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# Landau expansion for a metamagnetic model 

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#### Abstract

We performed a detailed Landau expansion of the free energy for a metamagnetic model considering terms up to twelfth order. We obtained explicit expressions for the coefficients as a function of the temperature and the ratio between ferro- and antiferromagnetic interactions. We showed that a naive analysis based on the signs of these coefficients cannot always give us sufficient guarantee about the correctness of the phase diagram of the model. In these cases it is necessary to resort to the full expression of the free energy in order to characterize the nature of the phase transition.


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## 1 Introduction

Kincaid and Cohen [1] presented an extensive theoretical review work concerning the mean field properties of the Ising metamagnetic model. They considered a twosublattice model with competing ferromagnetic (intrasublattice) and antiferromagnetic (intersublattice) interactions. They showed that depending on the value of the ratio between these interactions the phase diagram of the model can exhibit different types of critical points. Above a given value of this ratio the phase diagram displays a tricritical behavior, while below this ratio the phase diagram presents a critical endpoint and a bicritical endpoint. Although the tricritical point is observed in all known real metamagnets [2,3], the critical and the bicritical endpoints have not yet been observed in any real system. Monte Carlo simulations [4-9] and dynamical pair approximation [10] do not support the idea of a decomposition of the tricritical point into a critical and a bicritical endpoints. Selke [7] and Pleimling and Selke [8] showed that the specific heat and the magnetization of the layered metamagnets can exhibit some anomalies, but this fact was not sufficient to guarantee that the above cited decomposition of the tricritical point takes place. The anomalies are enhanced for very small values of the intralayer coupling and high coordinated interlayer coupling. However, the decomposition of the tricritical point is present both in meanfield and Monte Carlo simulations for other systems, such as in the three-dimensional Blume-Capel model [11].

In this paper we revisit the Ising metamagnetic model, but our interest now is to investigate the behavior of its Landau coefficients as a function of the temperature, mag-

[^0]netic field, and the ratio between ferro- and antiferromagnetic couplings. As it is well known the basic idea of the Landau theory of the continuous phase transitions is that at the transition point the order parameter of the system changes continuously from one phase to the other. In their work Kincaid and Cohen [1] performed a Landau expansion of the free energy for the Ising metamagnetic model, and by a careful analysis of the signs of the coefficients in the expansion foresaw the possibility of different phase diagrams, as exposed above. As we will show next, for suitable values of the ratio between the ferro- and antiferromagnetic couplings, a bicritical endpoint that would be expected in the phase diagram based only on the signs of Landau coefficients, is not really present.

In the next section we present the model for the layered metamagnet, the exact free-energy for the CurieWeiss version of the model system, and perform the corresponding Landau expansion. Our approach is somewhat different but results previously derived by Kincaid and Cohen [1] are reproduced for clarity. In Section 3 we present the phase diagram and calculate the critical points of the model. Finally, in Section 4, we present our conclusions.

## 2 Model

We have considered an Ising spin system on a cubic lattice, formed by two sublattices. The sublattices are chosen to be the alternating layers of the lattice. The exchange interaction between first neighboring spins on the same sublattice is of the ferromagnetic type, while the coupling between neighboring spins belonging to different sublattices is of the antiferromagnetic type. The Hamiltonian of
the model is

$$
\begin{equation*}
\mathcal{H}=-\frac{1}{2} \sum_{i, j} \sigma_{i} J_{i j} \sigma_{j}-\sum_{i} H_{i} \sigma_{i} \tag{1}
\end{equation*}
$$

where the first sum is over all pairs of nearest neighbors and $\sigma_{i}= \pm 1$. We choose $J_{i j}=J_{F}>0$ for spins interacting ferromagnetically inside the planes, and $J_{i j}=-J_{A}<0$ for spins interacting antiferromagnetically between adjacent planes. The second sum in equation (1) represents the interaction of the spins with the magnetic field. We considered two types of fields: the physical one, $H$, which is the same for all the lattice sites, and a staggered magnetic field of intensity $H_{s}$, which is directed up on one sublattice and down on the other. Let us introduce the sublattice magnetizations $m_{A}$ and $m_{B}$. The total magnetization $m$ and the staggered magnetization $m_{s}$ are defined by

$$
\begin{equation*}
m=\frac{m_{A}+m_{B}}{2}, \quad m_{s}=\frac{m_{A}-m_{B}}{2} \tag{2}
\end{equation*}
$$

