CORRELATION BETWEEN LATTICE CONSTANT AND MAGNETIC MOMENT IN 3d TRANSITION METAL ALLOYS

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ABSTRACT

An empirical relation between the lattice constant and the magnetic moment was found in binary solid solutions of 3*d* transition metals expressed as $a(x)=a_A(1-x)+ a_B x + C <|\mu|>$, where x is the atomic fraction, a_A, a_B and C are parameters, $<|\mu|>$ the average magnitude of atomic magnetic moments. It is shown that the equation holds for all possible combinations of 3*d* transition metals which form solid solutions over a considerably wide range of concentrations. The analysis of lattice constants at high temperatures leads to the conclusion that localized atomic moments are retained above *T*c in most 3d ferromagnetic alloys. The thermal expansion anomaly observed in the Invar alloy ($Fe_{65}Ni_{35}$) is explained as the result of collapse of localized moments above *T*c. The physical meanings of the parameters are discussed in terms of the atomic size. It is shown that expansion of the atomic size is caused by the formation of the localized moment.

INTRODUCTION

It is well known that 3d electrons, which are responsible for the magnetic moment of transition elements, partake in the cohesion of transition metals. Therefore, a correlation between the magnetic moment and the atomic distance may be expected. Little attention has been paid to this point so far. In this paper, an empirical relation between the lattice constant and the atomic magnetic moment of transition metal alloys is proposed. We shall show that the analysis of the lattice constant versus concentration curve (*L*-*C* curve) can provide information about the atomic magnetic moments in alloys and can serve as an experimental method to detect the existence of localized moments above the Curie temperature.

EMPIRICAL RELATION

The lattice constant of binary solid solutions varies linearly with atomic concentration in the first approximation (Vegard's law). When applied to metallic solid solutions, deviations from the law invariably appear. These deviations are caused by many factors. In the transition metal alloys, magnetic properties may be one of the important factors[1]. We have found an empirical relation between the lattice constant and the atomic moment[2]. The relation is given by a simple equation,

$$a(x)=a_A\cdot(1-x)+a_B\cdot x + C\langle|\mu|\rangle \qquad (1)$$

where $a(x)$ is the lattice constant of the solid solution $A_{1-x}B_x$, a_A, a_B and C are adjustable parameters and $\langle|\mu|\rangle$ the average magnitude of the atomic magnetic moments, which can be estimated according to the following rules. (i) For simple ferromagnetic alloys, $\langle|\mu|\rangle$ may be equated with μ_{s0}, the spontaneous atomic moment at 0 K. (ii) In some ferromagnetic alloys, which usually exhibit an exchange anisotropy and/or a rotational hysteresis loss, a part of atomic moments aligns antiferromagnetically to the bulk magnetization. In this case, $\langle|\mu|\rangle > \mu_{so}$. Modern experimental techniques such as neutron scattering and NMR may be helpful to estimate $\langle|\mu|\rangle$. (iii) It seems that $\langle|\mu|\rangle$ can be equated with the sublattice moment for antiferromagnetic alloys. However, the spin structure of antiferromagnetic disordered alloys is not so simple as that of antiferromagnetic insulators. They are occasionally lacking in a long range spin order and should rather be described as "spin glasses". Therefore, the sublattice moment determined by neutron diffraction is not always equal to $\langle|\mu|\rangle$. As for the case (ii), the estimation of $\langle|\mu|\rangle$ could be done with the help of neutron scattering or some other experiments. (iv) $\langle|\mu|\rangle =0$ for nonmagnetic alloys, namely Pauli paramagnetic alloys or alloys having no localized magnetic moments.

In any case, it is desirable to use the data at T = 0 K for both lattice constants and $\langle|\mu|\rangle$. Nevertheless, we are mostly concerned with lattice constants at room temperature (RT) because of a lack of data at T=0 K. Even in this case, better agreement is obtained by using the atomic moments at T=0 K for $\langle|\mu|\rangle$. However, if the alloy exhibits a thermal expansion anomaly below RT, we have to use the data at T=0 K for both $a(x)$ and $\langle|\mu|\rangle$. The effect of temperature on the lattice constant will be discussed later. We first demonstrate how well equation (1) holds for all possible combinations of 3d transition metals which make a solid solution over considerably wide range of concentrations.

ALLOYS OF FERROMAGNETIC METALS

bcc $Fe_{1-x}Co_x$: Fig. 1 shows the observed and the calculated lattice constants of bcc Fe-Co. As can be seen in the figure, the agreement is excellent. Moreover, it is worth noting a change of the lattice constant in the vicinity *x*=0.5 by atomic ordering. Generally speaking, the formation of a superlattice causes a shrinkage of the lattice. On the contrary, the lattice constant increases by atomic ordering in this case. This exceptional behavior may be explained as the result of increase of the magnetization due to atomic ordering.

