Ginzburg–Landau theory for the magnetic and structural transitions in La$_{1-y}$(C$_{1-x}$Sr$_x$)$_y$MnO$_3$

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Abstract
We present a phenomenological theory for the ferromagnetic transition temperature, the magnetic susceptibility at high temperatures, and the structural distortion in the La$_{1-y}$(C$_{1-x}$Sr$_x$)$_y$MnO$_3$ system. We construct a Ginzburg–Landau free energy that describes the magnetic and the structural transitions, and a competition between them. The parameters of the magnetic part of the free energy are derived from a mean-field solution of the magnetic interaction for arbitrary angular momentum. The theory provides a qualitative description of the observed magnetic and structural phase transitions as functions of Sr-doping level (x) for y = 0.25.

Keywords: magnetism, Ginzburg–Landau free energy, phase transitions

(Some figures may appear in colour only in the online journal)

1. Introduction

The compounds with colossal magnetoresistance (CMR) manganites, such as La$_{1-x}$Ca$_x$MnO$_3$ and La$_{1-x}$Sr$_x$MnO$_3$, have attracted a great deal interest in the last decades due to their rich fundamental physics and great potential to applications in magnetic memories and spintronics [1–5]. These compounds are strongly correlated electron systems with strong interplays among the charge, spin, orbital, and lattice degrees of freedom. In particular superexchange interactions combined with Hund rules that favor ferromagnetic order compete with Jahn–Teller-type electron-lattice distortions, leading to complex electronic, magnetic, and structural phase diagrams [6–11].

The CMR is a property of some materials, mostly manganese-based perovskite oxides, that enables them to dramatically change their electrical resistance in the presence of a magnetic field [12, 13]. Switching on a magnetic field helps to align neighboring spins, particularly near the the magnetic transition, where it is easier to line up the spins, and in this case the resistance is lower, because hopping from Mn$^{4+}$ to neighboring Mn$^{3+}$ is facilitated. The interaction responsible for the magnetic order is the double-exchange [14]. Specifically, in a nearly cubic environment, the Mn$^{4+}$ ions have each $t_{2g}$ orbital occupied by one electron forming a spin 3/2 (due to Hund rules) and the $e_g$ orbitals empty. The Mn$^{3+}$ ions have an additional electron in the more mobile $e_g$ orbitals forming total spin 2. This additional electron can move freely between parallel spin 3/2, while if the $t_{2g}$ spins are disordered, the $e_g$ electrons suffer a scattering mechanism due to partial frustration of the Hund rules. The double exchange also plays an important role in related perovskites, such as RuSr$_2$(Eu,Gd)Cu$_2$O$_y$ [15, 16].

In some CMR compounds, like (La,Ca)MnO$_3$ a coexistence of magnetic phases has been found, which is absent in others, like (La,Sr)MnO$_3$. This is likely to be related by the fact that the Ca compound presents a first order magnetic phase transition, while the transition is of second order in the Sr compound [17, 18]. In the series La$_{0.67}$(Ca$_{1-x}$Sr$_x$)$_{0.33}$MnO$_3$ a relationship between first or second order magnetic phase transitions...
transition and the crystalline structure has been found \[19\]. The crystalline structure depends on the Sr content. At room temperature, in absence of Sr \((x = 0)\), it is orthorhombic, while in absence of Ca \((x = 1)\) is rhombohedral. However, the origin of the first order magnetic transition is still unclear.

In a more recent work, the phase transitions in the \(\text{La}_{1-x}\text{(Ca}_{1-x}\text{Sr}_{x})\text{MnO}_{3}\) were experimentally studied for \(0.23 \leq x \leq 0.45\) \[20\]. In particular a phase diagram as a function of Sr content \(x\) for \(y = 0.25\) was constructed (figure 8 of \[20\], corresponding to figure 1 in this work). The replacement of Ca\(^{2+}\) ions with larger Sr\(^{2+}\) ones reduces the critical temperature of the structural transition from a high temperature rhombohedral phase to a low temperature orthorhombic phase. In addition, there is a magnetic transition from a high-temperature paramagnetic insulating phase to a low-temperature ferromagnetic metallic phase. The structural and magnetic transitions cross as a function of Sr content, in such a way that for low (high) \(x\) the magnetic transition temperature is lower (higher) and for \(x \approx 1\) the structural transition disappears. In addition, for low \(x\), the magnetic susceptibility \(\chi\) has a kink at the structural transition and \(\chi^{-1}\) changes slope (figure 4 of \[20\], corresponding to figure 2 in this work).

