


Article

The Forced Magnetostrictions and Magnetic Properties of Ni₂MnX (X = In, Sn) Ferromagnetic Heusler Alloys

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Received: 27 March 2020; Accepted: 23 April 2020; Published: 25 April 2020



Abstract: Experimental studies into the forced magnetostriction, magnetization, and temperature dependence of permeability in Ni₂MnIn and Ni₂MnSn 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 Ni₂MnIn and Ni₂MnSn, 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 Ni₂MnIn and $T_A = 447$ K, $T_0 = 279$ K for Ni₂MnSn, 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 Ni₂MnIn and other magnetic Heusler alloys. The p_C/p_S of Ni₂MnIn was 0.860. This is comparable with that of Co₂MnGa, 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):

$$\left(\frac{M}{M_S}\right)^4 = 1.20 \times 10^6 \times \left(\frac{T_C^2}{w_A T_A^3 p_S^4}\right) \times \left(\frac{H}{M}\right), \quad (1)$$

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 ω -space). The Lorentzian function of the k -space is proportional to $\chi(q = 0, \omega = 0)$. The half-width of this function, Δq , 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, Δq is shown in a dimension of the energy. Therefore, Δq 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 $T_A = \bar{A} q_B^2$, where q_B^2 indicates the effective zone boundary wave vector, and \bar{A} indicates the non-dimensional parameter, as shown in Equation (3.6) in reference [2]. Another parameter, T_0 , is a spectral distribution Γ_{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 $3d$ and $5f$ 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]:

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

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 ($T \ll T_C$) [1,13,15].

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

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

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

$$F_1 = \frac{2T_A^2}{15cT_0}. \quad (4)$$

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):

$$F_1 = \frac{N_0^3 (2\mu_B)^4}{k_B \zeta}, \quad (5)$$

where k_B indicates the Boltzmann factor, and ζ 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:

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

$$\left(\frac{T_C}{T_A}\right)^{5/6} = \frac{p_S^2}{5g^2C_{4/3}} \times \left(\frac{2T_C}{15cF_1}\right)^{1/2}. \quad (7)$$

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_{eff} and the spontaneous magnetic moment p_S can be expressed by a generalized Rhodes–Wohlfarth equation (Equation (3.47) in reference [1]):

$$\frac{p_{\text{eff}}}{p_S} = 1.4 \times \left(\frac{T_0}{T_C}\right)^{2/3}. \quad (8)$$

Equation (8) can be rewritten as

$$k_m = \left(\frac{p_{\text{eff}}}{p_S}\right) \times \left(\frac{T_C}{T_0}\right)^{\frac{2}{3}}. \quad (9)$$

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]:

$$(\Delta V/V) \propto M^4, \quad (10)$$

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

$$(\Delta V/V) = (\Delta L/L)_{//} + 2 \times (\Delta L/L)_{\perp}, \quad (11)$$

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_2MnIn and Ni_2MnSn alloys. These alloys are ferromagnetic Heusler alloys and do not cause martensitic transformation [20], in contrast to Ni_2MnGa 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_2MnIn and Ni_2MnSn alloys were synthesized from the constituent elements of Ni_2MnIn : Ni (4N), Mn (3N), In (4N); Ni_2MnSn : Ni (4N), Mn (4N), Sn(5N). The sample of Ni_2MnIn was prepared by induction melting under an Ar atmosphere. The sample of Ni_2MnSn was prepared by arc-melting in an Ar atmosphere. The product of Ni_2MnSn 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_2MnIn and Ni_2MnSn , 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].