In the last equation the staggered magnetization $m_{s}$ is the order parameter that is conjugate to the staggered field $H_{s}$. For the Curie-Weiss version of Hamiltonian (1), in which each spin interacts with all the others in the system, we can write the exact following expression for the Helmholtz free energy:

$$
\begin{align*}
f\left(m, m_{s}, T\right)= & -\frac{J_{-}}{2} m^{2}-\frac{J_{+}}{2} m_{s}^{2} \\
& +\frac{1}{4 \beta}\left[\left(1+m+m_{s}\right) \ln \left(1+m+m_{s}\right)\right. \\
& +\left(1+m-m_{s}\right) \ln \left(1+m-m_{s}\right) \\
& +\left(1-m+m_{s}\right) \ln \left(1-m+m_{s}\right) \\
& \left.+\left(1-m-m_{s}\right) \ln \left(1-m-m_{s}\right)\right] \tag{3}
\end{align*}
$$

where $\beta=1 / k_{B} T, k_{B}$ is the Boltzmann constant and $T$ is the absolute temperature. Under this approach, the layered and next nearest neighbour antiferromagnets become indistinguishable. We have also defined the new variables $J_{-}=J_{F}-J_{A}$ and $J_{+}=J_{F}+J_{A}$. From this expression we obtain the field equations:

$$
\begin{align*}
H_{s} & =\frac{\partial f}{\partial m_{s}}=-J_{+} m_{s}+\frac{1}{4 \beta} \ln \frac{\left(1+m+m_{s}\right)\left(1-m+m_{s}\right)}{\left(1+m-m_{s}\right)\left(1-m-m_{s}\right)},  \tag{4}\\
H & =\frac{\partial f}{\partial m}=-J_{-} m+\frac{1}{4 \beta} \ln \frac{\left(1+m+m_{s}\right)\left(1+m-m_{s}\right)}{\left(1-m+m_{s}\right)\left(1-m-m_{s}\right)} \tag{5}
\end{align*}
$$

The Landau expansion consists in developing the free energy in a power series of the order parameter, with coefficients depending only on field variables, for standard analysis[12]. So, we perform a Legendre transform on $f\left(m, m_{s}, T\right)$ in order to replace the magnetization $m$ by the magnetic field $H$. That is,

$$
\begin{equation*}
\Psi\left(T, H, m_{s}\right)=f\left(T, m, m_{s}\right)-H m \tag{6}
\end{equation*}
$$

where $m=m\left(T, H, m_{s}\right)$. The expansion takes the form,

$$
\begin{equation*}
\Psi\left(T, H, m_{s}\right)=\sum_{i=0}^{n} \Psi_{2 i} m_{s}^{2 i} \tag{7}
\end{equation*}
$$

where $\Psi_{j}=\Psi_{j}(T, H)$, and we have considered terms up to $n=6$. The expansion of $\Psi\left(T, H, m_{s}\right)$ around $m_{s}=0$ describes the phase transition between a paramagnetic phase, where $m_{s}=0$, and an antiferromagnetic phase, where the order parameter $m_{s}$ is different from zero. As we will discuss next, the expansion can also describe a phase transition between two antiferromagnetic phases, when the order parameter of both phases are close to zero. In any case, the critical behavior of the model is determined by the behavior of the coefficients of the expansion as a function of temperature, magnetic field and coupling constants. In order to obtain explicit expressions for the coefficients $\Psi_{j}$, we first note that they are proportional to the coefficients of $H_{s}$ :

$$
\begin{align*}
H_{s}\left(T, H, m_{s}\right) & =\frac{\partial \Psi}{\partial m_{s}} \\
& =2 \Psi_{2} m_{s}+4 \Psi_{4} m_{s}^{3}+6 \Psi_{6} m_{s}^{5}+\ldots \tag{8}
\end{align*}
$$

When we expand equation (4) in powers of $m_{s}$ we need to consider also $m=m\left(T, H, m_{s}\right)$. However, equation (5) shows that it is not possible to get an explicit expression for $m$, so the following expansion for $m$, due to symmetry, must be used:

$$
\begin{equation*}
m=\alpha_{0}+\alpha_{1} m_{s}^{2}+\alpha_{2} m_{s}^{4}+\alpha_{3} m_{s}^{6}+\ldots, \tag{9}
\end{equation*}
$$

where $\alpha_{i}=\alpha_{i}(T, H)$. Substituting this last expression in equation (5), we arrive at an expression of the form

$$
\begin{equation*}
H=\varphi_{0}+\varphi_{1} m_{s}^{2}+\varphi_{2} m_{s}^{4}+\varphi_{3} m_{s}^{6}+\ldots, \tag{10}
\end{equation*}
$$

where $\varphi_{i}=\varphi_{i}(T, H)$. But as $H, T$ and $m_{s}$ are the independent variables of the problem, we can write

$$
\begin{equation*}
\varphi_{0}=H \quad, \quad \varphi_{1}=\varphi_{2}=\varphi_{3}=\ldots=0 \tag{11}
\end{equation*}
$$

Then, we have the desired equations for $\varphi_{i}(T, H)$ that define the coefficients $\alpha_{i}$ in equation (9). Finally, using the equations $(4,8)$ we obtain the proper coefficients of the expansion. In the Appendix we give explicit expressions for these coefficients, in terms of the model parameters, on the critical plane, up to twelfth order. In Kincaid and Cohen's work [1], coefficients were included up to eighth order, but left as implicit functions of the magnetization expansion coefficients.

