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# The Curie temperature of thin ferromagnetic films

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**Abstract**

The thickness-dependent Curie temperature $T_c(d)$ of thin ferromagnetic films is calculated within the molecular field approximation ('Weiss mean field') of the Heisenberg model. Two higher mean field theories are applied to obtain a quantitative improvement of the results: the Oguchi cluster method and the 'constant coupling approximation' (CCA). Analytical expressions are derived from difference equations or eigenvalue problems with an unknown parameter which can be solved numerically. Explicit expressions for $T_c(d)$ can be given if the interaction is restricted to next neighbour monolayers only, for any value of the spin $S$ within the Weiss mean field and for $S = 1/2$ within the CCA. Effects of an enhanced interaction within the surface layers are briefly investigated. Calculated values of $T_c(d)$ for EuO are presented within the three models.

## 1. Introduction

The Heisenberg model is defined by the Hamiltonian $H = -\sum_{i,j} J_{ij}\hat{\mathbf{S}}_i\hat{\mathbf{S}}_j$ where $\hat{\mathbf{S}}_i$ describes a localized spin set in a lattice. Its mean field approximation is known to predict the basic characteristics of a ferromagnetic phase transition and also qualitatively reproduces many magnetic properties of thin films which can be regarded as a boundary problem within the model. Several numerical and analytical results were obtained by applying Heisenberg or Heisenberg-like models to thin films [1–4]. In this work higher mean field theories and the resulting quantitative deviations with regard to the prediction of the Curie temperature $T_c$ will be studied. The simple mean field theory will be called the 'Weiss mean field'. The first step is to study a two-atomic cluster in the effective field which will be called the 'Oguchi cluster method' [5–7]. Finally, both approaches are combined in the 'constant coupling approximation' [8, 7]. Results will be presented in the case of $S = 1/2$ as well as applied to europium oxide (EuO). We also discuss a change in the behaviour of $T_c(d)$ due to a stronger interaction at the surface layers.

Note that in a previous paper [9] it was shown that a Heisenberg film of finite thickness with isotropic exchange cannot show spontaneous magnetization, one has to implement a small magnetic anisotropy (Mermin–Wagner theorem [10]). It is, however, well-known that mean field type approaches, as we are going to present, are removing anisotropy, thereby violating the Mermin–Wagner theorem. Mean field theories do not consider magnons or spin waves which is surely a shortcoming of such approaches compared to, e.g., Green's function methods and the so-called Tyablikov approximation (random phase approximation) [11, 12]. However, the impact of anisotropy on the Curie temperature turns out to be small [11, 13]. Furthermore, the aim of this paper is to obtain handy analytical expressions for the transition temperature of film systems, independent of the somewhat arbitrary decoupling procedures of Green's function methods.

## 2. The Weiss mean field

We begin by introducing suitable dimensionless variables, the relative magnetization $m = M(T)/M(0)$, the temperature $t = k_B T/\hbar^2 J^{(1)}$ in terms of the exchange constant for next neighbours $J^{(1)}$ ($k_B$: Boltzmann constant) and the magnetic field $b = g\mu_B B/k_B T$ ($g$: Landé factor, $\mu_B$: Bohr magneton). No external field will be considered, hence the magnetic field only contains a contribution from the effective field. From the partition function in the Weiss model $Z_S = \sum_{S^z=-S}^{S} \exp(S^z b)$ it follows that $m = B_S(Sb(m))$ where $B_S$ is the Brillouin function.

