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Magnetic analysis of rapidly quenched LaFe alloys—a ferromagnetically coupled ferromagnetic cluster system: II. Analysis of the magnetization of as-quenched alloys

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Abstract. The magnetization of rapidly quenched LaFe alloys with less than 12 at.% Fe, reported in the preceding paper, was analysed. The alloy can be considered as a random set of ferromagnetic clusters with inter-cluster ferromagnetic coupling. It was demonstrated that the magnetization of an alloy for different external fields and temperatures can be expressed as a function of either $H/(T - T_C^{inter})$ in the super-paramagnetic region ($290 \leqslant T \leqslant 380$ K) or $(H + wM)/T$ in a slightly wider temperature range ($240 \leqslant T \leqslant 380$ K), provided that the temperature dependence of the magnetization of a cluster is taken into account. Here, T_C^{inter} is the asymptotic Curie temperature and w is the molecular-field constant for the inter-cluster coupling. The relations gave estimates of the average cluster size, 100–1500 Fe atoms, which depends not on the Fe concentration but on the quenching process. Magnetization in the ferromagnetic region, $5 < T < 250$ K, was analysed by the use of the molecular-field approximation with distributed cluster size and exchange coupling constant.

1. Introduction

Magnetic measurements of as-quenched LaFe alloys with Fe concentration less than 12%, reported in the preceding paper [1] (referred to as I hereafter), indicated that these are ferromagnetically coupled ferromagnetic cluster systems. The cluster is a metastable intermetallic compound of La and Fe, which becomes ferromagnetic below about 400 K. Inter-cluster coupling makes the total system ferromagnetic below about 250 K. Between these two Curie points, intra-cluster (T_C^{intra}) and inter-cluster (T_C^{inter}), the alloys are super-paramagnetic. Instability of the intermetallic compound above 500 K and the large moment of clusters below 400 K, however, prevent conventional analysis of the magnetization, such as a $1/\chi$ versus T plot, to obtain quantitative information on the magnitude of the coupling constants and the cluster size.

In this paper, we will report that the magnetization in the super-paramagnetic region of these alloys can be expressed uniquely by the effective ratio of the Zeeman to thermal energy, by either $b(T)H/k(T - T_C^{inter})$ or $b(T)(H + wM)/kT$. Here, H is the external

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Table 1. Magnetic parameters of LaFe rapidly quenched alloys.

	T_C^{intra}	T_C^{inter}	w	$\langle m^2\rangle/\langle m\rangle$ (μ_B)			
Specimen	(K)	(K)	(kOe g emu^{-1})	a	b	c	d
1-1	430	265	120	1430	1570	530	820
1-2	440	245	50	270	480	440	600
4	425	250	2.8	1900	3000	1580	1250
8	435	270	4.2	600	540	540	570
12	400	250	1.85	730	810	730	980

[a] From the linear part of the plot near the origin, figure 1(*d*).
[b] From the linear part of the plot near the origin, figure 3.
[c] From w and T_C^{inter}.
[d] From parameters tabulated in table 2.

magnetic field, T is the temperature, T_C^{inter} and w are the asymptotic Curie temperature and the molecular-field constant of the cluster system, respectively, M is the total moment of the specimen with Pauli paramagnetism subtracted, and $b(T)$ is a function of temperature that expresses the temperature dependence of the magnetization of a cluster.

The La–dilute Fe system is an example of macroscopically homogeneous random systems, without microscopic translational symmetry. Two kinds of atomic random systems have been investigated so far: structurally random or amorphous systems, and substitutionally random systems with different atoms distributed on the equivalent sites of the crystal. The present system is a randomly distributed set of clusters of high Fe concentration, with distributed size and inter-cluster distance, which are dispersed in a matrix of paramagnetic FCC β-La. Though the unit of this system is not atoms but much larger clusters, the alloys are macroscopically homogeneous and thermally equilibrated at room temperature. In a temperature range without long-range order of cluster magnetizations, the magnetization at different temperatures and external magnetic fields is expressed as a function of one variable, the ratio of the effective Zeeman energy to the effective thermal energy (see figure 3 below). In contrast to this, homogeneity of the system is rather doubtful in the ferromagnetic state below about 250 K. Magnetization below room temperature is discussed by the molecular-field approximation, assuming distributed molecular-field constant and cluster size.

