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Thickness dependence of the magnetization and the Curie temperature of ferromagnetic thin films

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The magnetic properties of thin ferromagnetic films are studied using a mean-field model The thickness dependent film Curie temperature $T_c(d)$ is calculated for different surface magnetic moments μ_s as well as for different lattice types and -faces For an enhanced μ_s the bulk Curie temperature $T_c(\infty)$ can be reached within a few layers or may even be exceeded The layer dependent magnetization curves $m_l(T)$ are calculated for a few systems, assuming Ising as well as Heisenberg type spins Thermal fluctuations are partly taken into account by an improved mean field theory The main features of our results seem to be presented in the presence of fluctuations.

The determination of the magnetic properties like the magnetization curves $m_l(T, d)$, the film Curie temperature $T_c(d)$, and the corresponding critical exponents of ferromagnetic thin films, sandwiches, and superlattices with a defined thickness d has become a very active field recently [1–3]. Especially the expected dimensionality crossover from a two-dimensional (2D) to a three-dimensional (3D) system with increasing film thickness was studied experimentally [4,5]. The physical quantities like the exchange coupling $\mathcal{J}$, the magnetic moments μ, the lattice anisotropy etc. may react quite differently on the reduction from a 3D-bulk to a 2D-monolayer system. In general the critical temperature is determined by local quantities like $\mathcal{J}$ and μ as well as by the global topology of the film The saturational behavior of $T_c(d)$ as predicted from scaling hypothesis [6] seems not to be universal especially for very thin films. For example $T_c(d)$ may reach the bulk Curie temperature $T(\infty)$ within a few atomic layers [7], or may even exceed $T_c(\infty)$ exhibiting a maximum at a small film thickness [8] In this thickness range $T_c(\infty) - T_c(d)$ is not only determined by the reduced coordination number, but depends also strongly on the surface/interface magnetic moments μ_s, which may deviate from the bulk value μ_b, as well as on the global topology of the film A number of theoretical studies obtained the possibility of an enhancement of μ_s by 20%–30% relative to μ_b for Fe free surfaces and interfaces and for Ni free surfaces [2,9], whereas for a Ni/Cu interface μ_s is reduced by roughly the same amount [10] Due to the roughness or intermixing effects μ_s may even vanish ("dead layer"). For the case of Co a flat surface/interface seems not to have such a large effect on μ_s [11]. Experimental observations support the enhancement of surface magnetic moments [12]. $T_c(d)$ has been calculated by high temperature series expansions [13] and spin-wave models [14] as well as by mean field type models [15,16]. In most cases a saturational behavior according to the scaling hypothesis was obtained, but also a different behavior of $T_c(d)$ due to different effective exchange couplings or magnetic moments for surfaces/interfaces.

In this paper we like to present a generalized mean field theory to calculate the magnetic properties of thin films. We will focus on the dependence of the magnetization curves $m_l(T, d)$ and the film Curie temperature $T_c(d)$ on the local

physical quantities like magnetic moments, coordination number, and type of spins (Ising or Heisenberg). The thin film is assumed to consist of completely filled and well ordered ferromagnetic layers (layer-by-layer growth), grown on a flat substrate. Thus, the effects due to vacancies, islands, steps or intermixings on the magnetic properties are not considered here. Because of the low dimensionality thermal fluctuations cannot be neglected. These are partly taken into account by an improved mean field model, the so called Kirkwood approximation [16,17]. It originates from the cumulant expansion of the free energy in terms of $(k_B T)^{-1}$, and can be applied to non-uniformly ordered phases as well as to Ising and Heisenberg type magnetic moments. Despite the itinerant nature of magnetism of the systems under consideration we assume a localized spin model. This is known to describe the magnetic properties of such systems qualitatively well. The effective exchange coupling J_{ij} between two magnetic moments μ_i and μ_j is approximately given by $\mu_i \mu_j \mathcal{J}_{ij}$. The interatomic exchange interaction $\mathcal{J}_{ij}$ is assumed here to be the same for all nearest neighbor pairs $\langle ij \rangle$ in the film. A possible deviation of $\mathcal{J}_{ij}$ in the neighborhood of surfaces/interfaces can also be taken into account by our model. The different lattice types and crystal faces are considered by different coordination numbers z_0 and z_1 in the same layer and between adjacent layers (bulk coordination number $z = z_0 + 2z_1$).