Motivated by these experimental results, in the present work we propose a simple Ginzburg–Landau free energy to describe both, the magnetic and structural transitions and also the kink in the magnetic susceptibility. Ginzburg–Landau functionals have been useful in different contexts. For example recently one of these functionals were used to show induced spin-triplet pairing at the interface between an antiferromagnetic state and a singlet superconductor \[21\]. Our results provide a semiquantitative explanation of the phase diagram and the magnetic susceptibility in the paramagnetic phase.

The paper is organized as follows. The free-energy functional is described in section 2. In section 3 we discuss the parameters of the free energy by comparison with experiment. In particular we discuss the form of the magnetic part of the free energy for arbitrary angular momentum. Section 4 contains the resulting phase diagram. In section 5 we show the dependence of the magnetic susceptibility with temperature for some values of \(x\), as in the experimental work \[20\]. Section 6 contains a summary.

### 2. Free energy

In order to describe a magnetic and structural transitions, we propose a Ginzburg–Landau functional in terms of two dimensionless order parameters: \(m = M/M_0\), where \(M\) is the magnetization and \(M_0\) the saturation magnetization and \(\delta\), proportional to the structural deformation (it is defined more precisely in section 3.3). As usual, we expand the free energy in terms of the two order parameters around \(m = 0\) and \(\delta = 0\) and consider the lowest order terms allowed by symmetry. The free energy can be written as

\[
F = -mh + A_m(T - T_m)m^2 + B_m m^4 + A_\delta(T - T_\delta)\delta^2 + B_\delta \delta^3 + C m^2 \delta^2,
\]

where the first three terms, with \(A_m > 0\) and \(B_m > 0\) determined below, correspond to the standard magnetic free energy in presence of a magnetic field \(h\). The fourth and fifth term, with \(A_\delta > 0\) and \(B_\delta > 0\), correspond to the structural transition. Finally, the last term, corresponds to the coupling between magnetic and structural degrees of freedom. As we shall show, the observed phase diagram indicates that \(C > 0\). This means that the magnetization inhibits the structural distortion and vice versa. Note that \(T_m(T_\delta)\) is the critical temperature of the magnetic (structural) transition in absence of distortion (magnetization).

In our functional we neglect gradient terms and intergradient interactions. These are naturally important in the presence of inhomogeneities due to compositional changes that may be present in the system \[22\]. However, as we show, the main aspects of the observed phase diagram can be explained without including these terms.

### 3. Determination of the coefficients of the Ginzburg–Landau free energy

#### 3.1. Coefficients of the purely magnetic part

In the mean-field approximation for a Heisenberg model with ferromagnetic nearest-neighbor interaction \(I\), the magnetic free energy can be written as

\[
F_m = -\frac{I m^2}{2(\gamma \mu_B)^2} - T S,
\]

where \(z\) is the number of nearest neighbors, \(S\) is the entropy, and \(T\) is the temperature. For models that include more interactions, \(I\) should be replaced by an appropriate sum including further neighbors. In any case, the value of this term can be obtained from the observed magnetic critical temperature and the details of the model do not affect our treatment.

Now we discuss the form of the entropy for a magnetic system composed of ions with angular momentum \(j\). Later we discuss the specific case of our system.

To obtain the entropy as a function of \(m\) for a given \(j\), we proceed as follows. In mean field, the magnetization is given by the Brillouin function \(B_h(T)\), where \(h = \gamma \mu_B h_{\text{eff}}\) is proportional to effective magnetic field \(h_{\text{eff}}\) acting on a particular site, which includes the external field \(h\) and the effect of the ordered magnetic moments of its neighbors proportional to \(m\). In addition, also the entropy in the mean-field approximation is given by the single-site entropy as a function of \(\tilde{h}/T\). The idea is to invert \(m\) as a function of \(h\) to obtain \(S\) as a function of \(m\) up to fourth order in \(m\).

Specifically expanding the corresponding functions in a Taylor series around of \(\tilde{h} = 0\) one has

\[
m \approx \alpha \tilde{h} + \beta \tilde{h}^3 \quad (3)
\]

\[
S \approx S_0 + \gamma \tilde{h}^2 + \zeta \tilde{h}^4, \quad (4)
\]

where the coefficients \(\alpha, \beta, \gamma\) and \(\zeta\) are given below. Inverting equation (3) one obtains \(\tilde{h}\) as a function of the magnetization.
\[ \tilde{h} \approx \eta m + \nu m^3. \]  

Replacing \( m \) from equation (3) in the equation (5) we obtain \( \eta = 1/\alpha \) and \( \nu = -\beta/\alpha^2 \).

