Surface-layer magnetization exponent of ferromagnetic semiconductors

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Abstract. Using a Green function technique combined with the transfer-matrix method for the study of surface phenomena, we have investigated a three-dimensional s–d(f) model with modified surface exchange. The surface layer-magnetization exponent is obtained as $\beta_s = 0.781 \pm 0.006$ (which is different compared with the bulk $\beta = 0.361 \pm 0.006$) and at the multicritical transition it is $\beta_{sm} = 0.171 \pm 0.016$. The critical exponents are consistent with results using the renorm-group and the Monte Carlo methods.

1. Introduction

It has been a point of interest to see the influence of the surface on the critical behaviour of ferromagnetic spin systems that exhibit a second-order magnetic phase transition (for a review see [1]). Though the critical phenomena near the magnetic phase transition temperature have been intriguing the scientific community for quite a long time, there have been only a few detailed studies made on the phase transitions that occur in magnetic semi-infinite systems and magnetic thin films [2–7, 20–22]. In physical systems one expects that the surface exchange will in general be not the same as in the bulk. Previous works have demonstrated that for sufficiently enhanced coupling in the surface layer there can be distinct transitions in the surface and in the bulk. Experiments [8] have shown that in the general case the Curie temperature of ultra-thin films is lower than the bulk Curie temperature and, as the thickness of an ultra-thin film decreases, its Curie temperature also decreases while the value obtained by Curie temperature measurements strongly depends on the surface situation. In some special cases the Curie temperature of ultra-thin films is higher than the bulk Curie temperature. For sufficient enhancement of the interaction between the surface spins, an ordered surface phase is expected to coexist with a disordered bulk phase [9]. It was shown by extensive Monte Carlo simulations [2] that the magnetization M_s in the surface layer vanishes at the critical point T_c with an exponent β_s different from the exponent β in the bulk. The thermal variation of M_s expresses a linear temperature dependence in the temperature region near T_c^b, as observed in many semi-infinite ferromagnetic systems both theoretically [5, 10] and experimentally [11]. The surface phase transition in anisotropic Heisenberg models was studied by Tsallis and Chame [20] within a real-space renormalization group, and by Figueiredo and de Moraes [21] using the Green function formalism and the random-phase approximation. The onset of surface ordering takes place when the surface magnons become more energetic than the bulk. In the limit where bulk and surface are both described by a pure Heisenberg model, there is no long-range surface magnetic order over the paramagnetic bulk, whatever the values of the exchange couplings are.

2. The model and the magnetic matrix Green function

The aim of the present paper is to calculate from the s–d model the critical exponent β_s for the surface localized-spin magnetization of semi-infinite ferromagnetic semiconductors (FMSs).

The Hamiltonian of the system is given by:

$$H = H + M + H_E + H_{ME}. \tag{1}$$

H_M is the Heisenberg Hamiltonian for the ferromagnetically ordered d electrons,

$$H_M = -\frac{1}{2} \sum_{l,\delta} J_{l,l+\delta} S_l S_{l+\delta} \tag{2}$$

where S_l and $S_{l+\delta}$ are the spin operators for the localized spins at sites l and $l + \delta$ in the semi-infinite system, the sum on δ is over nearest neighbours only and $J_{l,l+\delta}$ is the exchange interaction.

The problem with the simple Heisenberg model in the form (2) is that due to the Mermin–Wagner theorem [23] there is no solution showing collective magnetic order at finite temperatures $T > 0$. To steer clear of this obstacle there are two possibilities. First, one can apply a decoupling scheme to the Hamiltonian equation (2) which breaks the Mermin–Wagner theorem. The most common example in the case of the Heisenberg model would be a mean-field decoupling. For us, the main drawback of the mean-field decoupling is its incapability of describing critical properties. When choosing a better decoupling approximation to fulfil the Mermin–Wagner theorem, the original Heisenberg Hamiltonian equation (2) has to be extended to break the directional symmetry. The most common extensions are the introduction of an anisotropic exchange interaction $-D \sum_{ij} S_i^z S_j^z$ and/or the single-ion anisotropy $-D \sum_i (S_i^z)^2$. For the following we have chosen a single-ion anisotropy leaving us with the total Hamiltonian:

$$H_M = -\frac{1}{2} \sum_{l,\delta} J_{l,l+\delta} S_l S_{l+\delta} - D \sum_i (S_i^z)^2 \tag{3}$$

where D ($D < 0$) is the single-ion anisotropy parameter, which is typically smaller by some orders of magnitude than the Heisenberg exchange interaction, $|D| \ll J_{ij}$.

