The magnetisation and the correlation functions in thin diluted films (Ising model, $S=^{1}/_{2}$)

1990 J. Phys.: Condens. Matter 2 3955

(http://iopscience.iop.org/0953-8984/2/17/006)

J. Phys.: Condens. Matter 2 (1990) 3955–3966. Printed in the UK

The magnetisation and the correlation functions in thin diluted films (Ising model, $S = \frac{1}{2}$)

T Balcerzak†, J Mielnicki‡, G Wiatrowski† and A Urbaniak-Kucharczyk†

† Institute of Physics, University of Łódź, 90-236 Łódź, Nowotki 149/153, Poland

‡ Institute of Physics, Polish Academy of Sciences, 02-668 Warsaw, Al. Lotników 32, Poland

Received 11 April 1989, in final form 11 October 1989

Abstract. The thin diluted ferromagnetic film with SC symmetry is considered in the third-order Matsudaira approximation. The equations for the description of magnetisation and various correlation functions have been derived and the results of the numerical calculations have been presented. In particular the critical temperature, the critical concentration, the magnetisation and various correlation functions are discussed in detail for various film thicknesses and for several magnetic atom concentrations.

1. Introduction

The Matsudaira method of calculation of Ising ferromagnet properties, developed originally for planar and bulk materials [1], predicts much more accurate results than do other theories, e.g. the molecular-field approach, or theories where only the correlations between NNs were taken into account [2–7]. In this method the set of equations for magnetisation and the correlation functions was derived on the basis of general statistical equations [8, 9] and in the calculations of the magnetisation curve the various correlation functions of spin pairs with different spin distances were taken into account. The solution of the above set of equations can be obtained by iteration, where the equations for the correlation functions are treated as successive corrections taken into account in the basic equation for magnetisation. It can be shown that a sufficient improvement in the accuracy of the results is obtained when the correlation functions for spin pairs from about six coordination zones are taken into account with further correlations decomposed to the square of magnetisation.

This procedure corresponds to the third-order Matsudaira approximation. The significant improvement in the results obtained in this approximation in comparison with the results of methods where the correlations were neglected encouraged us to develop this method for the case of thin films.

In the first attempt a thin diluted film composed of only two atomic layers was considered [10].

In the present paper the Matsudaira method is applied to the description of a thin diluted film with an arbitrary film thickness and an arbitrary concentration of magnetic atoms.

0953-8984/90/173955 + 12 $03.50

As a result, first of all, a significant improvement in the critical temperature and the critical concentration have been obtained in comparison with the values resulting from the first-order approximation. Moreover, the temperature dependence of magnetisation and various correlation functions together with their profiles across the sample have been obtained.

2. The theory

The Ising Hamiltonian for a thin diluted film can be written in the form

$$\mathcal{H} = -\frac{1}{2}\sum_{\nu j,\nu' j'} J_{\nu j,\nu' j'}\mu_{\nu j}\xi_{\nu j}\mu_{\nu' j'}\xi_{\nu' j'} \tag{1}$$

where $\mu_{\nu j} = \pm 1$ is the spin variable, $J_{\nu j,\nu' j'}$ is the exchange integral which is assumed to be different from zero for NNs only ($J_{\nu j,\nu' j'} = J$ for $\nu' j' \in \nu j$), and $\xi_{\nu j} = 0$; 1 is the occupation operator of the lattice site (νj). The position of the lattice site in thin film (νj), is given by the j-vector in the νth atomic layer.

If there is a magnetic atom in the lattice site (νj) (the so-called conditional average), then on the basis of (1) we can write the generalised Callen equation

$$\mu_\nu \equiv \langle \mu_{\nu j}\rangle_{T,r} = \left\langle \tanh\left(\beta J \sum_{\nu' j'}^{z_\nu} \mu_{\nu' j'}\xi_{\nu' j'}\right)\right\rangle_{T,r} \tag{2}$$

where $\langle \ldots \rangle_{T,r}$ denotes both thermodynamical and configurational averages while z_ν is the coordination number of the lattice site (νj). This value depends on whether the lattice site is inside the sample or on the film surface.

Similarly, the general relation for the correlation functions can be written in the form

$$C_i^{\nu\nu'} \equiv \langle \mu_{\nu j}\mu_{\nu' j'}\rangle_{T,r} = \left\langle \mu_{\nu' j'} \tanh\left(\beta J \sum_{\nu_1 j_1}^{z_\nu} \mu_{\nu_1 j_1}\xi_{\nu_1 j_1}\right)\right\rangle_{T,r} \tag{3}$$

where we have assumed that the lattice sites (νj) and $(\nu' j')$ are occupied by magnetic atoms. In (3) we have introduced the abbreviated notation according to which $C_i^{\nu\nu'}$ denotes the two-spin correlation function with the considered spin pair located in ν and ν' atomic layers, while the subscript i is related to the spin-pair distance and denotes the number of the corresponding coordination zone (for instance $i = 2$ refers to the spin-pair distance equal to $a\sqrt{2}$ in the SC crystal with the lattice constant a). The position of spin pairs in the correlation functions considered in this work is shown in figure 1.