The ith layer magnetization $m_i(T, d)$ of a thin film with d layers is obtained by solving a system of d equations selfconsistently. For Ising-1/2 spins within the Bragg–Williams approximation these are given by

$$m_i = \tanh\left[\beta \mathcal{J} \mu_i \left(z_0 \mu_i m_i + z_1 \mu_{i+1} m_{i+1} + z_1 \mu_{i-1} m_{i-1}\right) - \Delta h_i\right], \tag{1}$$

$$\Delta h_i = (\beta \mathcal{J} \mu_i)^2 m_i \left[z_0 \mu_i^2 (1 - m_i^2) + z_1 \mu_{i+1}^2 (1 - m_{i+1}^2) + z_1 \mu_{i-1}^2 (1 - m_{i-1}^2)\right],$$

$i = 1, \ldots, d$, $\beta = 1/k_B T$, k_B Boltzmann's constant. The Kirkwood approximation causes the correction Δh_i of the molecular field. The surfaces/interfaces at $i = 1$ and $i = d$ are regarded by putting $\mu_0 = \mu_{d+1} = 0$. For $T \lesssim T_c(d)$ eq. (1) can be linearized leading to a $d \times d$ tridiagonal

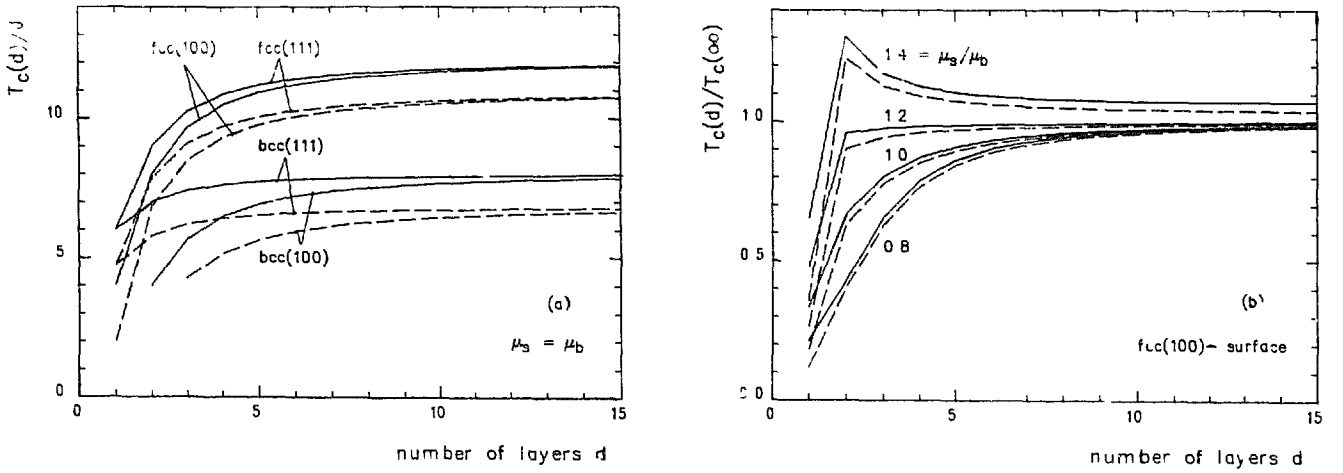

Fig. 1. (a) Thin film Curie temperature $T_c(d)$ in terms of the exchange coupling constant $\mathcal{J}$ as a function of the film thickness d. All magnetic moments are set equal to the bulk atomic magnetic moment μ_b. The curves refer to different lattice types and -faces: fcc (100) $z_0 = z_1 = 4$, fcc (111) $z_0 = 6$, $z_1 = 3$, bcc (100) $z_0 = 0$, $z_1 = 4$, bcc (111) $z_0 = 6$, $z_1 = 1$. We denote by z_0 and z_1 the number of nearest neighbors in the same layer and between adjacent layers. (b) $T_c(d)$ in terms of the bulk Curie temperature $T_c(\infty)$ for a fcc (100)-film ($z_0 = z_1 = 4$). The curves refer to different surface magnetic moments μ_s for both surfaces/interfaces as indicated in the figure. The magnetic moments in all other layers are set equal to the bulk value μ_b. In all cases Ising-1/2 spins are considered. Full lines: Bragg-Williams approximation, dashed lines: Kirkwood approximation.

matrix. Its largest eigenvalue determines $T_c(d)$ [14,15]. Similar expressions are obtained for general Ising and Heisenberg type spins.