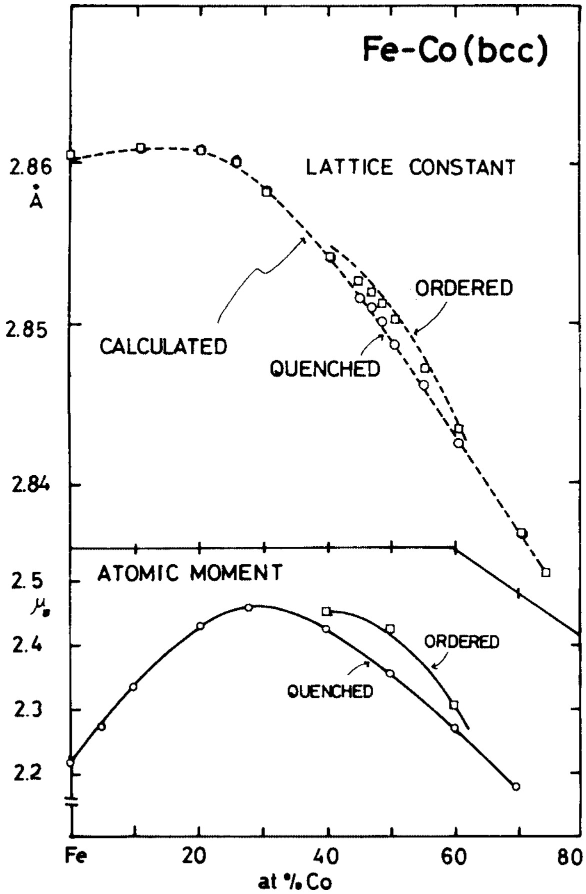

Fig. 1. The lattice constant of bcc Fe-Co at RT (Ref. 1, p.505).
---- represents calculated values with the parameters given in Table 1. <|μ|> =μ_{s0}[3] are also given.

fcc $Co_{1-x}Ni_x$: (Fig. 2) Both the lattice constant and the atomic moment varies linearly with concentration. In this case, however the parameters cannot be determined uniquely. It should be noted that the linear appearance of the *L-C* curve may be regarded as the evidence to show that contributions to the deviation from Vegard's law other than the magnetic one are negligible.

fcc $Fe_{1-x}Ni_x$: Since the alloys with $0.3 < x < 0.4$ have the thermal expansion anomaly known as the Invar anomaly, We have to use the lattice constant at 0 K. Fortunately, we can estimate it from the available data on the thermal expansion. The lattice constant at 0 K thus obtained is in good agreement with the calculated values as seen in Fig. 3. The deviation of the lattice constant from a straight line near the Invar region ($0.3 < x < 0.4$) can be explained as the result of the deviation of the magnetization from the Slater-Pauling curve.

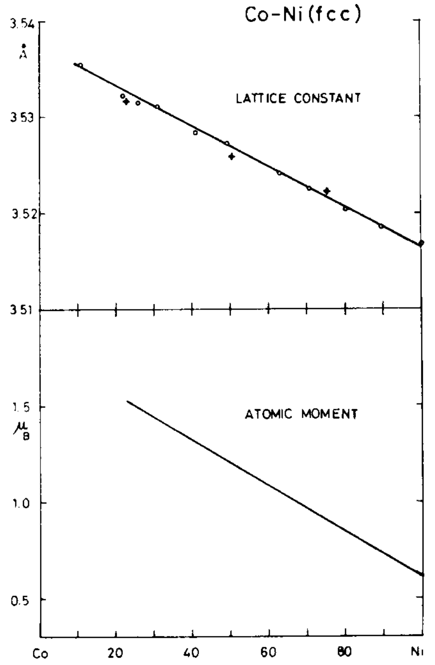

Fig. 2. The lattice constant at RT (Ref. 1, p.517) and the atomic moment at 0 K of fcc Co-Ni.

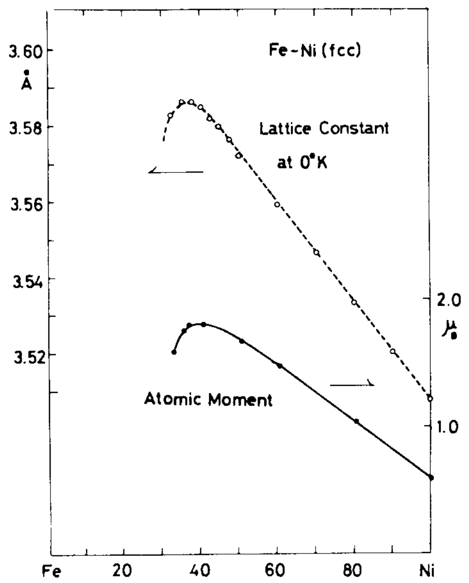

Fig. 3. The lattice constant[4)] and the atomic moments of fcc Fe-Ni at 0 K. ------ calculated value with the parameters given in Table 1.