The desired expansion of \( S \) as a function of the magnetization

\[ S \approx S_0 + \epsilon m^2 + \xi m^4, \]  

is obtained replacing \( \tilde{h} \) from equation (5) in equation (4) and matching with equation (6). We obtain \( \epsilon = \gamma/\alpha^3 \) and \( \xi = -2\beta/\alpha^4 + \zeta/\alpha^5 \). Thus \( S \) in terms of the magnetization becomes

\[ S = \frac{\gamma}{\alpha^3} m^2 + \left( -\frac{2\beta}{\alpha^4} + \frac{\zeta}{\alpha^5} \right) m^4. \]  

To calculate the values of the coefficients \( \alpha, \beta, \gamma, \) and \( \zeta \), we use well known expressions for the statistical theory of magnetism. The magnetization is expressed in terms of the Brillouin function \( B_j \)

\[ m = \frac{M}{M_0} = B_j(\tilde{h}/T) = \frac{(2j + 1)}{2j} \coth \left( j \frac{1}{2} \frac{\tilde{h}}{T} \right) - \frac{1}{2j} \coth \left( \frac{\tilde{h}}{2T} \right). \]  

Expanding the Brillouin function around \( \tilde{h} = 0 \), and truncating it to third order, the resulting magnetization reads

\[ m = \left( \frac{1 + j}{3T} \right) \tilde{h} - \frac{1}{90T^3} \left( 1 + 3j + 4j^2 + 2j^3 \right) \tilde{h}^3 \approx \alpha \tilde{h} + \beta \tilde{h}^3. \]  

This result allows us to obtain the values of \( \alpha \) and \( \beta \):

\[ \alpha = \frac{(1 + j)}{3T}, \]  

\[ \beta = -\frac{(1 + 3j + 4j^2 + 2j^3)}{90T^3}. \]

It remains to calculate \( \gamma \) and \( \zeta \). From the definition of the Helmholtz free energy (we use units with the Boltzmann constant \( k_B = 1 \))

\[ F = -T \ln(Z). \]

The entropy is given by

\[ S = -\frac{\partial F}{\partial T} = \ln(Z) + T \frac{1}{Z} \frac{\partial Z}{\partial T} \]  

which is expressed in terms of the partition function \( Z \)

\[ Z = \sum_{m=-j}^{j} e^{\frac{m\tilde{h}}{T}}. \]

Expanding \( S \) around \( \tilde{h} = 0 \), and truncating it to fourth order, the resulting entropy can be written as

\[ S = \ln(2j + 1) - \frac{j(1 + j)}{6T^2} \tilde{h}^2 + \frac{j(1 + 3j + 4j^2 + 2j^3)}{120T^4} \tilde{h}^4 \approx \gamma \tilde{h}^2 + \zeta \tilde{h}^4. \]  

Thus we obtain the values of the numbers \( \gamma \) and \( \zeta \)

\[ \gamma = -\frac{j(1 + j)}{6T^2}, \]  

\[ \zeta = \frac{j(1 + 3j + 4j^2 + 2j^3)}{120T^4}. \]

\( S_0 = \ln(2j + 1) \) is independent of \( m \) and will be dropped in what follows. Replacing \( \alpha, \beta, \gamma, \) and \( \zeta \) in the equation (7) and then in the equation (2), we obtain finally a expression to the magnetic free energy

\[ F_m = \frac{3}{2} \left( \frac{j}{1 + j} \right) (T - T_m) m^2 + \frac{9}{40} \frac{j(1 + 2j)^2}{(1 + j)^2} T_m^4, \]

where \( T_m = I j(j + 1)/3 \), is the usual mean-field expression for the magnetic transition temperature. This last result is a general expression for arbitrary \( j \). Note that for \( j = 1/2 \) the result is in agreement with previous results [23, 24]. The linear term in \( T - T_m \) is also present in the theory of [25]. From equation (18), the parameters \( A_m \) and \( B_m \) of the magnetic part of the free energy equation (1) are determined to be

\[ A_m = \frac{3}{2} \frac{j}{(1 + j)} \]  

\[ B_m(T) = \frac{9}{40} \frac{j(1 + 2j)^2}{(1 + j)^2} T = B'_m T. \]

For our system with \( y = 0.25 \), there is a mixture of 75% of Mn\(^{1+} \) and 25% of Mn\(^{4+} \) with total spin 2 and 3/2 respectively. Since the angular momentum is quenched in the system, we take the values of \( A_m \) and \( B_m \) from the corresponding average (0.75 the values for \( j = 2 \) plus 0.25 the values for \( j = 3/2 \)). Therefore we obtain

\[ A_m = 0.975 \]  

\[ B'_m = 0.208. \]