The averages $\langle \ldots \rangle_{T,r}$ on the RHSs, of (2) and (3) are calculated in two steps. The thermodynamic average $\langle \ldots \rangle_T$ is calculated by the cumulant method. In this method the cumulant averages $\langle \ldots \rangle_c$ of the order greater than 2 are neglected and as a result the multi-spin correlations can be decoupled to the two-spin correlations [1, 10]. The configurational average $\langle \ldots \rangle_r$ can be calculated on the basis of the general relation

$$\langle \exp(\alpha \xi_{\nu j})\rangle_r = c \exp \alpha + (1 - c) \tag{4}$$

where $c = \langle \xi_{\nu j}\rangle_r$ is the magnetic atom concentration in the sample.

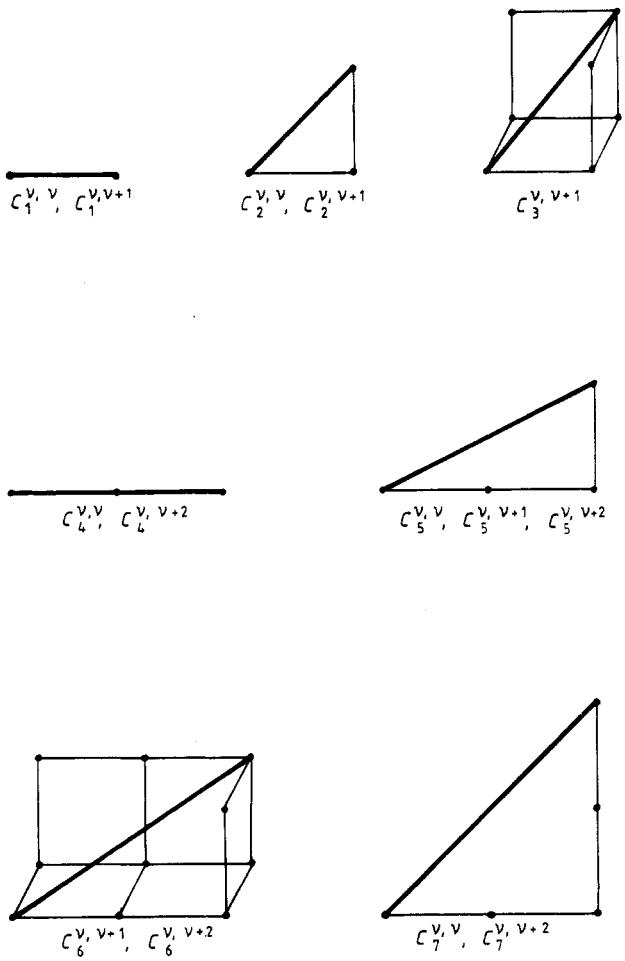

Figure 1. The position of spin pairs in the correlation function $C_i^{\nu,\nu'}$. The index i defines the coordination zone which is given by the corresponding spin-pair distance.

As a result, on the basis of equations (2) and (3), the sets of equations for magnetisation μ_ν and for various correlation functions have been obtained. For the magnetisation we have

$$\mu_\nu = B_1(\nu)\mu_{\nu-1} + 4B_2(\nu)\mu_\nu + B_3(\nu)\mu_{\nu+1} + B_4(\nu) \qquad \nu = 2, 3, \ldots, n-1 \qquad (5)$$

$$\mu_1 = 4B_2(1)\mu_1 + B_3(1)\mu_2 + B_4(1) \qquad \nu = 1 \qquad (6)$$

$$\mu_n = B_1(n)\mu_{n-1} + 4B_2(n)\mu_n + B_4(n) \qquad \nu = n. \qquad (7)$$

The coefficients $B_1(\nu), \ldots, B_4(\nu)$ depend on temperature, magnetic atom concentration and also on atomic layer magnetisation and correlations $C_2^{\nu\nu'}$ and $C_4^{\nu\nu'}$. They are listed in the appendix.

The correlation functions for NN spins are given by the relations

$$C_1^{\nu,\nu} = (2C_2^{\nu,\nu} + C_4^{\nu,\nu} + C_2^{\nu-1,\nu} + C_2^{\nu,\nu+1} + 1)b_1$$
$$+ [(2C_2^{\nu,\nu} + C_4^{\nu,\nu})(C_4^{\nu-1,\nu+1} + 3C_2^{\nu-1,\nu} + 3C_2^{\nu,\nu+1} + 1)$$

$$+ C_4^{\nu-1,\nu+1} + 3C_2^{\nu-1,\nu} + 3C_2^{\nu,\nu+1} + 6C_2^{\nu-1,\nu}C_2^{\nu,\nu+1} + 2(C_2^{\nu,\nu})^2$$
$$+ (C_4^{\nu,\nu})^2 - 6\mu_\nu^2\mu_{\nu-1}\mu_{\nu+1} - 6(\mu_{\nu-1} + \mu_{\nu+1})\mu_\nu^3 - 2\mu_\nu^4]b_2$$
$$+ [2(C_2^{\nu,\nu})^2 C_4^{\nu-1,\nu+1} + (C_4^{\nu,\nu})^2 C_4^{\nu-1,\nu+1}$$
$$+ (2C_2^{\nu,\nu} + C_4^{\nu,\nu})(C_4^{\nu-1,\nu+1} + 4C_2^{\nu-1,\nu}C_2^{\nu,\nu+1} + C_2^{\nu-1,\nu}$$
$$+ C_2^{\nu,\nu+1} - 4\mu_\nu^2\mu_{\nu-1}\mu_{\nu+1}) + 6C_2^{\nu-1,\nu}C_2^{\nu,\nu+1} - 8(C_2^{\nu-1,\nu}\mu_{\nu+1}$$
$$+ C_2^{\nu,\nu+1}\mu_{\nu-1})\mu_\nu^3 - 2C_4^{\nu-1,\nu+1}\mu_\nu^4 + 2(8\mu_\nu^2 - 3)\mu_\nu^2\mu_{\nu-1}\mu_{\nu+1}$$
$$- 2(\mu_{\nu-1} + \mu_{\nu+1})\mu_\nu^3]b_3 \tag{8}$$