ALLOYS OF NONFERROMAGNETIC METALS

bcc $V_{1-x}Cr_x$ and **bcc $Ti_{1-x}V_x$** : (Figs. 4 and 5) In these systems, one may assume $<|\mu|> = 0$, since they are Pauli paramagnetic over all concentrations. Therefore, the lattice constant is expected to obey Vegard's law. In fact, the observed lattice constant varies linearly with concentration within the scatter of the data points.

bcc $V_{1-x}Mn_x$: (Fig. 6) In the vanadium rich region, the lattice constant decreases linearly with x. This linear appearance breaks at about x=0.5 and the *L-C* curve deviates upwards for $x > 0.5$, which means a relative expansion of the lattice. On the other hand, the temperature dependence of the magnetic susceptibility is Pauli paramagnetic in the vanadium rich region. There are some experimental results indicating the formation of magnetic moment in the manganese rich region [6)] corresponding with the change of the slope in the *L-C* curve. From these observations, we may consider that $<|\mu|> = 0$ for $x < 0.5$ and $<|\mu|> \neq 0$ for $x > 0.5$. The change of the slope in the *L-C* curve can thus be explained on the basis of Eq. (1).

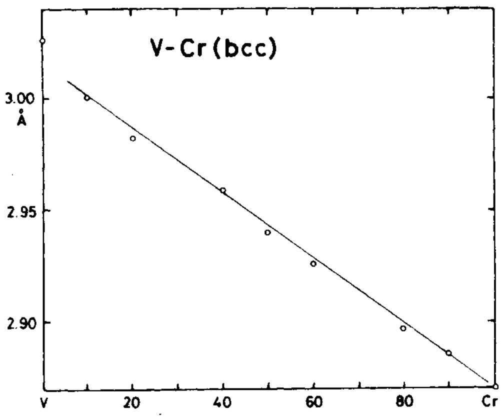

Fig. 4. The lattice constant at RT (Ref. 1, p.567) of bcc V-Cr.

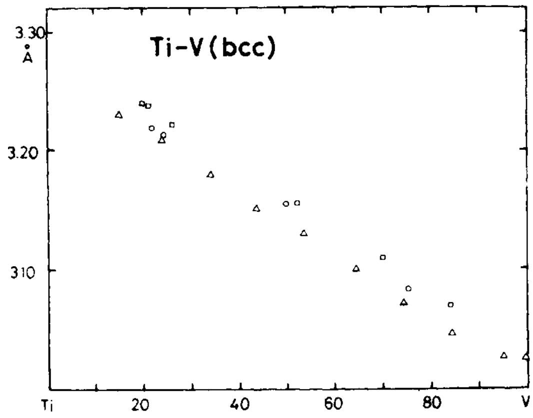

Fig. 5. The lattice constant of bcc Ti-V (Ref. 1, p. 875)

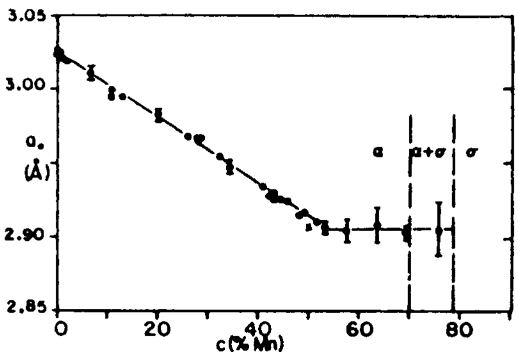

Fig. 6. The lattice constant of bcc V-Mn[6]

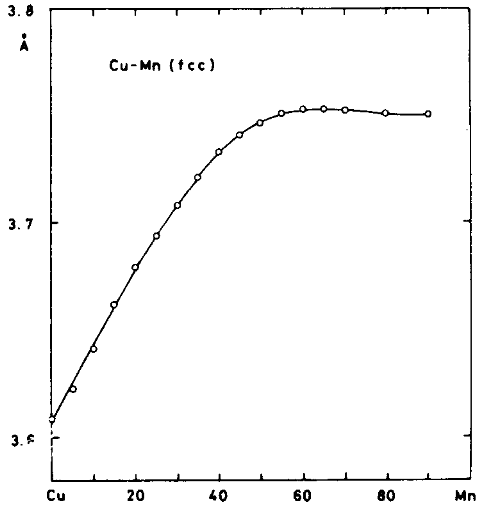

Fig. 7. The lattice constant of fcc Cu-Mn at RT (Ref. 1, p. 588).

fcc $Cu_{1-x}Mn_x$ (Fig. 7): Although intensive investigations have been done, the magnetic and the electronic structures are still not clear except at the two sides of the system. At the copper rich end, it is believed that Mn atoms have well defined localized moment with $S = 5/2$ or $\mu_{Mn} = 5\ \mu_B$. It has been revealed that Mn rich Mn-Cu alloys are antiferromagnetic with sublattice moment of about $2\mu_B$[7]. On the other hand, the lattice constant increases linearly with x in Cu rich region up to x= 0.4 and then the slope of the *L-C* curve becomes smaller and finally the lattice constant decreases with increasing *x*. Such a complicated behavior of the *L-C* curve might be explained by Eq. (1) as the result of the decrease of μ_{Mn} with increasing *x*.