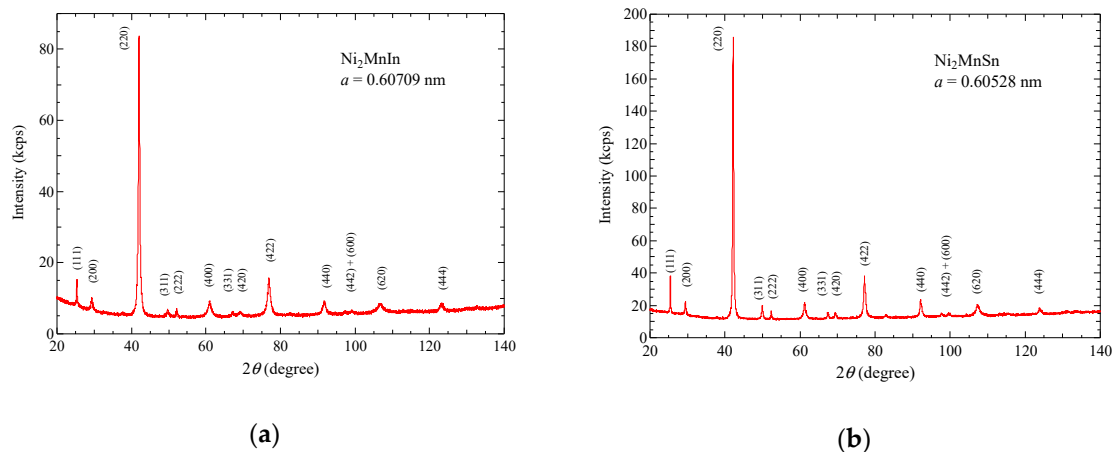


Figure 1. X-ray diffraction patterns of (a) Ni_2MnIn and (b) Ni_2MnSn . Parenthesis indicates the mirror indices.

3. Results and Discussion

3.1. Magnetic Field Dependence of Magnetization

Figure 2 shows the temperature dependence of the permeability P for (a) Ni_2MnIn and (b) Ni_2MnSn 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 Ni_2MnIn and Ni_2MnSn , 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].

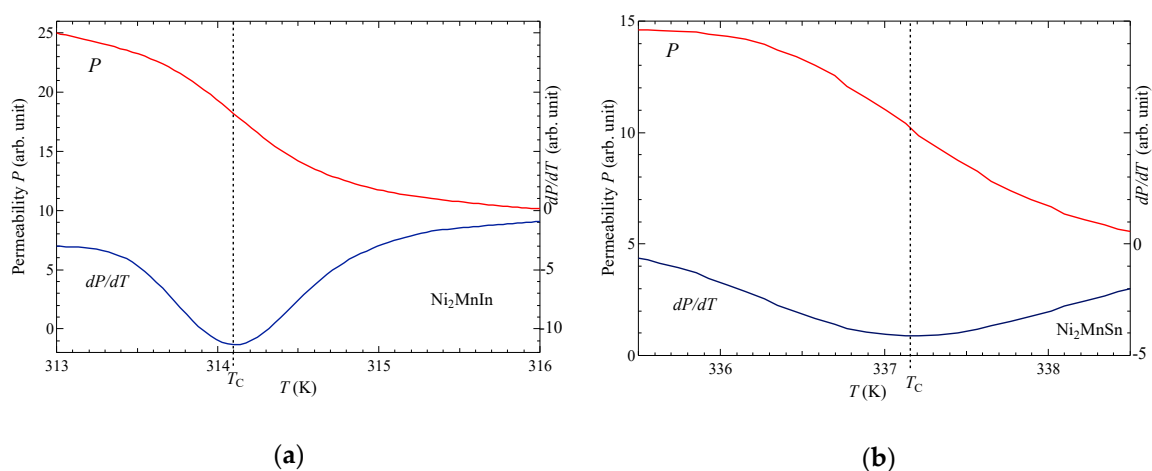


Figure 2. Permeability (P) and dP/dT (differential of the permeability in the temperature) of (a) Ni_2MnIn and (b) Ni_2MnSn around T_C . The dotted lines define T_C .

Figure 3 for (a) Ni_2MnIn and (b) Ni_2MnSn shows the plots of M^4 versus H/M at T_C . A good linearity can be seen at the origin at T_C . The magnetic field dependence of the magnetization indicates that $H \propto M^5$; therefore, the results agree with Takahashi's theory [1]. In former experimental investigations of

Ni_2MnGa -type Heusler alloys, such as $\text{Ni}_{2+x}\text{MnGa}_{1-x}$ ($0 \leq x \leq 0.04$) and $\text{Ni}_2\text{Mn}_{1-x}\text{Cr}_x\text{Ga}$ ($0 \leq x \leq 0.25$), 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].

