

STATISTICAL THEORY ON FORMATION OF FERROMAGNETIC SUPERLATTICE $\text{FeCr}(\alpha'_{1-1})$ IN IRON-CHROMIUM SYSTEM*

YOSHIRO IWAMA and SHUZO TAKEDA

Department of Metallurgy

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I. Introduction

It was discovered by the present authors and N. Nagai¹⁾²⁾ that there are formed, in Fe-Cr system, three ferromagnetic superlattices, $\text{Fe}_3\text{Cr}(\alpha'_{3-1})$, $\text{FeCr}(\alpha'_{1-1})$ and $\text{FeCr}_3(\alpha'_{1-3})$, as well as two meta-compounds, Fe_2Cr (τ -phase) and FeCr_2 (ω -phase), and also that these superlattices show various interesting behaviors caused by an overlapping of two co-operative phenomena, ordering and ferromagnetism. In this article, they have dealt theoretically with the statistics on formation of superlattice of $\text{FeCr}(\alpha'_{1-1})$ type, in particular, one of these superlattices, from the similar idea as was adopted by H. Sato³⁾ to general theory about ferromagnetic superlattices. The results obtained here agree fairly well with the experimental data which were reported in their last papers.¹⁾²⁾

II. Theoretical Treatment in General

The ordered structure of superlattice $\text{FeCr}(\alpha'_{1-1})$ was also elucidated, by one of the present authors and N. Nagai to be based upon a large unit cell, as shown in Fig. 1-(a). Let us suppose that this large unit cell consists of two kinds of atoms, A and B , whose atomic concentrations are expressed by θ and $1 - \theta$, respectively. This structure, constructed by 8 body-centered cubic unit cells, is considered to be made up by interpenetrating of two similar and mutually symmetrical sub-lattices, I and II , one of which is shown in Fig. 1-(b). If the sub-lattice I is filled up with only A or B atoms in the system of equiatomic composition AB , a perfectly ordered state is established.

On carrying out the statistical calculation, Bragg-Williams approximation was applied to order-disorder transformation, as well as Weiss approximation to ferromagnetism; these approximations are rough and in same order so that the computation becomes considerably easy. The complexity of the unit cell, however, obliges us to take account of the second nearest neighbor in addition to the direct neighbor. This fact is fairly significant, for, in a body-centered cubic structure, the atomic distance between the second nearest neighbors is not very larger than that between the direct neighbors. Furthermore, let us assume that the fraction of spins which are parallel to inner magnetic field and that of anti-parallel spins are independent of the sorts of atoms to which they belong; let these fractions be ρ and $1 - \rho$, respectively.

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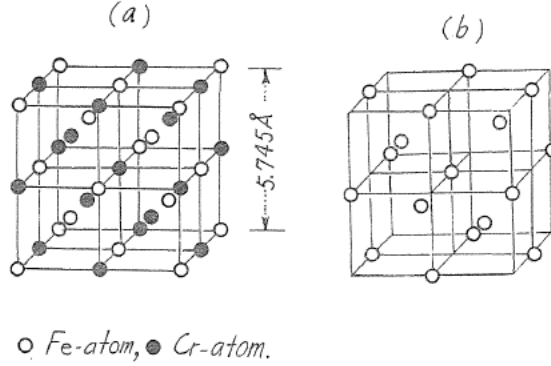


FIG. 1. (a) Ordered FeCr structure after Takeda and Nagai, (b) I-sites.

Now, let $\left[\frac{A_+}{I} \right]$, say, be the number of A atoms which are on I -sites and also have parallel spins. Then the following expressions are easily obtained from the definitions of s and ρ :

$$\left. \begin{aligned} \left[\frac{A_+}{I} \right] &= \frac{N}{2} \theta(1+s)\rho, & \left[\frac{B_+}{I} \right] &= \frac{N}{2} \{1-\theta(1+s)\}\rho, \\ \left[\frac{A_+}{II} \right] &= \frac{N}{2} \theta(1-s)\rho, & \left[\frac{B_+}{II} \right] &= \frac{N}{2} \{1-\theta(1-s)\}\rho, \\ \left[\frac{A_-}{I} \right] &= \frac{N}{2} \theta(1+s)(1-\rho), & \left[\frac{B_-}{I} \right] &= \frac{N}{2} \{1-\theta(1+s)\}(1-\rho), \\ \left[\frac{A_-}{II} \right] &= \frac{N}{2} \theta(1-s)(1-\rho), & \left[\frac{B_-}{II} \right] &= \frac{N}{2} \{1-\theta(1-s)\}(1-\rho), \end{aligned} \right\} \quad (1)$$