H_E represents the usual Hamiltonian of the conduction band electrons,

$$H_E = \sum_{l,\delta,\sigma} t_{l,l+\delta} a_{l\sigma}^+ a_{l+\delta,\sigma} \tag{4}$$

where $t_{l,l+\delta}$ is the hopping integral.

The most important term in (1) is the operator H_{ME} which couples the two subsystems (3) and (4) by an intraatomic exchange interaction I_i,

$$H_{ME} = -\sum_l I_l S_l s_l. \tag{5}$$

The spin operators s_l of the conduction electrons at site l can be expressed as $s_l^+ = a_{l+}^+ a_{l-}$, $s_l^z = (a_{l+}^+ a_{l+} - a_{l-}^+ a_{l-})/2$, where $a_{l\sigma}^+$ and $a_{l\sigma}$ are Fermi creation and annihilation operators at site l, respectively; $\sigma = \pm 1$ corresponds to spin-up and down states.

To study the magnetic excitations of the system we calculate the Green function,

$$G_{ij}(t) = \langle\langle S_i^+(t); S_j^-(0) \rangle\rangle. \tag{6}$$

On introducing the two-dimensional Fourier transform $G_{n_i n_j}(k_\parallel, \omega)$, one has the following form,

$$\langle\langle S_i^+; S_j^- \rangle\rangle_\omega = \frac{2\langle S_i^z \rangle}{N} \sum_{k_\parallel} \exp(i k_\parallel (r_i - r_j)) G_{n_i n_j}(k_\parallel, \omega) \tag{7}$$

where N is the number of sites in any of the lattice planes, r_i and n_i represent the position vectors of site i and the layer index, respectively, and $k_\parallel = (k_x, k_y)$ is a two-dimensional wave vector parallel to the surface. The summation is taken over the Brillouin zone.

We assume for simplicity only nearest-neighbour exchange interactions and take $J_{ij} = J_s$; $I_i = I_s$; $t_{ij} = t_s$; $\langle S_i^z \rangle = \langle S_s^z \rangle$; $\langle s_i^z \rangle = \langle s_s^z \rangle$ on the surface layer ($i = 1$) and $J_{ij} = J$; $I_i = I$, $t_{ij} = t$; $\langle S_i^z \rangle = \langle S^z \rangle$; $\langle s_i^z \rangle = \langle s^z \rangle$ in the bulk. As a result the equation of motion for the Green function (7) of the semi-infinite FMS for $T \leqslant T_c$ has the following matrix form:

$$(\omega - \mathbf{H})\mathbf{G}(k_\parallel, \omega) = \mathbf{R} \tag{8}$$

where $\mathbf{H}$, $\mathbf{G}$ and $\mathbf{R}$ are $\infty \times \infty$ matrices. $\mathbf{H}$ can be expressed as

$$\mathbf{H} = \begin{pmatrix} V & -k_1 & 0 & 0 & 0 & 0 & \dots \\ -k & U & -k & 0 & 0 & 0 & \dots \\ 0 & -k & Q & -k & 0 & 0 & \dots \\ 0 & 0 & -k & Q & -k & 0 & \dots \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \ddots \end{pmatrix}_{\infty \times \infty}$$

with

$$k_1 = J\langle S_s^z \rangle \qquad k = J\langle S^z \rangle$$

$$V = \frac{1}{2\langle S_s^z \rangle} \left(\frac{4J_s}{N} \sum_q (\gamma_q^s - \gamma_{k_\parallel - q}^s)(2\langle S_q^z S_{-q}^z \rangle_s + \langle S_q^- S_q^+ \rangle_s) + \frac{I_s}{N^2} \sum_{q,p} (\langle S_{q-p}^- a_{p+}^+ a_{q-} \rangle_s \right.$$

$$\left. + \langle S_{q-p}^z a_{p+}^+ a_{q+} \rangle_s + \langle S_{q-p}^z a_{p-}^+ a_{q-} \rangle_s) + 2(J + 2D)\langle S^z \rangle \langle S_s^z \rangle \right)$$

$$U = \frac{1}{2\langle S^z \rangle} \left(\frac{4J}{N} \sum_q (\gamma_q - \gamma_{k_\parallel - q})(2\langle S_q^z S_{-q}^z \rangle + \langle S_q^- S_q^+ \rangle) + \frac{I}{N^2} \sum_{q,p} (\langle S_{p-q}^- a_{p+}^+ a_{q-} \rangle) \right.$$