## 3 Phase diagram

The symmetry is broken when the sign of the coefficient of second order in the Landau expansion, equation (7), changes from a positive to a negative value, while all the
other coefficients remain positive. Therefore, from equation (26) of the Appendix, $\Psi_{2}=0$ gives

$$
\begin{equation*}
\alpha_{0 c}=\sqrt{1-t_{c}}, \tag{12}
\end{equation*}
$$

where $t_{c}=k_{B} T_{c} / J_{+}$is the reduced critical temperature and $\alpha_{0 c}=\alpha_{0}\left(T_{c}\right)$. If the last equation is introduced in the first relation of equation (11), one defines the critical line (see Eq. (25) of the Appendix):

$$
\begin{equation*}
h_{c}=-\frac{\epsilon-1}{\epsilon+1} \sqrt{1-t_{c}}+\frac{t_{c}}{2} \ln \frac{1+\sqrt{1-t_{c}}}{1-\sqrt{1-t_{c}}} \tag{13}
\end{equation*}
$$

where we have defined the reduced field $h_{c}=H_{c} / J_{+}$ and the interaction ratio $\epsilon=J_{F} / J_{A}$. The Néel temperature is defined for $h_{c}=0$. Then, in this case $t_{N}=1$, or $k_{B} T_{N}=J_{+}$.

It must be stressed that usually a point $\left(h_{c}, t_{c}\right)$ defined by equation (13) is a true critical point, when all the other higher order coefficients $\Psi_{i}\left(t_{c}, h_{c}\right), i \geq 4$ are positive. Therefore, it is interesting to investigate the sign of these coefficients. For instance, the fourth order coefficient $\Psi_{4}$ is

$$
\begin{equation*}
\Psi_{4}=\frac{J_{+}}{4 t_{c}^{2}}\left(\epsilon t_{c}-\epsilon+\frac{1}{3}\right) \tag{14}
\end{equation*}
$$

When $\Psi_{2}=0$ and $\Psi_{4}=0$, a tricritical point takes place if the higher order coefficients $\Psi_{i}, i \geq 6$, are all positive. If this condition is satisfied, we can write

$$
\begin{equation*}
\Psi_{4}=0 \quad \Rightarrow \quad t_{T}=1-\frac{1}{3 \epsilon} \tag{15}
\end{equation*}
$$

where $t_{T}$ is the temperature of the tricritical point. Inserting this temperature in equation (13) we find the corresponding tricritical field. Thus, for $t_{c}>t_{T}, \Psi_{4}>0$, and we have a continuous transition line. On the other hand, if $t_{c}<t_{T}, \Psi_{4}<0, \Psi_{6}>0$, a first order transition appears.

Let us now consider the next term in the expansion, $\Psi_{6}$, (see Eq. (35) in the Appendix). If the condition $\Psi_{2}=0$ is taken into account, we have

$$
\begin{array}{r}
\Psi_{6}=\frac{J_{+}}{2 t_{c}^{4}}\left[\left(-\frac{\epsilon^{3}}{12}+\frac{\epsilon^{2}}{2}+\frac{\epsilon}{4}\right) t_{c}^{2}+\left(\frac{\epsilon^{3}}{6}-\frac{5 \epsilon^{2}}{4}+\frac{1}{12}\right) t_{c}\right. \\
\left.-\frac{\epsilon^{3}}{12}+\frac{3 \epsilon^{2}}{4}-\frac{\epsilon}{4}-\frac{1}{60}\right] . \tag{16}
\end{array}
$$

Substituting in this equation $t_{c}$ by $t_{T}$ we obtain

$$
\begin{equation*}
\left[\Psi_{6}\right]_{t_{T}}=\frac{J_{+}}{\left(1-\frac{1}{3 \epsilon}\right)^{4}}\left(\frac{\epsilon}{27}-\frac{1}{45}\right) \tag{17}
\end{equation*}
$$

Therefore, if $\Psi_{2}=\Psi_{4}=0,\left[\Psi_{6}\right]_{t_{T}}=0$ for $\epsilon=3 / 5$. Thus, at $\epsilon=\frac{3}{5}$, we have the appearance of a higher order critical point [1], which separates two different types of critical behavior: for $\epsilon>\frac{3}{5}$, the phase diagram exhibits a continuous transition line and a first order transition line, which meet at a tricritical point. On the other hand, for $\epsilon<\frac{3}{5}$,


Fig. 1. Phase diagram in the plane $h$ versus $t$ for $\epsilon=0.8$. Full line, continuous phase transitions and dotted line, first order phase transitions. $A F$ and $P$ are the antiferromagnetic and paramagnetic phases, respectively. $A$ is a tricritical point.
the tricritical point is decomposed into a critical and a bicritical endpoints.

