ON STRUCTURALLY DISORDERED FERROMAGNETIC SYSTEMS

BY A. R. FERCHMIN

Institute of Physics, A. Mickiewicz University, Poznań*

A simple derivation of Kaneyoshi's formula for the magnetization of disordered dilute alloys is given by Handrich's approach. A possible application of this formula (in slightly modified form) to amorphous ferromagnetics is discussed. The critical concentration of amorphous binary magnetic alloys, below which ferromagnetism disappears, is predicted theoretically using a cluster variation method.

Recently, Kaneyoshi [1], using a rather sophisticated formalism of cumulant averages, derived expressions for dilute magnetic alloys reducing in the molecular field approximation to the following very simple formula (originally labeled 2.24–2.25):

$$\frac{\langle S \rangle}{S} = \frac{1}{2}\left\{ \tanh\left(\frac{1}{2kT}\,\alpha_+(c) \right) + \tanh\left(\frac{1}{2kT}\,\alpha_-(c) \right) \right\} \tag{1}$$

where

$$\alpha_\pm(c) = cJ_0\langle S \rangle \pm \langle S \rangle \left\{ c(1-c)\left(\frac{1}{N}\sum_k J_k^2 \right) \right\}^{\frac{1}{2}} \tag{2}$$

with $\langle S \rangle$ denoting the sample-averaged mean value of spin, J_k the Fourier-transform of the exchange integral, and c the concentration of magnetic atoms.

It is our aim to give a simple derivation of the formula (1) basing on Handrich's [2, 3] decoupling of higher order correlation functions of fluctuations. This is done in Section 1. Moreover, as Kaneyoshi used his results to explain certain properties of amorphous ferromagnetics, we shall discuss the modifications of formula (1), originally derived for a crystal, which are necessary when one wants to apply Eq. (1) to amorphous media (Section 2). The critical concentration of magnetic dilute amorphous alloys below which ferromagnetism disappears is calculated in Section 3 using the cluster variation method.

---

* Address: Instytut Fizyki, Uniwersytet im. A. Mickiewicza, Poznań, Grunwaldzka 6, Poland.

1. Derivation of equation (1) for crystalline alloys

For spin 1/2, the expectation value of the i-th spin $\bar{S}_i$ averaged $\langle \; \rangle$ over the sample is:

$$\frac{\langle \bar{S}_i \rangle}{S} = \left\langle \tanh \left(\frac{1}{2kT} \langle S \rangle \sum_j J_{ij} \right) \right\rangle. \tag{3}$$

We have assumed for simplification that, for the neighbours j of site i, $\langle \bar{S}_j \rangle = \langle S \rangle$. The sum runs over all **magnetic** neighbours of S_i; they are randomly distributed over a Bravais lattice. J_{ij} is the exchange integral between S_i and S_j. As before Eq. (2) the concentration of magnetic atoms is c. Pointed brackets $\langle \; \rangle$ denote averaging over the random distribution of atoms.

In order to simplify the notations, we shall discuss in detail the case of nearest neighbour interactions; however, we also derive expressions (7) and (8) for the general case involving interactions with **any** neighbours (*cf.* [4]). The i-th magnetic atom has, say, l magnetic nearest neighbours, so that in (2) $\Sigma_j J_{ij} = J_1 l$. We introduce the notation $l = \langle z \rangle + \Delta z_i$, where $\langle z \rangle$ denotes the mean value of l over the sample. We expand the right-hand side of Eq. (3) into a series in fluctuation terms and assume [2, 3]

$$\langle (\Delta z)^b \rangle = \begin{cases} 0 & \text{odd} \\ \langle (\Delta z)^2 \rangle^{b/2} & \text{for } b \\ & \text{even} \end{cases}$$

After re-summing we obtain (*cf.* [3])

$$\frac{\langle S \rangle}{S} = \frac{1}{2} \left\{ \tanh x \left(1 + \sqrt{\frac{\langle (\Delta z)^2 \rangle}{\langle z \rangle^2}} \right) + \tanh x \left(1 - \sqrt{\frac{\langle (\Delta z)^2 \rangle}{\langle z \rangle^2}} \right) \right\} \tag{4}$$

where

$$x = \frac{J_1 \langle S \rangle \langle z \rangle}{2kT}.$$

The probability that a given magnetic atom has l magnetic nearest neighbours in a lattice with co-ordination number z (and $z-l$ nonmagnetic ones) is

$$P(l) = c^l (1-c)^{z-l} \frac{z!}{l!(z-l)!}.$$

As well known, for the binomial distribution

$$\sum_{l=0}^{z} P(l) = 1; \quad \langle z \rangle = \sum_{l=0}^{z} l P(l) = cz;$$

$$\langle (\Delta z)^2 \rangle = \sum_{l=0}^{z} (l - \langle z \rangle)^2 P(l) = \sigma^2 = zc(1-c) \text{ (the variance)}.$$

