

Chemical and magnetic ordering in amorphous systems

Ludwik Dąbrowski

Institute of Nuclear Chemistry and Technology, Department of Structural Research, Dorodna 16, PL-03-195 Warsaw, Poland

(Received 27 July 1990)

A statistical method for the analysis of the chemical and magnetic ordering in multicomponent amorphous substances is developed. The model assumes that the amorphous state is that of a crystal with a high level of defects. The Hamiltonian allows for the existence of multiparticle and multispin interaction potentials. The magnetic interactions are of the Ising type. The spin value is not limited. Multiparticle radial distribution functions and multispin correlation functions are used for the description of the atomic and magnetic structure. The internal energy is represented as a low-temperature expansion.

I. INTRODUCTION

The fluctuations of the local atomic ordering in solid solutions cause fluctuations in the electronic structure, including the magnetic interactions. Thus, for instance, using Mössbauer spectroscopy, in Ref. 1 has been proved that in the case of iron these fluctuations appear in the first and second configuration shells. The inverse phenomenon—a change in the atomic ordering connected with the magnetic interactions—is also well known (Refs. 2 and 3). From a theoretical point of view, the problem is very complicated and, therefore, rather rarely undertaken, especially for amorphous substances. Naturally, the consideration of such problems needs certain model approximations.

In this paper we restrict ourselves to interactions of the Ising type (including the multispin interactions), which depend on the local atomic and spin configurations. Regarding the chemical interactions we assume the possibility of the existence of multiparticle interactions. All possible types of atomic and spin fluctuations were taken into account for the determination of the atomic and magnetic structure.

The description of amorphous substances is closely related to the structural model assumed. Our investigation is based on a model of a crystal with an arbitrary number of different structural defects. These defects are regarded as new kinds of atoms, and the difference between these and the real atoms is such that their total number is not fixed but is controlled by the conditions of thermal equilibrium. This model has been discussed in detail in a pre-

vious paper,⁴ but here we apply the method of a low-temperature expansion of the internal energy of the system so that our approximation can cover a larger temperature range.

II. LOW-TEMPERATURE EXPANSION

By a low-temperature range region we mean a range region, where the normalized (by kT) potentials of the chemical or magnetic interaction are much greater than unity.

The basic idea of the expansion proposed in this paper can be reduced to a definite operator transformation, which automatically leads to the renormalization of the terms in the expansion of the internal energy of the system.

Let us define the operator \hat{X}

$$\hat{X} = \frac{\hat{Z} - 1}{\hat{Z} + 1}, \quad (1)$$

where $\hat{Z} = \exp(-\hat{H}/kT)$ and \hat{H} is the Hamiltonian of the system; k is Boltzmann's constant and T is the absolute temperature. Taking into account the identities

$$\frac{1}{\hat{Z} + 1} = \frac{1}{2}(1 - \hat{X}), \quad (2)$$

$$\Delta Z = \frac{2}{1 - \hat{X}} - (1 + \langle \hat{Z} \rangle).$$

where $\Delta Z = \hat{Z} - \langle \hat{Z} \rangle$, we obtain

$$\hat{X} = \frac{\langle \hat{Z} \rangle - 1}{\langle \hat{Z} \rangle + 1} + \frac{1}{\langle \hat{Z} \rangle + 1} \left[\Delta Z (1 - \langle \hat{X} \rangle) + (\hat{X} - \langle \hat{X} \rangle) \left(1 + \langle \hat{Z} \rangle - \frac{2}{1 - \hat{X}} \right) \right]. \quad (3)$$

Using an expression for the internal energy given in Ref. 4 in the form

$$U = -kT \ln \langle \exp(-\hat{H}/kT) \rangle = -kT \ln \langle \hat{Z} \rangle, \quad (4)$$

we obtain

$$U = -kT \left[\ln \left\langle 1 + \hat{X} + \frac{2(\hat{X} - \langle \hat{X} \rangle)}{1 - \hat{X}} \right\rangle - \ln \langle 1 - \hat{X} \rangle \right]. \quad (5)$$

It is evident from Eq. (5) that the internal energy can be represented as a series in powers of the operator \hat{X} with re-normalized expansion coefficients (see Sec. III).