In the following we like to present our results for the film Curie temperature $T_c(d)$ as a function of the film thickness d and the layer magnetization curves $m_i(T)$. $T_c(d)$ is given in fig. 1a in terms of the exchange coupling constant J for four different lattice faces. In all cases the magnetic moments are set equal to the bulk value μ_b. For bcc-films $T_c(d)$ seems to saturate faster than fcc-films. In fig. 1b $T_c(d)$ is depicted relative to the bulk Curie temperature $T_c(\infty)$ for an fcc(100) – film ($z_0 = z_1 = 4$) and varying surface magnetic moments μ_s. μ_s is assumed to be the same on both surface/interfaces, whereas all other magnetic moments are set equal to μ_b. A different behavior of $T_c(d)$ is seen due to variation of μ_s. For an enhanced surface magnetic moment $T_c(d)$ can reach the bulk critical temperature $T_c(\infty)$ within a few layers. Such a behavior was found for bcc-Fe/Ag(100) [7]. If a still larger μ_s is assumed, $T_c(d)$ may even exceed $T_c(\infty)$ despite the reduced number of nearest neighbors. In this case $T_c(d)$ exhibits a maximum for a small film thickness d, as was experimentally observed for fcc-Fe/Cu(100) [8]. A saturational behavior of $T_c(d)$ is obtained for a reduced μ_s, according to experimental observations on Ni thin films [5] Similar results for $T_c(d)$ are also obtained for other lattice geometries and for both Ising and Heisenberg type spins. Note that except for a very small number of layers the consideration of thermal fluctuations through the Kirkwood approximation seem to have no large effect on $T_c(d)$.

Figs. 2 and 3 show the full magnetization curves $m_i(T)$ for a fcc (100)-film with 15 atomic layers Fig. 2a refers to Heisenberg spins with a reduced surface magnetic moment $\mu_s = 0.5\mu_b$ on both surface layers, whereas fig. 2b shows the same system for an enhanced $\mu_s = 1.5\mu_b$. In comparison the magnetization curves for the respective bulk systems are shown. It is obvious that for case (a) the magnetic order on the surface layers ($i = 1$) are stabilized by the stronger magnetized inner layers ($i = 5, 8$). However, for the case (b) of an enhanced surface magnetic moment μ_s the inner layers tend to approach the bulk ordering temperature $T_c(\infty)$, but are forced by the surface layers to preserve a weak magnetic ordering up to $T_c(d = 15) > T_c(\infty)$. For free surfaces ($d \to \infty$) the bulk will maintain its Curie temperature $T_c(\infty)$. In this case an ordered surface can exist on top of a