ALLOYS OF FERROMAGNETIC AND NONFERROMAGNETIC METALS

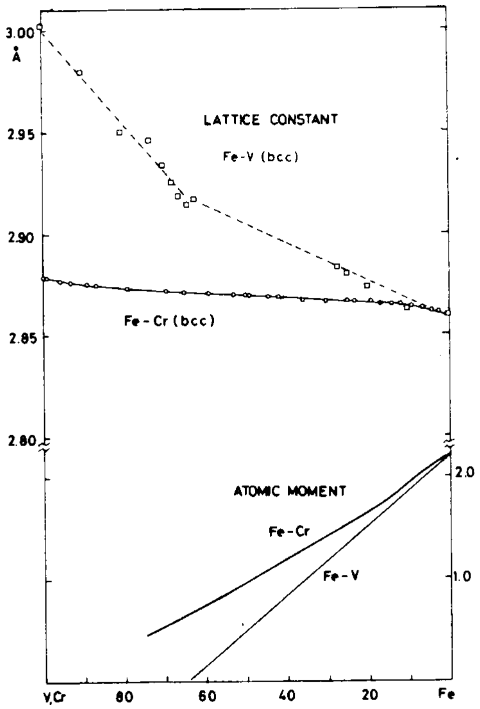

Fig. 8 The lattice constant of bcc Fe-V and bcc Fe-Cr at RT (Ref. 1, p 663 and p 533)
-------- Calculated value with the parameters given in Table 1.

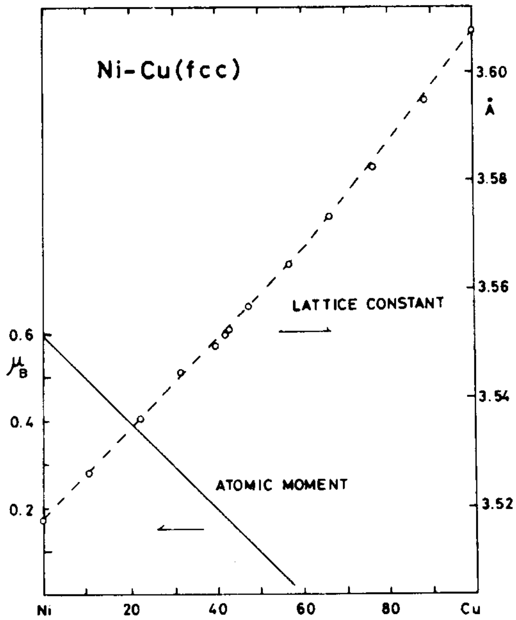

Fig. 9. The lattice constant at RT and the atomic moment at 0 K of fcc Ni-Cu.
------ calculated value with the parameters given in Table 1

bcc $Fe_{1-x}V_x$ and **fcc $Ni_{1-x}Cu_x$** (Figs. 8 and 9) : Fairly good agreement is obtained in these system with $<|\mu|> = \mu_{so}$. This indicates the absence of localized moments in the paramagnetic phase.

fcc $Mn_{1-x}Co_x$ (Fig. 10) : The equation holds fairly well between the lattice constant at RT and μ_{so}. A small discrepancy is observed at the concentrations where ferromagnetism disappears ($0.6 < x < 0.8$). The origin of this discrepancy may be explained as follows. It is considered that some of the atomic moments are antiferromagnetically aligned at the critical concentrations because an exchange anisotropy has been observed. Consequently, $<|\mu|>$ should be larger than the bulk magnetization, i.e. $<|\mu|> > \mu_{so}$, The agreement indicates absence of atomic moments in this concentration range. In fact, the magnetic susceptibility is Pauli paramagnetic. However, it was reported that the alloys with $x < 0.65$ becomes antiferromagnetic at low temperatures[10]. In order to discuss the effect of the antiferromagnetism on the lattice constant, the data at $T= 0$ are necessary.