$$\left. + \langle S_{q-p}^z a_{p+}^+ a_{q+} \rangle - \langle S_{q-p}^z a_{p-}^+ a_{q-} \rangle) + 2(J + 2D)\langle S_s^z \rangle \langle S^z \rangle + 2(J + 4D)\langle S^z \rangle^2 \right)$$

$$Q = \frac{1}{2\langle S^z \rangle} \left(\frac{4J}{N} \sum_q (\gamma_q - \gamma_{k_\parallel - q})(2\langle S_q^z S_{-q}^z \rangle + \langle S_q^- S_q^+ \rangle) + \frac{I}{N^2} \sum_{q,p} (\langle S_{p-q}^- a_{p+}^+ a_{q-} \rangle) \right.$$

$$\left. + \langle S_{q-p}^z a_{p+}^+ a_{q+} \rangle - \langle S_{q-p}^z a_{p-}^+ a_{q-} \rangle) + 4(J + 3D)\langle S^z \rangle^2 \right)$$

$$\gamma(k_\parallel) = \tfrac{1}{2}(\cos(k_x a) + \cos(k_y a)).$$

In order to obtain the solutions of the matrix equation (8) we use the transfer-matrix method [12, 13]. We define two infinite-dimensional column matrices, $\mathbf{G}_n$ and $\mathbf{R}_n$ with the elements given by $(\mathbf{G}_n)_m = G_{mn}$ and $(\mathbf{R}_n)_m = 2\langle S_n^z \rangle \delta_{mn}$, so that equation (8) yields

$$(\omega - \mathbf{H})\mathbf{G}_n = \mathbf{R}_n. \tag{9}$$

We introduce the two transfer functions:

$$T_1 = \frac{G_{2i+1,m}}{G_{2i,m}} \qquad T_2 = \frac{G_{2i,m}}{G_{2i-1,m}} \qquad (i \geqslant 1). \tag{10}$$

Substituting T_1 and T_2 into one column of (9), the following Green functions can be obtained:

$$G_{11} = \frac{1}{\omega - V + k_1 T_2} \tag{11}$$

$$G_{22} = \frac{1}{\omega - U + \left(k T_1 - \frac{k_1 k}{\omega - V} \right)} \tag{12}$$

$$G_{33} = \cfrac{1}{\omega - Q + \left(kT_2 - \cfrac{k^2}{\omega - U - \frac{k_1 k}{\omega - V}} \right)}$$ (13)

and so on, where

$$T_1 = \frac{1}{2k} \left(-\omega + U \pm \sqrt{\left(\omega - U - 4k^2 \frac{\omega - U}{\omega - Q} \right)} \right)$$ (14)

$$T_2 = \frac{1}{2k} \left(-\omega + Q \pm \sqrt{\left(\omega - Q - 4k^2 \frac{\omega - Q}{\omega - U} \right)} \right).$$ (15)

The correlation functions are calculated from the Green function using the spectral theorem.

As is well known, the poles of the Green functions gives the dispersion relation of the spin waves, therefore the SSW spectrum can be given from equation (11) by the following expression:

$$\omega_s - V(\omega_s) + k_1 T_2(\omega_s) = 0$$ (16)

which has to be numerically calculated. We obtain an equation of the seventh degree. It must be pointed out, however, that the seven roots obtained from (16) must be checked because extra expressions have been multiplied. We must make sure that the root does not make the expressions $(\omega_s - U)$, $(\omega_s - Q)$ etc vanish. As a matter of fact, only two of the seven roots satisfy these requirements and therefore represent the true SSW spectra, corresponding to the 'acoustic' and 'optical' branches of FMSs.

The so obtained two solutions for the SSW can be used for the evaluation of the relative localized-spin magnetization of the surface $\langle S_s^z \rangle$:

$$\langle S_s^z \rangle = (S + 0.5) \coth[(S + 0.5)\beta\omega_s] - 0.5 \coth(0.5\beta\omega_s).$$ (17)

However, the surface magnetization involves the SSWs, the bulk magnetization and spin waves, the surface and bulk conduction-electron magnetization and energy, so that all expressions must be solved self-consistently.