III. INTERNAL ENERGY OF THE SYSTEM

The assumed according to Ref. 4 model treats the amorphous substance as a crystal, whose sites are occupied by atoms or defects. This means that the Hamiltonian of the system can be expressed in a form analogical to the Hamiltonian of a crystal (Ref. 5), namely

$$\begin{aligned} \hat{H} = & - \sum_i \sum_{\mu} \gamma_i^{\mu} \hat{S}_i^{\mu} \hat{C}_i^{\mu} + \frac{1}{2} \sum_{i,j} \sum_{\mu,\nu} (-J_{ij}^{\mu\nu} \hat{S}_i^{\mu} \hat{S}_j^{\nu} + V_{ij}^{\mu\nu}) + \dots \\ & + \frac{1}{n!} \sum_{i,j,\dots,k} \sum_{\mu,\nu,\dots,\lambda} (-J_{ij\dots k}^{\mu\nu\dots\lambda} \hat{S}_i^{\mu} \hat{S}_j^{\nu} \dots \hat{S}_k^{\lambda} + V_{ij\dots k}^{\mu\nu\dots\lambda}) \hat{C}_i^{\mu} \hat{C}_j^{\nu} \dots \hat{C}_k^{\lambda} + \dots, \end{aligned} \quad (6)$$

where \hat{S}_i^{μ} and \hat{C}_i^{μ} are spin and concentration operators, respectively, $J_{ij\dots k}^{\mu\nu\dots\lambda}$ and $V_{ij\dots k}^{\mu\nu\dots\lambda}$ are magnetic and chemical interaction potentials.

The first term in Eq. (6) represents an interaction with an external magnetic field, the second represents two-particle and two-spin interactions, and the next, the general term, represents arbitrary multiparticle and multispin interactions accounting for the local atom and magnetic configurations.

Following Ref. 5 we introduce operators of the pseudoconcentration $\hat{C}_i^{\mu\omega}$, which satisfy the relations

$$\hat{S}_i^{\mu} = \sum_{\omega} S_{\omega}^{\mu} \hat{C}_i^{\mu\omega}, \quad (7)$$

where S_{ω}^{μ} represent the consecutive projections of the spin operator on the axis of quantization.

The expressions for \hat{Z} can thus be rewritten in the form of:

$$\begin{aligned} \hat{Z} = & 1 + \sum_i \sum_{\mu} \hat{C}_i^{\mu} \sum_{\omega} Z_{i,\omega}^{\mu} \hat{C}_i^{\mu\omega} + \frac{1}{2} \sum_{i,j} \sum_{\mu,\nu} \hat{C}_i^{\mu} \hat{C}_j^{\nu} \left[Z_{ij}^{\mu\nu} + \sum_{\omega} Z_{ij,\omega}^{\mu\nu} \hat{C}_i^{\mu\omega} + \sum_{\omega,\Omega} Z_{i,j,\omega\Omega}^{\mu\nu} \hat{C}_i^{\mu\omega} \hat{C}_j^{\nu\Omega} \right] \\ & + \dots \frac{1}{n!} \sum_{i,j,\dots,k} \sum_{\mu,\nu,\dots,\lambda} \hat{C}_i^{\mu} \hat{C}_j^{\nu} \dots \hat{C}_k^{\lambda} \left[Z_{ij\dots k}^{\mu\nu\dots\lambda} + \sum_{\omega} Z_{ij\dots k,\omega}^{\mu\nu\dots\lambda} \hat{C}_i^{\mu\omega} + \dots \right. \\ & \left. + \sum_{\omega,\Omega,\dots,\rho} Z_{ij\dots k,\omega\Omega\dots\rho}^{\mu\nu\dots\lambda} \hat{C}_i^{\mu\omega} \hat{C}_j^{\nu\Omega} \dots \hat{C}_k^{\lambda\rho} \right] + \dots, \end{aligned} \quad (8)$$

