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Critical Behavior at Surfaces of Strong Coupling Paramagnetic Systems Exhibiting a Paramagnetic-Ferrimagnetic Transition

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Abstract The purpose of this work is the investigation of the critical surface effects of two strongly coupled paramagnetic sublattices exhibiting a para-ferrimagnetic transition. The model is of Landau-Ginzburg type, whose bulk free energy is a functional of two kind of order parameters (local magnetizations) φ and ψ. This free energy involves, beside quadratic and quartic terms in both φ and ψ, a lowest-order coupling, $-C_o\varphi\psi$, where $C_o < 0$ is the coupling constant measuring the interaction between the two sublattices. Two terms $H\varphi$ and $H\psi$ are also introduced, to describe the interaction within an external magnetic field H. We introduce a surface free energy expanded in terms of the local order parameters. The magnetization at the surface are φ_s and ψ_s. We show, in particular, that the model can be reduced to an effective theory written in terms of the overall magnetization $\phi = \varphi + \psi$ and the associated fraction of magnetization $\eta_\alpha = \varphi/(\varphi + \psi)$. This formulation leads us to define an effective extrapolation length λ_S. We then derive all the critical properties of the system close to the critical temperature T_c. In particular, we determine the critical behavior of the overall surface magnetization $\phi_s = \varphi_s + \psi_s$, in terms of ϕ_b above and below T_c. The variations of ϕ_s with the magnetic field H, and when a surface field H_s is applied actually at T_c, are derived. We determine, also, the associated susceptibilities at the surface χ_s and $\chi_{s,s}$. The determination of the full profile of the magnetization close to the surface will be the subject of a future communication.

Keywords: *sublattices, coupling, paramagetism, ferrimagnetism, transition, critical surface effects*

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1. Introduction

Magnetic and structural properties of the so-called super-weak ferrimagnetic systems are a subject of a great deal of attention from theoretical and experimental point of view. This is due to their considerable importance, especially, in the domain of energy stocking (long life lithium batteries). They may exhibit a para-ferrimagnetic transition at a critical temperature T_c greater than room temperature. Among these, we can quote certain members of Heusler Pauli-paramagnetic alloys [1] based on the composition X_2YZ, with ($X = Pd, Cu$; $Y = T_i, V$ and $Z = Al, In, Sn$), and lamellar Curie-Weiss paramagnetic compounds [2], like $AM_xM'_{1-x}O_2$ ($0 \leq x < 1$) with ($A = Li, Na, K; M, M' = N, Co, \ldots$). The common feature of these materials is that they present a small magnetization at low temperature, in contrary to the usual ferrimagnetic materials.

To study the critical magnetic behavior of such materials, a continuous model based on the landau theory [3,4,5] has been successfully used by Neumann and co-workers [6]. Such a model assumes that the material consists of a lattices made up of two coupled Pauli or Curie-Weiss paramagnets sublattices [7,8], with respective local magnetizations φ and ψ. Above the critical temperature T_c both magnetizations vanish and the system is a paramagnet. Below this temperature an antiparallel configuration of the magnetization is favored, but with non-vanishing overall magnetization. One can say that the material exhibits a *ferrimagnetic* state. Quantitatively, this coupling manifests itself through the introduction of an extra term $-C_o\varphi\psi$ in the free energy. Negative values of the coupling constant C_o favor the anti-parallel alignment of the local moments φ and ψ, and ferrimagnetic order appears. Within the framework of this model the par-ferrimagnetic transition in the bulk arising from these materials was widely studied applying, first, from a mean-field point of view, the theory has been developed through numerical method [6], and through an exact analytic analysis [9,10,11]. Second, using the Renormalization-Group techniques [12,13] as in the case of usual para-ferromagnetic transition [14,15,16].

