Influence of plastic deformation on the magnetic transition temperature of L1₂-type materials

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The influence of plastic deformation on the magnetic transition temperature in L1₂-type materials is explained in terms of the modified slip-induced directional-order model with consideration of the magnetic interactions between the neighboring atoms. The calculation based on the pair-interaction model predicts that the magnetic transition temperature changes in proportion to the dislocation density at the early stage of plastic deformation and to the square root of it at the advanced stage. The numerical estimation is given for four alloys with the L1₂-type superstructure: i.e., Pt₃Fe, Pt₃Co, Fe₃Pt, and Ni₃Mn.

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I. INTRODUCTION

The effect of local atomic environment has been of considerable interest experimentally and theoretically in connection with the origin of a magnetic moment and the formation of ferromagnetism.¹ The studies on the composition dependence of disordered alloys or on the comparison between the chemically ordered and disordered alloys have been hitherto made to pursue the local environment effect on magnetic properties. Recently, it has been pointed out²⁻⁵ that the study on the correlation between the magnetic properties and plastic deformation must become an additional new way for the study of this subject.

Several remarkable magnetic phenomena due to plastic deformation were discovered in the chemically ordered alloys. The Pt₃Fe alloy is antiferromagnetic when ordered and ferromagnetic when disordered in the superlattice region.^{6,7} It was also discovered that in the long-range ordered Pt₃Fe alloy, which showed no ferromagnetism before plastic deformation, the application of plastic deformation caused it to become quite strongly ferromagnetic even at room temperature.⁶ It was found that in the ordered Ni₃Mn alloy, the saturation magnetization decreases with the amount of elongation, while in the ordered Pt₃Co alloy, the intensity of magnetization increases with the amount of compression.⁸

The modified slip-induced directional-order model^{9,10} has been applied to the cold-working effect on magnetization in the chemically ordered alloys. And, the transition of magnetic interactions caused by plastic deformation was elucidated by the dislocation theory with consideration of magnetic interactions between the neighboring atoms in the Pd₂MnSn Heusler alloy² and a series of L1₂-type materials such as Pt₃Fe, Pt₃Co, Ni₃Mn, Ni₃Fe, and Ni₃Al.³⁻⁵

The magnetic transition temperature θ_m is determined by the exchange interactions between atoms, which are influenced by the local environment. Actually, it is known¹ that θ_m in Pt₃Fe, Pt₃Co, Fe₃Pt, Ni₃Mn, etc., is sensitively

affected by the chemical order-disorder transition. The effect of atomic ordering on the Curie temperature T_C in Fe₃Pt (Ref. 11) and the composition dependence of T_C in the disordered Ni-Mn alloys¹² were interpreted by the pair-interaction model. In the present work, the influence of plastic deformation on θ_m is elucidated in the L1₂-type materials, by using the modified slip-induced directional-order model and the pair-interaction model. The effect of plastic deformation on θ_m is explained in terms of dislocation density and the type of superlattice dislocations. The numerical estimation will be given for four alloys with the L1₂-type superstructure: i.e., Pt₃Fe, Pt₃Co, Fe₃Pt, and Ni₃Mn.

II. CRYSTAL STRUCTURE AND PLASTIC DEFORMATION

The long-range ordered A₃B material of the L1₂-type has such the structure that A and B atoms occupy the face-centered and corner sites, respectively, in the face-centered cubic structure. Each B atom is surrounded by twelve A atoms at the first nearest neighbor (NN) (Table I). By plastic deformation, dislocations nucleate, slip over the glide plane, and make the atomic configuration change near the slip plane.

There is a repulsive force between two dislocations with the same Burgers vector, and the wider the distance between them, the more stable their interaction in usual crystals. The dislocation in the long-range ordered structure, however, creates an antiphase boundary (APB) over the glide plane after it has slipped and it is stable to make a pair with another dislocation. The separation between paired dislocations is given by the APB energy and the elastic interaction energy between individual dislocations. They are shown schematically in Fig. 1, where one perfect dislocation with $(a/2)[01\bar{1}]$ Burgers vector separates into two partial dislocations with $(a/6)\langle 211 \rangle$ Burgers vector. a is the lattice constant. Between perfect dislocations there exists an APB over the $\{111\}$ glide plane, while there exist an APB and stacking fault between partial dislocations. The separation of partial dislocations r_1 is expected to be much smaller than that of perfect disloca-

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TABLE I. Number of neighboring atoms around the host atoms in the A_3B materials with the $L1_2$ -type structure, before and after plastic deformation. There are two kinds of host A atoms with different number of the NN atoms near the APB.