### 3.2. Transition temperatures

As discussed in the introduction, the temperatures for both transitions depend of the Sr content. The same happens with the ‘bare’ transition temperatures \( T_{m} \) and \( T_s \), which correspond to a system with no coupling between magnetic and structural degrees of freedom \((C = 0)\). For small \( x \) one has \( T_s > T_m \). In this case, for \( h = 0 \), using equation (1), as the temperature is lowered, the first transition found is the structural one at the bare critical temperature \( T_s \). At lower temperatures, the order parameter \( \delta \) of the structural distortion is different from zero. For intermediate temperatures \( T'_m < T < T_s \), where \( T'_m < T_m \) is the real magnetic transition temperature, the value of \( \delta \) can be obtained minimizing the free energy of equation (1) for \( m = h = 0 \) with respect to \( \delta^2 \). This leads to

\[ \frac{\partial F}{\partial \delta^2} \bigg|_{m=0} = \frac{A_s(T - T_s) + 2B_s \delta^2}{2B_s} = 0 \rightarrow \delta^2 = \frac{A_s(T_s - T)}{2B_s}. \]

For temperatures greater than or equal to the magnetic transition temperature \((T > T'_m)\), we can replace the above obtained value of \( \delta \) (valid only for \( m = 0 \)) in the free energy. The resulting expression depends only on \( m \) and \( T_m \) can be obtained as usual from the requirement that the coefficient of \( m^2 \) vanishes. For \( m \to 0 \)

\[ \frac{\partial F}{\partial m^2} = A_m(T'_m - T_m) + C \frac{A_s(T - T'_m)}{2B_s} = 0. \]
From this condition, the magnetic transition temperature is

$$T'_m = \frac{T_m - \frac{A_m C}{2A_m B_m} T_\delta}{1 - \frac{A_m C}{2A_m B_m}}. \quad (25)$$

One can proceed in a similar way for the case in which $T_\delta < T_m$. In this case, the magnetic transition temperature takes place at the ‘bare’ transition temperature $T_m$ and the structural transition temperature is $T'_\delta < T_\delta$. The analysis is the same as before with corresponding parameters for structural and magnetic transitions interchanged. The result is

$$T'_\delta = \frac{T_\delta - \frac{A_m C}{2A_m B_m} T_m}{1 - \frac{A_m C}{2A_m B_m}}. \quad (26)$$

In this case however, since $B_m = B'_m T'_\delta$ (see equation (20)), $T'_\delta$ enters also the second member of equation (26) leading to a quadratic equation in $T'_\delta$. This is discussed below.

### 3.3. Structural coefficients of the free energy

Here we determine some relations between the different parameters of the free energy that come from experiment.

From the fit of the experimental data for the highest transition temperature as a function of the Sr concentration $x$ presented in the [20] we obtain

$$T_\delta = (712.06 - 668.60 x) \text{ K} \quad (27)$$

$$T_m = (284.15 + 59.86 x) \text{ K.} \quad (28)$$

The magnetic transition temperature given by equation (25) can be written in the form

$$T'_m = \frac{T_m - YT_\delta}{1 - Y} \quad \rightarrow Y = \frac{T'_m - T_m}{T_m - T_\delta} \quad (29)$$

with

$$Y = \frac{A_m C}{2A_m B_m}. \quad (30)$$

From the experimental data of the figure 8 in [20] for $x = 0 \rightarrow T'_m \approx 230 \text{ K}$. Using this value and equations (27)–(29) we obtain

$$Y = 0.112. \quad (31)$$

In a similar way the structural transition temperature given by equation (26) can be written as

$$T'_\delta = \frac{T_\delta - YT_m}{1 - Y'} \quad \rightarrow Y' \equiv \frac{Z}{T'_\delta} = \frac{T'_\delta - T_\delta}{T'_m - T_m} \quad (32)$$

with

$$Z = \frac{A_m C}{2A_m B'_m}. \quad (33)$$

Solving equation (32), the structural transition temperature can be expressed as

$$T'_\delta = \frac{Z + T_\delta}{2} + \sqrt{\left(\frac{Z + T_\delta}{2}\right)^2 - ZT_m}. \quad (34)$$