for $\nu = 2, \ldots, n-1$; for the surface correlations,

$$C_1^{1,1} = (2C_2^{1,1} + C_4^{1,1} + C_2^{1,2} + 1)a_1 + [(2C_2^{1,1} + C_4^{1,1})(3C_2^{1,2} + 1)$$
$$+ 3C_2^{1,2} + 2(C_2^{1,1})^2 + (C_4^{1,1})^2 - 6\mu_1^3\mu_2 - 2\mu_1^4]a_2$$
$$+ [(2C_2^{1,1} + C_4^{1,1})C_2^{1,2} - 2\mu_1^3\mu_2]a_3 \qquad (\nu = 1) \tag{9}$$

$$C_1^{n,n} = (2C_2^{n,n} + C_4^{n,n} + C_2^{n-1,n} + 1)a_1 + [(2C_2^{n,n} + C_4^{n,n})(3C_2^{n-1,n} + 1)$$
$$+ 3C_2^{n-1,n} + 2(C_2^{n,n})^2 + (C_4^{n,n})^2 - 6\mu_n^3\mu_{n-1} - 2\mu_n^4]a_2$$
$$+ [(2C_2^{n,n} + C_4^{n,n})C_2^{n-1,n} - 2\mu_n^3\mu_{n-1}]a_3 \qquad (\nu = n). \tag{10}$$

Equations (8)–(10) define the longitudinal correlations (both spins in the same layer). For perpendicular correlations (both spins in different layers) we have

$$C_1^{\nu,\nu+1} = (C_4^{\nu-1,\nu+1} + 4C_2^{\nu,\nu+1} + 1)b_1 + [2(C_2^{\nu,\nu} + C_4^{\nu,\nu})$$
$$\times (C_4^{\nu-1,\nu+1} + 2C_2^{\nu,\nu+1} + 1) + 4C_2^{\nu-1,\nu}(3C_2^{\nu,\nu+1} + 1)$$
$$- 4(3\mu_{\nu-1} + 2\mu_\nu)\mu_\nu^2\mu_{\nu+1}]b_2 + \{[2(C_2^{\nu,\nu})^2 + (C_4^{\nu,\nu})^2]$$
$$\times (C_4^{\nu-1,\nu+1} + 1) + 4(2C_2^{\nu,\nu} + C_4^{\nu,\nu})$$
$$\times [C_2^{\nu-1,\nu}(C_2^{\nu,\nu+1} + 1) - \mu_\nu^2\mu_{\nu-1}\mu_{\nu+1}] - 8[C_2^{\nu-1,\nu}\mu_{\nu+1}$$
$$+ (C_2^{\nu,\nu+1} + 1)\mu_{\nu-1}]\mu_\nu^3 + 2(8\mu_{\nu+1}\mu_{\nu-1}$$
$$- C_4^{\nu-1,\nu+1} - 1)\mu_\nu^4\}b_3 \qquad (\nu = 2, 3, \ldots, n-1) \tag{11}$$

and

$$C_1^{1,2} = (4C_2^{1,2} + 1)a_1 + [2(2C_2^{1,1} + C_4^{1,1})(2C_2^{1,2} + 1) - 8\mu_1^3\mu_2]a_2$$
$$+ [2(C_2^{1,1})^2 + (C_4^{1,1})^2 - 2\mu_1^4]a_3 \qquad (\nu = 1). \tag{12}$$

The temperature- and concentration-dependent coefficients a_1, a_2, a_3 and b_1, b_2, b_3 are given in the appendix.

For further correlations, i.e. if the lattice sites (νj) and $(\nu' j')$ are not the NNS, then on the basis of (3) we get the general relation

$$\langle \mu_{\nu,j}\mu_{\nu',j'}\rangle = B_1(\nu)\langle \mu_{\nu-1,j}\mu_{\nu',j'}\rangle + B_2(\nu)\langle(\mu_{\nu,1} + \mu_{\nu,2}$$
$$+ \mu_{\nu,3} + \mu_{\nu,4})\mu_{\nu',j'}\rangle + B_3(\nu)\langle \mu_{\nu+1,j}\mu_{\nu',j'}\rangle + B_4(\nu)\langle \mu_{\nu',j'}\rangle \tag{13}$$

which is analogous to equation (5) and reduces to this equation for $\mu_{\nu'j'} = 1$.