For a translation-invariant bulk material the effective field depends on the magnetization, $b = b(m)$, but in a thin film translation invariance is only given within each layer. Introducing $m_k$ as the magnetization in the $k$th monolayer one now has to solve

$$m_k = B_S(Sb_k(m_k, m_{k\pm1}, m_{k\pm2}, \ldots)), \qquad (1)$$

 

where the effective field in the $k$th layer is now dependent on the magnetizations in the neighbouring layers:

$$b_k = \frac{2S}{t}\Big(I_0 m_k + \sum_{j=1}^{\infty} I_j (m_{k+j} + m_{k-j})\Big) \tag{2}$$

with

$$I_j = \sum_{i=1}^{\infty} z_j^{(i)} \frac{J^{(i)}}{J^{(1)}}. \tag{3}$$

In this notation $z_j^{(i)}$ denotes the number of $i$th next neighbours ($i = 1$: next neighbours) in the $j$th adjacent monolayer ($j = 0$: same monolayer) and $J^{(i)}$ is the interaction strength of the respective localized moments which only depends on their distance. In practice, summation limits are restricted by the regarded lattice as well as by the order of neighbours one wishes to consider. Near the transition point $m_k$ and $b_k$ are small and (1) can be linearized. Using (2) one obtains a difference equation for $m_k$:

$$m_k = \tfrac{2}{3} S(S+1) t_c^{-1}\Big(I_0 m_k + \sum_{j=1}^{\infty} I_j (m_{k+j} + m_{k-j})\Big). \tag{4}$$

Applying the boundary conditions for a thin film consisting of $d$ monolayers, $m_{k\leqslant 0} = m_{k\geqslant d+1} = 0$, this can also be written as a Toeplitz-type matrix eigenvalue problem:

$$\begin{pmatrix} I_0 & I_1 & I_2 & I_3 & \dots & I_{d-1} \\ I_1 & I_0 & I_1 & I_2 & I_3 & \vdots \\ I_2 & I_1 & I_0 & \ddots & \ddots & I_3 \\ I_3 & I_2 & \ddots & \ddots & \ddots & I_2 \\ \vdots & \ddots & \ddots & \ddots & I_0 & I_1 \\ I_{d-1} & \dots & I_3 & I_2 & I_1 & I_0 \end{pmatrix} \begin{pmatrix} m_1 \\ m_2 \\ \vdots \\ m_{d-1} \\ m_d \end{pmatrix}$$
$$= \frac{t_c}{\frac{2}{3}S(S+1)} \begin{pmatrix} m_1 \\ m_2 \\ \vdots \\ m_{d-1} \\ m_d \end{pmatrix}. \tag{5}$$

The Curie temperature $T_c$ then follows from the maximal eigenvalue [14]. In the case that only $I_0$ and $I_1$ are non-vanishing, i.e. the interaction is restricted to next neighbouring monolayers only (note that this is not necessarily equal to next neighbouring atoms), the corresponding matrix is tridiagonal with an analytically known eigenvalue spectrum [15]. In this case one obtains

$$t_c(d) = \frac{2}{3} S(S+1)\Big(I_0 + 2I_1 \cos\Big(\frac{\pi}{d+1}\Big)\Big). \tag{6}$$

This is a slight generalization of the formula derived in [14].

## 3. The Oguchi cluster method

The Oguchi approximation is the simplest cluster method where the contribution of one next neighbour to the partition function is calculated exactly and the remaining next neighbours are described by means of an effective field [7, 5, 6] for which we now write $B_O$. The Hamiltonian of the pair located at the lattice sites $i$ and $j$ is hence given by

$$H_p = -2J^{(1)} \hat{\mathbf{S}}_i \hat{\mathbf{S}}_j - \frac{g\mu_B}{\hbar}(\hat{S}_i^z + \hat{S}_j^z) B_O. \tag{7}$$

Let $\hat{\mathbf{\Sigma}} = \hat{\mathbf{S}}_i + \hat{\mathbf{S}}_j$ denote the spin operator of the Oguchi cluster with the eigenvalues

$$\begin{aligned} \Sigma/\hbar &= 0, 1, \dots, 2S \\ \Sigma^z/\hbar &= -\Sigma, -\Sigma+1, \dots, \Sigma. \end{aligned} \tag{8}$$

The average total spin is given by

$$\langle \hat{\Sigma}^z \rangle/\hbar = Z_S^{-1} \frac{\partial}{\partial b_{O,k}} Z_S(t, b_{O,k}) \tag{9}$$

with the partition function

$$Z_S(t, b_{O,k}) = \frac{\exp(-t^{-1} 2S(S+1))}{\sinh(\frac{1}{2} b_{O,k})} \times \sum_{\Sigma=0}^{2S} \exp(t^{-1}\Sigma(\Sigma+1)) \sinh(b_{O,k}(\Sigma + \tfrac{1}{2})), \tag{10}$$

where the sum over $\Sigma^z$ has already been performed and layer dependence has been introduced by the index $k$.