In the next section, the experimental results reported in I are analysed by the scaling either of the temperature (as $T-T_C^{inter}$) or of the magnetic field (as $H+wM$). Magnetization in the low-temperature region is discussed in section 2.2. The method and the results of the analysis will be discussed in the last section.

A preliminary report of this investigation was made at the international conference 'Rare Earths '92', Kyoto [2].

2. Analysis of the experimental results

2.1. Magnetization above 290 K: super-paramagnetic region

As we described in I, as-quenched specimens show super-paramagnetic behaviour at room temperature but are ferromagnetic below about 250 K (see figure 6 and 9 of I). At the same time, thermomagnetic curves in an external magnetic field of 5 kOe indicate another transition at around 400 K (see figure 8 of I). These facts suggest that the system is composed of magnetic clusters, ferromagnetic below 400 K, and these clusters interact with each other ferromagnetically to form a macroscopic ferromagnet below room temperature.

Magnetization curves of these alloys at 5 K indicated the existence of a term proportional to the external field. This part was interpreted as Pauli paramagnetism of the La matrix (see equation (1) and table 1 in I). In the following discussion, this part of the magnetization is subtracted from the net moment. The possible temperature dependence of the Pauli susceptibility is ignored in the subtraction.

As is well known, magnetization of a super-paramagnetic system at any temperature and field, $M(T, H)$, is uniquely determined by a universal function of H/T, provided that the magnitude of the magnetization of each cluster is independent of both temperature and external field and inter-cluster coupling can be ignored [3]. This relation between M and H/T is easily explained by Langevin's theory of paramagnetism. Note that a distribution in cluster size does affect the functional form of the M versus H/T plot but does not violate the fact that M is determined uniquely by the ratio of Zeeman to thermal energy, or magnetic field to temperature, H/T. As is shown clearly in figure 1(a) for 8% Fe alloy, this is not the present case. The plot of M against H/T spreads two-dimensionally. This is an indication that the above-mentioned two conditions are not fulfilled. To recover a universal relation, we must take into account both intra- and inter-cluster coupling.