Let us now compare results from the numerical solutions of the full equations and predictions from the coefficients of the Landau expansion. Figures 1 and 2 show phase diagrams for $\epsilon=0.80$ and $\epsilon=0.50$, respectively, in which the coexistence lines were obtained numerically, by finding the minima of the free energy for fixed values of $\epsilon, t$ and $h$. For instance, from equations (4,5), and taking $H_{s}=0$, we can write the equations for the sublattice magnetizations:

$$
\begin{align*}
& m_{A}=(\epsilon+1)\left(h+\frac{\epsilon}{\epsilon+1} m_{B}-\frac{t}{2} \ln \frac{1+m_{B}}{1-m_{B}}\right)  \tag{18}\\
& m_{B}=(\epsilon+1)\left(h+\frac{\epsilon}{\epsilon+1} m_{A}-\frac{t}{2} \ln \frac{1+m_{A}}{1-m_{A}}\right) \tag{19}
\end{align*}
$$

For fixed values of $\epsilon, t$ and $h$, we determine the simultaneous solutions for the system of equations $(18,19)$. Then, these solutions are inserted in equation (6) in order to find the corresponding values of the free energy. We choose the solution that gives the lowest value for the free energy. In Figure 3, for comparison, we exhibit a plot of some Landau coefficients in the plane $\epsilon$ versus $t$. For example, the line corresponding to $\Psi_{4}=0$ divides the plane into regions when $\Psi_{4}>0$ and $\Psi_{4}<0$. Besides, along all these curves we have also put $\Psi_{2}=0$ so that the $\Psi_{4}=0$ describes the tricritical line on that plane. Also, the crossing point of the curves $\Psi_{4}=0$ and $\Psi_{6}=0$, with $\Psi_{8}>0$, gives the location of the multicritical point. For this point we have $\epsilon^{*}=0.6, t^{*}=0.4444$ and $h^{*}=0.6141$.

Figure 1 represents the phase diagram with a tricritical point. At higher temperatures, in the ordered phase, if we start to increase the field at fixed temperature, the staggered magnetization $m_{s}$ decreases continuously to zero into a paramagnetic phase, at the critical line. However, increasing the field at low temperatures, we find a value of the field for which a state with $m_{s}=0$ and $m_{s} \neq 0$ are


Fig. 2. The same legend as in Figure 1, but $\epsilon=0.50$. The inset shows the first order transiton lines $\Gamma$ and $\Lambda . C$ is a critical endpoint and $B$ is the bicritical endpoint.


Fig. 3. Behavior of some Landau coefficients as a function of $t$ and $\epsilon$. Full line, $\Psi_{4}=0$; dashed line, $\Psi_{6}=0$; dotted line, $\Psi_{8}=0$. At these lines we also have $\Psi_{2}=0$. We display regions separating positive from negative values. $X$ is a point for which $\epsilon=0.62$.
simultaneous minima of the free energy. In this case we find an ordinary first order transition point, where a paramagnetic state coexists with an antiferromagnetic state. This is represented in Figure 1 by the dotted line, and corresponds to $\Psi_{2}>0, \Psi_{4}<0$ and $\Psi_{6}>0$, as can be seen in Figure 3.

In Figure 2 we observe two different first order transition lines. On the $\Gamma$ line, paramagnetic and antiferromagnetic phases coexist, whereas on the $\Lambda$-line two ordered antiferromagnetic ordered phases coexist. In terms of the expansion coefficients (see Fig. 3), the critical line meets the $\Lambda$-line, at the critical endpoint C , somewhere inside the $\Psi_{6}<0$ region (with $\Psi_{2}=0, \Psi_{4}>0$ ), as seen in Figure 3. Continuations of the $\Gamma$-line $\left(\Psi_{2}>0, \Psi_{4}>0, \Psi_{6}<0\right.$, $\left.\Psi_{8}>0\right)$ and of the $\Lambda$ line $\left(\Psi_{2}<0, \Psi_{4}>0, \Psi_{6}<0\right.$ and $\Psi_{8}>0$ ), lie outside the $\epsilon-t$ critical plane exhibited in Figure 3.

Because the more complex behaviour occurs in a small region of the phase diagram, a good check consists in


Fig. 4. The same legend as in Figure 1, but $\epsilon=0.62$. Point 1 is a tricritical point and point 2 is on the critical line, very near the tricritical point. Just below point 2, in $A F$ phase, we have $\Psi_{2}<0, \Psi_{4}>0, \Psi_{6}<0$ and $\Psi_{8}>0$.
analysing the slope of the phase boundary at the tricritical point, given by

$$
\frac{\mathrm{d} h_{T}}{\mathrm{~d} t_{T}}=\frac{-1}{(\epsilon+1) \sqrt{1-t_{T}}}+\frac{1}{2} \ln \frac{1+\sqrt{1-t_{T}}}{1-\sqrt{1-t_{T}}}
$$

for $t>t_{T}$, which should agree with that found numerically at the first order transition line, for $t<t_{T}[13,14]$.

Although the topology of the phase diagram should be the same as that exhibited in Figure 1, where $\epsilon=0.8$, for any $\epsilon>\epsilon^{*}$, we must be careful in predicting the critical behavior based only on the signs of the Landau coefficients. In Figure 4, we plot the phase diagram for the value $\epsilon=0.62$. The point 1 in this figure is really a tricritical point: the right and left slopes of the phase boundary are the same at this point. However, let us consider the point 2 on the critical line, which is very near the tricritical point, indicated also in Figure 3 by the letter X. It can be seen that it lies in the region $\Psi_{4}>0, \Psi_{6}<0$ and $\Psi_{8}>0$ which could indicate the presence of a bicritical endpoint.

