5/6} = \frac{P_S^2}{5g^2C_{4/3}} \times \left( \frac{2T_C}{15cF_1} \right)^{1/2}.
\]

These equations use units of kOe and emu/g for the magnetic fields \( H \) and magnetization \( M \), respectively (p. 66 in reference [1]). The value of the magnetic fields \( H \) in 10 kOe is equal to the value in T (Tesla), and the value of magnetization \( M \) in emu/g is equivalent to the value in Am\(^2\)/kg.

As for the itinerant ferromagnets, the relation between the effective magnetic moment \( p_{\text{eff}} \) and the spontaneous magnetic moment \( p_S \) can be expressed by a generalized Rhodes–Wohlfarth equation (Equation (3.47) in reference [1]):

\[
p_{\text{eff}}/p_S = 1.4 \times \left( \frac{T_O}{T_C} \right)^{2/3}.
\]

Equation (8) can be rewritten as

\[
k_m = \left( \frac{p_{\text{eff}}}{p_S} \right) \times \left( \frac{T_C}{T_0} \right)^3.
\]

Therefore, if \( k_m = 1.4 \), Equation (9) is equal to Equation (8).

The other characteristic property of Takahashi’s theory is that the forced volume magnetostriction \( \Delta V/V \) and the magnetization \( M \) at \( T_C \) can be described as in reference [1]:

\[
(\Delta V/V) \propto M^4,
\]

where \( \Delta V/V \) can be derived by the following equation:

\[
(\Delta V/V) = (\Delta L/L)_{//} + 2 \times (\Delta L/L)_{\perp},
\]

where \( (\Delta L/L)_{//} \) and \( (\Delta L/L)_{\perp} \) are the forced linear magnetostriction parallel and perpendicular to an external magnetic field, respectively [18,19].

In this study, we selected Ni\(_2\)MnIn and Ni\(_2\)MnSn alloys. These alloys are ferromagnetic Heusler alloys and do not cause martensitic transformation [20], in contrast to Ni\(_2\)MnGa with a martensitic transformation temperature \( T_M \) of 195 K [21]. These alloys have \( L2_1 \)-type cubic crystal structure. We considered the magnetostriction and magneto-volume effects of these alloys. We measured the forced longitudinal magnetostriction \( (\Delta L/L)_{//} \) and \( (\Delta L/L)_{\perp} \), derived the forced volume magnetostriction \( \Delta V/V \) as shown by Equation (4), and evaluated the correlation between the magnetization and \( \Delta V/V \).

2. Materials and Methods

Polycrystalline Ni\(_2\)MnIn and Ni\(_2\)MnSn alloys were synthesized from the constituent elements of Ni\(_2\)MnIn: Ni (4N), Mn (3N), In (4N); Ni\(_2\)MnSn: Ni (4N), Mn (4N), Sn(5N). The sample of Ni\(_2\)MnIn was prepared by induction melting under an Ar atmosphere. The sample of Ni\(_2\)MnSn was prepared by arc-melting in an Ar atmosphere. The product of Ni\(_2\)MnSn was heated in vacuum at 1123 K for 3 days and then quenched in water. The results of the X-ray diffraction pattern (XRD, Ultima IV, Rigaku Co., Ltd., Akishima, Tokyo, Japan) indicated that these samples were single phase, as shown in Figure 1. The XRD results indicated that the crystal structure is \( L2_1 \) cubic, and lattice parameters \( a \) were 0.60709 nm and 0.60528 nm for Ni\(_2\)MnIn and Ni\(_2\)MnSn, respectively. A helium-free superconducting magnet at the High Field Laboratory for Superconducting Materials, Institute for Materials Research,
Tohoku University, and at the Center for Advanced High Magnetic Field Science, Osaka University was used for the magnetostriction measurements up to 5 T. The magnetization measurement at 4.2 K, which corresponds to the investigation of the magnetic field dependence of the magnetization at the ground state \((T \ll T_C)\) was performed by means of 30 T pulsed field magnet at the Center for Advanced High Magnetic Field Science, Osaka University. A detailed explanation of the experimental procedure has been given in previous studies [14–17].

![X-ray diffraction patterns of Ni2MnIn and Ni2MnSn. Parenthesis indicates the mirror indices.](image)

### 3. Results and Discussion

#### 3.1. Magnetic Field Dependence of Magnetization

Figure 2 shows the temperature dependence of the permeability \(P\) for (a) Ni2MnIn and (b) Ni2MnSn in a zero external magnetic field. The values of \(dP/dT\) shown in Figure 2 are the values of the differential of the permeability in the temperature. For Ni2MnIn and Ni2MnSn, the values of \(T_C\) were obtained from the peak of \(dP/dT\), which were 314 K and 337 K, respectively, using the same approach [14].

