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Repulsive magneto-structural interaction in the ferromagnetic shape memory alloys $Ni_2Mn_{1+x}In_{1-x}$

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ABSTRACT

A phenomenological theory is presented to study the multiferroic system in the shape memory alloys $Ni_2Mn_{1+x}In_{1-x}$ with the Heusler-type structure, where the phase transitions are characterized by the two order parameters, i.e., the martensitic distortion e_3 and the magnetization M. The Landau free energy is expanded in powers of e_3 and M with including the Zeeman energy and the energy increase by the uniaxial force on e_3. The resultant free energy is applied to analyze the experimental results, such as the phase diagram in the temperature–concentration plane and the reentrant ferromagnetism by increasing temperature. The magnetic fields are shown to cause the ferromagnetic–ferromagnetic or metamagnetic transitions as observed. Further, it is predicted that the uniaxial force can dominate the appearance of the ferromagnetism in some alloys. It is found that these exotic behaviors of this alloy system are all ascribed to the repulsive interaction between e_3 and M, which originates from their biquadratic term. Through the present analyses, it is verified that this repulsive interaction is expected in other alloy systems $Ni_2Mn_{1+x}X_{1-x}$ (X=Sn, Sb) and makes a contrast to the attractive interaction realized in another group of the alloy systems including $Ni_{2+x}Mn_{1-x}Ga$ and $Ni_2Mn_{1-x}Cu_xGa$.

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1. Introduction

In the last decade or so, some Heusler-type ferromagnetic shape memory alloys have attracted much attention of researchers, since they are multiferroic systems consisting of the martensitic distortion and the ferromagnetism and show various exotic phenomena [1–4]. Now, many experimental studies, such as those on the magnetic-field strain [5,6], giant magnetocaloric effect [4,7] and magnetoresistance [8,9] have been accumulated. Also the phase diagrams as a function of temperature and concentration of constituent elements of mixed alloys have been reported. Now, it has been established that those phase diagrams are spanned mainly by the paramagnetic-austenite (Para-A), paramagnetic-martensite (Para-M), ferromagnetic-austenite (Ferro-A), and ferromagnetic-martensite (Ferro-M) phases.

The phase diagram of $Ni_{2+x}Mn_{1-x}Ga$ was first investigated experimentally by Vasil'ev et al. [10] and Khovaylo et al. [11]. According to their results, the Para-A phase is adjacent to the Ferro-M phase on their phase boundary in the concentration region $0.18 \le x \le 0.27$. In this concentration region, therefore, the alloys exhibit the magnetostructural transition, i.e., the coupled transition of the ferromagnetism and the martensitic distortion, between the Para-A and Ferro-M phases. Such the magnetostructural transitions were observed also for $Ni_2Mn_{1-x}Cu_xGa$ with $0.23 \le x \le 0.3$ [12] and for $Ni_2MnGa_{1-x}Cu_x$ with $0.12 \le x \le 0.14$ [13]. For the end member Ni_2MnGa, the magnetization has been known to increase with the transition from the Ferro-A phase into the Ferro-M phase [14,15]. On the other hand, Sutou et al. obtained experimentally the phase diagrams of the alloy systems $Ni_2Mn_{1+x}X_{1-x}$ (X=In, Sn, and Sb) [16]. The phase diagrams of these alloy systems do not show the occurrence of the magnetostructural transition. Instead, alloys belonging to this group show reentrant ferromagnetisms with decreasing temperature in narrow concentration regions. Moreover, the magnetization in the Ferro-M phase was often found to be much smaller than that in the Ferro-A phase [16–18]. The properties of the two groups of alloy systems form a striking contrast to each other, although the observed phases are all characterized by the same order parameters.