The purpose of this work is to investigate the critical properties of the system at surfaces. Indeed, near a second order phase transition point, the correlation length or order parameters fluctuations becomes long-ranged, and hence the effect of surfaces on the bulk properties of the system is much more drastic. We will focus our attention to the situation where the surface still makes a small contribution to any bulk property only. Then it makes sense to split the free energy, in a bulk term proportional to the volume and a surface term proportional to the surface area. We first, reformulate the model in terms of an effective φ^4-theory using the fraction of the magnetization $\eta_\alpha = \varphi/\phi$, and the order parameter of interest is then the overall magnetization $\phi = \varphi + \psi$. Under these considerations the effective phenomenological constants of the free energy involve information about competition between the coupling C_o and temperature. The surface free energy is expanded in terms of the fraction of the magnetization at surface $\eta_s = \varphi_s/\phi_s$ with $\phi_s = \varphi_s + \psi_s$. This leads us to define an effective extrapolation length λ_s (see below). We determine, in particular, the overall magnetization at surface ϕ_s as function of λ_s, the bulk correlation length ξ_b^-, and the overall bulk magnetization ϕ_b. In addition to these quantities, ϕ_s depends, also, on the surface and bulk square masses A_s and A_b, that reflect the dependence on both temperature and coupling C_o. We determine, also, the local susceptibilities at the surface. The first is the response χ_s of a surface spin to a uniform field acting throughout the system, and the second is the response $\chi_{s,s}$ to a field acting in a surface.

The remainder of presentation proceeds as follows. Section to is devoted to a description of the used model. The reformulation of this latter as an effective φ^4-theory, and the investigation of the mean-field critical properties at surface of the system is the aim of Section 3. We draw some concluding remarks in section 4.

2. The Model

The physical system we consider here consists of two strongly coupled sublattices, of respective moments φ and ψ. For small moments and in the presence of an applied external magnetic field H, in a Landau approximation, the bulk free energy allowing to investigate the para-ferrimagnetic transition within this system writes [6,9,10]

$$\begin{aligned}\frac{F_b}{T} = \int d\vec{r}\Big\{&\frac{1}{2}c_\varphi\left(\nabla\varphi(\vec{r})\right)^2 + \frac{1}{2}c_\psi\left(\nabla\psi(\vec{r})\right)^2\\ &+\frac{a}{2}\varphi^2(\vec{r}) + \frac{A}{2}\psi^2(\vec{r}) - C_o\varphi(\vec{r})\psi(\vec{r})\\ &+\frac{u}{4}\varphi^4(\vec{r}) + \frac{v}{4}\psi^4(\vec{r}) - H(\vec{r})\left(\varphi(\vec{r})+\psi(\vec{r})\right)\Big\}.\end{aligned} \tag{2.1}$$

The squared gradient terms on the right-hand side of relation (2.1) traduce the spatial variations of order parameters φ and ψ. There, $\vec{r}$ stands for the d-dimensional position vector of the considered point. In relation (2.1), the coupling constants u and v are taken to be positive, to ensure the stability of the free energy. Coefficients a and A depend on temperature according to:

$$a(T) = a_o + a_1 T^2 > 0;\ A(T) = A_o + A_1 T^2 > 0, \tag{2.2A}$$

for a Pauli paramagnet [7,8], or

$$a(T) = \bar{a}(T-\theta_1);\ A(T) = \bar{a}(T-\theta_2), \tag{2.2b}$$

for a Curie-Weiss paramagnet [8,17]. a_o and a_1 appearing in relation $(2.2a)$, have a simple dependence in both free electron density and Fermi energy relative to the two sublattices [18]. In relation $(2.2b)$, the Curie-Weiss temperatures θ_1 and θ_2 are proportional to exchange integrals $J_1 > 0$ and $J_2 > 0$, inside the sublattices [12]. The extra term C_o in Eq. (2.1) represent the lowest-order coupling between the two sublattices. Such a term plays, in fact, the role of an internal magnetic field. For a negative coupling constant $C_o < 0$, an antiparallel configuration of magnetizations φ and ψ is favored, while $C_o > 0$ favors their parallel alignment. For Curie-Weiss materials like lamellar compounds [2,19], the coupling C_o is proportional to the exchange integral J_{12} between the two sublattices. In this work we are concerned only with negative values of C_o, in order to investigate the ferrimagnetic state of the system. $H(\vec{r})$ is a (suitable normalized) magnetic field. For a film of thickness L the generalization of Eq. (2.1) is

$$F/T = F_b/T + F_s/T, \tag{2.3}$$

where similarly the surface free energy F_s is expanded in terms of the local order parameters including terms up to second order-only

$$\begin{aligned}\frac{F_s}{T} = \int d\vec{\rho}\Big\{&\frac{1}{2}c_\varphi\lambda_\varphi^{-1}\left[\varphi^2(\vec{\rho},z=0)+\varphi^2(\vec{\rho},z=L)\right]\\ &+\frac{1}{2}c_\psi\lambda_\psi^{-1}\left[\psi^2(\vec{\rho},z=0)+\psi^2(\vec{\rho},z=L)\right]\\ &+c_{\varphi\psi}\lambda_{\varphi\psi}^{-1}\left[C_o\varphi(\vec{\rho},z=0)\psi(\vec{\rho},z=0)\right.\\ &\left.+C_o\varphi(\vec{\rho},z=L)\psi(\vec{\rho},z=L)\right]\\ &-H_s(\vec{\rho})[\varphi(\vec{\rho},z=0)+\psi(\vec{\rho},z=0)\\ &+\varphi(\vec{\rho},z=L)+\psi(\vec{\rho},z=L)]\Big\}.\end{aligned} \tag{2.4}$$