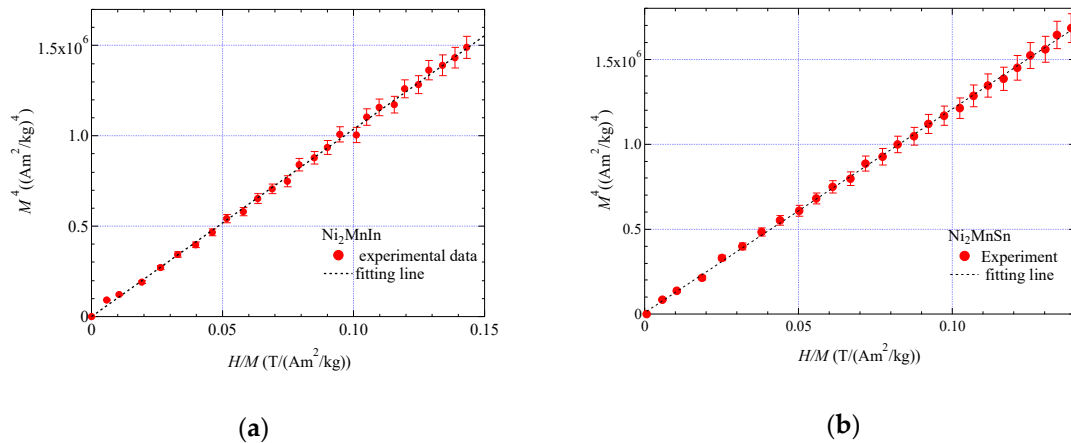


Figure 3. The magnetic field dependences of the magnetization, M^4 vs. H/M at T_C : (a) Ni_2MnIn ; (b) Ni_2MnSn . Dotted straight lines are linearly fitting lines.

Furthermore, we investigated the magnetization measurement at 4.2 K, which corresponds to the magnetization process that was performed at the ground state ($T \ll T_C$, $T/T_C \approx 1\%$). 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_2MnIn and (b) Ni_2MnSn [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).

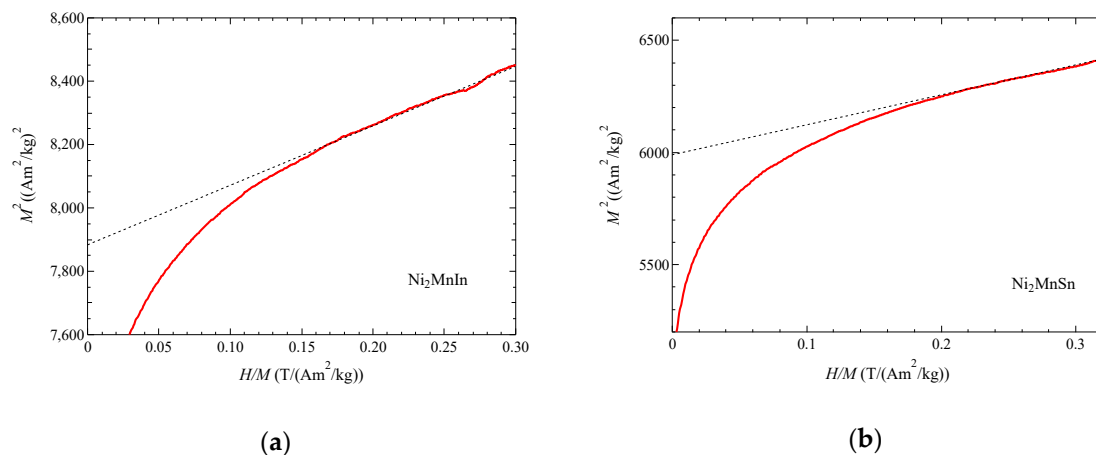


Figure 4. The magnetic field dependences of the magnetization, M^2 vs. H/M at 4.2 K: (a) Ni_2MnIn ; (b) Ni_2MnSn . Dotted straight lines are linearly fitting lines.

The obtained parameters, T_A and T_0 , are listed in Table 1. These results indicate that Takahashi's theory is applicable to Ni_2MnIn and Ni_2MnSn 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].

Table 1. Magnetic parameters of Ni₂MnX (X = Ga, In, Sn). The spontaneous magnetic moment, p_s ; effective moment, p_{eff} ; Curie temperature, T_C ; spin fluctuation parameter in k -space, T_A ; spin fluctuation parameter in energy space, T_0 . The parameter k_m was obtained from Equation (9), which was almost the same as $k_m = 1.4$. “This work T_C ” indicates the values obtained from the magnetization process measurements at T_C , and “This work 4.2 K” indicates the values obtained from the magnetization process measurements at 4.2 K.