	Neighboring atoms	At first NN		At second NN		At third NN		At fourth NN		At fifth NN		At sixth NN	
		A	B	A	B	A	B	A	B	A	B	A	B
Host atoms	Around A atom	8	4	6	0	16	8	12	0	16	8	8	0
	Around B atom	12	0	0	6	24	0	0	12	24	0	0	8
APB	Around A atom												
	at F site	8	4	6	0	16	8	12	0	16	8	8	0
	at C site	9	3	3	3	19	5	9	3	20	4	3	5
	Around B atom	11	1	3	3	21	3	3	9	20	4	4	4

tions r . These separations will take constant values. We will call these dislocations a -type superlattice dislocations. The a -type superlattice dislocations might be observable in the initial stage of plastic deformation.

Another type of superlattice dislocation can be introduced, which does not have a constant value as the separation r and does distribute independently free from the attractive force due to the APB energy. These superlattice dislocations are expected to be observable in the advanced stage of plastic deformation. They are called b -type superlattice dislocations in the present paper.

At first the effect of partial dislocations is neglected. The dislocations are created by plastic deformation; the first perfect dislocation makes the APB over the glide plane and the next dislocation makes the glide plane recover. The APB between these dislocations remains over the glide plane. Across the APB induced by cold-working over the glide plane, the number of each atom pair changes.

The superlattice dislocations are accompanied by the APB over the $\{111\}$ glide plane. The APB and the initial states are alternate over the glide plane after plastic deformation. The area of APB induced by plastic deformation is given by

$$A = \int_V r ds. \quad (1)$$

Here, r is the separation between two dislocations composing the APB and ds the segment of one of these dislocations.

FIG. 1. Schematic structure of superlattice dislocations over the (111) glide plane in the $L1_2$ -type structure. The $(a/2)[01\bar{1}]$ perfect dislocation separates into two partial dislocations with $(a/6)(211)$ Burgers vector.

The integral is performed over the entire specimen.

The number of B-B atom pairs per unit area in the $\{111\}$ plane, which are located at the first NN, is $S^2/(\sqrt{3}a^2)$. Here, S is the long-range order parameter before the specimen is plastically deformed. Then, the number of B-B atom pairs per unit volume is given⁹ by

$$n = \frac{S^2}{2\sqrt{3}a^2} \bar{r}\rho. \quad (2)$$

Here, $\bar{r}$ is the average of r and ρ the dislocation density, which is defined as the whole dislocation length per unit volume. The $\bar{r}$ takes a constant value r_0 at the early stage of plastic deformation where dislocations distribute in pairs (i.e., for the a -type dislocation). On the other hand, the $\bar{r}$ is a function of the dislocation density and its distribution for the b -type superlattice dislocation. Generally, if the dislocations distribute homogeneously, the $\bar{r}$ is given in terms of the dislocation density by $\bar{r} = 1/\sqrt{\rho}$.

Next, the effect of partial dislocations is considered. As shown in Fig. 1, one perfect dislocation separates into two partial dislocations over the (111) glide plane:

$$\frac{a}{2}(01\bar{1}) = \frac{a}{6}(11\bar{2}) + \frac{a}{6}(\bar{1}2\bar{1}). \quad (3)$$

The stacking fault and APB extend between the partial dislocations, as shown in Fig. 1. In regions (a) and (c), B-B atom pairs align along to $(a/6)[114]$ and $(a/6)[141]$, respectively. The number of B-B atom pairs appearing on the APB is given as

$$n = \frac{S^2}{2\sqrt{3}a^2} \bar{r}_1 \rho', \quad (4)$$

where $\bar{r}_1$ is the average of r_1 and ρ' the density of partial dislocations whose ρ' value is twice that of the perfect dislocation's ρ . $\bar{r}_1$ is expected to be much smaller than $\bar{r}$. The effect of partial dislocations would be neglected henceforth.

Plastic deformation produces the different atomic environment. A and B atoms exchange their sites with each other along the APB. Figure 2 shows the atomic arrangement around the APB of the $L1_2$ -type structure, in which the $(a/2)[\bar{1}10]$ dislocation slips over the (111) glide plane and places the APB there. A atoms exchange their sites with B atoms in both sides of the APB and B atoms on one side of the APB (at the corner sites in Fig. 2) become irregular for A atoms in the other side. On the (111) APB, there are two kinds of A atoms with different atomic environment. The first kind of A atoms

FIG. 2. Atomic arrangement of the L_{12} -type structure after the $(a/2)[\bar{1}10]$ dislocation slips over the (111) glide plane. The APB distributes over the (111) glide plane.