Since a thin film is anisotropic by its nature, the interaction partner can be either chosen from the same layer as the regarded atom or from an adjacent layer. In contrast to a bulk material these two approaches will now cause different results.

In the former case $\langle \hat{\Sigma}^z \rangle = 2\langle \hat{S}^z \rangle_k = 2\hbar S m_k$ and the effective Oguchi field $b_{O,k}$ differs from (2) only by the replacement $z_0^{(1)} m_k \to (z_0^{(1)} - 1) m_k$, as the contribution of one next neighbour has been included exactly. Hence we obtain a relation between the Oguchi effective field and the Weiss effective field:

$$b_{O,k} = \frac{2S}{t}\Big((I_0 - 1) m_k + \sum_{j=1}^{\infty} I_j (m_{k+j} + m_{k-j})\Big) = b_k - \frac{2S}{t} m_k. \tag{11}$$

The sum in (10) is finite and can be calculated for any value of $S$, but if $S$ is high the resulting expressions will be somewhat lengthy. However, after linearization for small $b_{O,k}$ one can always write

$$\langle \hat{\Sigma}^z \rangle/\hbar = f_S(t_c) b_{O,k} \tag{12}$$

with the function $f_S(t_c)$ to be calculated for the respective value of $S$. In the case of $S = 1/2$ one obtains $f_{1/2}(t_c) = 2/(3 + \exp(-\frac{2}{t_c}))$. Note that in the Weiss mean field method the approximation $f_S \approx$ const is made. Inserting (11) this leads to a new difference equation for $m_k$ instead of (4):

$$m_k = \frac{f_S(t_c)}{t_c}\Big((I_0 - 1) m_k + \sum_{j=1}^{\infty} I_j (m_{k+j} + m_{k-j})\Big). \tag{13}$$

On the other hand, if the cluster atoms belong to adjacent monolayers, it follows that $\langle \hat{\Sigma}^z \rangle = \hbar S(m_k + m_{k\pm1})$ and one has to substitute $z_1^{(1)} m_{k\pm1} \to (z_1^{(1)} - 1) m_{k\pm1}$ with the plus sign for

odd $k$ and the minus sign for even $k$, in analogy to the previous procedure. It now follows that

$$m_k = \frac{f_S(t_c)}{t_c}\left(I_0 m_k - \left(1+\frac{t_c}{2f_S(t_c)}\right)m_{k\pm1} + \sum_{j=1}^{\infty} I_j(m_{k+j}+m_{k-j})\right) \tag{14}$$

with the same choice of signs. Or in matrix notation (for even $d$):

$$\begin{pmatrix} I_0 & I_1' & I_2 & I_3 & \dots & I_{d-1} \\ I_1' & I_0 & I_1 & I_2 & I_3 & \vdots \\ I_2 & I_1 & I_0 & I_1' & \ddots & I_3 \\ I_3 & I_2 & I_1' & \ddots & \ddots & I_2 \\ \vdots & \ddots & \ddots & \ddots & I_0 & I_1' \\ I_{d-1} & \dots & I_3 & I_2 & I_1' & I_0 \end{pmatrix} \begin{pmatrix} m_1 \\ m_2 \\ \vdots \\ m_{d-1} \\ m_d \end{pmatrix} = \frac{t_c}{f_S(t_c)} \begin{pmatrix} m_1 \\ m_2 \\ \vdots \\ m_{d-1} \\ m_d \end{pmatrix} \tag{15}$$

with

$$I_1'(t_c) = I_1 - \left(1+\frac{t_c}{2f_S(t_c)}\right). \tag{16}$$

Equation (13) can be likewise written as an eigenvalue problem similar to (15) and (5) and diagonalized to obtain the Curie temperature. Note that the eigenvalues are now more complex functions of the transition temperature. In addition, the matrix in (15) is not of Toeplitz type any longer and some of its entries are themselves dependent on $t_c$, so instead of an eigenvalue problem one now rather has to solve $d$ equations self-consistently for the parameter $t_c$.