where N is the total number of atoms in this system, and s is the degree of long range order. The total number of atomic arrangements, $G(s, \rho)$, associated with given s and ρ , is given by

$$G(s, \rho) = \frac{(N/2)!}{\left[\frac{A_+}{I} \right]! \left[\frac{A_-}{I} \right]! \left[\frac{B_+}{I} \right]! \left[\frac{B_-}{I} \right]!} \cdot \frac{(N/2)!}{\left[\frac{A_+}{II} \right]! \left[\frac{A_-}{II} \right]! \left[\frac{B_+}{II} \right]! \left[\frac{B_-}{II} \right]!} \quad (2)$$

By means of Eqs. (1) and Stirling's formula, the logarithmic expression for Eq. (2) becomes as follows:

$$\begin{aligned} -\log G(s, \rho) &= \frac{N}{2} [\theta(1+s) \log \theta(1+s) + \theta(1-s) \log \theta(1-s) \\ &\quad + \{1-\theta(1+s)\} \log \{1-\theta(1+s)\} + \{1-\theta(1-s)\} \log \{1-\theta(1-s)\}] \\ &\quad + N\{\rho \log \rho + (1-\rho) \log (1-\rho)\}. \end{aligned} \quad (3)$$

Let the potential energy coefficients of atomic interactions between direct neighbors be $-V_{AA}$, $-V_{BB}$, $-V_{AB}$, and those of exchange interactions between parallel spins be $-J_{AA}$, $-J_{BB}$, $-J_{AB}$, where suffixes indicate the sorts of atoms

which form the interaction pair, and energy coefficients of exchange interactions between parallel and anti-parallel spins are defined to be zero. As for the second nearest neighbors, the coefficients are distinguished by priming. Furthermore, let the coordination number for the direct neighbors be $2z$, and that for the second nearest neighbors be z' . Then, $2z=8$ and $z'=6$, in the atomic configuration of FeCr above mentioned. And one half, z , of the direct neighbors to an atom on I -site lie on I -sites, another half, z , lie on II -sites, and the whole second nearest neighbors, z' , are on II -sites. It is the same as for neighbors to an atom on II -site.

Thus, the total energy $E(s, \rho)$ of the system for given s and ρ is written as follows:

$$\begin{aligned}
 E(s, \rho) = & - \left\{ \left[\frac{A_+}{I} \frac{A_+}{I} \right] + \left[\frac{A_+}{I} \frac{A_+}{II} \right] + \left[\frac{A_-}{I} \frac{A_-}{I} \right] + \left[\frac{A_-}{I} \frac{A_-}{II} \right] \right. \\
 & + \left. \left[\frac{A_+}{II} \frac{A_+}{II} \right] + \left[\frac{A_-}{II} \frac{A_-}{II} \right] \right\} (V_{AA} + J_{AA}) \\
 & - \left\{ \left[\frac{A_+}{I} \frac{A_-}{I} \right] + \left[\frac{A_+}{I} \frac{A_-}{II} \right] + \left[\frac{A_-}{I} \frac{A_+}{II} \right] + \left[\frac{A_+}{II} \frac{A_-}{II} \right] \right\} \cdot V_{AA} \\
 & - \left\{ \left[\frac{B_+}{I} \frac{B_+}{I} \right] + \left[\frac{B_+}{I} \frac{B_+}{II} \right] + \left[\frac{B_-}{I} \frac{B_-}{I} \right] + \left[\frac{B_-}{I} \frac{B_-}{II} \right] \right. \\
 & + \left. \left[\frac{B_+}{II} \frac{B_+}{II} \right] + \left[\frac{B_-}{II} \frac{B_-}{II} \right] \right\} (V_{BB} + J_{BB}) \\
 & - \left\{ \left[\frac{B_+}{I} \frac{B_-}{I} \right] + \left[\frac{B_+}{I} \frac{B_-}{II} \right] + \left[\frac{B_-}{I} \frac{B_+}{II} \right] + \left[\frac{B_+}{II} \frac{B_-}{II} \right] \right\} \cdot V_{BB} \\
 & - \left\{ \left[\frac{A_+}{I} \frac{B_+}{I} \right] + \left[\frac{A_+}{I} \frac{B_+}{II} \right] + \left[\frac{A_-}{I} \frac{B_-}{I} \right] + \left[\frac{A_-}{I} \frac{B_-}{II} \right] + \left[\frac{A_+}{II} \frac{B_+}{I} \right] \right. \\
 & + \left. \left[\frac{A_+}{II} \frac{B_+}{II} \right] + \left[\frac{A_-}{II} \frac{B_-}{I} \right] + \left[\frac{A_-}{II} \frac{B_-}{II} \right] \right\} (V_{AB} + J_{AB}) \\
 & - \left\{ \left[\frac{A_+}{I} \frac{B_-}{I} \right] + \left[\frac{A_+}{I} \frac{B_-}{II} \right] + \left[\frac{A_-}{I} \frac{B_+}{I} \right] + \left[\frac{A_-}{I} \frac{B_+}{II} \right] + \left[\frac{A_+}{II} \frac{B_-}{I} \right] \right. \\
 & + \left. \left[\frac{A_+}{II} \frac{B_-}{I} \right] + \left[\frac{A_-}{II} \frac{B_+}{I} \right] + \left[\frac{A_-}{II} \frac{B_+}{II} \right] \right\} \cdot V_{AB} \\
 & - \left\{ \left[\frac{A_+}{I} \frac{A_+}{II} \right]' + \left[\frac{A_-}{I} \frac{A_-}{II} \right]' \right\} (V'_{AA} + J'_{AA}) - \left\{ \left[\frac{A_+}{I} \frac{A_-}{II} \right]' + \left[\frac{A_-}{I} \frac{A_+}{II} \right]' \right\} \cdot V'_{AA} \\
 & - \left\{ \left[\frac{B_+}{I} \frac{B_+}{II} \right]' + \left[\frac{B_-}{I} \frac{B_-}{II} \right]' \right\} (V'_{BB} + J'_{BB}) - \left\{ \left[\frac{B_+}{I} \frac{B_-}{II} \right]' + \left[\frac{B_-}{I} \frac{B_+}{II} \right]' \right\} \cdot V'_{BB} \\
 & - \left\{ \left[\frac{A_+}{I} \frac{B_+}{II} \right]' + \left[\frac{A_-}{I} \frac{B_-}{II} \right]' + \left[\frac{A_+}{II} \frac{B_+}{I} \right]' + \left[\frac{A_-}{II} \frac{B_-}{I} \right]' \right\} (V'_{AB} + J'_{AB}) \\
 & - \left\{ \left[\frac{A_+}{I} \frac{B_-}{II} \right]' + \left[\frac{A_-}{I} \frac{B_+}{II} \right]' + \left[\frac{A_+}{II} \frac{B_-}{I} \right]' + \left[\frac{A_-}{II} \frac{B_+}{I} \right]' \right\} \cdot V'_{AB},
 \end{aligned}$$

where $\left[\frac{A_+}{I} \frac{B_-}{II} \right]$, etc. are the total numbers of such atomic pairs in direct neighbors that are formed between A atoms, lying on I -sites and having parallel spins, and B atoms on II -sites having anti-parallel spins, etc., and the primed notations are for those in the second nearest neighbors.

Taking Eqs. (1) in the above equation, we obtain

$$\begin{aligned}
 E(s, \rho) = & -Nz\{\theta^2 V_{AA} + (1-\theta)^2 V_{BB} + 2\theta(1-\theta)V_{AB}\} \\
 & - \frac{Nz'}{2}\{\theta^2 V'_{AA} + (1-\theta)^2 V'_{BB} + 2\theta(1-\theta)V'_{AB}\} \\
 & - \left[Nz\{\theta^2 J_{AA} + (1-\theta)^2 J_{BB} + 2\theta(1-\theta)J_{AB}\} \right. \\
 & \left. + \frac{Nz'}{2}\{\theta^2 J'_{AA} + (1-\theta)^2 J'_{BB} + 2\theta(1-\theta)J'_{AB}\} \right] \{\rho^2 + (1-\rho)^2\} \\
 & - \frac{Nz'}{2}\theta^2 w' s^2 - \frac{Nz'}{2}\theta^2 \alpha' s^2 \{\rho^2 + (1-\rho)^2\},
 \end{aligned}$$

where $w' = 2V'_{AB} - (V'_{AA} + V'_{BB})$, and $\alpha' = 2J'_{AB} - (J'_{AA} + J'_{BB})$.