where

$$\begin{aligned} Z_{i,\omega}^{\mu} &= \exp(\gamma_i^{\mu} S_{\omega}^{\mu} / kT) - 1, \\ Z_{ij}^{\mu\nu} &= \exp(-V_{ij}^{\mu\nu} / kT) - 1, \\ Z_{i,j,\omega}^{\mu\nu} &= \exp[(-V_{ij}^{\mu\nu} + \gamma_i^{\mu} S_{\omega}^{\mu}) / kT] - 1 - Z_{i,\omega}^{\mu} - Z_{ij}^{\mu\nu}, \\ Z_{i,j,\omega\Omega}^{\mu\nu} &= \exp[(J_{ij}^{\mu\nu} S_{\omega}^{\mu} S_{\Omega}^{\nu} - V_{ij}^{\mu\nu} + \gamma_i^{\mu} S_{\omega}^{\mu} + \gamma_j^{\nu} S_{\Omega}^{\nu}) / kT] - 1 - Z_{i,\omega}^{\mu} - Z_{j,\Omega}^{\nu} - Z_{ij}^{\mu\nu} - Z_{i,j,\omega}^{\mu\nu} - Z_{i,j,\Omega}^{\mu\nu}, \\ Z_{ij\dots k}^{\mu\nu\dots\lambda} &= \exp[(-V_{ij\dots k}^{\mu\nu\dots\lambda} - V_{ij}^{\mu\nu} - V_{jk}^{\nu\lambda} - V_{ki}^{\lambda\mu}) / kT] - 1 - Z_{ij}^{\mu\nu} - Z_{jk}^{\nu\lambda} - Z_{ki}^{\lambda\mu}. \end{aligned}$$

The arbitrary coefficient $Z_{ij\dots k,\omega\Omega\dots\rho}^{\mu\nu\dots\lambda}$, where there are n terms $i-k$ and m terms $\omega-\rho$, is constructed in the following way.

(1) Construct a sum, symmetric in all indices of the type i,μ,ω , of all possible expressions of the type $V_{ij\dots k}^{\mu\nu\dots\lambda} / kT$ and $J_{ij\dots k}^{\mu\nu\dots\lambda} S_{\omega}^{\mu} S_{\Omega}^{\nu} \dots S_{\rho}^{\lambda} / kT$, where there are p terms $i-k$ and q terms $\omega-\rho$, for which $2 \leq p \leq n$, $1 \leq q \leq m$.

(2) In the next step exponentiate of this sum, subtract 1 from the result as well as all possible combinations of the expressions $Z_{ij\dots k,\omega\Omega\dots\rho}^{\mu\nu\dots\lambda}$, where there are s terms $i-k$ and t terms $\omega-\rho$ for which the condition $s < n$, $t \leq m$, and $s + t < n + m$ holds.

In this way formula (8) gives the expansion of \hat{Z} in a series of the operators of concentration and pseudoconcentration. The structure of the expansion of $(\hat{Z})^n$, for n arbitrary, is the same:

$$(\hat{Z})^n = 1 + \sum_i \sum_{\mu} \hat{C}_i^{\mu} \sum_{\omega} Z_i^{(n)\mu\omega} \hat{C}_i^{\mu\omega} + \dots \frac{1}{n!} \sum_{i,j,\dots,k} \sum_{\mu,\nu,\dots,\lambda} \hat{C}_i^{\mu} \hat{C}_j^{\nu} \dots \hat{C}_k^{\lambda} \left[Z_{ij\dots k}^{(n)\mu\nu\dots\lambda} + \sum_{\omega} Z_{ij\dots k,\omega}^{(n)\mu\nu\dots\lambda} \hat{C}_i^{\mu\omega} + \dots \right] + \dots \quad (9)$$

After transformations it can easily be seen that all coefficients $Z_{ij\dots k,\omega\Omega\dots\rho}^{(n)\mu\nu\dots\lambda}$ in formula (9) can be calculated from the corresponding coefficients $Z_{ij\dots k,\omega\Omega\dots\rho}^{\mu\nu\dots\lambda}$ by means of the transformation $\exp(\dots) \rightarrow [\exp(\dots)]^n$. This rule will be used later for the determination of \hat{X} .