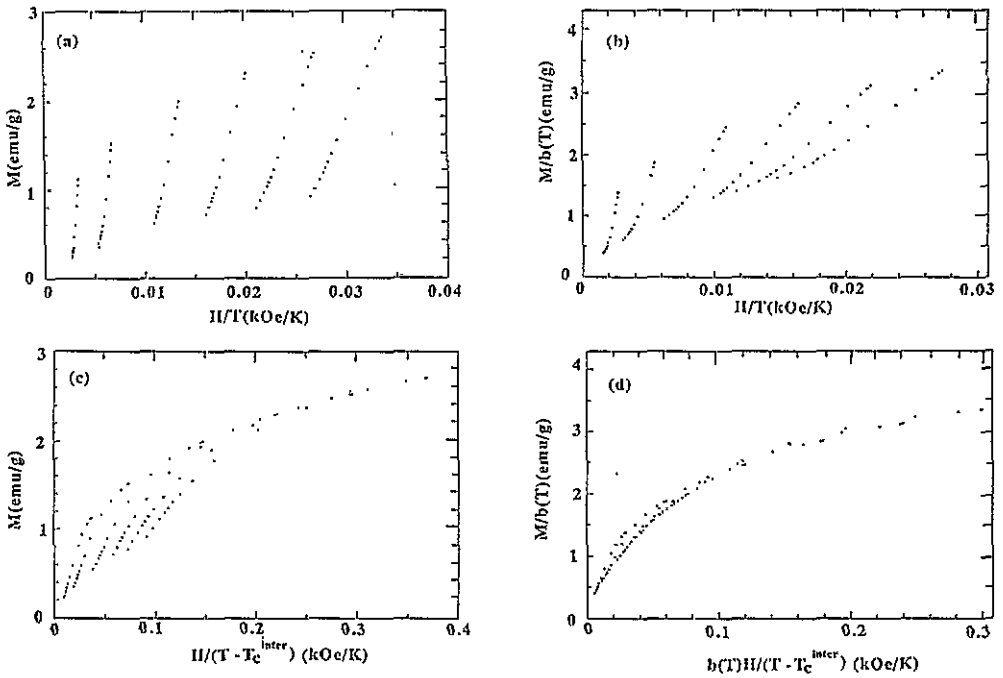

Figure 1. Plots of magnetization (with Pauli paramagnetism subtracted) of 8% Fe as-quenched alloy between 290 and 380 K, indicating finite intra- and inter-cluster coupling: (a) M against H/T from left to right, external magnetic field was 1, 2, 4, 6, 8 and 10 kOe, respectively; (b) $M/b(T)$ against H/T; (c) M against $b(T)H/(T-T_C^{\text{inter}})$; ($d$) $M/b(T)$ against $b(T)H/(T-T_C^{\text{inter}})$.

The effect of finite intra-cluster interaction becomes manifest through the temperature dependence of the magnetization of each cluster. For simplicity, we will approximate the dependence by one function, $b(T)$, for all the clusters:

$$m(T) = mb(T). \tag{1}$$

Here, $m(T)$ is the magnetization of a cluster at temperature T and m is that at 0 K. The function $b(T)$ is assumed to be the magnetization of a macroscopic ferromagnet composed of $S = 1$ spins, normalized by the saturation moment, and is calculated by the molecular-field approximation. Note that the magnetic moment of an Fe atom is estimated to be $2\mu_B$. Figure 2 shows the function $b(T)$ thus calculated. One parameter, the Curie temperature of a cluster, T_C^{intra}, is introduced here. The magnetization M of the specimen in the ordinate of figure 1, should be normalized by $b(T)$. At the same time, the magnetic field H in the abscissa should be modified as $b(T)H$. Physically, the variable of this plot is the ratio of Zeeman to thermal energy, and the former is proportional to $b(T)H$ in this case. As for inter-cluster interaction, we simply replaced thermal energy kT by $k(T - T_C^{inter})$ [4]. Another parameter, the Curie temperature of the whole system, T_C^{inter}, is introduced here. Then, the ratio of Zeeman to thermal energy in the abscissa is expressed by $b(T)H/(T - T_C^{inter})$.

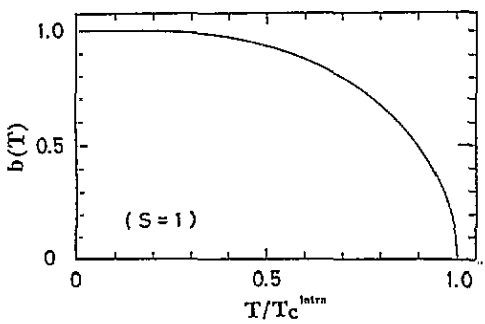

Figure 2. The temperature dependence of the magnetization of a ferromagnet composed of $S = 1$ spins, calculated by the molecular-field approximation. The magnetization is normalized by the saturation moment and the temperature is normalized by the Curie point of the system, T_C^{intra}. This function is denoted $b(T)$ in the text.

By these modifications, a universal relation is recovered once more by choosing the two parameters, T_C^{inter} and T_C^{intra}, appropriately (see figure 1(*d*) for the magnetization of the 8% Fe alloy at temperatures between 290 and 380 K). Figures 1(*b*) and (*c*) show the effect of intra- and inter-cluster couplings, T_C^{intra} and T_C^{inter}, separately. It is natural that the consideration of finite intra-cluster coupling mainly modifies data near T_C^{intra}, whereas that of finite inter-cluster coupling modifies those near T_c^{inter}. Such a plot is possible only for temperatures between T_C^{intra} and T_C^{inter}.

The two parameters, T_C^{inter} and T_C^{intra}, determined by the plot are tabulated in table 1. It is to be emphasized that the magnitudes of the two parameters, T_C^{inter} and T_C^{intra}, are almost the same for all the specimens.