We wish to study the system close enough to T_c we then neglect higher terms in F_s in Eq. (2.4). The linear term involves a field $H_s(\vec{\rho})$ acting on spins in the surface plane only, and the constants of the quadratic terms were written arbitrarily as $(c_i\lambda_i^{-1};\ i = \varphi, \psi\ or\ \varphi\psi)$, where the parameters λ_i have the dimension of a length and are called extrapolation lengths [20]. Next we consider the case where $H(\vec{r})$ and $H_s(\vec{\rho})$ are homogeneous and disregard variations of the magnetization within the layers, hence replacing $\varphi(\vec{r})$ and $\psi(\vec{r})$ by their averages over $\vec{\rho}$, which we denote simply by $\varphi(z)$ and $\psi(z)$. Equations (2.3) and (2.4) then become

$$\begin{aligned}\frac{F}{TS} = \int_0^L dz\Big\{&\frac{1}{2}c_\varphi\left(\frac{\partial\varphi}{\partial z}\right)^2 + \frac{1}{2}c_\psi\left(\frac{\partial\psi}{\partial z}\right)^2 + \frac{a}{2}\varphi^2(z)\\ &+\frac{A}{2}\psi^2(z) - C_o\varphi(z)\psi(z) + \frac{u}{4}\varphi^4(z)\\ &+\frac{v}{4}\psi^4(z) - H\left(\varphi(z)+\psi(z)\right)\Big]\Big\}\\ +\Big\{&\frac{1}{2}c_\varphi\lambda_\varphi^{-1}\left[\varphi^2(z=0)+\varphi^2(z=L)\right]\\ &+\frac{1}{2}c_\psi\lambda_\psi^{-1}\left[\psi^2(z=0)+\psi^2(z=L)\right]\\ &+c_{\varphi\psi}\lambda_{\varphi\psi}^{-1}\left[C_o\varphi(z=0)\psi(z=0)\right.\\ &\left.+C_o\varphi(z=L)\psi(z=L)\right]\\ &-H_s\left[\varphi(z=0)+\psi(z=0)\right.\\ &\left.+\varphi(z=L)+\psi(z=L)\right]\Big\}.\end{aligned} \tag{2.5}$$

Functional differentiation of Eq. (2.5) yields ($2S$ being the total surface area of the film)

$$a\varphi(z)+u\varphi^3(z)-C_o\psi(z)-c_\varphi\left(\frac{\partial^2\varphi}{\partial z^2}\right)=H, \quad (2.6a)$$

$$A\psi(z)+v\psi^3(z)-C_o\varphi(z)-c_\psi\left(\frac{\partial^2\psi}{\partial z^2}\right)=H, \quad (2.6b)$$

for which the surface terms in Eq. (2.5) supply the boundary conditions

$$\frac{\partial\varphi}{\partial z}-\frac{\varphi}{\lambda_\varphi}-\frac{c_{\varphi\psi}}{c_\varphi\lambda_{\varphi\psi}}\psi=-\frac{H_s}{c_\varphi}, z=0, \quad (2.7a)$$

$$\frac{\partial\psi}{\partial z}-\frac{\psi}{\lambda_\varphi}-\frac{c_{\varphi\psi}}{c_\psi\lambda_{\varphi\psi}}\varphi=-\frac{H_s}{c_\psi}, z=0, \quad (2.7b)$$

and

$$\frac{\partial\varphi}{\partial z}+\frac{\varphi}{\lambda_\varphi}+\frac{c_{\varphi\psi}}{c_\varphi\lambda_{\varphi\psi}}\psi=\frac{H_s}{c_\varphi}, z=L, \quad (2.8a)$$

$$\frac{\partial\psi}{\partial z}+\frac{\psi}{\lambda_\varphi}+\frac{c_{\varphi\psi}}{c_\psi\lambda_{\varphi\psi}}\varphi=\frac{H_s}{c_\psi}, z=L. \quad (2.8b)$$