Alloy	p_s ($\mu_B/f. u.$)	p_{eff} ($\mu_B/f. u.$)	T_C (K)	T_A (K)	T_0 (K)	k_m	Reference
Ni ₂ MnGa	3.93	4.75	375	563	245	1.61	[15] $T = T_C$
Ni ₂ MnGa	3.93	4.75	375	556	254	1.57	[15] $T = 5$ K
Ni ₂ MnIn	4.40 ¹	4.69 ²	314	351	255	1.23	This work T_C
Ni ₂ MnIn	4.40 ¹	4.69 ²	314	332	296	1.11	This work 4.2 K
Ni ₂ MnSn	4.05 ¹	5.00 ²	337	461	271	1.42	This work T_C
Ni ₂ MnSn	4.05 ¹	5.00 ²	337	432	286	1.37	This work 4.2 K

¹ [23], ² [20].

3.2. Correlation between Magnetization and Forced Magnetostriction

In this subsection, we describe the investigations of forced magnetostrictions for Ni₂MnIn and Ni₂MnSn, 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) Ni₂MnIn and (b) Ni₂MnSn. 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 Ni₂MnGa-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 $(\Delta L/L) \propto M^4$, 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 $L2_1$ -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.

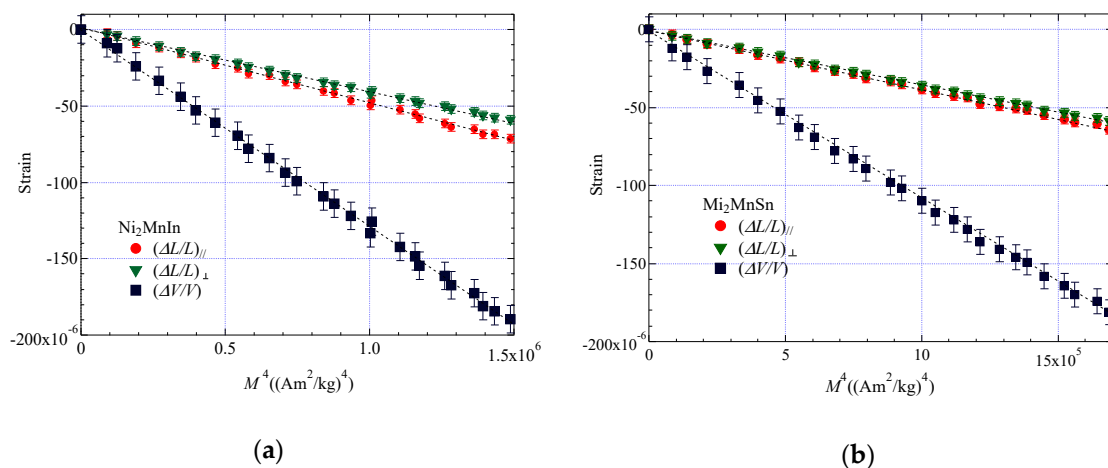


Figure 5. Forced magnetostriction vs. M^4 at T_C : (a) Ni₂MnIn; (b) Ni₂MnSn at T_C . Dotted straight lines are linearly fitting lines.

In a previous study, we measured the magnetostrictions of Ni₂MnGa-type and Heusler alloys at T_C and proved that $\Delta V/V$ is proportional to the valence electron per atom, e/a [17]. As for Ni₂MnGa,

Ni₂MnIn, and Ni₂MnSn, the e/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 Ni₂MnGa < Ni₂MnSn < Ni₂MnIn. The values of M^4 for Ni₂MnGa and Ni₂MnIn are comparable. The K of Ni₂MnIn is smaller than that of Ni₂MnGa. Therefore, Ni₂MnIn is softer than that of Ni₂MnGa. It is conceivable that the strain grows larger for a softer alloy. Then, the $\Delta V/V$ of Ni₂MnIn is larger than that of Ni₂MnGa. The value of M^4 for Ni₂MnSn is larger than that of Ni₂MnGa. Moreover, from the results of K , Ni₂MnSn is softer than Ni₂MnGa. Therefore, the $\Delta V/V$ of Ni₂MnSn is larger than that of Ni₂MnGa.