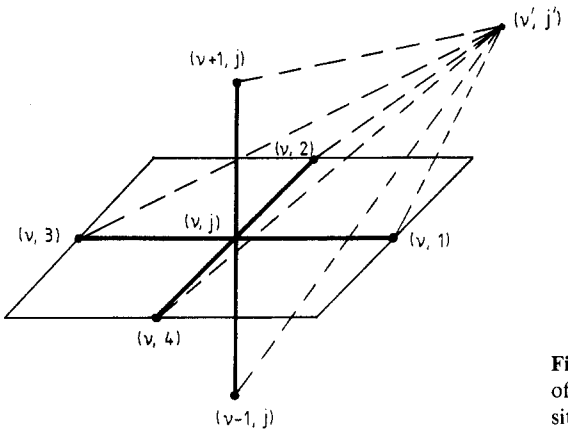

Figure 2. The spin positions used in the derivation of the correlation functions $\langle \mu_{\nu j} \mu_{\nu' j'} \rangle$. The lattice site $(\nu' j')$ is not the NN of the site (νj).

If the lattice site (νj) is on the film surface, $\nu = 1$ (or $\nu = n$), then in (13) the correlation $\langle \mu_{\nu-1,j} \mu_{\nu' j'} \rangle$ (or $\langle \mu_{\nu+1,j} \mu_{\nu' j'} \rangle$) does not appear.

With the use of (13) and figures 1 and 2 the respective equations for successive correlations $C_i^{\nu,\nu'}$ in the framework of the third Matsudaira approximation (where $C_i^{\nu,\nu'} = \mu_\nu \mu_{\nu'}$ for $i > 7$) can be easily derived.

It can be noted here that from equations (11) and (12) some asymmetry of perpendicular correlations results, for instance $C_1^{1,2} \neq C_1^{n-1,n}$. The reason for this asymmetry in the asymmetrical form of the identity (3) for non-uniform systems. However, as we have assumed symmetrical boundary conditions in the thin-film sample, this asymmetry should be removed and below we shall consider the mean correlations

$$\bar{C}_i^{\nu,\nu'} = \frac{1}{2}\Bigg[\Bigg\langle \mu_{\nu' j'} \tanh\Bigg(\beta J \sum_{(\nu_1 j_1) \in (\nu j)} \mu_{\nu_1 j_1} \xi_{\nu_1 j_1}\Bigg)\Bigg\rangle_{T,r} + \Bigg\langle \tanh\Bigg(\beta J \sum_{(\nu'_1 j'_1) \in (\nu' j')} \mu_{\nu'_1 j'_1} \xi_{\nu'_1 j'_1}\Bigg) \mu_{\nu j} \Bigg\rangle_{T,r}\Bigg] \tag{14}$$

which in our notation can also be written as

$$\bar{C}_i^{\nu,\nu'} = \tfrac{1}{2}(C_i^{\nu,\nu'} + C_i^{\nu',\nu}). \tag{15}$$

The formulae for $C_1^{\nu',\nu}$ can be derived in analogous forms to (11) and (12) from equations symmetrical to (3). This in fact corresponds simply to the following change in the indices in equations (11) and (12):

$$\begin{aligned} &\nu - 1 \rightarrow \nu' + 1 \\ &\nu \rightarrow \nu' \\ &\nu' \rightarrow \nu \\ &\nu + 1 \rightarrow \nu - 1. \end{aligned} \tag{16}$$

The solutions $\mu_\nu (\nu = 1, \ldots, n)$ of (5)–(7) can be found for a given film thickness n by iteration with successive correlation functions taken into account; hence finally the numerical values of μ_ν and of various correlations are obtained for a given temperature and concentration c. The results of these calculations are presented and discussed in the next section.

Table 1. The k_BT_c/J-values for thin films with SC symmetry obtained in the first- and third-order approximations.

n	First-order approximation	Third-order approximation	Series [13]
1	3.090	2.609	2.269
2	4.081	3.637	
3	4.479	4.070	
4	4.678	4.290	
5	4.791	4.419	
10	4.984	4.646	
∞ (bulk)	5.073	4.718	4.510 (4.428 [14])

3. Numerical results and discussion

Initially the first-order approximation was considered. In this approximation all correlations are decoupled and hence are presented as the products of corresponding magnetisations μ_ν. Thus, thin-film magnetisation can be obtained only with the use of equations (5)–(7) and after calculations we obtained the same results as those presented in [11, 12].

Then, on the basis of the results of the first-order approximation the correlations $\bar{C}_1^{\nu,\nu'}$ can be found, with all further correlations decoupled. Once $\bar{C}_1^{\nu,\nu'}$ are known, then also $\bar{C}_2^{\nu,\nu'}$ and $\bar{C}_4^{\nu,\nu'}$ can be obtained and in this way we can calculate successive correlations on the basis of previously calculated ones for nearer spins.

In the approximation corresponding to the third-order Matsudaira approximation we calculated correlations up to $C_7^{\nu,\nu}$ and $\bar{C}_7^{\nu,\nu+2}$. Then, with these correlations known, we can successively correct the correlations $\bar{C}_6^{\nu,\nu'}$, $\bar{C}_5^{\nu,\nu'}$, . . . up to finally $\bar{C}_1^{\nu,\nu'}$. On the basis of all these correlations the magnetisation has been calculated once more, which in turn enables further correction of correlations. This, in turn, enables calculation of new magnetisation values. This procedure is repeated until the difference between successive results for μ_ν becomes sufficiently small.