There are no further complications if the amount of monolayers $d$ is even. Suppose however that $d$ is odd, $d = 2n+1$. If $n$ is even, there will be $n$ pairs of coupled monolayers and one 'surplus' monolayer that cannot be characterized in this way, see figure 1. One is then again forced to choose interaction partners for its atoms from the layer itself. Furthermore, its position within the film can be theoretically freely chosen. The physically sensible choice would be the middle (i.e. the $(n+1)$th) monolayer since this preserves the rotational invariance of the film.

If $n$ is odd, preservation of the rotational invariance means that there will be three of such surplus monolayers. Again, the physically sensible setting would be to choose them as the middle (i.e. the $n$th, $(n+1)$th and $(n+2)$th) layers because one obtains a higher precision in respect to the surface layers, which essentially determine the properties of a thin film.

This means that for $d = 2n+1$ odd we obtain (15), but with one or three deviating lines

$$m_l = \frac{f_S(t_c)}{t_c}\left((I_0-1)m_l + \sum_{j=1}^{\infty} I_j(m_{l+j}+m_{l-j})\right) \tag{17}$$

according to (13), where $l = n$ for $n$ even and $l = n, n+1, n+2$ for $n$ odd.

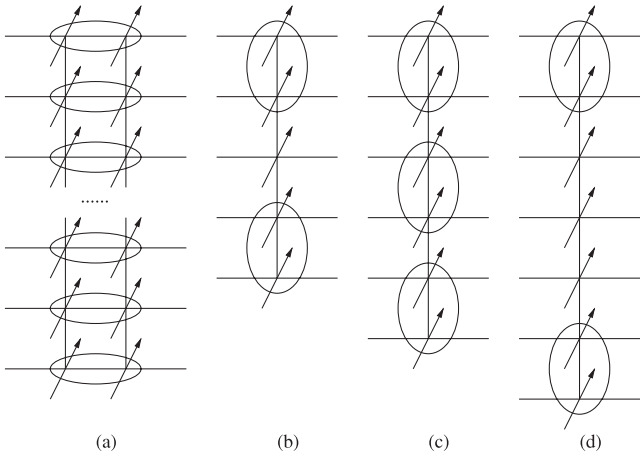


**Figure 1.** Schematic profile of a thin film with different possibilities of pair clusters: (a) pair clusters within monolayers (any desired amount); (b) odd amount of monolayers with one uncoupled; (c) even amount of monolayers, all coupled; (d) odd amount of monolayers with three uncoupled preserving rotational symmetry.

## 4. The constant coupling approximation (CCA)

In the previous models two different self-consistency equations for the magnetization have been derived: (1) for one atom and (9) for two atoms. The corresponding self-consistent expression in the CCA [8, 7] can be most quickly arrived at by combining them considering the respective physical situations, i.e. by using the connection (11) between their effective fields. We closely follow the procedure presented in [7] and apply it to films.