It is important to recall that some previous results [9] can be obtained from Eqs. (2.6), for the overall bulk magnetization $\phi_b=\varphi_b+\psi_b$ and the overall bulk susceptibility χ_b of a homogeneous system where $\left(\frac{\partial^2\varphi}{\partial z^2}\right)$ and $\left(\frac{\partial^2\psi}{\partial z^2}\right)$ can be omitted. The first important result is that the critical temperature occurs at [9]

$$a(T_c)A(T_c)=C_o^2 \Leftrightarrow C_o^2-a(T)A(T)\sim(T-T_c). \quad (2.9)$$

Just below the critical temperature $T<T_c$, the overall magnetization ϕ_b is given by

$$\phi_b \cong \pm\left(\frac{1}{\sqrt{av+uA^3/C_o^2}}-\frac{1}{\sqrt{Au+va^3/C_o^2}}\right)\sqrt{C_o^2-aA} \quad (2.10)$$
$$\cong\sqrt{(T-T_c)}\;.T\underset{T<T_c}{\rightarrow}T_c, H=0,$$

and the overall susceptibility is given by (superscripts + and – distinguish temperatures above and below T_c)

$$\chi_b^+=\frac{a+A+2C_o}{aA-C_o^2}\cong\chi_o^+(T-T_c)^{-1};T\underset{T>T_c}{\rightarrow}T_c, H=0, \quad (2.11a)$$

$$\chi_b^-=\frac{1}{2}\frac{a+A+2C_o}{C_o^2-aA}\cong\chi_o^-(T-T_c)^{-1}; \quad (2.11b)$$
$$T\underset{T<T_c}{\rightarrow}T_c, H=0.$$

We recall also that the correlation length is given by [9]

$$\xi\cong\left|C_o^2-aA\right|^{-1/2}\cong|t|^{-1/2};t=(T-T_c)/T. \quad (2.12)$$

3. Effective Theory and Critical Behavior at Surface

We focus, for the moment, our attention to a semi-infinite system, $L\to\infty$; in this case the phase transition in the bulk of the film occurs at precisely the critical temperature T_c as for a fully infinite system, and we may replace the second boundary condition, Eqs. (2.8), by

$$\begin{cases}\left.\frac{\partial\varphi}{\partial z}\right|_{z\to\infty}=0,\ \varphi(z\to\infty)=\varphi_b,\\ \left.\frac{\partial\psi}{\partial z}\right|_{z\to\infty}=0,\ \psi(z\to\infty)=\psi_b.\end{cases} \quad (3.1)$$

We can obtain the magnetizations at the surface $\varphi_s=\varphi(z=0)$ and $\psi_s=\psi(z=0)$ multiplying respectively Eqs. $(2.6a)$ and $(2.6b)$, by $\left(\frac{\partial\varphi}{\partial z}\right)$ and $\left(\frac{\partial\psi}{\partial z}\right)$ respectively, and integrating over z from zero to infinity, whereby the boundary conditions Eqs. (2.7) and (3.1), can be used. This leads to the following two coupled equations

$$\begin{aligned}&\frac{a}{2}\left(\varphi_b^2-\varphi_s^2\right)+\frac{u}{4}\left(\varphi_b^4-\varphi_s^4\right)\\&-C_o\int_0^\infty\psi\frac{\partial\varphi}{\partial z}dz+\frac{1}{2}c_\varphi\left(\frac{\partial\varphi}{\partial z}\right)^2\\&=H(\varphi_b-\varphi_s),\end{aligned} \quad (3.2a)$$

$$\begin{aligned}&\frac{A}{2}\left(\psi_b^2-\psi_s^2\right)+\frac{v}{4}\left(\psi_b^4-\psi_s^4\right)\\&-C_o\int_0^\infty\varphi\frac{\partial\psi}{\partial z}dz+\frac{1}{2}c_\psi\left(\frac{\partial\psi}{\partial z}\right)^2\\&=H(\psi_b-\psi_s).\end{aligned} \quad (3.2b)$$