As was mentioned above, a distribution in cluster size does not affect the existence of the universal relation itself but does affect its functional form. Indeed, data points in a plot like that in figure 1(*d*) for different specimens do not lie on an identical curve nor on a Langevin function. Though it is impossible to determine the distribution function of the cluster size from the functional form, the initial slope of the plots in figure 1(*d*) which we denote K_T, gives information on the average size of the cluster.

According to Langevin's theory of paramagnetism modified as described above, the magnetization of a specimen in an external field H at temperature T is expressed as

$$M(H,T) = \int mb(T)L(X)f(m)\,dm \tag{2}$$

with

$$X = mb(T)H/k(T - T_C^{inter}). \tag{3}$$

Here, $L(X)$ is the Langevin function and $f(m)$ is the distribution function of clusters, i.e. the number of clusters with magnetization between m and $m + dm$ at 0 K. For small X, the Langevin function can be expanded as usual:

$$M(H,T) = \int mb(T)\frac{mb(T)H}{3k(T - T_C^{inter})} f(m)\,dm \tag{4}$$

giving

$$\frac{M(H,T)}{b(T)} = \frac{b(T)H}{3k(T - T_C^{inter})} \int m^2 f(m)\,dm. \tag{5}$$

Note that the curve starts from the origin. The initial slope is given as

$$K_T = \frac{1}{3k}\int m^2 f(m)\,dm \equiv \frac{\langle m^2\rangle}{3k} N_c. \tag{6}$$

Here, N_c is the number of clusters. Since $M(T=0) = \int m f(m)\,dm \equiv \langle m\rangle N_c$, we have

$$\frac{\langle m^2\rangle}{\langle m\rangle} = \frac{3kK_T}{M(T=0)}. \tag{7}$$

Here $\langle m\rangle$ and $\langle m^2\rangle$ are mean and mean-square magnetizations over the cluster size, respectively. The values of $\langle m^2\rangle/\langle m\rangle$, deduced from the plots in figure 1(*d*), are tabulated in table 1, column a.

There is another way to account for the inter-cluster coupling constant and evaluate average cluster size. According to the molecular-field approximation, the effective magnetic field H_{eff} is expressed as

$$H_{eff} = H + wM(H,T). \tag{8}$$

Here, H is the external magnetic field and w is the molecular-field constant. Plots of $M(H,T)/b(T)$ against $b(T,H)H_{eff}/T$ should be universal if the molecular-field approximation is valid. As is shown in figure 3 for 8% Fe alloy, this was proved in the present case. In contrast to the normalization of T by $(T - T_C^{inter})$, the molecular-field approximation is not restricted to the paramagnetic region. The temperature region of the data plotted in figure 3 is extended down to 240 K. Data below 240 K do not show such a universal relation: i.e. the ordinary molecular-field approximation is not valid in the temperature region of the long-range magnetic ordering of clusters. The estimated molecular-field constants are tabulated in table 1.

The initial slope of the plot in figure 3 gives another estimate of the average cluster size: equation (7) with K_T replaced by K_H. The results are tabulated in table 1, column b.

In the molecular-field approximation, the Curie point of the whole system is determined by the following equation:

$$w\chi_0(T = T_C^{inter}) = w\int \frac{m^2 b(T_C^{inter})^2}{3kT_C^{inter}} f(m)\,dm = \frac{wb(T_C^{inter})^2}{3kT_C^{inter}}\langle m^2\rangle N_c = 1. \tag{9}$$

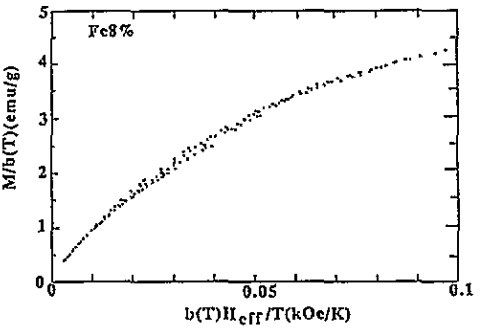

Figure 3. Plot of $M/b(T)$ against $b(T)H_{\rm eff}/T$ for 8% Fe as-quenched alloys, for the experimental data between 240 and 380 K. $H_{\rm eff} = H + wM$, where H is the external field. Molecular-field constants w are tabulated in table 1.