TABLE II. Probabilities of atom pairs at the first NN distance in three kinds of atomic states for the A_3B alloys with the L_{12} -type superstructure.

$i-j$ atom pairs	Probabilities of $i-j$ atom pairs		
	Ordered state	APB	Disordered state
A-A	1/2	25/48	9/16
A-B	1/2	11/24	3/8
B-B	0	1/48	1/16

P_{ij} depends on the alloy composition c and the long-range order parameter S : for instance, $P_{AA} = (9 - S^2)/16$, $P_{AB} = (3 + S^2)/8$ and $P_{BB} = (1 - S^2)/16$ in the ordered A_3B alloy with S , whereas $P_{AA} = (1 - c)^2$, $P_{AB} = 2c(1 - c)$, and $P_{BB} = c^2$ in the perfectly disordered $A_{1-c}B_c$ alloy. Table II shows the probabilities of three kinds of atom pairs at the first NN on the APB, in the perfectly ordered and disordered states of the L_{12} -type superstructure. This table shows that the atomic arrangement on the APB stands in a transitional state from the ordered to disordered state.

With plastic deformation advancing, the number of A-A atom pairs increases at the same rate as that of B-B atom pairs, whereas the number of A-B atom pairs decreases at the twofold rate of the case of B-B atom pairs. Since the total number of atom pairs per unit volume is $n_0 = 24/a^3$, the P_{ij} values in the A_3B crystal after plastic deformation are expressed by

$$P_{AA} = \frac{9 - S^2}{16} + \frac{n}{n_0}, \quad (6)$$

$$P_{AB} = \frac{3 + S^2}{8} - \frac{2n}{n_0}, \quad (7)$$

and

$$P_{BB} = \frac{1 - S^2}{16} + \frac{n}{n_0}. \quad (8)$$

Before plastic deformation, the magnetic transition temperature of fully ordered A_3B alloy and/or compound is written as

$$\theta_m(O) = \frac{T_{AA} + T_{AB}}{2}. \quad (9)$$

On the other hand, the magnetic transition temperature in disordered state $\theta_m(D)$ is given by

$$\theta_m(D) = \frac{9}{16}T_{AA} + \frac{3}{8}T_{AB} + \frac{1}{16}T_{BB}. \quad (10)$$

The application of cold work makes APB over the glide plane, across which the atoms exchange their own sites with each other (Table I). With plastic deformation advancing, the probabilities, P_{ij} ($i, j = A$ or B) change according to Eqs. (6)–(8). Thus, the magnetic transition temperature of the plastically deformed A_3B alloy and/or compound will be expressed by

$$\theta_m = S^2\theta_m(O) + (1 - S^2)\theta_m(D) + \frac{S^2 a \bar{r}}{3\sqrt{3}} [\theta_m(D) - \theta_m(O)]. \quad (11)$$

As described in the Sec. II, the $\bar{r}\rho$ is rewritten as $\bar{r}\rho = r_0\rho$ at the early stage of plastic deformation and as

occupy the F sites (at the face-centered sites in Fig. 2), where they have the same number of the A and B atoms as those of the normal state in their each NN; although, the arrangement of the atoms around the host A atoms changes in the first, third, and fifth NN (Table I). The second kind of A atoms occupy the C sites (the corner site in Fig. 2) and have more neighboring A atoms than those of the normal state in the first, third, and fifth NN but the B atoms appear in the second, fourth, and sixth NN of the host A atoms at the C sites (Table I). The probability that the A atoms occupy the C sites is 1/3.

III. MAGNETIC TRANSITION TEMPERATURE AND PLASTIC DEFORMATION

There are many alloys with the L_{12} -type structure which show the different magnetic behaviors in the chemically ordered and disordered states. The neighboring environments of each atom are different in these states. The magnetic interactions between atoms are intimately related with the atomic environment. The Pt_3Fe alloy, for instance, is antiferromagnetic in the fully ordered state, while the coexistence of ferromagnetic and antiferromagnetic states is observed in the disordered state.^{6,7} As another example, the Ni_3Mn alloy is known: the Curie temperature of this L_{12} -type alloy is remarkably reduced by disordering,¹³ suggesting the existence of competing ferromagnetic and antiferromagnetic interactions in the disordered state.¹⁴

The magnetic couplings change by plastic deformation and then the magnetic transition temperature θ_m will be influenced. For simplicity, it is assumed that θ_m in the plastically deformed alloy is determined by the pair-interaction model. This model was adopted for the study on the Curie temperatures in the ordered and disordered Fe_3Pt (Ref. 11) and disordered Ni-Mn alloys.¹² According to this model, θ_m is represented by the sum of first NN pair interactions: i.e.,

$$\theta_m = P_{AA}T_{AA} + P_{AB}T_{AB} + P_{BB}T_{BB}, \quad (5)$$

where P_{ij} ($i, j = A$ or B) means the relative number of $i-j$ atom pairs and T_{ij} the pair interaction parameter being proportional to the interaction energy ($-2J_{ij}S_iS_j$) of first NN $i-j$ atom pairs.