Making the sum of Eqs $(3.2a)$ and $(3.2b)$, we have then

$$\begin{aligned}&\frac{a}{2}\left(\varphi_b^2-\varphi_s^2\right)+\frac{A}{2}\left(\psi_b^2-\psi_s^2\right)+\frac{u}{4}\left(\varphi_b^4-\varphi_s^4\right)\\&+\frac{v}{4}\left(\psi_b^4-\psi_s^4\right)-C_o(\varphi_b\psi_b-\varphi_s\psi_s)\\&+\frac{1}{2}c_\varphi\left(\frac{\partial\varphi_s}{\partial z}\right)^2+\frac{1}{2}c_\psi\left(\frac{\partial\psi_s}{\partial z}\right)^2\\&=H((\varphi_b-\varphi_s)+(\psi_b-\psi_s)).\end{aligned} \quad (3.3)$$

$$\eta_b=\frac{\varphi_b}{\varphi_b+\psi_b};1-\eta_b=\frac{\psi_b}{\varphi_b+\psi_b}, \quad (3.4a)$$

$$\eta_s=\frac{\varphi_s}{\varphi_s+\psi_s};1-\eta_s=\frac{\psi_s}{\varphi_s+\psi_s}, \quad (3.4b)$$

where η_b and η_s can be viewed as the fractions of bulk and surface magnetizations relatively to the two sublattices. With help of these changes and using relation (2.7), Eq. (3.3) becomes

$$\begin{aligned}&\phi_b^4\left(\frac{u}{4}\eta_b^4+\frac{v}{4}(1-\eta_b)^4\right)\\&+\phi_b^2\left(\frac{a}{2}\eta_b^2+\frac{A}{2}(1-\eta_b)^2-C_o\eta_b(1-\eta_b)\right)\\&-\phi_s^4\left(\frac{u}{4}\eta_s^4+\frac{v}{4}(1-\eta_s)^4\right)\\&-\phi_s^2\left(\frac{a}{2}\eta_s^2+\frac{A}{2}(1-\eta_s)^2-C_o\eta_s(1-\eta_s)\right)\\&+\frac{1}{2}c_s\left(\frac{\phi_s}{\lambda_s}-\frac{H_s}{c_s}\right)^2=H(\phi_b-\phi_s),\end{aligned}\quad(3.5)$$

where c_s is an effective constant and λ_s the effective extrapolation length, which write

$$c_s=c_\varphi\eta_s^2+c_\psi(1-\eta_s)^2,\quad(3.6a)$$

and

$$\lambda_s=\frac{c_\varphi\eta_s^2+c_\psi(1-\eta_s)^2}{\frac{c_\varphi}{\lambda_\varphi}\eta_s^2+\frac{c_\psi}{\lambda_\psi}(1-\eta_s)^2+2\frac{c_{\varphi\psi}}{\lambda_{\varphi\psi}}\eta_s(1-\eta_s)}.\quad(3.6b)$$

It is important to note that we can write the model as an effective φ^4-theory in terms of the fraction of magnetizations $\eta_\alpha=\varphi/(\varphi+\psi)$ and the overall magnetization $\phi=(\varphi+\psi)$. Indeed, under these considerations the free energy (2.5) reduces to

$$\begin{aligned}\frac{F}{TS}=\int_0^L dz\Bigg\{&\frac{1}{2}c_\alpha\left(\frac{\partial\phi}{\partial z}\right)^2+\frac{A_\alpha}{2}\phi^2(z)\\&+\frac{B_\alpha}{4}\phi^4(z)-H\phi(z)\Bigg\}+F_s,\end{aligned}\quad(3.7)$$

with

$$A_\alpha=a\eta_\alpha^2+A(1-\eta_\alpha)^2-2C_o\eta_\alpha(1-\eta_\alpha),\quad(3.8a)$$

$$B_\alpha=u\eta_\alpha^4+v(1-\eta_\alpha)^4,\quad(3.8b)$$

$$c_\alpha=c_\varphi\eta_\alpha^2+c_\psi(1-\eta_\alpha)^2,\quad(3.8c)$$

and

$$\begin{aligned}F_s=&\frac{1}{2}\frac{c_s}{\lambda_s}\left[\phi^2(z=0)+\phi^2(z=L)\right]\\&-H_s\left[\phi(z=0)+\phi(z=L)\right],\end{aligned}\quad(3.9)$$

where c_s and λ_s are those parameters given by Eqs. $(3.6a-b)$. Functional differentiation of Eq. (3.7) yields an equation due to Ginsburg Landau [21] and familiar from theory of superconductivity, but with the effective phenomenological parameters A_α, B_α and c_α. Notice that these latters contain all information about the system (like dependence in temperature, coupling…).