By setting $\rho = (1+m)/2$, or $\rho^2 + (1-\rho)^2 = (1+m^2)/2$, where the introduced variable m corresponds to the relative magnetization of this system, we obtain more simplified expression as follows:

$$\begin{aligned}
 E(s, m) = & E(0, 0) - N\left\{\left(\frac{z}{2}J_{AA} + \frac{z'}{4}J'_{AA}\right)\theta^2 \right. \\
 & \left. + \left(\frac{z}{2}J_{BB} + \frac{z'}{4}J'_{BB}\right)(1-\theta)^2 + 2\left(\frac{z}{2}J_{AB} + \frac{z'}{4}J'_{AB}\right)\theta(1-\theta)\right\}m^2 \\
 & - N\left\{\frac{z'}{2}\theta^2 w' + \frac{z'}{4}\theta^2 \alpha'\right\}s^2 - N\frac{z'}{4}\theta^2 \alpha' s^2 m^2,
 \end{aligned} \quad (4)$$

where $E(0, 0)$ expresses the sum of the terms which imply neither the variable s nor m .

From Eqs. (3) and (4), the free energy of this system is written by

$$F(s, m) = E(s, m) - kT \log G(s, m), \quad (5)$$

where k is Boltzmann's constant.

For given temperature T and concentration θ , the stable state will correspond to the minimum value of free energy. So, from $\frac{\partial F}{\partial s} = 0$ and $\frac{\partial F}{\partial m} = 0$, we can derive the equilibrium values of the degree of order s and the relative magnetization m . From these conditions,

$$z'\theta(2w' + \alpha' + \alpha'm^2)s = kT \left\{ \log \frac{1+s}{1-s} + \log \frac{1-\theta(1-s)}{1-\theta(1+s)} \right\}, \quad (6)$$

and

$$\begin{aligned}
 & \{(2zJ_{AA} + z'J'_{AA})\theta^2 + (2zJ_{BB} + z'J'_{BB})(1-\theta)^2 \\
 & + 2(2zJ_{AB} + z'J'_{AB})\theta(1-\theta) + z'\theta^2 \alpha' s^2\}m = kT \log \frac{1+m}{1-m}.
 \end{aligned} \quad (7)$$

III. Determination of Parameters

It is necessary, first of all, for solving the simultaneous equations (6) and (7), to give suitable numerical values to the implied parameters. If the system is

heated or cooled considerably rapidly, the degree of order s will be unable to follow up the equilibrium value for each temperature owing to the retardation of atomic movement, while the magnetization m will be not so because the reversal of spins will take place instantly. Under such condition, therefore, the variable s in Eq. (7), which is related to the magnetization change, may be assumed to be nearly constant. Then, the ferromagnetic Curie points θ , which would be measured at such experiments, are given by

$$\theta = \frac{1}{2k} \{ (2zJ_{AA} + z'J'_{AA})\theta^2 + (2zJ_{BB} + z'J'_{BB})(1-\theta)^2 + 2(2zJ_{AB} + z'J'_{AB})\theta(1-\theta) + z'\theta^2\alpha's^2 \}, \quad (8)$$

where s is the degree of order of specimen before heating. The fact that Eq. (8) expresses a parabola in θ - θ diagram agrees well with the experimental results; that is, Fe-Cr alloys of various compositions quenched in water from $1,000^\circ$ show their ferromagnetic Curie points on rapid heating, which lie on a parabola in θ - θ diagram, as shown in Fig. 2. Moreover, let us assume that during rapid heating such as $5^\circ/\text{min.}$, the change of atomic ordering occurs little, and so the degree of order of alloys remains unchanged in such experiments. By comparing the experimental data, obtained on alloys of various compositions perfectly annealed or quenched, with the calculated values derived from Eq. (8), where we put $s=1$ or 0, and $z=4$, $z'=6$, we can obtain the following values of parameters:

$$4J_{AA} + 3J'_{AA} = -543k, \quad 4J_{BB} + 3J'_{BB} = 1,042k, \quad 4J_{AB} + 3J'_{AB} = 944k,$$

and

$$\alpha' = 2J'_{AB} - (J'_{AA} + J'_{BB}) = 320k. \quad (9)$$

In Fig. 2, three full lines denote the θ - θ curves for $s=0$, $s=0.45$ and $s=1$, respectively, calculated by using Eqs. (8) and (9). While, solid circles, crosses and open circles show ferromagnetic Curie points observed experimentally on alloys, quenched in water from $1,000^\circ$, slowly cooled in furnace from $1,200^\circ$ and perfectly annealed at about 500° , respectively; and hence they correspond to A_2 , A'_2 (intermediate) and A'_2 points, respectively. In this figure, the T_c points, obtained by measurements of specific heat and electric resistance at elevated temperatures as well as thermo-magnetic analysis, are also inserted for reference. Here, the Curie points of perfectly ordered phase, A'_2 points, lying at higher temperatures than their T_c points, correspond to the superheated ones, obtainable only through markedly rapid heating.

Though these calculations were carried out under many rough assumptions, their results agree well with the experimental ones, except within the composition range with more than about 60% Cr, in which meta-compound ω -phase or superlattice $\text{FeCr}_3(\alpha'_{1-3})$ is formed. It is also noticeable that the degree of order develops up to about $0.4 \sim 0.5$, even on furnace-cooled specimens; this fact suggests that the relaxation time of ordering in this system is comparatively small. It can be verified by the fact that specimens, even on cooling from higher temperature with a comparatively rapid rate, show somewhat dilatation due to ordering.

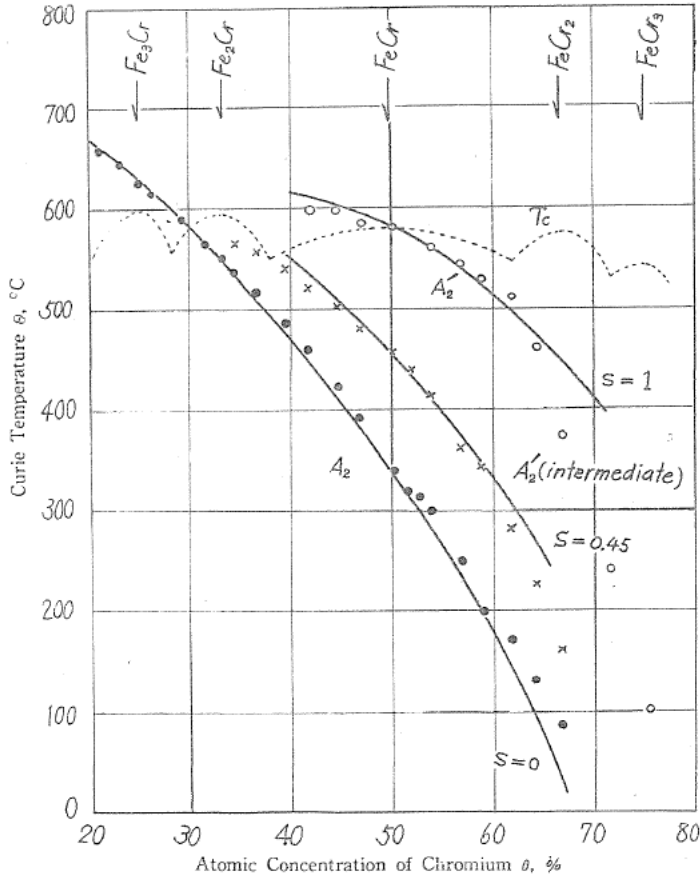


FIG. 2. θ - θ curves of Fe-Cr alloys.