![Permeability \((P)\) and \(dP/dT\) (differential of the permeability in the temperature) of Ni2MnIn and Ni2MnSn around \(T_C\). The dotted lines define \(T_C\).](image)
Ni$_2$MnGa-type Heusler alloys, such as Ni$_{2+y}$MnGa$_{1-x}$ ($0 \leq x \leq 0.04$) and Ni$_2$Mn$_{1-y}$Cr$_y$Ga ($0 \leq y \leq 0.25$), Takahashi’s theory has also been adapted successfully [11,14–17]. The spin fluctuation parameter in $k$-space, $T_A$, and in energy space, $T_0$, has been calculated from the magnetization process at $T_C$ using Equations (3) and (4) by Takahashi’s theory [1].

Furthermore, we investigated the magnetization measurement at 4.2 K, which corresponds to the magnetization process that was performed at the ground state ($T << T_C$, $T/T_C \approx 1\%$). Figure 4 plots the magnetic field dependences of the magnetization, $M^2$ versus $H/M$, which corresponds to the Arrott plot at 4.2 K for (a) Ni$_2$MnIn and (b) Ni$_2$MnSn [22]. These plots indicated that $M^2$ was proportional to $H/M$ in high magnetic fields and could be appreciable to Equation (3) of Takahashi’s theory [1]. Then, $T_A$ and $T_0$ were obtained by means of Equations (3)–(7).

The obtained parameters, $T_A$ and $T_0$, are listed in Table 1. These results indicate that Takahashi’s theory is applicable to Ni$_2$MnIn and Ni$_2$MnSn alloys. The experimental results followed the relation of $(\Delta V/V) \propto M^4$, which is correct in Equation (10), proposed by Takahashi’s theory [1].
3.2. Correlation between Magnetization and Forced Magnetostriction

In this subsection, we describe the investigations of forced magnetostrictions for Ni$_2$MnIn and Ni$_2$MnSn, and the correlation between forced volume magnetostriction and magnetization is discussed. In order to consider the relevance between magnetization and forced magnetostriction, we examined the magnetostriction in the magnetic fields and at $T_{C}$. Figure 5 shows the external magnetic field dependence of the forced magnetostriction for (a) Ni$_2$MnIn and (b) Ni$_2$MnSn. The forced volume magnetostriction $\Delta V/V$ was derived using Equation (11). For both alloys, the obtained $\Delta V/V$ was proportional to the fourth power of the $M$, $(\Delta V/V) \propto M^4$, and crossed the origin, $(M^4, \Delta V/V) = 0$, as indicated by the dotted linearly fitting line. This result is consistent with other Ni$_2$MnGa-type Heusler alloys [14,15,17]. Faske et al. conducted an experimental investigation into the magnetization $M$ and magnetostriction $\Delta V/V$ of LaFe$_{11.6}$Si$_{1.4}$ [12]. They found the relationship between $\Delta V/V$ and $M$ as $(\Delta V/V) = M^4$, and crossed the origin, and they suggested that the experimental results of $\Delta V/V$ and $M$ were in accordance with Takahashi’s theory [1]. As for renowned weak ferromagnet MnSi [8], Takahashi suggested that the relationship between $\Delta L/L$ and $M$ is $(\Delta L/L) \propto M^4$ [1]. Not only weak ferromagnet but also $L_2$-type cubic Heusler alloys, and LaFe$_{11.6}$Si$_{1.4}$ (NaZn13-type structure), which has a more complex structure, are in accordance with Takahashi’s theory.

![Figure 5](image-url)

**Figure 5.** Forced magnetostriction vs. $M^4$ at $T_{C}$: (a) Ni$_2$MnIn; (b) Ni$_2$MnSn at $T_{c}$. Dotted straight lines are linearly fitting lines.