$$A_\alpha\phi(z)+B_\alpha\phi^3(z)-c_\alpha\left(\frac{\partial^2\phi}{\partial z^2}\right)=H,\quad(3.10)$$

for which the surface term in Eq. (3.7) supply the boundary conditions

$$\frac{\partial\phi}{\partial z}-\frac{\phi}{\lambda_s}=-\frac{H_s}{c_s},\ z=0,\quad(3.11a)$$

$$\frac{\partial\phi}{\partial z}+\frac{\phi}{\lambda_s}=\frac{H_s}{c_s},\ z=L.\quad(3.11b)$$

From Eq. (3.10) one obtains the standard results for the bulk magnetization $\phi_b=\varphi_b+\psi_b$ and the overall susceptibility χ_b, for a homogeneous system where $\left(\frac{\partial^2\phi}{\partial z^2}\right)$ can be omitted

$$\phi_b=\sqrt{-\frac{A_b}{B_b}}\cong(T-T_c)^{\frac{1}{2}},T\to T_c^-,H=0,\quad(3.12)$$

with

$$A_b=a\eta_b^2+A(1-\eta_b)^2-2C_o\eta_b(1-\eta_b),\quad(3.13a)$$

$$B_b=u\eta_b^4+v(1-\eta_b)^4.\quad(3.13b)$$

The fraction of bulk magnetization η_b is that given by relation $(3.4a)$. Note that close to the critical temperature T_c, the effective phenomenological parameter A_b behaves as: $A_b\cong(T-T_c)$. Indeed at $T\cong T_c$, we have: $aA\cong C_o^2$, which can be written as de product of the two following equations

$$a\eta_b=C_o(1-\eta_b)\Leftrightarrow\eta_b=\frac{C_o}{a+C_o},\quad(3.14a)$$

$$A(1-\eta_b)=C_o\eta_b\Leftrightarrow\eta_b=\frac{A}{A+C_o}.\quad(3.14b)$$

Reporting these expressions of η_b in relation $(3.13a)$, one obtains $A_b\cong(aA-C_o^2)\cong(T-T_c)$. The overall susceptibility χ_b above and below T_c, can be obtained directly by taking the first derivative of Eq. (3.10) with respect of the magnetic field H, and setting $H=0$, we find

$$\chi_b^+=A_b^{-1}\cong\chi_o^+(T-T_c)^{-1},T\to T_c^+,H=0,\quad(3.15a)$$

$$\chi_b^-=\frac{1}{A_b+3B_b\phi_b^2}=-\frac{1}{2A_b}\cong\chi_o^-(T-T_c)^{-1},\quad T\to T_c^-,H=0.\quad(3.15b)$$

Expressions of the correlation length ξ_b above and below T_c can also be extracted

$$\xi_b^+=\sqrt{\frac{c_b}{A_b}}\cong\xi_o^+(T-T_c)^{-\frac{1}{2}},T\to T_c^+,H=0,\quad(3.16a)$$

$$\begin{aligned}\xi_b^-=&\sqrt{\frac{c_b}{A_b+3B_b\phi_b^2}}=\sqrt{\frac{c_b}{2A_b}}\\&\cong\xi_o^-(T-T_c)^{-\frac{1}{2}},T\to T_c^-,H=0,\end{aligned}\quad(3.16b)$$

where $c_b = c_\varphi \eta_b^2 + c_\psi(1-\eta_b)^2$. Remark that the behavior of these bulk quantities Eqs. (3.12), (3.15) and (3.16), obtained from the effective theory are equivalent to those derived above from the theory written as two coupled order parameters and given by Eqs. (2.10) − (2.12). It is also important to note that for a semi-infinite system, $L \to \infty$, we may replace the boundary condition Eq. $(3.11b)$, by

$$\left.\frac{\partial \phi}{\partial z}\right|_{z\to\infty} = 0, \phi(z\to\infty) = \phi_b \qquad (3.17)$$

and the magnetization at the surface is given by: $\phi_s = \phi(z=0)$. Multiplying Eq. (3.10) by $\left(\frac{\partial \phi}{\partial z}\right)$ and integrating over z from zero to infinity, whereby the boundary conditions Eqs. $(3.11a)$ and (3.17), can be used. We obtain directly the equation (3.5) which writes

$$\frac{B_b}{4}\phi_b^4 + \frac{A_b}{2}\phi_b^2 - \frac{B_s}{4}\phi_s^4 - \frac{A_s}{2}\phi_s^2 + \frac{1}{2}c_s\left(\frac{\phi_s}{\lambda_s} - \frac{H_s}{c_s}\right)^2 = H(\phi_b - \phi_s). \qquad (3.18)$$