In table 1 the values of the reduced critical temperature k_BT_c/J calculated in the first- and third-order Matsudaira approximations are given for several films with different film thickness parameters n. For $n = 1$ (the single plane) and for $n = \infty$ (bulk) the values of k_BT_c/J obtained are compared with the most accurate results known from the literature [13]. As can be seen, the k_BT_c/J-values resulting from the third-order approximation are much closer to the exact values than those obtained in the first-order approximation. From table 1 it is also seen that the critical temperature is particularly strongly dependent on the film thickness for small n-values. Next, the critical concentration $c_0(n)$ for various film thicknesses given by the parameter n has been calculated in the first- and third-order approximation (table 2).

As can be seen, $c_0(n)$ also strongly depends on n for small film thicknesses. It can be noted also that our results for $n = 1$ (single plane) and for $c = \infty$ are the same as those obtained in [1].

In figure 3 the magnetisation profiles for film composed of five atomic layers are given for several values of the reduced temperature T/T_c and for two values of concentration c.

Table 2. The critical concentration values for films with various numbers n of atomic planes.

n	$c_0(n)$		
	First-order approximation	Third-order approximation	From [15]
1	0.4284	0.4860	0.57
2	0.3479	0.3805	
3	0.3245	0.3496	
4	0.3140	0.3360	
5	0.3082	0.3286	
10	0.2981	0.3153	
∞ (bulk)	0.2929	0.3110	0.33

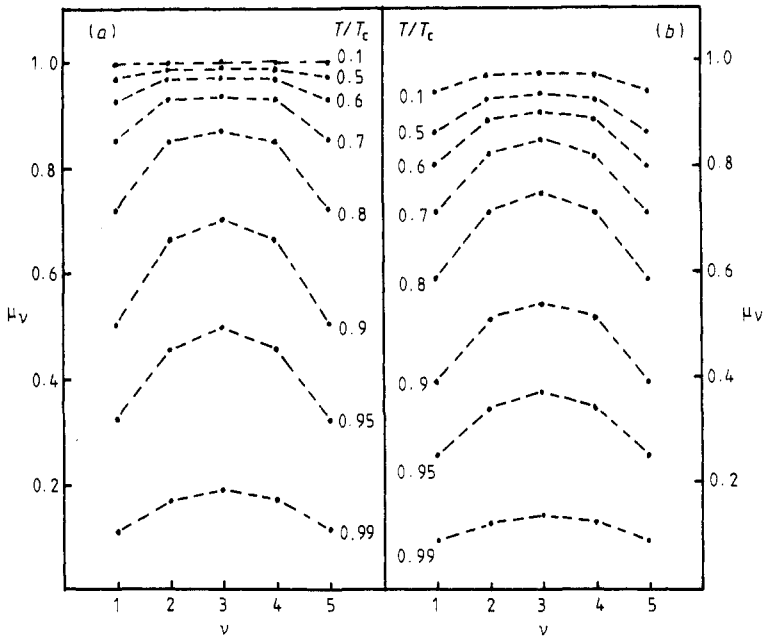

Figure 3. The magnetisation profiles for a film composed of five atomic planes: (*a*) the perfect crystal case ($c = 1$); (*b*) the diluted crystal with $c = 0.5$.

Figure 3(*a*) presents the results for $c = 1$ (with $k_B T_c/J = 4.419$) while in figure 3(*b*) the results obtained for $c = 0.5$ (with $k_B T_c/J = 1.617$) are shown.

These profiles and all further results were obtained in the third-order Matsudaira approximation, where the correlation functions up to the seventh coordination zone have been taken into account.

In figures 4 and 5 some of the correlation functions calculated for the same film thickness ($n = 5$) are given.

In figure 4(*a*) the profiles of several longitudinal correlations ($C_i^{\nu,\nu}$, $i = 1, 2, 4$ and 7) calculated for $T = 0.9T_c$ are shown while figure 4(*b*) presents some of the perpendicular correlations ($\bar{C}_i^{\nu-1/2,\nu+1/2}$, $i = 1,2,3,6$). The values for these last correlations