Indeed, this analysis can be applied to any value of $\epsilon$ in the range $0.6<\epsilon<0.631$. The value $\epsilon=0.631$ is the largest that can be obtained satisfying the condition $\Psi_{6}<0$. Based only on the signs of the Landau expansion, we would expect to find in the neighborhood of $X$ a line of first order transition points, as the line $\Lambda$ in Figure 2. However, a careful examination of the minima of the free energy in this region did not support the presence of a bicritical endpoint.

The original theory of Landau of phase transitions was devised to explain continuous phase transitions to the fourth order in the order parameter, but can be extended to take also account of the first order transitions for very small values of the order parameter. For the ordinary Ising model, the Landau expansion around its critical point, renders all coefficients higher than second order positive, and the critical point is well defined without ambiguity. However, when the coefficients exhibit different signs, we must be careful in disregarding those we believe are
irrelevant [15]. For instance, when we considered Figure 1, for $\epsilon=0.8$, the tricritical point appeared, and we had $\Psi_{2}=\Psi_{4}=0$ and $\Psi_{6}>0$. However, as we can see in Figure $3, \Psi_{8}<0$. Indeed, in Figure 3 the $\Psi_{8}>0$ condition occupies a very small region of the plane $\epsilon$ versus $t$. The same happens with the coefficients $\Psi_{10}$ and $\Psi_{12}$, which also exhibit regions of positive and negative values. We can conclude that, in the case of the tricritical point it is sufficient to consider terms up to six order, whereas for the case of the bicritical endpoint, terms of eighth order must be taken into account. The inclusion of the coefficients $\Psi_{10}$ and $\Psi_{12}$ will have the only effect to produce new local minima in the free energy, but for large values of the order parameter $m_{s}$. However, as we have seen in the discussion of Figure 4, in some cases a full account of the free energy is better than solely trust in the signs of the corresponding Landau expansion.

As a final comment concerning the phase diagram of the metamagnetic model, we call attention to the curious maximum observed in the phase boundary, which is predicted by the mean field theory. For $\epsilon<0.6959$ the continuous transition line has a maximum, that is, $\mathrm{d} h_{c} / \mathrm{d} t_{c}=0$ (see Fig. 2). Thus, it is possible for a fixed value of the magnetic field, to cross the phase boundary from a disordered paramagnetic phase to an ordered antiferromagnetic phase by just increasing the temperature. $\mathrm{FeBr}_{2}$ metamagnet $[16,17]$ exhibits this type of phase diagram. Besides this behavior, the $\mathrm{FeBr}_{2}$ metamagnet also displays other interesting properties in its phase diagram as can be seen in the recent neutron scattering studies $[18,19]$.

## 4 Conclusion

We investigated the critical behavior of an Ising metamagnetic model in the mean field approximation. We expanded the free energy of the model in powers of its order parameter, and determined the Landau coefficients of the model up to twelfth order. We plotted these coefficients in the plane $t$ (reduced temperature) versus $\epsilon$ (ratio of exchange couplings), and based on the signs of these coefficients, we constructed the phase diagrams for different values of $\epsilon$. We showed that, for some range of values of the parameter $\epsilon$, a crude analysis of the signs of the coefficients would not be sufficient to foresee the nature of the critical point. In such cases, we need to do a careful investigation of the full free energy in order to find its minima in the selected region.

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## Appendix: The coefficients of the Landau expansion

Let's consider a general logarithmic term of equations (4, 5). Introducing the expansion for $m$, equation (9),
we can write

$$
\begin{gather*}
\ln \left(1+a m_{s}+b m\right)=\ln \left(1+b \alpha_{0}\right)+A_{0} m_{s}+\left(-\frac{A_{0}^{2}}{2}+A_{1}\right) m_{s}^{2} \\
+\left(\frac{A_{0}^{3}}{6}-A_{0} A_{1}\right) m_{s}^{3}+\left(A_{2}-\frac{A_{1}^{2}}{2}-\frac{A_{0}^{4}}{4}+A_{0}^{2} A_{1}\right) m_{s}^{4} \\
+\left(\frac{A_{0}^{5}}{5}+A_{0} A_{1}^{2}-A_{0}^{3} A_{1}+A_{0} A_{2}\right) m_{s}^{5} \\
+\left(A_{3}-A_{1} A_{2}+\frac{A_{1}^{3}}{3}+A_{0}^{2} A_{2}-\frac{3 A_{0}^{2} A_{1}}{2}+A_{0}^{4} A_{1}-\frac{A_{0}^{6}}{6}\right) m_{s}^{6} \\
\quad+\left(-A_{0} A_{3}+2 A_{0} A_{1} A_{2}-A_{0}^{3} A_{2}\right. \\
\left.\quad+A_{0} A_{1}^{3}+2 A_{0}^{3} A_{1}^{2}-A_{0}^{5} A_{1}+\frac{A_{0}^{7}}{7}\right) m_{s}^{7} \\
+C_{8} m_{s}^{8}+C_{9} m_{s}^{9}+C_{10} m_{s}^{10}+C_{11} m_{s}^{11}+\ldots, \tag{20}
\end{gather*}
$$