$\bar{\rho} = \sqrt{\rho}$ at the advanced stage. Then, in the advanced stage of plastic deformation where the *b*-type superlattice dislocations are the main distributors, θ_m of the $L1_2$ -type ordered alloy, when $S = 1$, is expressed by

$$\theta_m = \theta_m(O) + \frac{a[\theta_m(D) - \theta_m(O)]}{3\sqrt{3}}\sqrt{\rho}. \quad (12)$$

Malmström *et al.*¹⁵ have theoretically shown that the magnetic interaction between Fe or Mn atoms with the first NN distance in alloys dominates over those with other distances. Therefore, the pair-interaction model, which gave the present formulation, would be reasonable so far as the deformation advances homogeneously. Lastly, the numerical estimation for some $L1_2$ -type ordered alloys is carried out by using the experimental values of a : $\theta_m(O)$ and $\theta_m(D)$.

A. Pt_3Fe

The fully ordered Pt_3Fe alloy is antiferromagnetic with the Néel temperature of $T_N = 90$ K (Ref. 7). Figure 3 shows the magnetic structure of this alloy determined by the neutron diffraction measurements, where the magnetic interaction in the second NN Fe-Fe atom pairs is antiferromagnetic and Pt atoms have no magnetic moment. In the plastically deformed alloy, there appear Fe-Fe atom pairs at the first NN across the APB over the glide plane. These Fe-Fe atom pairs bring about the strong ferromagnetism. According to Bacon and Crangle,⁷ the cold-worked powder specimen with the stoichiometric component becomes ferromagnetic; the saturation magnetization extrapolated to 0 K is $M(0) = 640$ emu/cm³; the Curie temperature is 425 K. X-ray examination also showed that the superlattice reflections were not observed in the powdered specimen. And the annealing not only restored the superlattice reflection but also removed the ferromagnetism.

If $\theta_m(O) = -90$ K and $\theta_m(D) = 425$ K are adopted on the basis of their experimental results,^{6,7} θ_m of the plastically deformed Pt_3Fe alloy is shown as a function of ρ over the range between 10^{11} and 10^{14} cm⁻² by using Eq. (12). Here, $a = 3.864$ Å (Ref. 16) is adopted. This relation is illustrated

FIG. 3. Atomic arrangement and magnetic structures for the ideal stoichiometric Pt_3Fe alloy. Open circles represent iron atoms and shaded circles represent platinum atoms.

FIG. 4. Relation between the magnetic transition temperature θ_m and the dislocation density ρ in the plastically deformed Pt_3Fe alloy. It is estimated for the advanced stage of plastic deformation where the *b*-type superlattice dislocations are the main distributors.

for the *b*-type superlattice dislocations, because this range of ρ corresponds to the advanced stage of plastic deformation (Fig. 4). If this analysis is extended to the higher region of ρ , the critical value of ρ where the long-range magnetic order transfers from antiferro- to ferromagnetism (i.e., θ_m becomes zero) is 5.5×10^{14} cm⁻². We assume that the magnitude of magnetic moment of Fe and Pt atoms at their "irregular" sites which appear along the APB is the same as that at their regular sites [i.e., $\mu_{Fe} = 3.3 \mu_B$ and $\mu_{Pt} = 0.0 \mu_B$ (Ref. 7)]. Then the magnetization for the critical value of ρ will be $M(0) = 185$ emu/cm³, after the relation of the magnetization in the plastically deformed Pt_3Fe alloy: $M(0) = 4n\mu_{Fe}/3$ (Ref. 3).

B. Pt_3Co and Fe_3Pt

The Pt_3Co alloy is ferromagnetic in both the ordered and disordered states. According to the polarized neutron investigation on this alloy,¹⁷ the magnetic moment per unit cell is $2.43 \mu_B$ and $2.65 \mu_B$ in the ordered and disordered states, respectively. Simpson and Tredgold¹⁸ have found that the chemical order-disorder transition makes the Curie temperature change considerably. These results suggest that the interatomic exchange interactions are important in this alloy.

FIG. 5. Relation between the magnetic transition temperature θ_m and the dislocation density ρ in the plastically deformed Pt_3Co alloy. The *b*-type superlattice dislocations are the main distributors.