Phenomenological analyses of the multiferroic system were made for the phase diagram of $Ni_{2+x}Mn_{1-x}Ga$, which belongs to the first group, by Vasil'ev et al. [10] and Khovaylo et al. [11]. They proposed that the magnetostriction (or e_3-linear) term in the

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assumed free energy can cause the magnetostructural transition seen in its phase diagram. Recently, Kataoka et al. [12] and Endo et al. [13], respectively, studied phenomenologically the phase diagrams of $Ni_2Mn_{1-x}Cu_xGa$ and $Ni_2MnGa_{1-x}Cu_x$, which also belong to the first group. They assumed the magnetostriction term to be too small to cause the observed magnetostructural transition, and considered a biquadratic term of e_3 and M for the interaction between e_3 and M. As the result, it was found that the magnetostructural transition occurs in the case where the biquadratic term gives an attraction between e_3 and M for which the coexistence of non-vanishing e_3 and M is favorable. On the other hand, such analysis has not yet been done on any alloy system belonging to the second group.

In this paper, therefore, we analyze on the basis of a Landau theory the phase diagram and magnetic properties of the alloy system $Ni_2Mn_{1+x}In_{1-x}$ belonging to the second group, which were already studied experimentally by Kanomata et al. [17] and by Umetsu et al. [19], respectively. This analysis will clarify that the e_3–M interaction in the alloys $Ni_2Mn_{1+x}In_{1-x}$ is repulsive, in contrast to the attractive interaction in the alloys belonging to the first group. For the obtained repulsive interaction, the temperature dependence of the order parameters and the effects of the magnetic field and uniaxial force on the order parameters will be calculated and compared with experimental results. On the basis of these calculations, we will find the reason why the two groups of alloy systems exhibit so strongly different properties.

2. Phenomenological free energy of the system under external fields

The order parameters to describe the properties of $Ni_2Mn_{1+x}In_{1-x}$ are the martensitic distortion e_3 and the magnetization **M**, being same as in $Ni_2Mn_{1-x}Cu_xGa$. As in $Ni_2Mn_{1-x}Cu_xGa$, it is assumed that the magnetization **M** always directs to a crystal axis, say the z-axis, for the positive cubic anisotropy constant $K_1>0$ and the martensitic distortion e_3 is a tetragonal distortion defined by $e_3=(2e_{zz}-e_{xx}-e_{yy})/\sqrt{6}$ with the strain along the i-axis, e_{ii}. It is further assumed that the external fields, i.e., magnetic field H and uniaxial force (pressure $P>0$ or tension $P<0$) are applied to the crystal along the z-axis. After making a parallel argument to that in [12], the phenomenological free energy per unit volume as a function of e_3 and $M=|\mathbf{M}|$ is written as follows:

$$F_{\rm tot}=\frac{1}{2}c_2e_3^2+\frac{1}{3}A_3e_3^3+\frac{1}{4}I_4e_3^4+\frac{1}{2}\chi^{-1}M^2+\frac{1}{4}J_1M^4+\frac{1}{2}\left(G_2+\frac{2}{\sqrt{6}}G_4\right)e_3^2M^2+\frac{1}{2}Q_1e_3^2M^4+e_3P-MH, \quad (1)$$

where c_2, A_3 and I_4 are to be expressed in terms of various elastic constants, χ^{-1} and J_1 originate from the spin exchange energy, and G_2, G_4, and Q_1 represent the magnitudes of interactions between e_3 and M. In the above $F_{\rm tot}$, the magnetostriction term linear to e_3 and an interaction term proportional to $e_3^3M^2$ were already neglected by assuming their minor roles, whereas the term of $e_3^2M^4$ was taken into account by preserving its contribution to determination of M in the Ferro-M phase of $Ni_2Mn_{1+x}In_{1-x}$.

The coefficients in the terms on the right hand side of Eq. (1) are regarded as constants independent of concentration x and temperature T except for c_2 and χ^{-1}. As usually, the simplest x- and T-dependence of c_2 and χ^{-1} are assumed as follows:

$$c_2=\frac{c_{\rm m}}{T_{\rm M}(1)}[T-T_{\rm M}(x)], \quad (2)$$

$$\chi^{-1}=\frac{c_{\rm f}}{T_{\rm C}(0)}[T-T_{\rm C}(x)], \quad (3)$$

with

$$T_{\rm M}(x)=T_{\rm M}(0)+[T_{\rm M}(1)-T_{\rm M}(0)]x, \quad (4)$$

$$T_{\rm C}(x)=T_{\rm C}(0)+[T_{\rm C}(1)-T_{\rm C}(0)]x, \quad (5)$$

where $c_{\rm m}$ and $c_{\rm f}$ are positive constants independent of x and T, and $T_{\rm M}(x)$ and $T_{\rm C}(x)$ are fictitious (or sometimes real) second order transition temperatures of e_3 and M, respectively.