In a previous study, we measured the magnetostrictions of Ni$_2$MnGa-type and Heusler alloys at $T_{C}$ and proved that $\Delta V/V$ is proportional to the valence electron per atom, $e/\alpha$ [17]. As for Ni$_2$MnGa,
Ni$_2$MnIn, and Ni$_2$MnSn, the $c/a$ were all the same value as 7.500. Therefore, we compared the forced volume magnetostriction $\Delta V/V$ and its mechanical parameter, bulk modulus $K$ [14,15]. The forced volume magnetostriction $\Delta V/V$ at 5 T and bulk modulus $K$ are listed in Table 2. The $K$ is inversely proportional to Young’s modulus. Therefore, as $K$ becomes smaller, it softens more. The order of $\Delta V/V$ at 5 T is Ni$_2$MnGa < Ni$_2$MnSn < Ni$_2$MnIn. The values of $M^4$ for Ni$_2$MnGa and Ni$_2$MnIn are comparable. The $K$ of Ni$_2$MnIn is smaller than that of Ni$_2$MnGa. Therefore, Ni$_2$MnIn is softer than that of Ni$_2$MnGa. It is conceivable that the strain grows larger for a softer alloy. Then, the $\Delta V/V$ of Ni$_2$MnIn is larger than that of Ni$_2$MnGa. The value of $M^4$ for Ni$_2$MnSn is larger than that of Ni$_2$MnGa. Moreover, from the results of $K$, Ni$_2$MnSn is softer than Ni$_2$MnGa. Therefore, the $\Delta V/V$ of Ni$_2$MnSn is larger than that of Ni$_2$MnGa.

### Table 2. The forced volume magnetostriction $\Delta V/V$ at 5 T and the bulk modulus.

| Alloy     | $\Delta V/V$ at 5 T | $M^4$ ((Am$^2$/kg)$^4$) at 5 T | Bulk Modulus $K$ (GPa) | $K$ ($\Delta V/V$) (J/m$^3$) |
|-----------|----------------------|---------------------------------|-------------------------|-------------------------------|
| Ni$_2$MnGa | $152 \times 10^{-6}$ | $1.52 \times 10^6$             | 166                     | $2.52 \times 10^{-2}$        |
| Ni$_2$MnIn | $190 \times 10^{-6}$ | $1.49 \times 10^6$             | 137                     | $2.60 \times 10^{-2}$        |
| Ni$_2$MnSn | $182 \times 10^{-6}$ | $1.69 \times 10^6$             | 143                     | $2.60 \times 10^{-2}$        |

1 [14,15]. 2 [24]. 3 [25].

The units of $M^4$ and $K$ are defined by (Am$^2$/kg)$^4$ and Pa, respectively; $\Delta V$ and $V$ are measured in m$^3$; $K$ is also defined in N/m$^2$. The $K\Delta V$ is in the dimension of Pa·m$^3$ = (N/m$^2$)·m$^3$ = Nm = J. Therefore, $K(\Delta V/V)$ is in J/m$^3$. Here, we defined the parameter $E_K$ in J/m$^3$. The $\Delta V/V = E_K/K$. This equation indicates that the forced volume magnetostriction $\Delta V/V$ is inversely proportional to bulk modulus $K$. The $K(\Delta V/V)$ is also listed in Table 2. This is almost the same value. This result also indicates that $\Delta V/V$ is inversely proportional to $K$.

### 3.3. Spin Polarization of Ni$_2$MnGa-Type Heusler Alloys

In this subsection, we consider the magnetism of Ni$_2$MnGa-type Heusler alloys by comparing the spontaneous magnetic moment at the ground state, $p_s$, and paramagnetic magnetic moment, $p_C$.

The relation between $p_{eff}$ and $p_C$ is described as:

$$p_{eff} = \sqrt{p_C(p_C + 2)}.$$  \hspace{1cm} (12)