Here, χ_0 *is the magnetic susceptibility of the system without coupling.* Then,

$$\frac{\langle m^2\rangle}{\langle m\rangle} = \frac{1}{wb(T_{\rm C}^{\rm inter})^2}\frac{3kT_{\rm C}^{\rm inter}}{M(T=0)}. \tag{10}$$

Equation (10) gives another estimate of the average cluster size, which is also tabulated in table 1, column c.

These three estimates of the cluster size give the same order of magnitude, between 270 and $3000\mu_{\rm B}$, or 130 and 1500 Fe atoms, not dependent on the Fe concentration. In the case of the specimen of 1% (1–1) and 4% alloys, estimated cluster sizes in columns a and b deviate very much from that in column c. Extrapolation of the data at high temperatures, as in figure 1(d), to go through the origin seems rather difficult. Indeed, if we ignore the condition of going through the origin, the slopes of the data points give cluster size estimations in much better coincidence with column c: 530 and $550\mu_{\rm B}$ for the 1% (1–1) and 1320 and $1900\mu_{\rm B}$ for the 4% alloy by the use of K_T and K_H, respectively. The assumption of equation (1), namely identical $T_{\rm C}^{\rm intra}$ for all the clusters, does not seem good in these alloys. A very small amount of iron precipitation is suspected.

2.2. *Magnetization below 300 K: ferromagnetic region*

Two examples of thermomagnetic curves of as-quenched alloys below 450 K are shown in figure 4 for several external field strengths. Note that the curves for different external field strengths seem only to be shifted vertically by almost the same amount. Magnetization does not saturate in an external field of the order of 10 kOe even at 10 K or lower temperatures. The existence of free small magnetic moments, or a distribution in the coupling constants as well as the cluster size, should be introduced to explain such a characteristic. We will apply the molecular-field approximation to analyse the experimental results on this random system.

One point to be made here is that the molecular-field coefficient, λ_{ij}, between the ith and jth magnetic moments should be symmetrical:

$$\lambda_{ij} = \lambda_{ji}. \tag{11}$$

In order to avoid complexity due to too many parameters, the following assumptions are made:

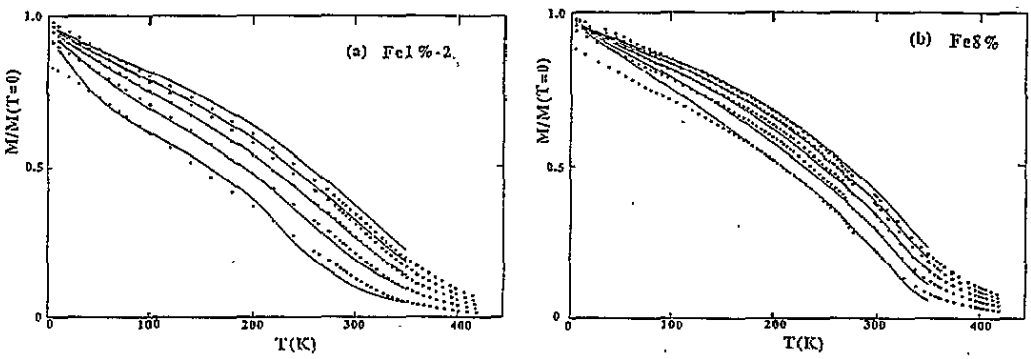

Figure 4. Thermomagnetic curves of 1% (1-2) and 8% Fe as-quenched alloys. External field was 10, 8, 6, 4 and 2 kOe, from top to bottom. Full curves are results of calculation described in the text.

(i) Clusters are divided into several groups with fixed size and coupling, instead of a continuous distribution function as in equation (2).

(ii) To fulfill equation (11) with the simplest approximation, the molecular-field coupling constant between clusters i and j is expressed as the product of two parameters indigenous to each cluster:

$$\lambda_{ij} = \lambda(i)\lambda(j). \tag{12}$$

There is no justification for this assumption except that it is the simplest. If the coupling between clusters, separated by non-magnetic La lattice, is due to homogeneous polarization of the conduction electron of La, however, equation (12) can be naturally accepted. In practice, two cluster sizes, $m(i)$, and two coupling constants, $\lambda(i)$, thus $2 \times 2 = 4$ kinds of clusters as a total, are assumed.