Thus, Eq. (3) takes the following form:

$$\frac{\langle S \rangle}{S} = \frac{1}{2} \left\{ \tanh \frac{1}{2kT} \, \Psi_+(c) + \tanh \frac{1}{2kT} \, \Psi_-(c) \right\} \tag{5}$$

where

$$\Psi_\pm(c) = zcJ_1\langle S \rangle \pm \langle S \rangle \sqrt{zc(1-c)J_1^2} . \tag{6}$$

On taking into account also interactions J_i with z_i i-th neighbours [4], we obtain

$$\frac{\langle S \rangle}{S} = \frac{1}{2} \left\{ \tanh \frac{1}{2kT} \, \lambda_+(c) + \tanh \frac{1}{2kT} \, \lambda_-(c) \right\} \tag{7}$$

where

$$\lambda_\pm = c \Big(\sum_{i=1}^{p} J_i z_i \Big) \langle S \rangle \pm \langle S \rangle \sqrt{c(1-c) \sum_{i=1}^{p} J_i^2 z_i} . \tag{8}$$

Our $\Sigma_i J_i z_i$ and $\Sigma_i J_i^2 z_i$ correspond to Kaneyoshi's J_0 and $\frac{1}{N} \sum_k J_k^2$, respectively. We recognize therefore in Eqs (7) and (8) Kaneyoshi's formula (1). Thereby we have established the equivalence of Handrich's and Kaneyoshi's approaches to the question — in the limits of known approximations. The generalization to arbitrary spins can be derived easely.

2. Magnetization of amorphous ferromagnetics

We shall now consider a magnetic metal, pure but amorphous. The structure of amorphous metals is very like that of the liquid state. It can be described by the radial distribution function (RDF) in conjunction with the statistical distribution $f(l)$ of the coordination number l fluctuating about its mean value $\langle z \rangle$ [5]. If we assume after Ref. [3] that very roughly $J(r) = J$ for nearest neighbours, our further considerations can run parallel to those of Section 1. We have, however, to keep in mind that now the number l of nearest neighbours cannot exceed a maximum value z_{max}, as before, but contrary to the previous case l cannot be less than $z_{min} \neq 0$. We approximate the distribution $f(l)$ by a binomial distribution $P'(l) = P(l - z_{min}) = c^{l-z_{min}} (1-c)^{z_{max}-l} L! / (l - z_{min})! (z_{max} - l)!$, (where $L = z_{max} - z_{min}$), adjusted to the first two moments of the distribution $f(l)$ and the same minimum coordination number z_{min}. Given $f(l)$, one thus obtains the values of the three parameters: z_{min}, L and c (the latter, however, can no longer be interpreted as a concentration!). Eq. (4) still holds in the present case if we substitute $\langle z \rangle = z_{min} + cL$ and $\langle (\Delta z)^2 \rangle = Lc(1-c)$ (this is to be compared with cz and $zc(1-c)$, respectively, for a crystalline dilute alloy).[1] Thus, a direct application of Kaneyoshi's theory to noncrystalline magnetics (as he seems to do [1]) requires the above assumptions (or their equivalent), mainly the existence of a minimum coordination number z_{min}.

---

[1] For comparison, we refer to Eqs (7) and (8) rather than to Eqs (1) and (2) because of difficulty in defining the Fourier-transform J_k in a non-crystalline material.

214

3. Critical concentration of an amorphous dilute binary alloy

Hitherto, we refrained from considering in detail the dependence of the exchange integral $J(r)$ on the interatomic distance r and assumed a single constant value of J in every coordination sphere. As a matter of fact, a main difficulty in the interpretation of experimental data on amorphous ferromagnetics resides in our unsatisfactory knowledge of this dependence. It is worthwhile, therefore, to search for effects connected with ferromagnetism, but n o t with the value of the exchange integral. It is well known that the critical concentration c_c of magnetic atoms in a non-magnetic host below which the ferromagnetism of a crystalline binary alloy disappears, does not depend on the value of J [6, 7], but only on the co-ordination number of the crystal and on the range of J. We ask whether this is true for amorphous alloys also.

So far, we used the molecular field approximation; as well known [1, 6], this approximation does not yield any critical concentration except for $c = 0$. Therefore, we apply the so-called "cluster variation method" [6] to an amorphous magnetic alloy containing one non-magnetic and one magnetic component (as in Section 1, of spin $S = 1/2$ and concentration c). As pointed out in Section 2, the number of nearest neighbours l varies from one atom to another, and so does the interatomic distance. The number of l-neighboured atoms can be described by a probability $f(l)$ of finding them in the sample. Now, we define nearest neighbours as those atoms (magnetic or not) which lie closer to a given atom than the range r_m of magnetic interaction, within which the exchange integral $J(r)$ is appreciably different from zero (Fig. 1). Let their mean number be $\langle z \rangle_m$. This "magnetic coordination number" must not necessarily be the same (at least in "liquidlike" structures)
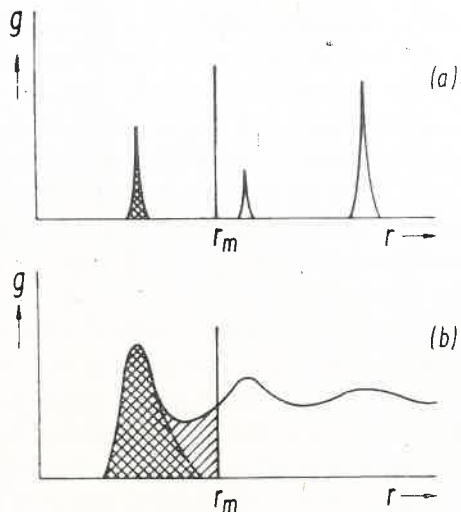