where

$$
\begin{align*}
C_{8}= & A_{4}-A_{1} A_{3}-\frac{A_{2}^{2}}{2}+A_{0}^{2} A_{3}+A_{1}^{2} A_{2}-3 A_{0}^{2} A_{1} A_{2} \\
& +\frac{A_{1}^{4}}{4}+A_{0}^{4} A_{2}+2 A_{0}^{2} A_{1}^{3}-\frac{5 A_{0}^{4} A_{1}^{2}}{2}+A_{0}^{6} A_{1}-\frac{A_{0}^{8}}{8},  \tag{21}\\
C_{9}= & \frac{A_{0}^{9}}{9}-A_{0}^{7} A_{1}+3 A_{0}^{5} A_{1}^{2}-\frac{10 A_{0}^{3} A_{1}^{2}}{3} \\
& +A_{0} A_{1}^{4}-A_{0}^{5} A_{2}+4 A_{0}^{3} A_{1} A_{2}-3 A_{0} A_{1}^{2} A_{2} \\
& +A_{0} A_{2}^{2}-A_{0}^{3} A_{3}+2 A_{0} A_{1} A_{3}-A_{0} A_{4},  \tag{22}\\
C_{10}= & -\frac{A_{0}^{10}}{10}+A_{0}^{8} A_{1}-\frac{7 A_{0}^{6} A_{1}^{2}}{2}+5 A_{0}^{4} A_{1}^{3}-\frac{5 A_{0}^{2} A_{1}^{4}}{2} \\
& +\frac{A_{1}^{5}}{5}+A_{0}^{6} A_{2}-5 A_{0}^{4} A_{1} A_{2}+6 A_{0}^{2} A_{1}^{2} A_{2}-A_{1}^{3} A_{2} \\
& -\frac{3 A_{0}^{2} A_{2}^{2}}{2}+A_{1} A_{2}^{2}+A_{0}^{4} A_{3}-3 A_{0}^{2} A_{1} A_{3}+A_{1}^{2} A_{3} \\
& -A_{2} A_{3}+A_{0}^{2} A_{4}-A_{1} A_{4}+A_{5}, \tag{23}
\end{align*}
$$

$$
\begin{align*}
C_{11}= & \frac{A_{0}^{11}}{11}-A_{0}^{9} A_{1}+4 A_{0}^{7} A_{1}^{2}-7 A_{0}^{5} A_{1}^{3}+5 A_{0}^{3} A_{1}^{4} \\
& -A_{0} A_{1}^{5}-A_{0}^{7} A_{2}+6 A_{0}^{5} A_{1} A_{2}-10 A_{0}^{3} A_{1}^{2} A_{2} \\
& +4 A_{0} A_{1}^{3} A_{2}+2 A_{0}^{3} A_{2}^{2}-3 A_{0} A_{1} A_{2}^{2}-A_{0}^{5} A_{3} \\
& +4 A_{0}^{3} A_{1} A_{3}-3 A_{0} A_{1}^{2} A_{3}+2 A_{0} A_{2} A_{3} \\
& -A_{0}^{3} A_{4}+2 A_{0} A_{1} A_{4}-A_{0} A_{5} \tag{24}
\end{align*}
$$

In these equations we have defined that $a= \pm 1, b= \pm 1$, and

$$
A_{0}=\frac{a}{1+b \alpha_{0}} ; \quad A_{i}=\frac{b \alpha_{i}}{1+b \alpha_{0}}
$$

$$
\begin{align*}
\alpha_{5 c}= & \frac{-(\epsilon+1) \sqrt{1-t}}{3840 t^{9}}\left[\left(105 \epsilon^{8}-1050 \epsilon^{7}+4080 \epsilon^{6}-6150 \epsilon^{5}-3570 \epsilon^{4}+16890 \epsilon^{3}\right.\right. \\
& \left.+13680 \epsilon^{2}+2790 \epsilon+105\right) t^{4}+\left(-420 \epsilon^{8}+4830 \epsilon^{7}-22650 \epsilon^{6}+48310 \epsilon^{5}-15690 \epsilon^{4}\right. \\
& \left.-94150 \epsilon^{3}-31150 \epsilon^{2}+2610 \epsilon+790\right) t^{3}+\left(630 \epsilon^{8}-8190 \epsilon^{7}+44680 \epsilon^{6}-119482 \epsilon^{5}\right. \\
& \left.+104720 \epsilon^{4}+156270 \epsilon^{3}-4880 \epsilon^{2}-12470 \epsilon+2\right) t^{2}+\left(-420 \epsilon^{8}+6090 \epsilon^{7}-37730 \epsilon^{6}\right. \\
& \left.+119434 \epsilon^{5}-154330 \epsilon^{4}+-85490 \epsilon^{3}+41450 \epsilon^{2}+4510 \epsilon-1034\right) t+105 \epsilon^{8} \\
& \left.-1680 \epsilon^{7}-11620 \epsilon^{6}-42112 \epsilon^{5}+68990 \epsilon^{4}+6000 \epsilon^{3}-18380 \epsilon^{2}+2080 \epsilon+257\right] . \tag{33}
\end{align*}
$$