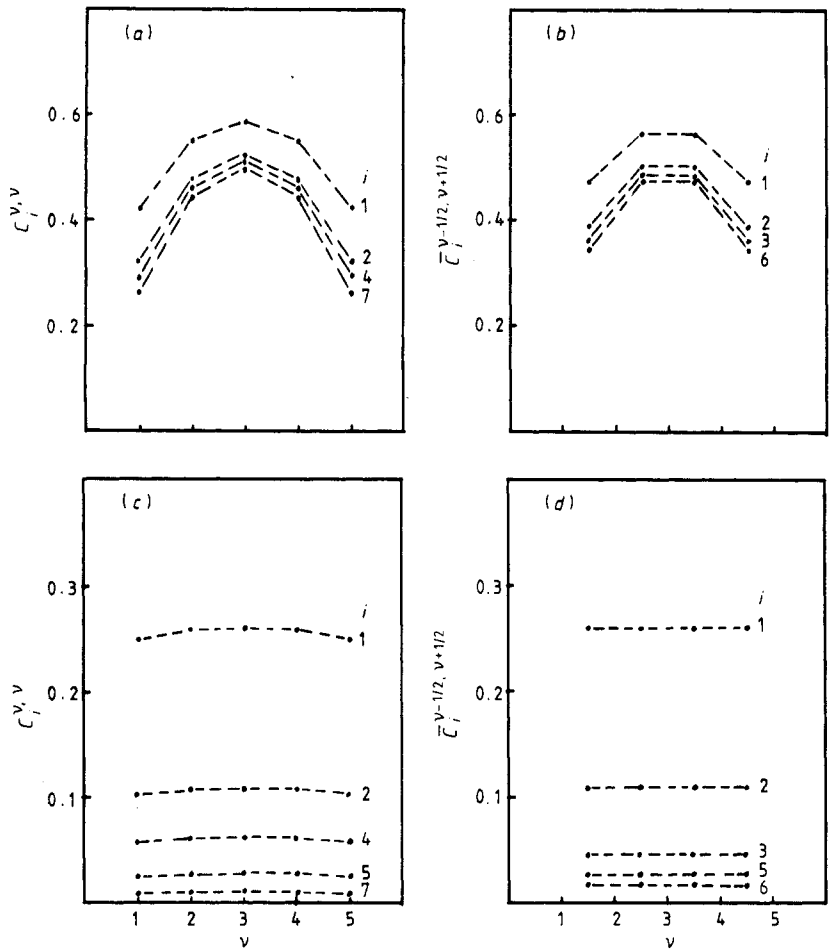

Figure 4. The profiles of some correlation functions across thin film with $n = 5$ (third-order approximation). (*a*) Longitudinal correlation functions $C_i^{\nu,\nu}$ ($i = 1, 2, 4$ and 7) at the reduced temperature $T/T_c = 0.9$. (*b*) Perpendicular correlation functions $\bar{C}_i^{\nu-1/2,\nu+1/2}$ ($i = 1, 2, 3$ and 6) at $T/T_c = 0.9$. (*c*) Longitudinal correlations $C_i^{\nu,\nu}$ ($i = 1$, 2, 4, 5 and 7) at $T = T_c$. (*d*) Perpendicular correlations $\bar{C}_i^{\nu-1/2,\nu+1/2}$ ($i = 1, 2, 3, 5$ and 6) at $T = T_c$.

are given for non-integer indices, e.g. for $\nu = 1.5$ we have correlation of the type $\bar{C}_i^{1,2}$, and so on.

As can be seen from figures 4(*a*) and 4(*b*) the correlations strongly depend on the ν-value. This behaviour is similar to the changes in the magnetisation across the film found at the same temperature (figure 3). The calculations made for low temperatures reveal that all correlation profiles flatten for $T \rightarrow 0$. The flattened correlation profiles are obtained also at $T = T_c$, as is shown in figures 4(*c*) and 4(*d*). In these figures, besides the correlations given in figures 4(*a*) and 4(*b*), correlations $C_5^{\nu,\nu}$ and $\bar{C}_5^{\nu-1/2,\nu+1/2}$ are also shown. At the critical temperature the differences between the correlations of different ranges reach the largest values. Above T_c the correlation profiles remain flat and the differences between correlations gradually decrease, as all correlations tend to zero for the infinite temperature.

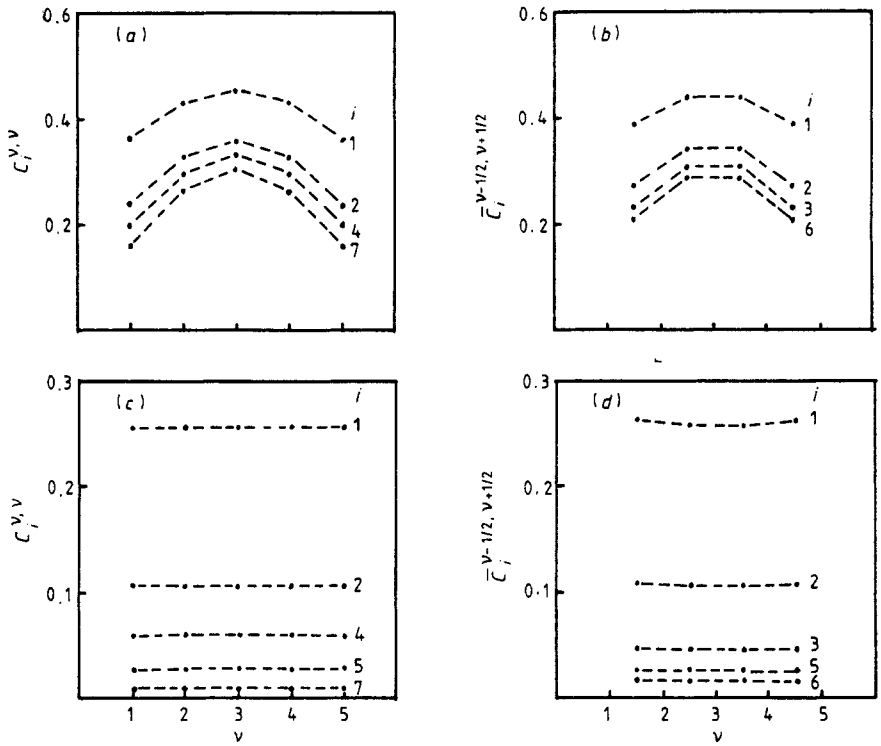

Figure 5. The same correlation functions as in figure 4 but for concentration $c = 0.5$ ($n = 5$).