In order to obtain the conduction-electron magnetization $\langle s_i^z \rangle$ we must define the one-electron Green function $g_{ij\sigma}(E) = \langle\langle a_{i\sigma}; a_{j\sigma}^+ \rangle\rangle$. The equation of motion is given by

$$[E + 0.5\sigma I_i \langle S_i^z \rangle] g_{ij\sigma} = \delta_{ij} + \sum_\delta t_{i,i\delta} g_{i+\delta, j\sigma}(E).$$ (18)

Analogously to the previous calculation of the magnetic Green function (6) we obtain for the surface conduction-electron energy:

$$E_s = N_s + kT_2(E_s)$$ (19)

with

$$N_s = -0.5\sigma I_s \langle S_s^z \rangle + t_s \gamma(k_\parallel)$$ (20)

$$T_2 = \frac{1}{2k}(E_s - N_s \pm \sqrt{(E_s - N_s)^2 - 4k^2}).$$ (21)

$t_s = -W_s$ and $t = -W$, where W_s and W are the conduction band width on the surface layer and in the bulk, respectively. Equation (19) can be used for the calculation of the surface conduction-electron magnetization:

$$\langle s_s^z \rangle = \frac{n_+ - n_-}{2}$$ (22)

where n_+ and n_- are the numbers of conduction electrons in the spin-up and spin-down bands, respectively [14]. The expressions (16), (17), (19) and (22) form a closed system of self-consistent equations, whose solution leads to these four quantities on the surface. So, through the renormalized conduction-electron energy and the conduction-electron magnetization $\langle s_s^z \rangle$ we take into account the t-dependence of the SSW spectrum (16) and our theoretical results can be applied to narrow-band FMSs ($W \ll IS$) such as $CdCr_2Se_4$ as well a to wide-band FMSs ($W \gg IS$) such as EuO.

3. The critical exponent of the surface magnetization

The critical exponent β_s describing the order parameter on the surface $\langle S_s^z \rangle$ is defined as [15]

$$\langle S_s^z \rangle = A \left(\frac{T_c - T}{T_c} \right)^{\beta_s} \text{ for } T \to T_c^-. \tag{23}$$

β_s is independent of the magnitude of the spin, but dependent on the dimensionality of the lattice. The temperature dependence of $\langle S_z^z \rangle$ from equation (16) was calculated numerically using parameters appropriate to the ferromagnetic chalcogenide spinels $CdCr_2Se_4$ [16]: $J = 0.0001$ eV, $I = 0.5$ eV, $t = 0.1$ eV, $T_c = 132.5$ K. The single-ion anisotropy which plays the mere role of keeping the magnetization at finite temperatures was chosen as $D/J = 0.01$. The thermal variation of $\langle S_s^z \rangle$ [5] expresses the linear temperature dependence in the temperature region near T_c^b, as observed in many semi-infinite ferromagnetic systems both theoretically [10] and experimentally [11]. For $I_s = 0.2I$ we obtain that the bulk magnetization is always greater compared with the surface magnetization, i.e. $T_c^b > T_c^s$, whereas for $I_s = 2I$ we get $T_c^s > T_c^b$ [5]. Previous work [1] has shown that for sufficiently enhanced coupling in the surface layer $J_s > J_{sm}$ (J_s is the surface exchange) there can be distinct transitions in the surface and in the bulk. At $J_s = J_{sm}$ there is a 'special' (multicritical) point where one has the unusual situation that there is simultaneous criticality of two-dimensional (2D) surface layer correlations and bulk 3D correlations. We obtain similar effects for the s–d exchange coupling constant, too. We have calculated β_s for a three-dimensional cubic lattice for $J_s = 0.2J$, $I_s = 0.2I$ and $t_s = 0.5t$ in the temperature range $5 \times 10^{-4} < |T_c - T/T_c| < 5 \times 10^{-3}$ from

$$\ln \langle S_s^z \rangle = \ln A + \beta_s \ln(1 - T/T_c) \tag{24}$$

and obtained the following value, which is different compared with the bulk $\beta = 0.361 \pm 0.006$ [17],

$$\beta_s = 0.781 \pm 0.006 \qquad A = 1.706 \pm 0.006. \tag{25}$$

The critical exponent $\beta_s = 0.781$ is independent of I_s/I in the case when $I_s/I < 1$. Then β_s steadily decreases with increasing of I_s/I until, at bout $I_s/I \approx 2$ it is $\beta_s = 0.101 \pm 0.006$, i.e. it becomes less than the two-dimensional value $\beta(2D) = 0.125$. Clearly, this variation of β_s with I_s/I is due to crossover associated with the multicritical point at I_{sm} [2]. For $I_s > I_{sm}$ the surface orders at $T_c^s > T_c^b$ and hence $\beta_s < \beta(2D)$. The calculations show that $T_c^s > T_c^b$ at least for $I_s/I \approx 1.2 \pm 0.06$ with $\beta_{sm} = 0.171 \pm 0.016$. The obtained values for β_s and β_{sm} are consistent with renorm-group predictions [18, 19] and with the Monte Carlo method [2] for ferromagnetic systems.

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