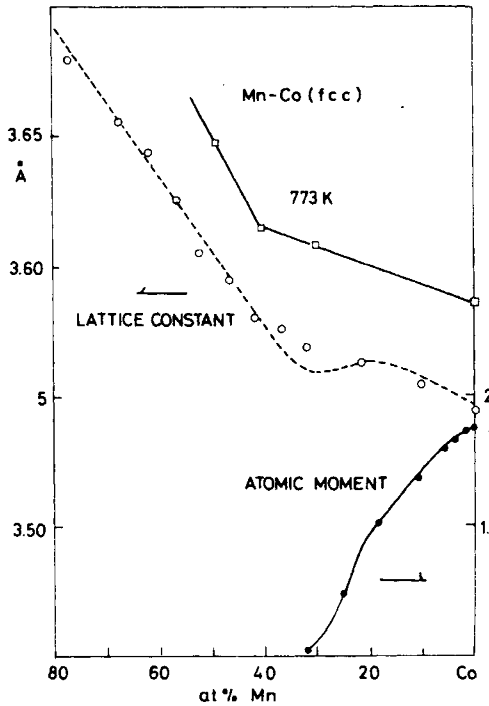

Fig. 10. The lattice constant at RT (o), (Ref. 1, p.510) and at 773 K (□)[8] of fcc Co-Mn.
---- calculated value with the parameters given in Table 1.

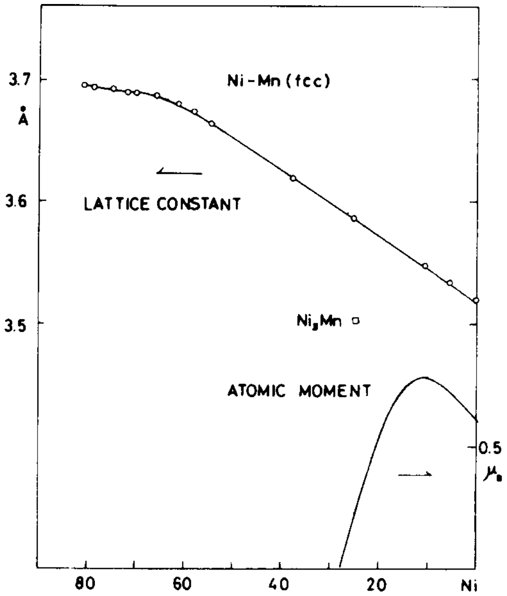

Fig. 11. The lattice constant at RT and atomic moment at 0 K of disordered Ni-Mn.
□ represents the atomic moment of ordered Ni_3Mn [12].

bcc $Cr_{1-x}Fe_x$ (Fig. 8) : The magnetization versus concentration curve of this system is similar to that of Fe-V, while the appearance of the *L-C* curve is quite different. The equation does not hold if one takes simply $<|\mu|> = \mu_{s0}$. In this system, however, it is believed that an iron atom has a localized atomic moment of about 2 μ_B[11)] in the nonferromagnetic region. Assuming μ_{Fe} =2 μ_B and μ_{Cr}= 0 over all concentrations, we may write $<|\mu|> =2x$. Then, it is expected that $<|\mu|>$ and consequently the lattice constant varies linearly with concentration in rough agreement with experimental observations. Strictly speaking, the *L-C* curve deviates slightly from a straight line at two sides of the system. Atomic moments on Cr atoms could be the cause of this behavior.

fcc $Ni_{1-x}Mn_x$ (Fig. 11): The equation does not hold if $<|\mu|>$ is equated with μ_s. This apparent disagreement may be explained on the same basis as above, that is, the magnitude of atomic moments on each atom would remain unchanged in the concentration range where the *L-C* curve is linear ($0 < x < 0.6$). The sharp decrease of the spontaneous magnetization in the region $0.1 < x < 0.3$ might be attributable to antiferromagnetic alignment of surviving localized moments. It should be noted that no difference is observed in the lattice constants between the atomically ordered and disordered states at the composition of Ni_3Mn despite the remarkable difference of the magnetization. The antiferromagnetic spin coupling is supported from recent experimental results such as neutron scattering[13)] and NMR[14)]. The magnitude of the Mn moment differs slightly between the two sides of the system. In the Ni rich region, Mn atoms have an atomic moment of about 3 μ_B [12)] and in the Mn rich region about 2 μ_B[15)]. The decrease of the slope of the *L-C* curve for $x > 0.6$ might be attributable to the decrease of the Mn moment.

Table I Values of parameters

system A - B	lattice structure	a_A (A)	a_B (A)	C (A/u_B)	temperature
Fe-Co	bcc	2.788	2.757	0.0324	RT
Fe-V	bcc	2.775	3	0.0386	RT
Fe-A1	bcc	2.788	3.03	0.0355	RT
Fe-Ni	fcc	3.551	3.489	0.0317	0K
Ni-Cu	fcc	3.506	3.607	0.0191	RT
Co-Mn	fcc	3.464	3.75	0.045	RT

The parameters thus obtained are listed in Table 1. One should note that the parameter C has a roughly constant value of about 0.03 Å/μ_B. It is interesting to compare the values of a_A or a_B which are determined for different systems with the same structure. For example, the three values of a_{Fe} which are determined for bcc Fe-Co, bcc Fe-V and bcc Fe-Al are 2.788 Å, 2.775 Å and 2.788 Å, respectively. It is worth noting that these three values are very close. This indicates a_A (or a_B) has a definite physical meaning.