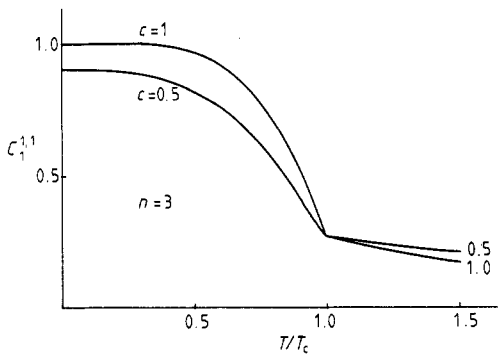

Figure 6. The temperature dependence of the surface correlation $C_1^{1,1}$ between NN spins for $c = 1$ and $c = 0.5$ ($n = 3$).

In figure 5 the same correlations are illustrated for $c = 0.5$. As the consequence of dilution some flattening of correlation profiles and increased difference between their values for different distances i is observed in the ferromagnetic region.

In figure 6 the temperature dependence of the correlation function between NN spins, with both spins on the film surface has been presented for the case of $n = 3$ and for two concentration values, i.e. for $c = 1$ ($k_B T_c/J = 4.070$) and for $c = 0.5$ (with $k_B T_c/J = 1.432$).

It is seen that with increase in the alloy dilution some flattening of the $C_1^{1,1}$ versus temperature curve is observed, i.e. with decreasing c-value $C_1^{1,1}$ becomes smaller in

the ferromagnetic region, while it increases above T_c for corresponding reduced temperatures.

The crossing of the correlation function of the diluted and of the pure system exactly at T_c (as seen in figure 6) is only due to the reduced temperature scale and does not occur when the absolute scale with different $k_B T_c/J$-values is adopted.

The calculations made for other n- and c-values confirm the general behaviour of the correlation functions in thin diluted films discussed above.

To summarise, it can be stated that the Matsudaira method, although rather sophisticated, can be successfully applied also in the case of thin films. As a result, not only are the magnetisation curve, critical temperature and concentration significantly improved but also some additional information concerning the behaviour of the correlation functions is obtained. In particular, the directional dependence of these functions in thin films is predicted. It can be noted also that, with the correlation functions known, other film properties such as for instance the magnetic susceptibility, the specific heat or the magnetic contribution to electrical resistivity can be calculated.

Acknowledgment

This research was supported by CPBP 01.08.

Appendix. The list of coefficients in equations (5)–(13)

For $\nu = 2, 3, \ldots, n-1$ we have

$$B_1(\nu) = b_1 + 2b_2(C_2^{\nu,\nu} + C_4^{\nu,\nu} + 2C_2^{\nu,\nu+1}) + b_3[2(C_2^{\nu,\nu})^2 + (C_4^{\nu,\nu})^2 + 4(2C_2^{\nu,\nu} + C_4^{\nu,\nu})C_2^{\nu,\nu+1} - 2\mu_\nu^4 - 8\mu_\nu^3\mu_{\nu+1}] \tag{A1}$$

$$B_2(\nu) = b_1 + b_2(2C_2^{\nu,\nu} + C_4^{\nu,\nu} + 3C_2^{\nu-1,\nu} + 3C_2^{\nu,\nu+1} + C_4^{\nu-1,\nu+1}) + b_3[(2C_2^{\nu,\nu} + C_4^{\nu,\nu})(C_2^{\nu-1,\nu} + C_2^{\nu,\nu+1} + C_4^{\nu-1,\nu+1}) + 6C_2^{\nu-1,\nu}C_2^{\nu,\nu+1} - 2\mu_\nu^3(\mu_{\nu-1} + \mu_{\nu+1}) - 6\mu_\nu^2\mu_{\nu-1}\mu_{\nu+1}] \tag{A2}$$

$$B_3(\nu) = b_1 + 2b_2(2C_2^{\nu,\nu} + C_4^{\nu,\nu} + 2C_2^{\nu-1,\nu}) + b_3[2(C_2^{\nu,\nu})^2 + (C_4^{\nu,\nu})^2 + 4(2C_2^{\nu,\nu} + C_4^{\nu,\nu})C_2^{\nu-1,\nu} - 2\mu_\nu^4 - 8\mu_\nu^3\mu_{\nu-1}] \tag{A3}$$

$$B_4(\nu) = -4b_2(3\mu_\nu^2\mu_{\nu-1} + 2\mu_\nu^3 + 3\mu_\nu^2\mu_{\nu+1} + 2\mu_\nu\mu_{\nu-1}\mu_{\nu+1}) - 4b_3[(2C_2^{\nu,\nu} + C_4^{\nu,\nu} + 6C_2^{\nu,\nu+1} - 4\mu_\nu^2)\mu_\nu^2\mu_{\nu-1} + 2(C_2^{\nu-1,\nu} + C_2^{\nu,\nu+1} + C_4^{\nu-1,\nu+1})\mu_\nu^3 + (2C_2^{\nu,\nu} + C_4^{\nu,\nu} + 6C_2^{\nu-1,\nu} - 4\mu_\nu^2)\mu_\nu^2\mu_{\nu+1} + 2(2C_2^{\nu,\nu} + C_4^{\nu,\nu})\mu_\nu\mu_{\nu-1}\mu_{\nu+1} - 16\mu_\nu^3\mu_{\nu-1}\mu_{\nu+1}]. \tag{A4}$$