As in [12], we measure the various quantities in Eqs. (1)–(5) in their respective units as follows:

$$t=T/T_{\rm C}(0), \quad (6)$$

$$t_{\rm m}(x)=T_{\rm M}(x)/T_{\rm C}(0)=t_{\rm m}(0)+[t_{\rm m}(1)-t_{\rm m}(0)]x, \quad (7)$$

$$t_{\rm c}(x)=T_{\rm C}(x)/T_{\rm C}(0)=1+[t_{\rm c}(1)-1]x, \quad (8)$$

$$\overline{e}_3=e_3/\sqrt{c_{\rm m}/I_4}, \quad (9)$$

$$\overline{M}=M/\sqrt{c_{\rm f}/J_1}, \quad (10)$$

$$\overline{P}=P/(c_{\rm f}^2/J_1)\sqrt{I_4/c_{\rm m}}, \quad (11)$$

$$\overline{H}=H/c_{\rm f}\sqrt{c_{\rm f}/J_1}, \quad (12)$$

and

$$\overline{F}_{\rm tot}(\overline{e}_3,\overline{M})=F_{\rm tot}(e_3,M)/(c_{\rm f}^2/J_1). \quad (13)$$

Then, Eq. (1) is simplified as follows:

$$\overline{F}(\overline{e}_3,\overline{M})=\frac{1}{2}[t-t_{\rm c}(x)]\overline{M}^2+\frac{1}{4}\overline{M}^4+r\left\{\frac{1}{2t_{\rm m}(1)}[t-t_{\rm m}(x)]\overline{e}_3^2+\frac{2}{3}a\overline{e}_3^3+\frac{1}{4}\overline{e}_3^4\right\}+\frac{1}{2}g\overline{e}_3^2\overline{M}^2+\frac{1}{2}q\overline{e}_3^2\overline{M}^4+\overline{e}_3\overline{P}-\overline{M}\overline{H}, \quad (14)$$

where the new coefficients were defined by

$$r=(c_{\rm m}/c_{\rm f})^2(J_1/I_4),\ a=(1/2\sqrt{c_{\rm m}I_4})A_3,$$

$$g=\frac{c_{\rm m}}{c_{\rm f}I_4}\left(G_2+\frac{2}{\sqrt{6}}G_4\right),\quad \text{and}\quad q=\frac{c_{\rm m}}{J_1I_4}Q_1. \quad (15)$$

The biquadratic term of $\overline{e}_3$ and $\overline{M}$ on the right hand side of Eq. (14) affects directly their transition temperatures. It has been already seen in [12] that the martensitic and ferromagnetic states are attractive to each other for $g<0$ to cause the magnetostructural transition. Contrary, the term with $g>0$ gives the repulsion between the two states. The term with $q>0$ also contributes to this repulsion. For the repulsive interaction, the phase diagram and the temperature dependence of the order parameters are expected to be drastically changed from those for the attractive interaction. In the following, we confine ourselves to the repulsive interaction, which will be shown to be realized in $Ni_2Mn_{1+x}In_{1-x}$.

3. Phase diagram

In this section, the phase diagram of $Ni_2Mn_{1+x}In_{1-x}$ in the t–x plane is calculated and compared with the phase diagram observed by Kanomata et al. [17]. The phases observed in this alloy system, Para-A, Para-M, Ferro-A, and Ferro-M, are, respectively, labeled as I, II, III, and IV for convenience of the following equations. Moreover, the temperatures of the transition from the phase N to the phase N′ by increasing temperature are denoted by $t_{\rm c,N'\leftrightarrow N}(x)$ in the unit of $T_{\rm C}(0)$, N and N′ being any of I, II, III, or IV. The transitions accompanying the martensitic transition are always of the first order, while the ferromagnetic transitions which do not accompany the martensitic transition are always of the second order. By taking into account these facts, various

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