Full lines show ferromagnetic Curie temperature curves calculated for $s=0$, $s=0.45$ and $s=1$, respectively. Solid circles, crosses and open circles are Curie points obtained experimentally on quenched, furnace-cooled and perfectly annealed specimens, respectively, by Takeda and Nagai. Dotted line shows T_c points observed experimentally also by them.

Besides, we adopted $w' = 407k$ as a parameter of ordering energy to fit the calculated results for the experimental data.

For examples, let us choose the cases of $\theta = 50\%$ and $\theta = 35\%$ in Eqs. (6) and (7), then

$$\left. \begin{aligned} (1,700 + 480m^2)s &= T \log \frac{1+s}{1-s}, \\ (1,194 + 480s^2)m &= T \log \frac{1+m}{1-m}, \end{aligned} \right\} \text{ for } \theta = 0.50,$$

and

$$\left. \begin{aligned} (2,382 + 672 m^2) s &= T \left\{ \log \frac{1+s}{1-s} + \log \frac{0.65 + 0.35 s}{0.65 - 0.35 s} \right\}, \\ (803 + 118 s^2) m &= T \log \frac{1+m}{1-m}, \end{aligned} \right\} \quad \text{for } \theta = 0.35.$$

These simultaneous equations can be easily solved graphically. The s - T and m - T curves, obtained by these calculations, for 50 at.% Cr and 35 at.% Cr compositions, are shown by full lines in Figs. 3-(a), (b) and 4-(a), (b), respectively. Furthermore, the abnormal change in specific heat accompanying the changes of s and m can be calculated by $\Delta C_p = \frac{1}{N} \frac{dE}{dT}$. The ΔC_p - T curves thus calculated are shown by full lines in Figs. 3-(c) and 4-(c), respectively.

IV. Comparison with Experiments

In Figs. 3-(b), (c) and 4-(b), (c), the m - T and ΔC_p - T curves observed by experiments are also inserted for comparison by dotted or broken lines, respectively, for alloys of the respective compositions. With these figures, we shall compare the results of calculations with those of experiments.

IV-1. The case of an alloy with 50 at.% Cr

From the calculated results (full-lined in Fig. 3-(a), (b)), it is seen that in equilibrium condition the degree of order of an alloy with 50 at.% Cr vanishes completely at 577°, its T_c point; while the vanishing of magnetization precedes at 432° prior to the former one. This preceded magnetic transformation brings out a sudden drop in degree of order, and hence, a knick on s - T curve (Fig. 3-(a)). Generally speaking, on such a ferromagnetic superlattice, a drop in degree of order s is accompanied by a decrease in magnetization m , and vice versa. Owing to such a mutual co-operation, therefore, it is characteristic that the behavior of the preceded vanishing of m , namely, the slope of m - T curve near Curie point, is much steeper than that of an ordinary ferromagnetic material such as iron. This fact could be actually proved by magnetic analysis for ordered alloys with 30~50% Cr.

As seen on dotted line in Fig. 3-(b), however, the observed m - T curve for this alloy, on heating with a rate of 2°/min., shows no preceded ferromagnetic Curie point prior to T_c point, but its Curie point at about 580° which is nearly equal to its T_c point. This discrepancy is considered to be attributed to the sluggishness of atomic movement in the temperature range from about 400 to 500°, below its T_c point; in fact, the atomic movement is so sluggish that s is almost unchangeable before its critical temperature T_c is approached, and thus this delays the decrease in m , accordingly giving rise to a superheating of its Curie point up to its T_c point, as shown in Fig. 2.

In Fig. 3-(c), the calculated ΔC_p - T curve (full-lined) shows two separated peaks corresponding to ferromagnetic Curie point and T_c point of this alloy, respectively. In the observed ΔC_p - T curve (dot-lined), however, these two peaks are united, showing one steep peak at about 500°, which can be attributed to the sluggishness of atomic diffusion, as above mentioned. Here, both the observed abnormal change in specific heat amounting to about 10 R/mol and the observed total transformation energy agree also well with the calculated ones.