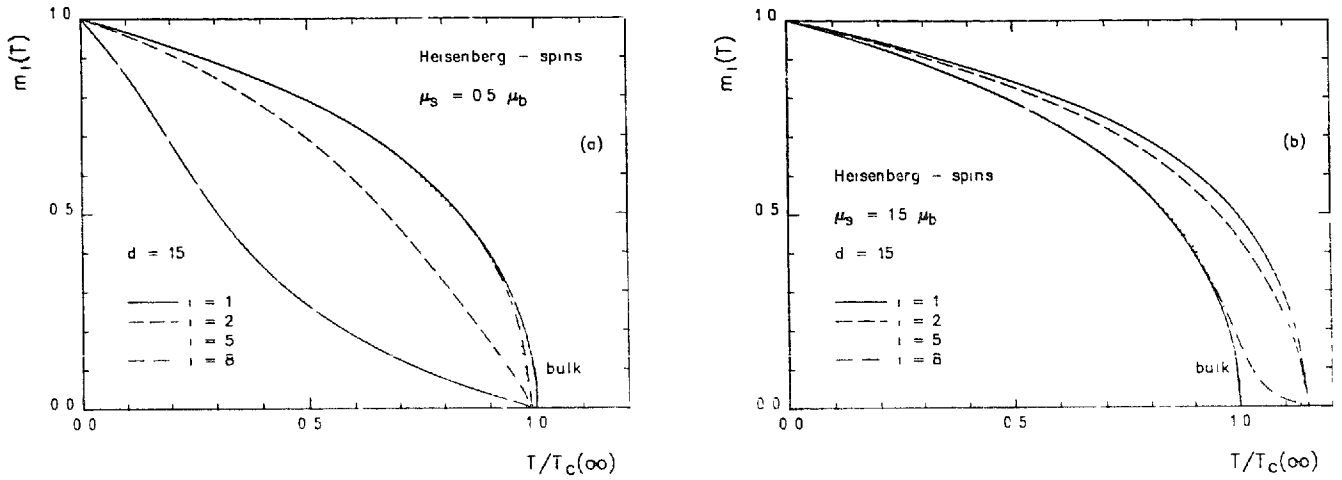

Fig 2. Magnetization curves $m_i(T)$ ($i = 1, 2, 5, 8$) within the Bragg–Williams approximation for a fcc (100)-film with 15 atomic layers and Heisenberg type magnetic moments Different surface magnetic moments on both surfaces are assumed (a) $\mu_s = 0.5\mu_b$. (b) $\mu_s = 1.5\mu_b$ All other magnetic moments are assumed to be equal to the bulk value μ_b In comparison the magnetization curve for the bulk system is shown

disordered bulk, if the effective surface exchange coupling is large enough [15]. This property is experimentally observed for Gd surfaces [18].

The magnetization curves of fig. 3a are calculated for Ising-1/2 spins, assuming a fcc (100)-film with 15 layers and a reduced $\mu_s = 0.5\mu_b$. Whereas the qualitative features compared with fig. 2a are quite similar, the main difference is the exponential decay of $m_l(T)$ for $T \gtrsim 0$ in contrast to the linear decay for the Heisenberg type magnetic moments. Spin wave theory yields for the bulk the well known $T^{3/2}$-law of $m(T)$ in this temperature region, which is also observed experimentally [19]. For thin films an almost linear decay is expected, which may also be deduced from spin wave theory. The Heisenberg spins also exhibit this feature (see fig. 2), but this accordance seems to be accidental, because a linear decrease of $m(T)$ is also obtained for Heisenberg spins in bulk systems. Despite the incorrect temperature dependence we think that our model is useful to obtain the qualitative behavior of the magnetization curves for thin films, because the familiar spin wave theory is valid only for $T \lesssim T_c/3$.

In fig. 3b the influence of thermal fluctuations through the Kirkwood approximation are shown for the Ising spin system of case (a). As mentioned above, the consideration of this fluctuations seems to have no large influence on the magnetic properties of such thick films ($d = 15$). The spread of the magnetization curves between the surface ($l = 1$) and the central layer ($l = 8$) is slightly reduced. This is in accordance with the observed reduction of the modulated region in the phase diagram of a different Ising spin system due to thermal fluctuations [17]. The main features of our results seem to be preserved in the presence of fluctuations.