Replacing the experimental values $x \simeq 0.7, T'_\delta = 200 \text{ K}$ of the [20] in equations (27) and (28) and then in the equation (32) we obtain

$$Z = 69.9 \text{ K.} \quad (35)$$

Without loss of generality, we can assume that $\delta$ is normalized to 1 for the largest possible value at $x = 0, T = 0$ in the absence of magnetization. In this case, equation (23) leads to

$$1 = \frac{A_\delta T_{\delta o}}{2B_\delta}, \quad (36)$$

where $T_{\delta o} = T_\delta (x = 0) = 712.06 \text{ K}$. Finally, from the combination of equations (21), (22), (30), (31), (35) and (36) we obtain

$$A_\delta = 2.563 \quad (37)$$

$$B_\delta = 912.6 \text{ K} \quad (38)$$
\[ C = 77.61 \text{ K.} \] (39)

4. Phase diagram

With the coefficients obtained above we can calculate the magnetic and structural transition temperatures and compare them with the experimental results.

In figure 1 we present the phase diagram, temperature versus Sr concentration \( x \), for the free energy described by equation (1) and the parameters obtained as described in the previous section. The four regions of the phase diagram are characterized by the vanishing or not of the magnetic \((m)\) and structural \((\delta)\) order parameters. At high temperatures one has \( m = 0 \) and \( \delta = 0 \) corresponding to the paramagnetic rhombohedral phase. For low enough temperatures and low Sr content the system is in the ferromagnetic orthorhombic phase. For low Sr content, for which \( \delta = 0 \) and \( m \neq 0 \), these two phases meet at a tricritical point with the paramagnetic orthorhombic phase \((m = 0 \text{ and } \delta = 0)\) and the ferromagnetic rhombohedral one \((m \neq 0 \text{ and } \delta = 0)\).

The phase diagram is in good agreement with the experimental one (figure 8 of [20]). Although several parameters were derived from experiment, it was not obvious that the proposed free energy functional describes the system.

5. Magnetic susceptibility

In this section, we discuss the magnetic susceptibility \( \partial M / \partial h \) in the region of low Sr content, for which \( T_\delta > T_m \). It has been measured experimentally [20]. One expects a kink at \( T = T_\delta \).

For \( T > T_\delta \) we need to consider only the magnetic free energy under a magnetic field \( h \), the first three terms of the free energy of the system equation (1)

\[ F = -mh + A_m(T - T_m)m^2 + B_m m^4. \] (40)

When \( h \to 0 \), \( m \) is linear with \( h \). Therefore, minimizing the free energy with respect to \( m \) and then deriving it with respect to \( h \) we obtain

\[ \chi_0 = \frac{\partial m}{\partial h} = \frac{K}{T - T_m}, \quad K = \frac{1}{2A_m}. \] (41)

For \( T_m < T < T_\delta \) we have to add the coupling between magnetic and structural order parameters. Since \( m \to 0 \) we can use equation (23) for the value of \( \delta \) and proceed in a similar way as above to obtain

\[ \chi_0 = \frac{K'}{T - T_m}, \quad K' = \frac{1}{2A_m} - \frac{c_4 \delta}{h}. \] (42)

The magnetic susceptibility is \( \chi = M_0 \chi_0 \). In figure 2 we plot \( \chi^{-1} \) versus \( T \) for \( x = 0 \). Clearly \( \chi^{-1} \) present a change of slope at the transition temperature \( T_\delta \). The inverse of the susceptibility for \( T > T_\delta \) extrapolates to \( T_m \), while for \( T < T_\delta \) it extrapolates to \( T_m' \). This kink is also observed experimentally (figure 4 of [20]). Experimentally a positive curvature of \( \chi^{-1} \) is observed at small temperatures which is beyond the reach of our theory.

6. Summary and discussion

We have presented a Ginzburg–Landau theory to describe experimentally observed magnetic and structural transitions in \( \text{La}_{1-x}(\text{Ca}_{1-x} \text{Sr})_x\text{MnO}_3 \). For the magnetic part of the free energy, we derive the parameters from a generalization to arbitrary angular momentum \( j \) (or spin in the angular momentum is quenched) of previous results for \( j = 1/2 \) [23]. This might be useful for future use of magnetic free-energy functionals.

The theory provides an explanation of the observed phase diagram and kinks in the magnetic susceptibility versus temperature in the system for \( y = 0.25 \) as reported by Alejandro et al [20].

We have considered only second-order phase transitions. For other compositions, for example \( y = 2/3 \) [19], first-order transitions and phase segregation was observed. Explanation of these phenomena would require a generalization of the theory. First-order transitions were discussed theoretically in [24].

In any case, we believe that our results provide a qualitative understanding of the effects of the competition between structural and magnetic order in these systems.

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