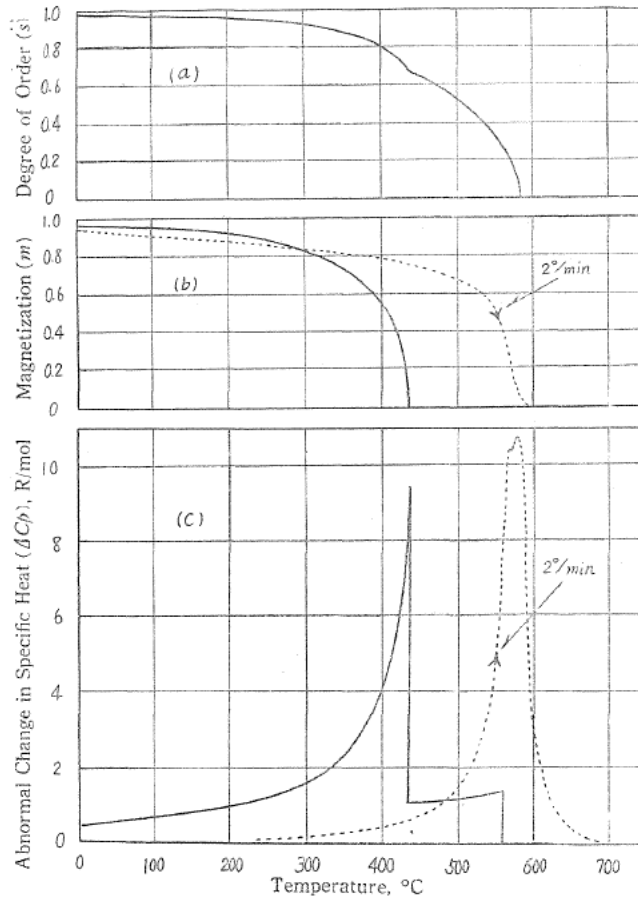


FIG. 3. Calculated (full-lined) and experimental (dot-lined) s - T (a), m - T (b) and ΔC_p - T (c) curves for an alloy with 50 at.% Cr.

IV-2. The case of an alloy with 35 at.% Cr

Our calculations, shown by full lines in Fig. 4-(a), (b) and (c) are carried out for an alloy with 35 at.% Cr, but the experimental m - T curves (dot- and broken-lined in Fig. 4-(b)) are for that with 35 wt.% Cr and the experimental ΔC_p - T curve (dot-lined in Fig. 4-(c)) is for that with 34 wt.% Cr, taken both from the work of one of the present authors and Nagai. Cr contents of the latter two specimens, used in the experiments, are not precisely equal to 35 at.% Cr referred in our calculation, but these slight discrepancies will be, in practice, negligible with regard to inaccuracy about the theoretical treatment and the experimental procedure.

Now, the calculated s - T curve drops precipitously down to zero at 515°, with which the m - T curve drops suddenly to 0.25 influenced by the drop in s and then m reaches zero at 530°. In this case, again, the retardation of atomic diffusion must be in question. Yet, the delay of ferromagnetic Curie point is not so striking for this alloy. Because its Curie point of perfectly ordered state is higher than its