The effective field on a cluster j is expressed as

$$H_{\text{eff}}(j) = H + \lambda(j) \sum \lambda(i)\overline{m(i)}f(i). \tag{13}$$

Here, $\bar{Z}$ is the thermal average of quantity Z and $f(i)$ is the number of clusters of class i. Then, the magnetization at any field and temperature can be calculated by self-consistency equations, equations (13)–(15):

$$M(H,T) = \sum \overline{m(j)}f(j) = \sum m(j)b(T)L(X_j)f(j) \tag{14}$$

$$X_j = m(j)b(T)H_{\text{eff}}(j)/kT. \tag{15}$$

We will introduce new parameters,

$$F(i) = \frac{m(i)f(i)}{\sum m(j)f(j)} \tag{16}$$

$$\Lambda(i) = \lambda(i)[M(T=0)]^{1/2} \tag{17}$$

$M(T=0) = \sum m(j)f(j)$ is the saturation moment of the specimen at 0 K and $F(i)$ is the fraction of the magnetization of class i clusters at 0 K. Equations (13) and (14) are

rewritten as

$$H_{\text{eff}}(j) = H + \Lambda(j) \sum_i \Lambda(i) b(T) L(X_i) F(i) \tag{18}$$

$$\frac{M(H,T)}{M(T=0)} = \sum_j m(j) b(T) L(X_j) F(j). \tag{19}$$

Equations (14), (18) and (19) are solved numerically, by minimizing the sum of the absolute values of the differences between experimental and calculated magnetizations, $M(H, T)$. There are seven parameters, two 'm's, two 'Λ's and three 'F's (note the condition $\sum F = 1$). We determined 'F's for given 'm' and 'Λ's first, and then 'm's and 'Λ's were optimized. The results are shown in figure 4 by full curves and the determined parameter values are tabulated in table 2. Average cluster sizes, $\langle m^2 \rangle / \langle m \rangle$, are calculated from the determined F values and tabulated in table 1, column d.

Table 2. Parameter values determined as described in the text.

		Cluster			
Specimen		1	2	3	4
1-1	m (μ_B)	60	60	870	870
	Λ (kOe$^{1/2}$)	1×10^{-5}	10.7	10.7	1×10^{-5}
	F	0.334	0.139	0.368	0.159
1-2	m (μ_B)	7	7	600	600
	Λ (kOe$^{1/2}$)	1×10^{-5}	5.4	5.4	1×10^{-5}
	F	0.067	0.000	0.619	0.314
4	m (μ_B)	20	20	1250	1250
	Λ (kOe$^{1/2}$)	0.45	7.8	7.8	0.45
	F	0.040	0.001	0.614	0.345
8	m (μ_B)	6	6	570	570
	Λ (kOe$^{1/2}$)	1.5	9.3	9.3	1.5
	F	0.041	0.002	0.403	0.554
12	m (μ_B)	180	180	1170	1170
	Λ (kOe$^{1/2}$)	1×10^{-5}	5.5	5.5	1×10^{-5}
	F	0.062	0.127	0.648	0.163

The model calculation seems to have reproduced the qualitative characteristics of the experiment, though the crude assumptions prevented a quantitative explanation.

3. Discussion

3.1. On the analysis in the super-paramagnetic region

The analysis of the magnetization of rapidly quenched LaFe alloys in the last section demonstrated that the as-quenched alloys form ferromagnetically coupled magnetic cluster systems. The magnetization M in the super-paramagnetic state at any temperature and external magnetic field was expressed uniquely by the ratio of Zeeman to thermal energy, if

effective temperature ($T - T_C^{inter}$) or effective field ($H + wM$) was introduced. At the same time, the temperature dependence of the magnetization of each cluster should be taken into account. By this process, the Curie temperatures of inter- and intra-cluster coupling were quantitatively evaluated and the average size of the clusters was estimated.