Expanding \hat{X} , defined by formula (1), in power series we obtain

$$\hat{X} = 1 + 2 \sum_{n=1}^{\infty} (-1)^n (\hat{Z})^n. \quad (10)$$

Keeping in mind formulas (8) and (9) we obtain

$$\hat{X} = \sum_i \sum_{\mu} \hat{C}_i^{\mu} \sum_{\omega} X_{i,\omega}^{\mu} \hat{C}_i^{\mu\omega} + \dots + \frac{1}{n!} \sum_{i,j,\dots,k} \sum_{\mu,\nu,\dots,\lambda} \hat{C}_i^{\mu} \hat{C}_j^{\nu} \dots \hat{C}_k^{\lambda} \left[X_{ij\dots k}^{\mu\nu\dots\lambda} + \sum_{\omega} X_{ij\dots k,\omega}^{\mu\nu\dots\lambda} \hat{C}_i^{\mu\omega} + \dots + \sum_{\omega,\Omega,\dots,\rho} X_{ij\dots k,\omega\Omega\dots\rho}^{\mu\nu\dots\lambda} \hat{C}_i^{\mu\omega} \hat{C}_j^{\nu\Omega} \dots \hat{C}_k^{\lambda} \right]. \quad (11)$$

where $X_{ij\dots k,\omega\Omega\dots\rho}^{\mu\nu\dots\lambda} = 2 \sum_{n=1}^{\infty} (-1)^n Z_{ij\dots k,\omega\Omega\dots\rho}^{(n)\mu\nu\dots\lambda}$.

The power series, defining $X_{ij\dots k,\omega\Omega\dots\rho}^{\mu\nu\dots\lambda}$ in expression (11), can be summed, and as a result we obtain directly, for the first three coefficients,

$$X_{i,\omega}^{\mu} = \tanh(\gamma_i^{\mu} S_{\omega}^{\mu} / 2kT), \quad X_{ij}^{\mu\nu} = \tanh(-V_{ij}^{\mu\nu} / 2kT), \quad X_{i,\omega}^{\mu\nu} = \tanh[(-V_{ij}^{\mu\nu} + \gamma_i^{\mu} S_{\omega}^{\mu}) / 2kT] - X_{i,\omega}^{\mu} - X_{ij}^{\mu\nu}. \quad (12)$$

Further, it is again easy to note that these coefficients $X_{ij\dots k,\omega\Omega\dots\rho}^{\mu\nu\dots\lambda}$ can be obtained formally from the coefficients $Z_{ij\dots k,\omega\Omega\dots\rho}^{\mu\nu\dots\lambda}$, defined in expression (8), through the transformations $Z \rightarrow X$ and $\exp(\dots) - 1 \rightarrow \tanh[(\dots)/2]$. This rule holds also for all remaining coefficients $X_{ij\dots k,\omega\Omega\dots\rho}^{\mu\nu\dots\lambda}$.

This algorithm allows the coefficients under consideration to be obtained in a recursive way, which is equivalent to the diagram technique widely used in the theory of ordering.

IV. FREE ENERGY

The internal energy is defined by formulas (5) and (11). For the configuration, entropy formulas given in Ref. 5 are valid, namely,

$$S = \sum_n (S_a^n + S_m^n), \quad (13)$$

where S_a^n and S_m^n are the n particle and n spin entropies, respectively. They are of the following form:

$$S_a^n = -\frac{k}{n!} \sum_{i,j,\dots,k} \sum_{\mu,\nu,\dots,\lambda} p_{ij\dots k}^{\mu\nu\dots\lambda} \ln \left[\frac{p_{ij\dots k}^{\mu\nu\dots\lambda}}{p_{ij\dots k}^{\mu\nu\dots\lambda} - g_{ij\dots k}^{\mu\nu\dots\lambda}} \right], \quad (14)$$