If the pair cluster is situated within the same monolayer one has to insert $\langle\hat{\Sigma}^z\rangle = 2\hbar S m_k = 2\hbar S B_S(Sb_k)$ on the left side of (9) and consequently (12) to obtain a new self-consistency equation within the CCA instead of (1) and (9). Linearizing both sides for small $m_k$ one arrives at the difference equation

$$m_k = (1-\tfrac{2}{3}S(S+1)f_S^{-1}(t_c)) \times \left(I_0 m_k + \sum_j I_j(m_{k+j}+m_{k-j})\right). \tag{18}$$

Comparing this with the similar equation for pure Oguchi clusters (13) one notices that the preceding factor now only depends on $f_S$. In the $S = 1/2$ case, where $f_{1/2}$ contains only one exponential $t_c$-dependent term, this allows an explicit solution for $t_c$ if the eigenvalue problem can be solved analytically. This is again the case when all $I_j$ except for $I_0$ and $I_1$ vanish and one obtains a short analytical formula

$$t_c(d) = 2\left(\ln\left(\frac{I_0+2I_1\cos\left(\frac{\pi}{d+1}\right)}{I_0-4+2I_1\cos\left(\frac{\pi}{d+1}\right)}\right)\right)^{-1} \qquad (S=1/2), \tag{19}$$

which for $d\to\infty$ approaches the bulk formula derived in [8].

If the pair cluster involves two monolayers, one has to put $\langle\hat{\Sigma}^z\rangle = \hbar S(m_k+m_{k\pm1}) = \hbar S(B_S(Sb_k)+B_S(Sb_{k\pm1}))$ with the choice of signs as before. Inserting this into (12) and

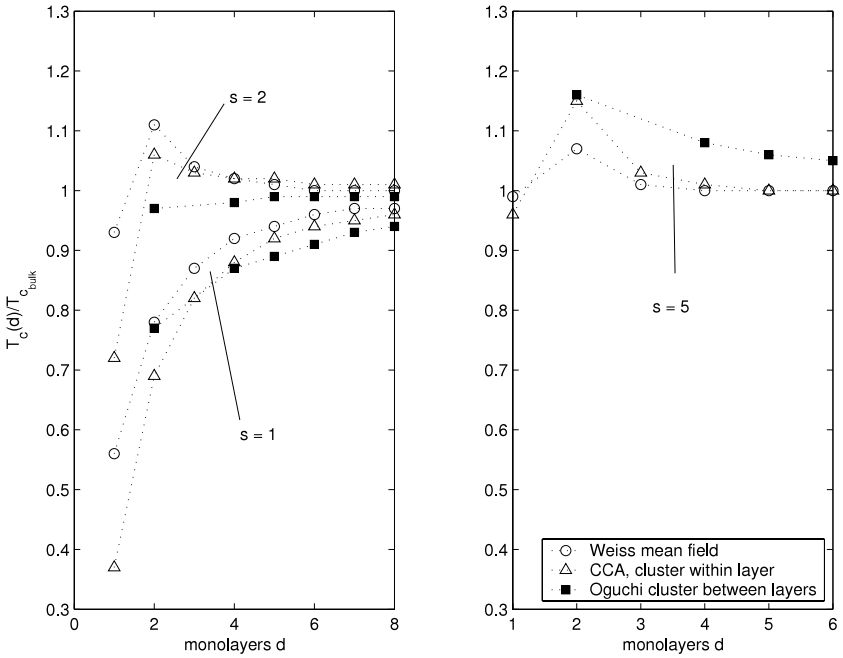


**Figure 2.** Calculated values $T_c(d)/T_c(\infty)$ with an enhanced surface interaction of the form $J^{(i)}_{sf.} = sJ^{(i)}$. $T_c(\infty)$ was set to the value determined for a sufficiently large $d$. Assumed parameters: sc(100) film of $d$ monolayers with coordination numbers $z_0^{(1)} = 4$, $z_0^{(2)} = 4$, $z_1^{(1)} = 1$, $z_1^{(2)} = 4$, furthermore $J^{(2)}/J^{(1)} = 0.25$, $S = 1/2$.

abbreviating $\sigma = \frac{2}{3}S(S+1)$ this leads to

$$\begin{aligned} 2SI_0 f_S m_k = {} & \sigma(I_0 + I_1)m_k + (\sigma(I_0 + I_1) \\ & - 2Sf(I_1 - 1))m_{k\pm1} + (I_1(\sigma - 2Sf_S) + \sigma I_2)m_{k\mp1} \\ & + \sum_{j=2}^{\infty} I_j(\sigma - 2Sf_S)(m_{k\pm j} + m_{k\mp j}) \\ & + \sum_{j=2}^{\infty}(I_{j-1}m_{k\pm j} + I_{j+1}m_{k\mp j}). \end{aligned} \tag{20}$$