To solve this latter we first assume positive surface energy, $(\lambda_s > 0)$. Then a spontaneous magnetization ϕ_s at the surface exists only for $T < T_c$, where a spontaneous magnetization also exists in the bulk. Equation (3.18) yields $(H = H_s = 0)$

$$\phi_s^4 - \frac{2}{B_s}\left(\frac{c_s}{\lambda_s^2} - A_s\right)\phi_s^2 + \frac{1}{3B_s}\left(\frac{c_b}{\left(\xi_b^-\right)^2} - A_b\right)\phi_b^2 = 0 \quad (3.19)$$

where A_s and B_s are those surface parameters given by

$$A_s = a\eta_s^2 + A(1-\eta_s)^2 - 2C_o\eta_s(1-\eta_s), \qquad (3.20a)$$

$$B_s = u\eta_s^4 + v(1-\eta_s)^4. \qquad (3.20b)$$

The solution of Eq. (3.19) writes then

$$\phi_s^2 = \frac{k_s}{B_s} - \sqrt{\left(\frac{k_s}{B_s}\right)^2 - \frac{1}{3}\frac{k_b}{B_s}\phi_b^2}, \qquad (3.21)$$

which for $T \to T_c^-$ reduces to

$$\phi_s \cong \left(\frac{1}{6}\frac{k_b}{k_s}\right)^{\frac{1}{2}}\phi_b \cong \left(\frac{1}{6}\frac{k_b}{k_s}\right)^{\frac{1}{2}}(T - T_c), \qquad (3.22)$$

where k_s and k_b are given by

$$k_s = \frac{c_s}{\lambda_s^2} - A_s, \qquad (3.23a)$$

$$k_b = \frac{c_b}{\left(\xi_b^-\right)^2} - A_b. \qquad (3.23b)$$

Expression (3.22) shows that the magnetization at surface vanishes linearly at the critical temperature T_c. Notice that for a vanishing coupling constant $(C_o = 0)$ where the two sublattices are decoupled, the ratio (k_b/k_s) reduces to (λ_s/ξ_b^-) which is the result relative to the usual $\varphi^4 - theory$ [20]. It is also straightforward to obtain the variation of the bulk magnetization with the magnetic field actually at T_c (where $H_s = 0$). Indeed at $T = T_c$ the temperature parameter $A_b \cong 0$, from Eq. (3.10) one obtain

$$\phi_b = \left(\frac{H}{B_b}\right)^{\frac{1}{3}} = \left(\frac{H}{u\eta_b^4 + v(1-\eta_b)^4}\right)^{\frac{1}{3}}, H \to 0^+ \quad (3.24a)$$

and using Eq. $(3.16b)$, ϕ_s writes then

$$\phi_s = \left(\frac{B_b^{-\frac{1}{3}}}{2k_s}\right)^{\frac{1}{2}} H^{\frac{2}{3}}, H \to 0^+, \qquad (3.24b)$$

In contrast, if a surface field is applied only exactly at T_c, it cannot induce any magnetization in the bulk and the magnetization at the surface is simply given by

$$\phi_s \cong \frac{1}{\lambda_s k_s}\left(1 + \sqrt{\frac{A_s}{c_s}}\lambda_s\right)H_s. \qquad (3.25)$$

Next we obtain the susceptibilities at the surface, considering the linear response of ϕ_s with respect to both H and H_s, above and below T_c. Inserting $\phi_b = \chi_b^+ H$ in Eq. (3.18), and using Eq. (3.15a) for $T > T_c$ and $H_s = 0$, yields

$$\phi_s \cong \left(\frac{1}{k_s}\right)^{\frac{1}{2}} A_b^{-\frac{1}{2}} H \cong \left(\frac{1}{k_s}\right)^{\frac{1}{2}} (T - T_c)^{-\frac{1}{2}} H, \quad (3.26a)$$

$$\chi_s^+ \cong \left(\frac{1}{k_s}\right)^{\frac{1}{2}} (T - T_c)^{-\frac{1}{2}}. \qquad (3.26b)$$

and using $\phi_b \cong \sqrt{\frac{A_b}{B_b}} + \chi_b^- H$, for $T < T_c$ and close to T_c, yields

$$\phi_s \cong \left(-\frac{1}{2A_b k_s}\right)^{\frac{1}{2}} H \cong \left(-\frac{1}{2k_s}\right)^{\frac{1}{2}} (T_c - T)^{-\frac{1}{2}} H, \quad (3.27a)$$

$$\chi_s^- \cong \left(-\frac{1}{2k_s}\right)^{\frac{1}{2}} (T_c - T)^{-\frac{1}{2}}. \qquad (3.27b)$$