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)	$K \cdot (\Delta V/V)$ (J/m ³)
Ni ₂ MnGa	152×10^{-6} ¹	1.52×10^6 ¹	166 ²	2.52×10^{-2}
Ni ₂ MnIn	190×10^{-6}	1.49×10^6	137 ²	2.60×10^{-2}
Ni ₂ MnSn	182×10^{-6}	1.69×10^6	143 ³	2.60×10^{-2}

¹ [14,15], ² [24], ³ [25].

The units of M^4 and K are defined by (Am²/kg)⁴ and Pa, respectively; ΔV and V are measured in m³; K is also defined in N/m². The $K\Delta V$ is in the dimension of Pa·m³ = (N/m²)·m³ = Nm = J. Therefore, $K \cdot (\Delta V/V)$ is in J/m³. Here, we defined the parameter E_K in J/m³. 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 $K \cdot (\Delta V/V)$ 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 Ni₂MnGa-Type Heusler Alloys

In this subsection, we consider the magnetism of Ni₂MnGa-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:

$$p_{eff} = \sqrt{p_C(p_C + 2)}. \quad (12)$$

The p_C is obtained from the Curie constant and it is non-dimensional, $C = N_0\mu_{eff}^2/3k_B = N_0p_{eff}^2\mu_B^2/3k_B = N_0p_C(p_C + 2)\mu_B^2/3k_B$. 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 Ni₂MnGa-type Heusler alloys are listed in Table 3. Bocklage et al. performed point contact Andreev reflection (PCAR) spectroscopy on Ni₂MnIn [26]. The obtained polarization value P_0 was 35%. The p_C/p_s of Ni₂MnIn was 0.860. Both Co₂VGa and Co₂MnGa are known as typical HMAs. The P_0 values were 75% and 48% for Co₂VGa and Co₂MnGa, respectively [27]. The p_C/p_s values of Co₂VGa and Co₂MnGa 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 Ni₂MnSn was obtained by theoretical calculations [25]. The obtained P_0 was about 10%, which indicates that the spin polarization of Ni₂MnSn is smaller than that of Ni₂MnIn. Then, the p_C/p_s of Ni₂MnSn was almost 1. Even at low temperature, Ni₂MnIn and Ni₂MnSn take an L2₁-type cubic structure. On the contrary, Ni₂MnGa 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 Ni_2MnGa 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 $p_{\text{eff}} = \sqrt{p_C(p_C + 2)}$.

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
Ni_2MnGa	375	3.93	4.75	3.85	0.980	[16,20]
Ni_2MnIn	314 *	4.4	4.69	3.78	0.860	* This work, [20]
Ni_2MnSn	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 Ni_2MnIn and Ni_2MnSn alloys. These alloys are ferromagnetic Heusler alloys and do not cause martensitic transformation [20], in contrast to Ni_2MnGa 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 $(\Delta L/L)_{\parallel}$ 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 .

Author Contributions: Y.A. and T.K. (Takeshi Kanomata) prepared the samples; T.S., H.N., Y.N., M.H., and T.K. (Takeshi Kanomata) conceived and designed the experiments; J.Y., T.K. (Takumi Komori), H.N., Y.N., M.H., and T.S. performed the experiments; J.Y., T.K. (Takumi Komori), and T.S. analyzed the data; T.S., J.Y., T.K. (Takumi Komori), Y.N., M.H., T.K. (Takeshi Kanomata), and Y.A. wrote the paper. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Acknowledgments: The authors thank Mitsuo Okamoto and Fumihiko Morioka of Ryukoku University for helping to create the apparatus. The authors thank Koji Fukushima and Taishi Yasuda of Tohoku Gakuin University. This project is partly supported by the Ryukoku Extension Center (REC) at Ryukoku University. This research was also carried out at the High Field Laboratory for Superconducting Materials, Institute for Materials Research, Tohoku University, and at the Center for Advanced High Magnetic Field Science in Osaka University under the Visiting Researchers' Program of the Institute for Solid State Physics, the University of Tokyo.

Conflicts of Interest: The authors declare no conflict of interest.

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