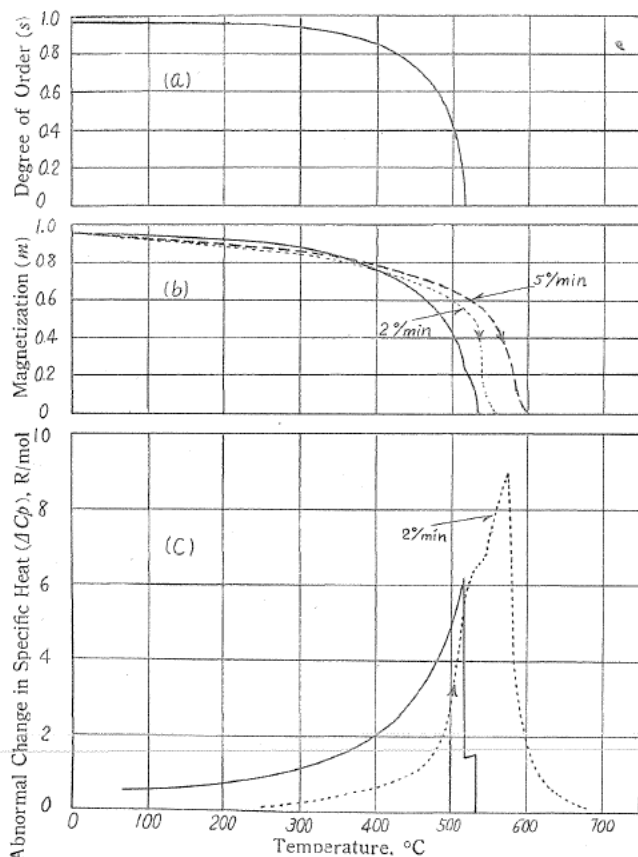


FIG. 4. Calculated s - T (a), m - T (b) and ΔC_p - T (c) curves for an alloy with 35 at.% Cr (full-lined), m - T curves obtained experimentally for an alloy with 35 wt.% Cr (dot- and broken-lined in (b)), and experimental ΔC_p - T curve for an alloy with 34 wt.% Cr (dot-lined in (c)).

T_c point, which is nearly equal to that of the alloy with 50 at.% Cr; so that the atomic rearrangement is sufficiently vigorous near its T_c point, before its Curie point is not reached. Thus, the observed m - T curve, measured with a slow heating rate of $2^\circ/\text{min.}$, agrees well with the theoretical curve, as seen on dotted line in Fig. 4-(b). However, being measured with a rapid heating rate of $5^\circ/\text{min.}$, its Curie point is found at higher temperature, 590° , due to the retardation of atomic diffusion, as seen on broken line in Fig. 4-(b).

Also as for ΔC_p - T curve, the calculated result agrees well with the experimental one, as seen in Fig. 4-(c), particularly from the view-point of two neighboring peaks. Though the observed peaks are removed to slightly higher temperature than in the theoretical curve, their agreement is considered to be sufficiently good, if the unfavorable experimental conditions and our rough assumptions are taken account of.

IV-3. The case of alloys with more than 50 at.% Cr

For alloys with more than 50 at.% Cr, their ferromagnetic Curie points are much lower than their T_c points, because with increasing Cr content, the former point is lowered more strikingly than the latter one. The theory predicts, therefore, that there appear two individually separated peaks on C_p - T curve, which correspond to Curie and T_c points, respectively. Fig. 5 shows the C_p - T curve for an alloy with 55 wt.% Cr, measured by one of the present authors and Nagai, which agrees well with the theoretical prediction. Because of the retardation of atomic diffusion, the peak at the lower temperature should be taken for the ferromagnetic Curie point of the ordered alloy; in fact, this temperature on the observed C_p - T curve is 520° in accordance with the Curie point theoretically obtained on the ordered alloy from Fig. 2.

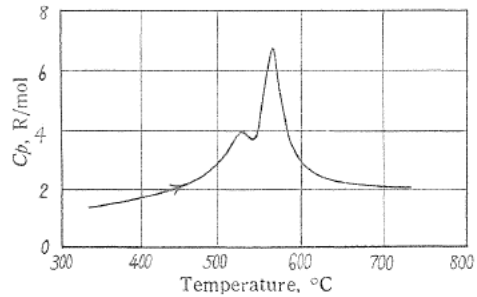


FIG. 5. C_p - T curve for an alloy with 55 wt.% Cr after Takeda and Nagai.

V. Summary

The results of the present research may be summarized as follows:—

Based on the experimental work of one of the present authors and N. Nagai on superlattices in Fe-Cr system, the statistical calculation for ferromagnetic superlattice $\text{FeCr}(\alpha'_{-1})$ was carried out. Bragg-Williams approximation and Weiss approximation were, according to H. Sato, applied to order-disorder transformation and ferromagnetism, respectively. The problem of large unit cell in this case was solved by taking account of the second nearest neighbors besides the direct neighbors. Of the parameters which were implied in the calculation, those for ferromagnetism were determined from the experimental data about ferromagnetic Curie points of various alloys under probable assumptions. The results obtained by calculation were compared with the experimental data in respect of m - T and ΔC_p - T curves. If the retardation of atomic diffusion during heating is considered, the theoretical predictions agree very well with the experimental results.

In conclusion, the present authors wish to express their hearty thanks to N. Nagai who offered them many detailed experimental data about Fe-Cr alloys. Their thanks are also due to the Grant of the Ministry of Education in Aid for Fundamental Scientific Research as well as the Hattori Public Service Foundation which gave them financial aid.

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