For $\nu = 1$,

$$B_2(1) = a_1 + a_2(2C_2^{1,1} + C_4^{1,1} + 3C_2^{1,2}) + a_3[(2C_2^{1,1} + C_4^{1,1})C_2^{1,2} - 2\mu_1^3\mu_2] \tag{A5}$$

$$B_3(1) = a_1 + 2a_2(2C_2^{1,1} + C_4^{1,1}) + a_3[2(C_2^{1,1})^2 + (C_4^{1,1})^2 - 2\mu_1^4] \tag{A6}$$

$$B_4(1) = -4a_2(2\mu_1^3 + 3\mu_1^2\mu_2) - 4a_3[2C_2^{1,2}\mu_1^3 + (2C_2^{1,1} + C_4^{1,1} - 4\mu_1^2)\mu_1^2\mu_2]. \tag{A7}$$

For $\nu = n$,

$$B_1(n) = a_1 + 2a_2(2C_2^{n,n} + C_4^{n,n}) + a_3[2(C_2^{n,n})^2 + (C_4^{n,n})^2 - 2\mu_n^4] \tag{A8}$$

$$B_2(n) = a_1 + a_2(2C_2^{n,n} + C_4^{n,n} + 3C_2^{n-1,n}) + a_3[(2C_2^{n,n} + C_4^{n,n})C_2^{n-1,n} - 2\mu_n^3\mu_{n-1}] \tag{A9}$$

$$B_4(n) = -4a_2(3\mu_n^2\mu_{n-1} + 2\mu_n^3) - 4a_3[(2C_2^{n,n} + C_4^{n,n} - 4\mu_n^2)\mu_n^2\mu_{n-1} + 2C_2^{n-1,n}\mu_n^3]. \tag{A10}$$

The temperature- and concentration-dependent coefficients a_1, a_2, a_3 and b_1, b_2, b_3 are given by the relations

$$a_1 = (c^5/16)(T_5 + 3T_3 + 2T_1) + [c^4(1-c)/2](T_4 + 2T_2) + [3c^3(1-c)^2/2] \times (T_3 + T_1) + 2c^2(1-c)^3T_2 + c(1-c)^4T_1 \tag{A11}$$

$$a_2 = (c^5/16)(T_5 - T_3 - 2T_1) + [c^4(1-c)/4](T_4 - 2T_2) + [c^3(1-c)^2/4](T_3 - 3T_1) \tag{A12}$$

$$a_3 = (c^5/16)(T_5 - 5T_3 + 10T_1) \tag{A13}$$

$$b_1 = (c^6/32)(T_6 + 4T_4 + 5T_2) + [5c^5(1-c)/16](T_5 + 3T_3 + 2T_1) + [5c^4(1-c)^2/4](T_4 + 2T_2) + [5c^3(1-c)^3/2] \times (T_3 + T_1) + [5c^2(1-c)^4/2]T_2 + c(1-c)^5T_1 \tag{A14}$$

$$b_2 = (c^6/32)(T_6 - 3T_2) + [3c^5(1-c)/16](T_5 - T_3 - 2T_1) + [3c^4(1-c)^2/8](T_4 - 2T_2) + [c^3(1-c)^3/4](T_3 - 3T_1) \tag{A15}$$

$$b_3 = (c^6/32)(T_6 - 4T_4 + 5T_2) + [c^5(1-c)/16](T_5 - 5T_3 + 10T_1) \tag{A16}$$

where

$$T_n \equiv \tanh(nJ/k_BT). \tag{A17}$$

References

[1] Matsudaira N 1973 *J. Phys. Soc. Japan* **35** 1593
[2] Smart J S 1966 *Effective Field Theories of Magnetism* (Philadelphia, PA: W B Saunders)
[3] Oguchi T and Obokata T 1969 *J. Phys. Soc. Japan* **27** 1111
[4] Kaneyoshi T, Fittipaldi I P and Beyer H 1980 *Phys. Status. Solidi* b **102** 393
[5] Kaneyoshi T 1985 *Z. Phys.* B **50** 35
[6] Benayad N, Benyoussef A and Boccara N 1985 *J. Phys. C: Solid State Phys.* **18** 1899
[7] Balcerzak T, Bobák A, Jaščur M, Mielnicki J and Wiatrowski G 1987 *Phys. Status Solidi* b **143** 261

[8] Callen H B 1963 *Phys. Lett.* **4** 161
[9] Suzuki M 1965 *Phys. Lett.* **19** 267
[10] Wiatrowski G Mielnicki J, Balcerzak T and Urbaniak-Kucharczyk A 1988 *J. Magn. Magn. Mater.* **73** 89
[11] Wiatrowski G, Balcerzak T, Wojtczak L and Mielnicki J 1986 *Phys. Status Solidi* b **138** 189
[12] Mielnicki J, Balcerzak T and Wiatrowski G 1987 *J. Magn. Magn. Mater.* **65** 27
[13] Domb C 1974 *Phase Transitions and Critical Phenomena* vol 3, ed C Domb and M S Green (London: Academic) p 173
[14] Frank B and Mitran O 1977 *J. Phys. C: Solid State Phys.* **10** 2641
[15] Charap S H 1962 *Phys. Rev.* **126** 1393