FIG. 6. Relation between the magnetic transition temperature θ_m the dislocation density ρ in the plastically deformed Fe_3Pt alloy. The b -type superlattice dislocations are the main distributors.

It was found⁸ in the ordered Pt_3Co alloy that the disordering due to plastic deformation causes the magnetization to enhance and the increase of magnetization was roughly proportional to the amount of plastic deformation. By plastic deformation, the area of APB increases over the glide plane and each atom exchanges its site with each other near the APB. According to Eq. (12), θ_m in the plastically deformed Pt_3Co alloy is calculated as a function of the dislocation density over the range between 10^{11} and 10^{14} cm^{-2} . The results are shown in Fig. 5, where $a = 3.831 \text{ \AA}$ (Ref. 19) and the Curie temperatures in the ordered and disordered states are $\theta_m(O) = 285 \text{ K}$ and $\theta_m(D) = 482 \text{ K}$, respectively.¹⁸

The Fe_3Pt alloy also is ferromagnetic in both the ordered and disordered states. Mizoguchi *et al.*¹¹ measured the Curie temperatures of the ordered and disordered $\text{Fe}_{3-x}\text{Pt}_{1+x}$ alloys with $x = 0, 0.1$, and 0.2 , and discussed the atomic ordering effect on T_C in terms of the pair-interaction model. According to their analyses based on Eq. (5), the pair-interaction parameters of $\text{Fe}_{3-x}\text{Pt}_{1+x}$ alloys are $T_{AA} = -705 \text{ K}$, $T_{AB} = 1567 \text{ K}$, and $T_{BB} = 1214 \text{ K}$. Thus, a considerable decrease of the magnetic transition temperature caused by plastic deformation will be expected also in the Fe_3Pt alloy with $\theta_m(O) = 431 \text{ K}$ and $\theta_m(D) = 267 \text{ K}$. Figure 6 shows the calculated results of θ_m from Eq. (12) for the Fe_3Pt alloy with the L1_2 -type superstructure, where $a = 3.74 \text{ \AA}$ (Ref. 20) is adopted.

C. Ni_3Mn

The Mn-Mn coupling has been investigated by many investigators in a Ni_3Mn alloy.^{13,14,16} Mn atoms coupled ferromagnetically at the second NN distance in the ordered state. On the other hand, the neutron diffraction²¹ and NMR²² studies on disordered Ni-Mn alloys have shown that there is a complicated state resulting from competing magnetic interactions, which may explain the observed spin-glass-like properties such as displaced hysteresis loops.²³ There are positive exchange interactions in the Ni-Ni or Ni-Mn atom pairs at the first NN distance, whereas the exchange interaction in the first NN Mn-Mn atom pairs is very strongly negative.^{12,24,25} In the fully ordered Ni_3Mn alloy, there are no second NN Mn-Mn atom pairs and no Mn atoms having the magnetic moment of the antiparallel direc-

FIG. 7. Relation between the magnetic transition temperature θ_m and the dislocation density ρ in the plastically deformed Ni_3Mn alloy. The b -type superlattice dislocations are the main distributors.

tion to the magnetization. After plastic deformation, Mn-Mn atom pairs appear at the first NN across the APB over the glide plane (Table I).

It was found that the saturation magnetization decreases with increasing plastic deformation. The decrease of magnetization during plastic deformation was understood with consideration of Mn-Mn couplings.^{5,8} The chemical order-disorder transition markedly changes the Curie temperature in this alloy. T_C of fully ordered Ni_3Mn alloy is 750 K (Ref. 26), whereas it lowers to 132 K in the disordered state.¹³ In Fig. 7, the magnetic transition temperature of Ni_3Mn alloy is shown versus the dislocation density over the range 10^{11} – 10^{14} cm^{-2} , where $a = 3.59 \text{ \AA}$ (Ref. 27) is adopted and the b -type superlattice dislocations are main distributors.

IV. CONCLUSION

The influence of plastic deformation on the magnetic properties has not been studied in the long-range ordered materials with a few exceptions. And the effect of plastic deformation have been avoided by annealing as the complicated phenomena. But it may be possible to study the magnetic localized phenomena in the new aspects by use of the modified slip-induced directional-order model.

The change of the magnetic transition temperature by plastic deformation is proportional to the dislocation density at the early stage of plastic deformation and to the square root of it at the advanced stage in the L1_2 -type materials. Moreover, its change is predicted to be proportional to the difference between the magnetic transition temperatures in completely ordered and disordered states. The present results might also be useful for the estimation of the dislocation density in the cold-worked specimen.

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