The temperature dependence of the magnetization of each cluster, $b(T)$, was assumed the same, for all clusters in an alloy. The molecular-field approximation is applied to estimate magnetization at any temperature. As was shown in figures 1 and 3, the result of this approximation was rather good up to about $0.9T_C^{intra}$: i.e. clusters have well defined Curie temperature, T_C^{intra}, which was almost the same for all samples. These two points are the basis of our argument in I that a metastable ferromagnetic intermetallic compound exists between La and Fe.

For the atomically random system without microscopic translational symmetry, it has been established experimentally that a well define second-order magnetic phase transition can exist. In some cases, the magnetic equation of state was determined near the critical point [5, 6]. This means that the specimen is homogeneous, or the physical characteristics such as the Curie point are the same in all portions. In other words, translational symmetry must be recovered macroscopically. Unfortunately, the condition of macroscopic homogeneity has not been clarified. It depends not only on the atomic configuration of the specimen but also on the temperature or the external field [6]. In general, however, we can safely assume that the system tends to be homogeneous at higher temperatures.

One example is the Curie–Weiss law. This law is a universal relation between the magnetization, magnetic field and temperature that expresses the magnetic state of the specimen as a whole. The asymptotic Curie temperature is determined by an average of the exchange coupling constants between spins, and the slope of the $1/\chi$ versus T plot is given by the mean square of the magnetic moments. In other words, the statistical average of the exchange constants and the mean-square moments are meaningful to understand magnetic properties of the system.

In the present case, too, the linear portion near the origin of the plots in figure 1(d) is just the Curie–Weiss law. A specific point here is that the universal relation is not limited to the linear part of the magnetization curves. This is the first discovery of this, as far as we know. Because the Zeeman energy of each cluster, different cluster by cluster, is not small compared with the thermal energy, total magnetization saturates with increasing external field. Still, total moment is expressed by one variable, $b(T)H/(T - T_C)$.

3.2. On the molecular-field approximation

The Curie–Weiss law can be deduced by the molecular-field approximation. In contrast to the scaling of thermal energy as $T - T_C$, the molecular-field approximation, $H \rightarrow H + wM$, is not limited to the temperature range higher than T_C, in principle. In the present case, however, experimental data plotted in figure 3 are only those above 240 K. Data for lower temperatures do not locate on a universal curve. Inapplicability of the molecular-field approximation suggests that the system is not homogeneous at temperatures lower than about $0.9T_C^{inter}$. The system cannot be described by one parameter, the molecular-field constant in an average. Distribution in the molecular-field constant and cluster size is necessary to analyse magnetization below 240 K.

Another candidate of the model for the present systems is the correlated spin glass [7]. We have ignored the effect of the magnetic anisotropy, however, partly because there is no evidence of the blocking of the magnetization. The magnetization curves at 5 K (figure 7 of I) do not saturate as a linear function of $H^{-1/2}$, as expected for a correlated spin glass, as well as for the one-dimensional pinning model of magnetization [8].

As is shown in figure 4, the qualitative characteristics of thermomagnetic curves are reproduced by the 2×2 kinds of cluster model, except at lowest temperatures where calculated magnetization does not depend so much on the external field. The crudeness of the molecular-field approximation, of the assumption for the coupling constants, to be given as products of parameters determined for two coupled clusters (equation (12)), and of only two kinds of cluster size and coupling constant in place of continuous distribution function, should be the cause of the absence of quantitative agreement. More fundamentally, the adequacy of the model of a coupled cluster system is questionable at temperatures much lower than both T_C^{inter} and T_C^{intra}, where weak coupling cannot be ignored compared with thermal fluctuation.

Parameter values tabulated in table 2 suggest the following characteristics of the system. (i) Cluster size and inter-cluster coupling as well as the distribution of them are dependent on the quenching conditions, which cannot be controlled perfectly. (ii) The number of strongly coupled small clusters is small in all cases, except for the specimen 1-1: small clusters are relatively free from other clusters. (iii) Weakly coupled small clusters are also scarce in 1% (1-2), 4% and 8% Fe alloys.

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