LATTICE CONSTANT AT HIGH TEMPERATURES

It is interesting to see whether Eq. (1) holds or not at high temperatures or above the Curie temperature *T*c. However, we encounter a difficulty in estimating <|μ|>. According to the simple itinerant picture, it is possible that <|μ|> = 0 above *T*c since the polarization of 3*d* bands and accordingly that of the Wannier functions at each atomic sites should vanish. On the other hand, on the basis of the localized spin model, <|μ|> would remain finite even above *T*c. Bearing this in mind, we can expect to get information about the validity of different models from the appearance of the *L*-*C* curves at high temperatures.

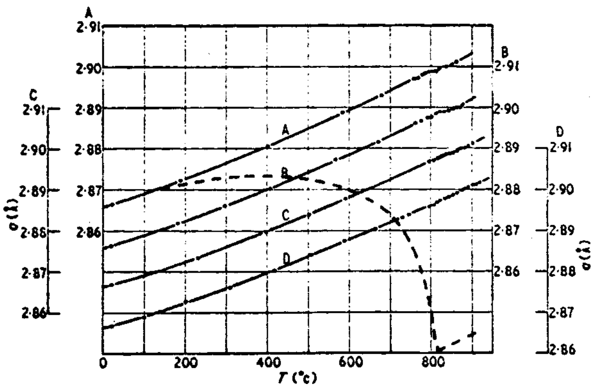

Fig. 12. Lattice constant a plotted against temperature for bcc Fe-Co. Co content: A 3% B 6%,c 9%, D 12%.
----- represents the expected change of lattice constant on the assumption that the magnetic term decreases with temperature.

Firstly, let consider the lattice constant of bcc Fe-Co alloys. According to our analysis at a low temperature, the magnetic term *C*<|μ|> is so large that a notable thermal expansion anomaly should be observed around T_c if <|μ|> decreased with the decreasing bulk magnetization as illustrated in Fig. 12. On the contrary, such an anomaly has not been observed. From this fact, it was previously considered that the deviation from Vegard's law could not be ascribed to a magnetic origin. However, noting that the agreement at room temperature is too good to be accidental, we should rather conclude that <|μ|> remains constant over a wide temperature range including *T*c and that thermal demagnetization is caused by fluctuations of localized atomic moments. One can describe such a situation in terns of Wannier functions which are still polarized above *T*c within a certain time interval. In this sense, we may say that the localized spin picture is a better description of this phenomenon. Similar behavior is observed in the *L-C* curve of fcc Co-Mn alloys at a high temperature, namely 773 K, as seen in Fig. 9.

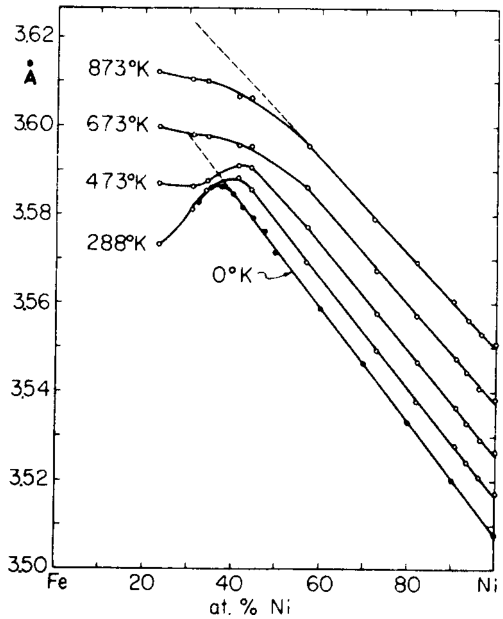

Fig. 13. The lattice constant of fcc Fe-Ni at various temperatures.

Particular attention should be paid to the *L-C* curve of fcc $Fe_{1-x}Ni_x$ alloys in connection with the Invar problem. As seen in Fig. 13, the lattice constant at 870 K, where the alloys are paramagnetic over all concentrations, varies linearly with x for $x > 0.6$ and the increase of its value from that at 0 K may well be explained by the ordinary thermal expansion due to lattice vibrations. This behavior indicates that the magnetic term $C\langle|\mu|\rangle$ remains unchanged above *T*c. Actually, no thermal expansion anomaly attributable to a magnetic origin is observed over this concentration range. For $0.4 < x < 0.6$, the lattice constant at 870 K deviates downwards from a straight line, indicating the reduction of the magnetic term. At 0 K, however, the lattice constant and μ_{so} are still increasing linearly with increasing iron composition over this range. This behavior may be explained as follows. Both iron and nickel atoms have full moments, namely $\mu_{Fe} = 2.8\ \mu_B$ and $\mu_{Ni} = 0.6\ \mu_B$ at 0 K for $0.4 < x < 1.0$. Therefore, $\langle|\mu|\rangle$, which may be written as $\langle|\mu|\rangle = 2.8\,(1-x) + 0.6\,x$, is linear in x. Above T_c, they still have full moments for $0.6 < x < 1.0$ but for $x < 0.6$, the magnitude of the atomic moments is reduced by raising temperature, which results in the thermal expansion anomaly observed in this region. Thus, the anomalous thermal expansion of the Invar alloy could be explained as the result of the reduction of the magnitude of the atomic moments[17]. This is in agreement with the result of neutron scattering[18].