In conclusion we presented a simple model for the magnetic properties of thin ferromagnetic films assuming only systems with well ordered and completely filled atomic layers. For very thin films the surface magnetic moment μ_s depends also on the film thickness d, whereas we used a constant μ_s for the calculations presented here. In addition our model allows for the consideration of next nearest neighbor (nnn-) magnetic couplings. This seems especially important for the case of bcc (100)-layers, which have no nearest neighbors in the same plane ($z_0 = 0$), thus leading to a vanishing $T_c(d)$ for $d = 1$. However, already bcc (100)-Fe monolayers on Ag [7] and Au [4] were observed to be ferromagnetic, which can be obtained by nnn-interactions.

The magnetic properties of thin films are not only determined by local quantities like the magnetic moments, but are also affected strongly by the overall film topology. For film thicknesses in

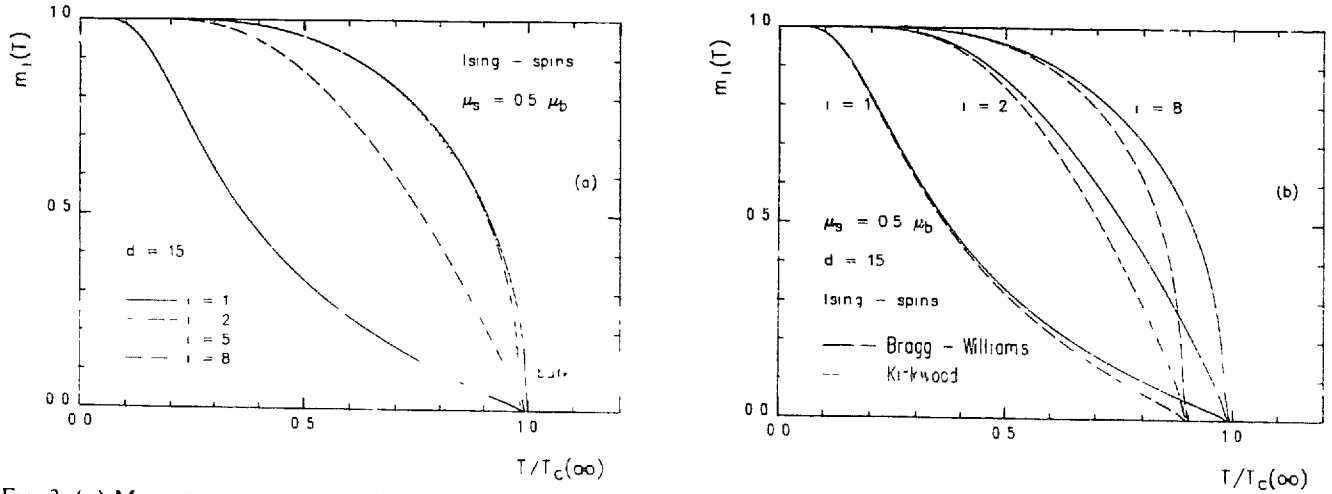

Fig. 3. (a) Magnetization curves $m_l(T)$ for the same system indicated in fig. 2a assuming Ising 1/2-type magnetic moments. (b) Comparison of the magnetization curves $m_l(T)$ ($l = 1, 2, 8$) calculated within the Bragg–Williams approximation (full lines) and within the Kirkwood approximation (dashed lines). The same system is in (a) is assumed. $T_c(\infty)$ refers to the bulk Curie temperature calculated with the Bragg–Williams approximation.

the monolayer range $T_c(d)$ depends on a variety of influences like vacancies, noncomplete layers, substrate roughness, steps, etc. Especially important is the role of intermixing due to roughness or thermal effects on the magnetic moment. In this case the interface region will behave as an alloy of a nonmagnetic and a magnetic compound. For example, a Co monolayer on Cu almost preserves its magnetic moment μ_{Co}, whereas μ_{Co} vanishes for Co in a Cu matrix [20]. As a consequence, different preparation conditions seem to cause partly contradicting experimental observations on the same system [3].

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