Similarly, one can obtain the response to the local field H_s (at $H = 0$). Above the critical temperature and from Eq. (3.18), surface magnetization ϕ_s and the associated susceptibility $\chi_{s,s}^+$ are given by

$$\phi_s \cong \frac{\lambda_s}{c_s}\left(\frac{1}{1 - \lambda_s\sqrt{\frac{A_s}{c_s}}}\right)H_s, T > T_c, \qquad (3.28a)$$

and then

$$\chi_{s,s}^{+} = \left(\frac{\partial \phi_s}{\partial H_s}\right) \cong \frac{\lambda_s}{c_s}\left(\frac{1}{1-\lambda_s\sqrt{\frac{A_s}{c_s}}}\right), T > T_c. \quad (3.28b)$$

Below and close to the critical temperature $(T < T_c)$, Eq. (3.18) writes

$$\left(\frac{c_s}{\lambda_s^2} - A_s\right)\phi_s^2 - 2\frac{H_s}{\lambda_s}\phi_s + \frac{H_s^2}{c_s} - \frac{A_b^2}{B_b} = 0, \quad (3.29)$$

and the solution of this equation is given by

$$\phi_s = \frac{1}{k_s}\left(\frac{H_s}{\lambda_s} \pm \sqrt{\frac{A_s}{c_s}H_s^2 + \frac{A_b^2}{B_b}k_s}\right), T < T_c, \quad (3.30a)$$

this reduces, for a small surface field H_s, to

$$\phi_s = \frac{1}{k_s}\left(\frac{H_s}{\lambda_s} \pm \sqrt{\frac{A_b^2}{B_b}k_s}\right), \; T < T_c, \quad (3.30b)$$

and then

$$\chi_{s,s}^{-} = \left(\frac{\partial \phi_s}{\partial H_s}\right) \cong \frac{1}{\lambda_s k_s}, \; T < T_c. \quad (3.30c)$$

The above results, show clearly that the dependence of ϕ_s, χ_s and $\chi_{s,s}$ on the shift to critical temperature $(T - T_c)$ differs from the behavior of the corresponding bulk properties: while $\chi_b \sim |T - T_c|^{-1}$ we have $\chi_s \sim |T - T_c|^{-1/2}$, and $\chi_{s,s}$ even remains finite at T_c.

4. Concluding Remarks

We have investigated, in this work, the critical properties at surface within superweak ferrimagnetic materials undergoing a para-ferrimagnetic transition. We have, first, shown that the free energy governing the system can be written as an effective φ^4-theory in terms of the overall magnetization $\phi = (\varphi + \psi)$. The phenomenological parameter multiplying the quadratic and quartic terms are, in this case, functional of the fraction of the magnetization $\eta = \varphi/(\varphi + \psi)$. This leads to an effective extrapolation length λ_s appearing in the surface energy term. Through this formulation we have derived, in particular, the overall surface magnetization ϕ_s. We have shown that, in addition of the extrapolation λ_s and the bulk correlation length ξ_b^-, the surface magnetization ϕ_s depend also on the effective bulk and surface temperature parameters A_b and A_s. We have also determined the surface susceptibilities χ_s and $\chi_{s,s}$, considering the linear response with respect to both the external magnetic field H and the local surface field H_s, above and below critical temperature T_c. We have found that the critical exponents remain the same as those relative to the usual φ^4-theory, but the amplitudes are changed. We note that a work dealt with the order parameter profiles near surface is in progress.

The mean-field theory (MFT) of critical behavior in the bulk is known to be inaccurate for systems below their marginal dimensionality. There is no reason whatsoever to assume that the MFT for the critical behavior of surfaces is any more accurate than for the bulk. To have non-classical exponents and their associated scaling laws, one has to use appropriate homogeneity assumptions such as finite size scaling theory. Such a work is also in progress.

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