The $p_C$ is obtained from the Curie constant and it is non-dimensional, $C = N_0\mu eff^2/3k_B = N_0p_{eff}^2/3k_B = N_0(p_C(p_C + 2))\mu eff^2/3k_B$. The $p_C/p_s$ is 1 for the local-moment ferromagnetism. For the weak itinerant electron ferromagnetism, the $p_C/p_s$ is larger than 1 [1]. On the contrary, many Heusler alloys have a $p_C/p_s$ value smaller than 1 [16]. As for the itinerant electron magnets, the minority-spin electrons band has a gap at the Fermi level $E_F$ and indicates semi-metallic or insulating bands. On the contrary, the Fermi level intersects the majority-spin electrons band and represents metallic bands. The $p_C/p_s < 1$ indicates that the spin polarization occurs, and these alloys can be classified as half-metallic alloys (HMFA). The $p_S$ and $p_C$ for Ni$_2$MnGa-type Heusler alloys are listed in Table 3. Bocklage et al. performed point contact Andreev reflection (PCAR) spectroscopy on Ni$_2$MnIn [26]. The obtained polarization value $P_0$ was 35%. The $p_C/p_S$ of Ni$_2$MnIn was 0.860. Both Co$_2$VGa and Co$_2$MnGa are known as typical HMAs. The $P_0$ values were 75% and 48% for Co$_2$VGa and Co$_2$MnGa, respectively [27]. The $p_C/p_S$ values of Co$_2$VGa and Co$_2$MnGa were 0.70 and 0.80, respectively. The results for these three alloys indicate that the alloy with a larger spin polarization showed a smaller $p_C/p_S$ value. The spin polarization of Ni$_2$MnSn was obtained by theoretical calculations [25]. The obtained $P_0$ was about 10%, which indicates that the spin polarization of Ni$_2$MnSn is smaller than that of Ni$_2$MnIn. Then, the $p_C/p_S$ of Ni$_2$MnSn was almost 1. Even at low temperature, Ni$_2$MnIn and Ni$_2$MnSn take an $L_2_1$-type cubic structure. On the contrary, Ni$_2$MnGa causes martensitic transformation at $T_M = 195$ K, and below this temperature, $14 M$ structure was realized [28]. In the martensitic phase, the spin polarization was
19.72% [24]. Webster et al. analyzed the magnetic moment obtained by the saturation magnetization measurement, where \( p_S = 4.17 \) [29]. Then, the \( p_{sat}/p_s \) was 0.92, which is smaller than 1 and deviated from 1 (local moment magnetism). The spin polarization of Ni₂MnGa affected the deviation of the \( p_{sat}/p_s \) value.

Table 3. Magnetic parameters of ferromagnetic Heusler alloys. \( p_C \) indicates the magnetic moment at the paramagnetic phase. The relationship between \( p_{eff} \) and \( p_C \) is defined by the equation of \( p_{eff} = \sqrt{p_C(p_C + 2)} \).

| Sample     | \( T_C \) (K) | \( p_S \) (\( \mu_B/\text{f.u.} \)) | \( p_{eff} \) (\( \mu_B/\text{f.u.} \)) | \( p_C \) (\( \mu_B/\text{f.u.} \)) | \( p_C/p_S \) | Reference          |
|------------|----------------|-------------------------------------|----------------------------------------|-------------------------------------|----------------|---------------------|
| Ni₂MnGa    | 375            | 3.93                                | 4.75                                   | 3.85                                | 0.980          | [16,20]             |
| Ni₂MnIn    | 314 *          | 4.4                                 | 4.69                                   | 3.78                                | 0.860          | * This work, [20]   |
| Ni₂MnSn    | 337 *          | 4.05                                | 5.00                                   | 4.10                                | 1.01           | * This work, [20]   |

Takahashi’s theory can be applied even to the ferromagnetic Heusler alloy, which has a spin polarization, and further study is needed to clarify the origin of the magnetism and its physical properties.

4. Conclusions

In this article, we investigated the itinerant magnetism of Ni₂MnIn and Ni₂MnSn alloys. These alloys are ferromagnetic Heusler alloys and do not cause martensitic transformation [20], in contrast to Ni₂MnGa with a martensitic transformation temperature \( T_M \) of 195 K [21]. These alloys have an \( L2_1 \)-type cubic crystal structure even at low temperature. We considered the magnetostriction and magneto-volume effects of these alloys. We measured the forced longitudinal magnetostriction \( (\Delta L/L)_// \) and \( (\Delta L/L)_\perp \), and we derived the forced volume magnetostriction \( \Delta V/V \). The correlation between the magnetization \( M \) and \( \Delta V/V \) is \( (\Delta L/L) \propto M^4 \), and the linear fitting line crossed the origin for both alloys. These results were confirmed by Takahashi’s theory [1]. From the magnetization results at \( T_C \) and 4.2 K, the spin fluctuation parameters were \( T_A \) in \( k \)-space and \( T_0 \) in energy space. The obtained \( k_m \) parameter of the generalized Rhodes–Wohlfarth equation was around 1.4. This result accorded with Takahashi’s theory. We considered the results of the examinations and theoretical calculations. We concluded that Takahashi’s theory can apply even to the ferromagnetic Heusler alloy, which has a spin polarization. We compared the forced volume magnetostriction \( \Delta V/V \) and its mechanical parameter, bulk modulus \( K \), and found that \( \Delta V/V \) is inversely proportional to \( K \).

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