Fig. 1. Numerical density g of atoms in function of interatomic distance r, schematically: (a) for a crystal, (b) for an amorphous medium, compared with the hypothetical range of the exchange integral $J(r)$, very schematically ($J(r) = 0$ for $r > r_m$). The cross-wise shaded area is related to z_{str} — the usual coordination number, the cross-wise plus line-shaded area up to $r = r_m$ is related to $\langle z \rangle_m$. For a crystal, as obvious from (a), $\langle z \rangle_m = z_{str}$ (the areas in graphs (a) and (b) are n o t to scale mutually)

as the "structural" coordination number z_{str}, as would in fact be the case if the magnetic interaction covered neighbours situated near "interstitial" positions between first and second neighbours. A justification for using the nearest neighbours approximation can be found in Felsch's [9] and Hasegawa's [10] results, who established experimentally that the magnetic properties of amorphous alloys depend essentially on local ordering only. The Curie temperature[2] as obtained from the relation $\chi^{-1} = 0$ (χ — susceptibility) is:

$$T_c = \frac{\langle J\rangle_{am}}{k}\left[\log\frac{(\langle z\rangle-1)c+1}{(\langle z\rangle-1)c-1}\right]^{-1}.\qquad(9)$$

Here, the exchange integral is averaged over the radial distribution function and the coordination number over its statistical distribution $f(l)$. We believe that $\langle z\rangle = \langle z\rangle_{am}$ should be equal $\langle z\rangle_m$ rather than z_{str}.

We see that T_c, in an amorphous magnetic, depends on $\langle J\rangle_{am}$ and $\langle z\rangle_{am}$. For $\langle J\rangle_{am} = J_{cryst}$ and for e.g. $\langle z\rangle_{am}$ smaller than z_{cryst}, one would have $T_{c\,am} < T_{c\,cryst}$[3]. We find the critical concentration c_c by noting that $c \to c_c$ as $T_c \to 0$ in the preceding formula. Indeed, c_c does not depend on the value of the exchange integral, but only on one parameter, $\langle z\rangle$. The latter could be estimated from structural (e.g. X-rays) investigations, were one to assume $\langle z\rangle_m = z_{str}$.

The only studies of amorphous magnetic dilute alloys near the critical concentration are due to the Keck Laboratory group [8, 10] for PdFeSi alloy, which is a three-component system, whereas our considerations concerned binary alloys only. Further experimental work would therefore be welcome.

The author wishes to express his deepest gratitude to Professor L. Néel for reception at his Laboratoire d'Electrostatique et de Physique du Métal at Grenoble, where this work was begun. Also, acknowledgement is due to Dr B. K. Chakraverty for his discussions and for the initial impulse to study amorphous ferromagnets, as well as to Docent Dr L. Kowalewski and Docent Dr H. Cofta, who moreover critically read the manuscript.

REFERENCES

[1] T. Kaneyoshi, *Prog. Theor. Phys.*, **44**, 328 (1970).
[2] K. Handrich, *Phys. Status Solidi*, **32**, K55 (1969).
[3] K. Handrich, S. Kobe, *Acta Phys. Polon.*, **A38**, 819 (1970).
[4] T. G. Blocker, F. G. West, *Phys. Letters*, **28A**, 487 (1969).
[5] L. V. Woodcock, *Z. Naturforsch.*, **26a**, 287 (1971).
[6] H. Sato, A. Arrott, R. Kikuchi, *J. Phys. Chem. Solids*, **10**, 19 (1959).
[7] M. Coopersmith, R. Brout, *J. Phys. Chem. Solids*, **17**, 254 (1961).
[8] C. C. Tsuei, P. Duwez, *J. Appl. Phys.*, **37**, 435 (1966).
[9] W. Felsch, *Z. Angew. Phys.*, **29**, 217 (1970); **30**, 275 (1970).
[10] R. Hasegawa, *J. Appl. Phys.*, **41**, 4096 (1970).

---

[2] Here, unlike Sections 1 and 2, the influence of structural fluctuations is not taken into account.

[3] The formulas valid for a crystal are similar, the only difference residing in the nonoccurrence of the averaging brackets $\langle\ \rangle$; cf. [6].