$$S_m^n = -\frac{k}{n!} \sum_{i,j,\dots,k} \sum_{\mu,\nu,\dots,\lambda} p_{ij\dots k}^{\mu\nu\dots\lambda} \sum_{\omega,\Omega,\dots,\rho} P_{ij\dots k,\omega\Omega\dots\rho}^{\mu\nu\dots\lambda} \ln \left[\frac{P_{ij\dots k,\omega\Omega\dots\rho}^{\mu\nu\dots\lambda}}{P_{ij\dots k,\omega\Omega\dots\rho}^{\mu\nu\dots\lambda} - G_{ij\dots k,\omega\Omega\dots\rho}^{\mu\nu\dots\lambda}} \right],$$

where $p_{ij\dots k}^{\mu\nu\dots\lambda}$, $g_{ij\dots k}^{\mu\nu\dots\lambda}$, $P_{ij\dots k,\omega\Omega\dots\rho}^{\mu\nu\dots\lambda}$, and $G_{ij\dots k,\omega\Omega\dots\rho}^{\mu\nu\dots\lambda}$ are the probabilities and correlation functions of chemical and magnetic clusters, respectively.

The lattice sums in the expressions (11) and (14) are defined explicitly only in the case of an ideal crystal. In our case the limits of the coordination spheres are somewhat diffuse and the number of atomic sites in each of them is not fixed. Therefore, we must use integration instead of summation. Let us introduce the following notations

$$C^{\mu} = N^{\mu} / N, \quad C^{\kappa} = N^{\kappa} / N, \quad N = \sum_{\mu} N^{\mu} + \sum_{\kappa} N^{\kappa}, \quad \eta = \left[\sum_{\mu} N^{\mu} \right] / N, \quad (15)$$

where N^{μ} and N^{κ} are the common numbers of atoms of the type μ and defects of the type κ in the system.

From the definitions (15) and the condition of the isotropy of the amorphous substance as a whole, it follows that

$$\sum_{\kappa} C^{\kappa} = 1/\eta - 1, \quad \langle \hat{C}_i^{\mu} \rangle = C^{\mu}, \quad \langle \hat{C}_i^{\mu\omega} \rangle = C^{\mu\omega}. \quad (16)$$

Taking into account formulas (5), (11), (15), and (16) we obtain, finally,

$$\begin{aligned}
U = -\frac{2kT}{\eta} & \left[\sum_{\mu} C^{\mu} \sum_{\omega} X_{\omega}^{\mu} C^{\mu\omega} + \dots \right. \\
& + \frac{1}{n!} \sum_{\mu, \nu, \dots, \lambda} \frac{1}{V_{\nu}} \dots \frac{1}{V_{\lambda}} \\
& \times \int_0^{\infty} \dots \int_0^{\infty} p^{\mu}(\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda}) \left\{ X^{\mu}(\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda}) + \sum_{\omega} X_{\omega}^{\mu}(\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda}) C^{\mu\omega} \right. \\
& \left. + \dots + \sum_{\omega, \Omega, \dots, \rho} X_{\omega\Omega \dots \rho}^{\mu}(\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda}) P_{\omega\Omega \dots \rho}^{\mu}(\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda}) \right\} \\
& \left. \times d\mathbf{r}^{\nu} \dots d\mathbf{r}^{\lambda} \right]. \tag{17}
\end{aligned}$$

where the coefficients $X_{\omega\Omega \dots \rho}^{\mu}(\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda})$ are constructed from the corresponding coefficients $X_{ijk, \omega\Omega \dots \rho}^{\mu\nu \dots \lambda}$ by means of the transformation $V_{ij \dots k}^{\mu\nu \dots \lambda} \rightarrow V^{\mu}(\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda})$ and $J_{ij \dots k}^{\mu\nu \dots \lambda} \rightarrow J^{\mu}(\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda})$. Here $p^{\mu}(\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda})$ denotes the probability for forming a multiparticle cluster, composed of atoms of the type $\nu \dots \lambda$ placed at the points, defined by the vectors $\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda}$, along with an atom μ (placed at the beginning of the coordination system). $P_{\omega\Omega \dots \rho}^{\mu}(\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda})$ describes the probability of forming the corresponding magnetic cluster. The normalization constants $V_{\nu} \dots V_{\lambda}$ have the meaning of average volumes occupied by the atoms or defects, respectively. The summation is carried out over all types of atoms and defects.