Now, we collect all the terms in the expansions, equations $(4,5)$, and we use the conditions established in equation (11). The first relation, $\varphi_{0}=H$ gives

$$
\begin{equation*}
h=-\frac{\epsilon-1}{\epsilon+1} \alpha_{0}+\frac{t}{2} \ln \frac{1+\alpha_{0}}{1-\alpha_{0}} \tag{25}
\end{equation*}
$$

where $h=H / J_{+}, t=k_{B} T / J_{+}$and $\epsilon=J_{F} / J_{A}$. In this way we obtain the expression for the coefficient of second order

$$
\begin{equation*}
\Psi_{2}=-J_{+} / 2+\frac{1}{4 \beta}\left(\frac{1}{1+\alpha_{0}}+\frac{1}{1-\alpha_{0}}\right) . \tag{26}
\end{equation*}
$$

If we put $\Psi_{2}=0$, the coefficient $\alpha_{0 c}$ at the criticality is given by

$$
\begin{equation*}
\alpha_{0 c}=\sqrt{1-t_{c}} \tag{27}
\end{equation*}
$$

The line of critical points $\left(h_{c}, t_{c}\right)$, is obtained by putting $\alpha_{0 c}$ in equation (25).

The other coefficients $\alpha_{i}$ are calculated in a similar manner and we use the expression for $\alpha_{0 c}$ obtained above. For instance, the coefficient $\varphi_{1}$ is given by
$\varphi_{1}=J_{-} \alpha_{1}-\frac{1}{2 \beta}$

$$
\begin{equation*}
\times\left[\frac{\alpha_{1}}{1+\alpha_{0}}-\frac{1}{2} \frac{1}{\left(1+\alpha_{0}\right)^{2}}+\frac{\alpha_{1}}{1-\alpha_{0}}+\frac{1}{2} \frac{1}{\left(1+\alpha_{0}\right)^{2}}\right] . \tag{28}
\end{equation*}
$$

Putting $\varphi_{1}=0$ we get,

$$
\begin{equation*}
\alpha_{1 c}=\frac{-(\epsilon+1) \sqrt{1-t}}{2 t} . \tag{29}
\end{equation*}
$$

The other coefficients are:

$$
\begin{align*}
\alpha_{2 c}= & \frac{-(\epsilon+1) \sqrt{1-t}}{8 t^{3}}\left[\left(-\epsilon^{2}+4 \epsilon+1\right) t+\epsilon^{2}-6 \epsilon+1\right],  \tag{30}\\
\alpha_{3 c}= & \frac{(\epsilon+1) \sqrt{1-t}}{48 t^{5}}\left[\left(-3 \epsilon^{4}+18 \epsilon^{3}-30 \epsilon^{2}-30 \epsilon-3\right) t^{2}\right. \\
& +\left(6 \epsilon^{4}-46 \epsilon^{3}+114 \epsilon^{2}+30 \epsilon-8\right) t-3 \epsilon^{4} \\
& \left.+28 \epsilon^{3}-90 \epsilon^{2}+12 \epsilon+5\right],  \tag{31}\\
\alpha_{4 c}= & \frac{(\epsilon+1) \sqrt{1-t}}{384 t^{7}}\left[\left(15 \epsilon^{6}-120 \epsilon^{5}+339 \epsilon^{4}-192 \epsilon^{3}\right.\right. \\
& \left.-723 \epsilon^{2}-264 \epsilon-15\right) t^{3}+\left(-45 \epsilon^{6}+430 \epsilon^{5}\right. \\
& \left.-1567 \epsilon^{4}+1924 \epsilon^{3}+2165 \epsilon^{2}+46 \epsilon-73\right) t^{2} \\
& +\left(45 \epsilon^{6}-500 \epsilon^{5}+2205 \epsilon^{4}-3856 \epsilon^{3}-1417 \epsilon^{2}\right. \\
& +612 \epsilon+31) t-15 \epsilon^{6}+190 \epsilon^{5}-977 \epsilon^{4}+2148 \epsilon^{3} \\
& \left.-97 \epsilon^{2}-322 \epsilon+33\right],  \tag{32}\\
& \text { See equation (33) above. }
\end{align*}
$$