## 5. Enhancement of the surface interaction

One can emphasize the role of the surface layers in the discussed models by modifying the interaction within them. This can be achieved by scaling every exchange constant on the surface (i.e. multiplicatively appearing before $m_1$ and $m_d$) by a surface factor $s$: $J^{(i)}_{sf.} = sJ^{(i)}$. This closely follows the approach in [1] and very similar results are obtained.

With growing $s$ the Curie temperature will apparently also grow beyond the bulk value where one can assume that no such surface effects exist. But it stands to reason to consider the function $T_c(d)/T_c(\infty)$, i.e. carrying out the limit $d \to \infty$ and keeping the surface effect, even though it will not be the real bulk temperature. With growing $s$, $T_c(d)/T_c(\infty)$ will at first become almost a constant because the stronger interaction will compensate the reduced coordination number, then exhibit a maximum at $d = 2$ where there are no weaker intermediate layers to force the surface layers into the paramagnetic phase. For very large $s$ it will become a constant again because the surface interaction will dominate for every $d$.

This behaviour is shown in figure 2 for an sc(100) film. We note that when varying $s$ for the different mean field theories, the maximum at $d = 2$ typically remains below 1.3. In [16] a maximum $T_c(d = 3)/T_c(d = 9) = 1.4$ was measured for an fcc-Fe film.

## 6. Comparison of results

The Weiss mean field theory is known to predict higher values for $T_c$ than the experimental results. They are lowered by the application of higher mean field theories [7]. Similarly for a thin film, corrections obtained from numerical solutions for the parameter $t_c$ of (15) and analogous matrix representations of (13), (18) and (20) lead to lower values of both $T_c(d)$ and $T_c(d)/T_c(\infty)$ than those obtained within the Weiss model ((5) and (6)). The qualitative behaviour is unchanged, $T_c(d)$ starts with small values for small $d$ because of a reduced coordination number and approaches the bulk value for $d \to \infty$. This has been calculated numerically within the Heisenberg and Ising models [1–4] and experimentally measured for different materials [17–21, 16].

A local maximum of $T_c(d)/T_c(\infty)$ can be achieved with higher interaction within the surface layers which is assumed not to vanish when $d$ approaches infinity [1, 16]. A genuinely oscillatory behaviour of $T_c$ in the shape of a damped sinusoidal has been predicted [22] and also measured [23], but occurs due to a Ruderman–Kittel–Kasuya–Yoshida (RKKY) type of exchange coupling in itinerant ferromagnets which are not considered here, we restrict ourselves to local-moment systems.

Corrections from higher mean field theories are more significant in the case of small coordination numbers and

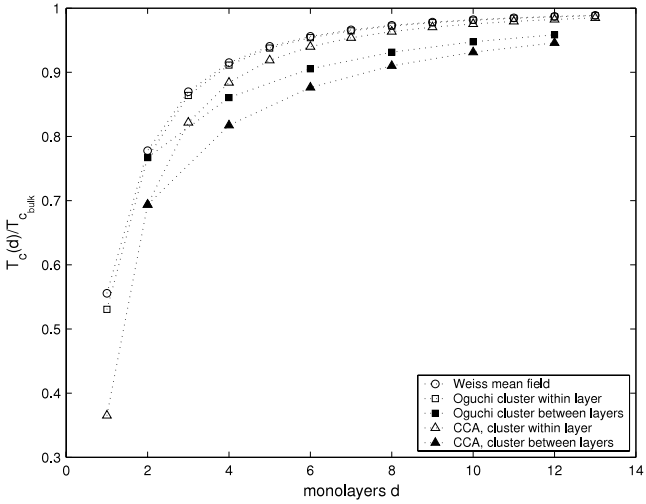


**Figure 3.** Comparison of the calculated values $T_c(d)/T_c(\infty)$ within the Weiss mean field theory, the Oguchi cluster method and the constant coupling approximation (CCA). Assumed parameters: sc(100) film of $d$ monolayers with coordination numbers $z_0^{(1)} = 4$, $z_0^{(2)} = 4$, $z_1^{(1)} = 1$, $z_1^{(2)} = 4$, furthermore $J^{(2)}/J^{(1)} = 0.25$, $S = 1/2$.

