Phase separation and enhanced charge-spin coupling near magnetic transitions

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The generic changes of the electronic compressibility in systems which show magnetic instabilities is studied. It is shown that, when going into the ordered phase, the compressibility is reduced by an amount comparable to the its original value, making charge instabilities also possible. We discuss, within this framework, the tendency towards phase separation of the double exchange systems, the pyrochlores, and other magnetic materials.

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I. INTRODUCTION

The theoretical possibility of phase separation (PS) in magnetic systems was first discussed in connexion to the Hubbard model for itinerant ferromagnetism and antiferromagnetism [1]. Recent arguments suggest that PS is also likely in other magnetic materials, such as those in which magnetism is due to double exchange interactions [2–6]. Spins polarons, which can be viewed as a manifestation of PS on a small scale, have been analyzed in relation to the pyrochlores [1]. Finally, there is an extensive literature on PS in two dimensional doped antiferromagnets (see, for instance [10–15]), although there is no definitive consensus on its existence.

A variety of different experiments show features which are compatible with electronic phase segregation, at least on small scales, near magnetic transitions. For instance, near an antiferromagnetic transition there is ample evidence for the formation of charged stripes, both in the cuprates [6] and in the nickelates [7]. In the ferromagnetic manganites, various experiments suggest the existence of polarons near the Curie temperature [8], which, as mentioned above, can be viewed as phase separation on small scales. In addition, and also in the ferromagnetic manganites (i.e. La₁₋ₓCaₓMnO₃, x ≈ 1/3), inhomogeneous textures [9] and hysteretic effects [2] have been reported near Tc. Note that we are discussing the relatively simple case of the optimally doped, ferromagnetic compounds, avoiding the complications which arise between the competition between antiferromagnetism, charge ordering and double exchange in these materials. Finally, there is evidence for inhomogeneous textures in ferromagnetic pyrochlores [21]. The purpose of this paper is to show the theoretical foundations which make likely the existence of electronic phase segregation near magnetic transitions. We do not pretend to give an exhaustive list of experimental evidence which support this view.

In the following, we analyze how a magnetic transition influences the electronic compressibility. A general framework is presented in the next section. Simple applications to variations of the Hubbard model are presented in section III. Then, it is shown that the same framework predicts the existence of PS in the pyrochlores (section IV). Section V includes in the same framework the known results about PS in double exchange models, in the light of the present framework. Section VI analyzes the role of long range interactions, and some physical quantities likely to be affected by a reduced compressibility. Finally, the main conclusions of our work are presented in section VII.

II. GENERAL FEATURES OF THE ELECTRONIC COMPRESSIBILITY NEAR A MAGNETIC PHASE TRANSITION.

The formation of an ordered phase induces a decrease in the free energy of the material, usually called the condensation energy. Because of it, quantities which depend on the variation of the free energy with temperature, such as the specific heat, show anomalous, non analytic behavior at the critical temperature. For instance, the specific heat shows an abrupt reduction at Tc in mean field theories. The phase transition can also be tuned by varying n, the electronic content (per unit cell), in many systems leading to phase diagrams like that shown in fig. 2. Thus, one expects an anomalous dependence of the free energy on electronic density as the phase boundary is crossed by changing the electronic concentration.

We can get a simple estimate of the effect by using a standard Ginzburg-Landau expansion for the free energy:

\[ \mathcal{F}(s) = \frac{c}{2} \int (\nabla s)^2 d^D r + \frac{a}{2} \int (T - T_c(n)) ds^2 d^D r + \frac{b}{4} \int s^4 d^D r + \mathcal{F}_n(n) \]

where s is the electronic magnetization and n is the electronic density. We now neglect spatial fluctuations, and obtain a mean field approximation to \( \mathcal{F} \):
We obtain:

\[ \mathcal{F}_{MF} = \frac{a[T - T_c(n)]}{2} s^2 + \frac{b}{4} s^4 + \frac{(n - n_0)^2}{2\kappa_0} \]  

(2)

where we have expanded the dependence of the free energy of the paramagnetic phase on \( n \). \( \kappa_0 \) is the (scaled) specific heat. Moreover, it is reasonable to expect that the origin of this discontinuity is the same as that in the critical region are taken into account. When \( T < T_c \), the magnetization is:

\[ M \sim \frac{n - n_0}{2\kappa_0} \]  

(3)

Let us now fix the temperature \( T \) and expand this expression around the density \( n_c \) such that \( T = T_c(n_c) \). We obtain:

\[ \mathcal{F}_{MF} \approx -\frac{a^2}{4b} \left( \frac{\partial T_c}{\partial n} \right)^2 (n - n_c)^2 + \frac{(n - n_0)^2}{2\kappa_0} \]  

(4)

And by taking derivatives, we have:

\[ \tilde{\kappa}^{-1} \equiv \frac{\partial^2 \mathcal{F}_{MF}}{\partial n^2} = \left\{ \begin{array}{ll}
\frac{1}{\kappa_0} & T_c < T \\
-\frac{a^2}{2b} \left( \frac{\partial T_c}{\partial n} \right)^2 + \frac{1}{\kappa_0} & T < T_c
\end{array} \right. \]  

(5)

The compressibility has a jump at the transition. The origin of this discontinuity is the same as that in the specific heat. Moreover, it is reasonable to expect that this anomaly will be enhanced when fluctuations in the critical region are taken into account.