A procedure of integration, related to formulas (14) gives the following expressions for the entropy:

$$\begin{aligned}
S_a^n &= -\frac{kN}{\eta} \frac{1}{n!} \sum_{\mu, \nu, \dots, \lambda} \frac{1}{V_{\nu}} \dots \frac{1}{V_{\lambda}} \\
& \times \int_0^{\infty} \dots \int_0^{\infty} p^{\mu}(\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda}) \ln \left[1 + \frac{g^{\mu}(\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda})}{p^{\mu}(\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda}) - g^{\mu}(\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda})} \right] d\mathbf{r}^{\nu} \dots d\mathbf{r}^{\lambda}, \\
S_m^n &= -\frac{kN}{\eta} \frac{1}{n!} \sum_{\mu, \nu, \dots, \lambda} \frac{1}{V_{\nu}} \dots \frac{1}{V_{\lambda}} \\
& \times \int_0^{\infty} \dots \int_0^{\infty} p^{\mu}(\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda}) \\
& \times \sum_{\omega, \Omega, \dots, \rho} P_{\omega\Omega \dots \rho}^{\mu}(\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda}) \\
& \times \ln \left[1 + \frac{G_{\omega\Omega \dots \rho}^{\mu}(\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda})}{P_{\omega\Omega \dots \rho}^{\mu}(\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda}) - G_{\omega\Omega \dots \rho}^{\mu}(\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda})} \right] d\mathbf{r}^{\nu} \dots d\mathbf{r}^{\lambda}, \tag{18}
\end{aligned}$$

where $g^{\mu}(\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda})$ and $G_{\omega\Omega \dots \rho}^{\mu}(\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda})$ are the corresponding atomic and magnetic correlation functions. The other notations are the same as in formulas (17).

Formulas (17) and (18) are basic for the calculation of the parameters of the atomic and magnetic ordering—in our case η , $g^{\mu}(\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda})$, and $G_{\omega\Omega \dots \rho}^{\mu}(\mathbf{r}^{\nu} \dots \mathbf{r}^{\lambda})$. Their values are defined by the minimization of the free energy.

V. CONCLUSIONS

It should be mentioned that if magnetic and chemical interactions coexist in a given system, the critical temperatures of the magnetic transformation and of the order-disorder transformation may differ significantly. For this reason it is very important that the expansions in the chemical and magnetic potentials should hold simultaneously in a given temperature range. For this purpose a low-temperature expansion procedure was introduced. This possibility was not taken into account in our previous paper.⁴

It is well known that the number of the diagrams, widely used in the theory of ordering, rapidly increase with the consecutive approximations. The latter strongly restricts the achievement of the desired accuracy. Furthermore, this is valid for such complex systems as discussed above. In Ref. 4 we proposed, and in this paper we generalized, a new technique [see formula (12)], which gives a simpler way for the calculation of the coefficients of the internal energy in a recursive way. This method essentially increases the number of the expansions accounted for in real calculations, which leads to a better accuracy of the results obtained.

ACKNOWLEDGMENTS

The author is thankful to Professor J. Leciejewicz for his valuable comments and suggestions. This work was sponsored by the Central Research Program No. 01.09.

- ¹G. K. Wertheim, V. Jaccarino, J. H. Wernick, and D. N. E. Buchman, *Phys. Rev. Lett.* **12**, 25 (1964).
- ²F. Mejia-Lira and J. Urias, *Phys. Rev. B* **24**, 5270 (1981).
- ³V. A. Volkov, S. I. Masharov, A. F. Ribalko, and N. I. Timofeev, *Fiz. Met. Metalloved.* **60**, 262 (1985).
- ⁴L. Dąbrowski, *Z. Phys. B* **77**, 421 (1989).
- ⁵L. Dąbrowski, *J. Magn. Magn. Mater.* **73**, 184 (1988).