**Table 1.** Comparison of the calculated values $T_c(d)$ for EuO within the Weiss mean field theory, the Oguchi cluster method and the constant coupling approximation (CCA) choosing (a) pair cluster within the same layer, (b) pair cluster within adjacent layers. Lattice parameters: fcc(100) film with coordination numbers $z_0^{(1)} = 4$, $z_0^{(2)} = 4$, $z_1^{(1)} = 4$, $z_1^{(2)} = 1$; EuO parameters: $S = 7/2$, $\hbar^2 J^{(1)}/k_B = 0.606K$, $\hbar^2 J^{(2)}/k_B = 0.119K$ [24]. The values in brackets are flawed because not all monolayers can be coupled by pair clusters (see figure 1).

| $T_c(d)$ (K) for EuO, fcc(100) | | | | | |
|---|---|---|---|---|---|
| $d$ | Weiss m.f. | Oguchi m.f. (a) | Oguchi m.f. (b) | CCA (a) | CCA (b) |
| 1 | 30.4 | 29.2 | — | 22.7 | — |
| 2 | 57.2 | 56.4 | 56.4 | 50.0 | 51.7 |
| 3 | 68.2 | 67.6 | — | 61.1 | — |
| 4 | 73.6 | 73.1 | 62.3 | 66.6 | 55.8 |
| 5 | 76.7 | 76.2 | (63.4) | 69.6 | (56.9) |
| 6 | 78.6 | 78.0 | 64.7 | 71.5 | 58.3 |
| 7 | 79.8 | 79.3 | (67.0) | 72.8 | (60.6) |
| 8 | 80.7 | 80.1 | 65.9 | 73.6 | 59.3 |
| $\infty$ | 83.9 | | 83.4 | | 76.9 |

small values of $S$. In addition, if the pair cluster couples two monolayers it will lead to higher corrections if the corresponding amount of next neighbours between monolayers (i.e. $z_1^{(1)}$) is small. Hence, a high difference is expected for a simple cubic lattice with the total coordination number $z = 6$ and especially an sc(100) film where $z_1^{(1)} = 1$. This is shown for an arbitrary choice of exchange constants in figure 3.

When referring to real substances a question arises about the influence of the electronic bandstructure on the magnetic properties. The investigation in [13] reveals that the electronic structure will exclusively influence the exchange parameters $J^{(1)}$ and $J^{(2)}$ which are accurately known from the experiment [24, 25] and cannot be derived within the underlying theories. Therefore we use the experimental values, as it is also done in [13].

The material EuO (fcc, $S = 7/2$) is usually regarded as a prototypical Heisenberg ferromagnet. The results of a calculation by the abovementioned procedure for an fcc(100) film are shown in table 1. Because of the suitable lattice parameters with vanishing $I_{i\geqslant 2}$ (see the commentary above the table), equation (6) may be used for the Weiss mean field. The correction from higher mean field theories is not very significant for small $d$, as compared to figure 3, but with the pair cluster coupling adjacent monolayers a much slower saturation behaviour is obtained. For the transition temperature of a bulk material the Weiss mean field yields 83.9 K, the Oguchi approximation adds just a small correction to 83.4 K while the CCA predicts 76.9 K. Experimentally, $T_c$ was measured to be between 69 and 70 K for EuO [24, 26]. It is to be expected that the calculated film values will deviate from the measured results accordingly. Overall, mean field theories applied to thin films seem to yield good results with comparatively little effort and simple formulae which has to be seen as their advantage.

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