Finally, with these coefficients $\alpha_{i}$, and with help of equations $(4,8)$ we can write the remaining coefficients in the Landau expansion up to order 12:

$$
\begin{equation*}
\Psi_{4}=\frac{J_{+}}{4 t^{2}}\left(\epsilon t-\epsilon+\frac{1}{3}\right) \tag{34}
\end{equation*}
$$

$$
\Psi_{6}=\frac{J_{+}}{2 t^{4}}\left[\left(-\frac{\epsilon^{3}}{12}+\frac{\epsilon^{2}}{2}+\frac{\epsilon}{4}\right) t^{2}+\left(\frac{\epsilon^{3}}{6}-\frac{5 \epsilon^{2}}{2}+\frac{1}{12}\right) t\right.
$$

$$
\begin{equation*}
\left.-\frac{\epsilon^{3}}{12}+\frac{3 \epsilon^{2}}{2}-\frac{\epsilon}{4}-\frac{1}{60}\right] \tag{35}
\end{equation*}
$$

$$
\begin{align*}
\Psi_{8}= & \frac{J_{+}}{1344 t^{6}}\left[\left(21 \epsilon^{5}-168 \epsilon^{4}+378 \epsilon^{3}+504 \epsilon^{2}+105 \epsilon\right) t^{3}+\left(-63 \epsilon^{5}+595 \epsilon^{4}\right.\right. \\
& \left.-1778 \epsilon^{3}-966 \epsilon^{2}+161 \epsilon+35\right) t^{2}+\left(63 \epsilon^{5}-686 \epsilon^{4}+2506 \epsilon^{3}+168 \epsilon^{2}-385 \epsilon\right. \\
& \left.+14) t-21 \epsilon^{5}+259 \epsilon^{4}-1106 \epsilon^{3}+294 \epsilon^{2}+119 \epsilon-25\right]  \tag{36}\\
\Psi_{10}= & \frac{J_{+}}{5760 t^{8}}\left[7+645 \epsilon-2685 \epsilon^{2}-2055 \epsilon^{3}+12405 \epsilon^{4}-4497 \epsilon^{5}\right. \\
& +705 \epsilon^{6}-45 \epsilon^{7}+\left(-177-270 \epsilon+8925 \epsilon^{2}-6240 \epsilon^{3}-34695 \epsilon^{4}\right. \\
& \left.+14682 \epsilon^{5}-2565 \epsilon^{6}+180 \epsilon^{7}\right) t+\left(129-1800 \epsilon-6105 \epsilon^{2}+23010 \epsilon^{3}\right. \\
& \left.+33615 \epsilon^{4}-17484 \epsilon^{5}+3465 \epsilon^{6}-270 \epsilon^{7}\right) t^{2}+\left(105+1110 \epsilon-2745 \epsilon^{2}\right. \\
& \left.-19800 \epsilon^{3}-12585 \epsilon^{4}+8910 \epsilon^{5}-2055 \epsilon^{6}+180 \epsilon^{7}\right) t^{3}+(315 \epsilon \\
& \left.\left.+2610 \epsilon^{2}+5085 \epsilon^{3}+1260 \epsilon^{4}-1611 \epsilon^{5}+450 \epsilon^{6}-45 \epsilon^{7}\right) t^{4}\right]  \tag{37}\\
\Psi_{12}= & \frac{J_{+}}{253440 t^{10}\left[\left(2833-14267 \epsilon-118140 \epsilon^{2}+534820 \epsilon^{3}+52030 \epsilon^{4}\right.\right.} \\
& -1563738 \epsilon^{5}+782452 \epsilon^{6}-180620 \epsilon^{7}+21945 \epsilon^{8}-1155 \epsilon^{9}+(-4224 \\
& +101871 \epsilon-17160 \epsilon^{2}-1923900 \epsilon^{3}+1759560 \epsilon^{4}+5081274 \epsilon^{5}-3009336 \epsilon^{6} \\
& \left.+768900 \epsilon^{7}-101640 \epsilon^{8}+5775 \epsilon^{9}\right) t+\left(-7986-98406 \epsilon+694320 \epsilon^{2}+1702800 \epsilon^{3}\right. \\
& -5600100 \epsilon^{4}-5964684 \epsilon^{5}+4473216 \epsilon^{6}-1288320 \epsilon^{7}+187110 \epsilon^{8} \\
& \left.-11550 \epsilon^{9}\right) t^{2}+\left(7832-61138 \epsilon-696300 \epsilon^{2}+624800 \epsilon^{3}+6174740 \epsilon^{4}\right. \\
& \left.+2876148 \epsilon^{5}-3171652 \epsilon^{6}+1057760 \epsilon^{7}-170940 \epsilon^{8}+11550 \epsilon^{9}\right) t^{3}+(3465 \\
& +61545 \epsilon+8580 \epsilon^{2}-1376100 \epsilon^{3}-2827770 \epsilon^{4}-368610 \epsilon^{5}+1048740 \epsilon^{6} \\
& \left.-423060 \epsilon^{7}+77385 \epsilon^{8}-5775 \epsilon^{9}\right) t^{4}+\left(10395 \epsilon+128700 \epsilon^{2}+437580 \epsilon^{3}\right. \\
+ & \left.\left.441540 \epsilon^{4}-60390 \epsilon^{5}-123420 \epsilon^{6}+65340 \epsilon^{7}-13860 \epsilon^{8}+1155 \epsilon^{9}\right) t^{5}\right] \tag{38}
\end{align*}
$$

See equations (36, 37, 38) above.

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