ATOMIC SIZE MODEL

The change of the lattice constant in a solid solution $A_{1-x}B_x$ can be described in terms of the mean atomic size *d* expressed as

$$F\,a(x) = d = d_A\cdot(1-x)+d_B\cdot x, \qquad (2)$$

where d_A and d_B are atomic sizes of pure elements and *F* is a structure factor which depends on the lattice structure. $F = 1$ for simple cubic, $F = \sqrt{2}/2$ for fcc and $F= \sqrt{3}/2$ for bcc. We will show that Eq. (1) can be derived from extension of this idea. The procedure will give a certain physical meaning to the parameters a_A, a_B and *C* in Eq. (1). One shoud bear in mind, however, that the concept of atomic size does not necessarily have physical reality but may simply be a convenient representation of the atomic distance. We assume that constituent atoms take different states with the atomic moment $\mu i_{A/B}$ and the atomic size $d^i_{A/B}$. The lattice constant and the meem atomic moment can be given by

$$a(x) = (1-x)\sum p_A^i(x)\, d_A^i + x\sum p_B^i(x)\, d_B^i \qquad (3)$$

$$\langle|\mu|\rangle = (1-x)\sum p_A^i(x)\, \mu_A^i + x\sum p_B^i(x)\, \mu_B^i \qquad (4)$$

where $p^i_{A/B}(x)$ is the probability of finding an A/B atom in the *i* th state. Eq. (1) can be derived from these equations if the following conditions are fulfilled.

Case I : If $d^i_{A/B} = d^0_{A/B} + k\cdot\mu^i_{A/B}$, a(x) can be given by

$$\begin{aligned} Fa(x) &= (1-x)\left\{\sum p_A^i(x)\right\}d_A^0 + x\left\{\sum p_B^i(x)\right\}d_B^0 \\ &\quad + k\left\{(1-x)\sum p_A^i(x)\mu_A^i + x\sum p_B^i(x)\mu_B^i\right\} \\ &= (1-x)d_A^0 + xd_B^0 + k\langle|\mu|\rangle \end{aligned} \qquad (5)$$

Comparing with Eq. (1), we get $a_A = d_A/F$, $a_B = d_B/F$ and $C = k/F$

Case II. (Pseudo Ternary Alloy Model) If the element A has two states and B has an only one state, $a(x)$ and $\langle|\mu|\rangle$ are given by

$$F\ a(x) = (1-x)\left\{p_A^I(x)d_A^I + p_A^{II}(x)d_A^{II}\right\} + x\ d_B^0 \qquad (6)$$

$$\langle|\mu|\rangle = (1-x)\left\{p_A^I(x)\mu_A^I + p_A^{II}(x)\mu_A^{II}\right\} + x\mu_B^0 \qquad (7)$$

Noting $p_A^I(x) + p_A^{II}(x) = 1$, we can eliminate $p_A^I(x)$ and $p_A^{II}(\mathbf{x})$ from Eqs. (6) and (7). Then we get Eq. (1) with

$a_A = \{d_A^{II} - K\mu_A^{II}\}/F$, $a_B = \{d_B^0 - K\mu_B^0\}/F$ and $C=K/F$,

where $K=\{d_A^{II} - d_A^I\}/\{\mu_A^{II} - \mu_A^I\}$.

One should note that, in this case, the atomic size of individual atoms is not necessarily a linear function of the atomic moment. Some other experimental information is necessary to determine which case is realized in actual alloys.

It is possible to estimate $d^i_{A/B}$ for some special cases. For example, in bcc $Fe_{1-x}Co_x$ alloys, it has been revealed by neutron scattering that the individual atomic moments remain constant over the concentration range $x > 0.5$, where both the lattice constant and the magnetic moment vary linearly with concentration[21]. We may assume that each element takes only one state with $\mu_{Fe} = 3\mu_B$ and $\mu_{Co} = 1.8\ \mu_B$ over this range. By extrapolating the linear region $0.5 < x < 0.7$ to pure iron, we can estimate the atomic size of bcc Fe in the state of $\mu_{Fe}=3\ \mu_B$. $d_{Fe} = 2.492$ Å is thus obtained. Pure iron may be another state with $\mu_{Fe} = 2.2\ \mu_B$ and $d_{Fe} = 2.477$ Å. Since iron atoms are thought to be nonmagnetic in V rich Fe-V, we can estimate the atomic size of nonmagnetic iron. In this case, evidently $d_{Fe}{}^0 = \sqrt{3}/2\ a_{Fe}$, where a_{Fe} is the parameter of Eq. (1) for bcc Fe-V system. The atomic sizes thus obtained are listed in Table 2 with other examples estimated in a similar way.

The expansion of the atomic size with the atomic moment is plotted in Fig. 14. It should be emphasized that the rate of the expansion is almost same for different elements. It appears that the expansion is a linear function of the atomic moment, corresponding to the Case I. However, the number of data points is not adequate to make a definite conclusion.

Table II Atomic sizes of some elements in different atomic states

element and structure	atomic moment	atomic size (A)	expansion (%)	system
bcc Fe	0	2.403	0	Fe-V
	0	2. 408	0	Fe-Al
	2.2	2.477	2.88	pure Fe
	3	2.492	3.53	Fe-Co
fcc Fe	0	2.521	0	Fe-Mn
	2.8	2.574	2.1	Fe-Ni
fcc Co	0	2.464	0	Co-Mn
	1.8	2.507	2.3	pure Co
fcc Mn	0	2.59	0	after Weiss19
	2.4	2.69	2.8	
	4.5	3.82	6.4	

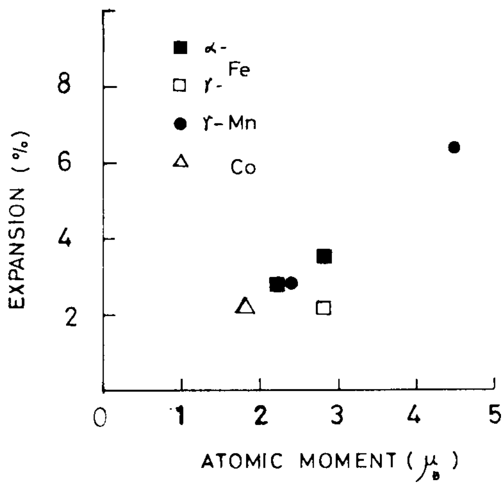

Fig. 14. Expansion of atomic size with atomic moment. The atomic sizes of gamma-Mn in different states are estimated by Weiss[22].

So far, we have referred only to binary solid solutions of 3*d* metals with cubic structure. The equation may be more widely applicable, for example, one of the constituents is not a 3*d* metal. Nice agreement is obtained in bcc Fe-Al alloys as seen in Fig. 15. We may apply the relation to pseudo-binary alloys. The peculiar appearance of the *L-C* curve of cubic Laves compounds such as $Y(Fe_{1-x}Co_x)_2$[20] may be explained by adding a magnetic term. For alloys or metallic

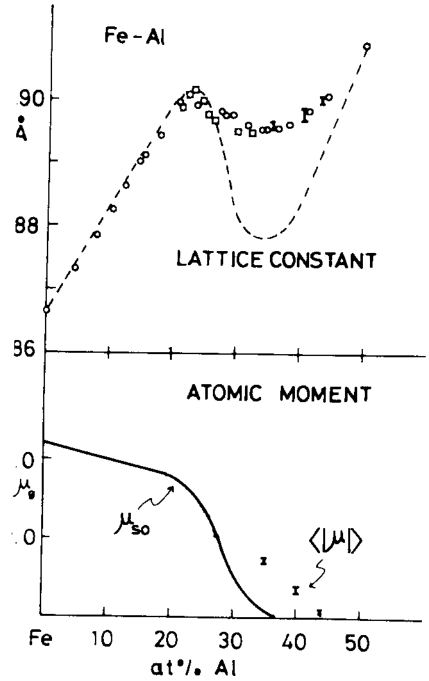

Fig. 15. The lattice constant of bcc Fe-Al at RT (Ref. 1, p.344).
------ calculated values with the parameters given in Table 1, where μ_{s0} is used for $<|\mu|>$, Bars represent the calculated lattice constant with $<|\mu|>$ which are estimated from the distribution of the internal magnetic fields acting on Fe nuclei (bars in the lower figure).

compounds with non-cubic structure, a similar relation might be found between the atomic volume and the magnetic moment.

Presumably, the magnetic term $C<|\mu|>$ is attributable to volume magneto-striction. However, theoretical[21)] and experimental[22)] investigations predict quadratic dependence of the spontaneous volume magneto-striction on the magnetic moment, while the magnetic term of Eq. (1) is linear with respect to the magnetic moment. This contradiction may be removed by invoking the pseudo-ternary alloy model with an assumption

$$d_A^i = d_A^0 + k\,(\mu_A^i)^2.$$

Or there might be another mechanism to cause a volume expansion accompanied by the formation of localized moments. It is possible that the attractive force between magnetized atoms is weaker than that between nonmagnetic atoms. Theoretical investigation on this subject are desirable.

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