Alternatively, and keeping \( T \) fixed, one can minimize eq. (4) first with respect to \( n \). By expanding \( T_c(n) \) around \( n_0 \), we find:

\[ n \approx \left\{ \begin{array}{ll}
0 & T_c < T \\
0 + \frac{a\kappa_0^2}{2} \frac{\partial T_c}{\partial n} & T < T_c
\end{array} \right. \]  

(6)

and, inserting the value of \( n \) in the free energy when \( T < T_c \):

\[ \mathcal{F} \approx \frac{a[T_c(n_0) - T]}{2} s^2 + \frac{b}{4} s^4 - \frac{a^2\kappa_0}{8} \left( \frac{\partial T_c}{\partial n} \right)^2 s^4 \]  

(7)

The dependence of \( T_c \) on density leads to a negative quartic term in the dependence of the free energy on the magnetization. When \( b/4 - (a^2\kappa_0)(\partial T_c/\partial n)^2/8 < 0 \), the magnetic transition becomes first order. This condition is equivalent to saying that the effective compressibility, defined in eq. (4), becomes negative. Thus, PS near the transition can be thought of as arising from the transmutation of a continuous phase transition into a first order one by the introduction of an additional field, the density \( n \), which is a well known possibility in statistical mechanics [22]. The new feature found in a magnetic transition is that the correction to the compressibility can easily be comparable to the initial compressibility. The latter is determined by the density of states at the Fermi level in the paramagnetic phase. In typical magnetic systems, the transition is driven by a coupling constant which is of the order of the inverse of the density of states. Finally, the dependence of the critical temperature on the electronic density depends on the change of the coupling constant with variations in the density of states, which is of the same order (see examples below). Thus, no fine tuning of parameters is required to obtain corrections to the compressibility of the order of the compressibility itself.

III. PHASE SEPARATION IN HUBBARD LIKE MODELS

A. Ferromagnetic transitions

We first consider a one band Hubbard model with nearest neighbor ferromagnetic exchange couplings:

\[ \mathcal{H} = \sum_{k,s} \epsilon_k c_{k,s}^\dagger c_{k,s} + \sum_i U n_{i,\uparrow} n_{i,\downarrow} \]  

(8)

where \( i \) is a site index, \( n_{i,s} = \langle c_{i,s}^\dagger c_{i,s} \rangle \), \( \delta_i = \langle c_{i,s}^\dagger \delta_{s,s'} c_{i,s'} \rangle \) and \( \delta_{s,s'} \) are Pauli spin matrices. This model (with appropriate additions) has been invoked as a reasonable starting point for the study of itinerant ferromagnetism [23]. Within a mean field approximation, the Stoner criterium gives a ferromagnetic instability for \( \mathcal{U}(\epsilon_F) \geq 1 \), where \( \mathcal{U}(\epsilon) \) is the density of states per spin at energy \( \epsilon \). Let us assume that in the paramagnetic phase there are \( n_0 \) electrons per site, with chemical potential \( \mu_0 \):

\[ n_0 = 2 \int_{\mu_0}^{\epsilon} \mathcal{D}(\epsilon) \mathcal{d}\epsilon \]  

(9)

If we shift the two spin bands by \( \pm \delta \), the induced polarization \( s = n_{i,\uparrow} - n_{i,\downarrow} \) satisfies:

\[ \delta = \frac{s}{2\mathcal{D}(\mu_0)} + \frac{\mathcal{D}^2(\mu_0) s^3}{16\mathcal{D}^5(\mu_0)} - \frac{\mathcal{D}''(\mu_0) s^3}{48\mathcal{D}^3(\mu_0)} \]  

(10)

where \( \mathcal{D}^\prime \) and \( \mathcal{D}'' \) stand for the derivatives with respect to energy of the density of states. Because of the lack of electron-hole symmetry when these derivatives are finite, the chemical potential in the polarized state is shifted:

\[ \mu - \mu_0 = -\frac{\mathcal{D}''(\mu_0) \delta^2}{2\mathcal{D}(\mu_0)} + \frac{\mathcal{D}''(\mu_0) \mathcal{D}''(\mu_0) \delta^2}{4\mathcal{D}^2(\mu_0)} - \frac{\mathcal{D}''(\mu_0) \delta^4}{8\mathcal{D}^3(\mu_0)} \]  

(11)

The ground state energy, at zero temperature, can be written as:

\[ \mathcal{F}_s = E_0 + \frac{s^2}{4\mathcal{D}(\mu_0)} + \frac{\mathcal{D}^2(\mu_0) s^4}{64\mathcal{D}^5(\mu_0)} - \frac{\mathcal{D}''(\mu_0) s^4}{192\mathcal{D}^4(\mu_0)} \]  

(12)
where \( E_0 = 2 \int \mu \epsilon \mathcal{D}(\epsilon) \, d\epsilon \). This is the Ginzburg-Landau expansion needed to study the phase transition as function of electronic density at zero temperature. The system becomes ferromagnetic when \( \mathcal{D}(\mu_c)U \geq 1 \). We can expand the quadratic term in the magnetization around \( n_c = 2 \int \mu \mathcal{D}(\epsilon) \, d\epsilon \) as:

\[
\frac{1}{4} \left[ U - \frac{1}{D(\mu)} \right] \approx \frac{D'(\mu_c)(n-n_c)}{8D^4(\mu_c)} \tag{13}
\]

which leads to the following scaled inverse compressibility, as the transition is approached from the ordered side:

\[
\tilde{\kappa}^{-1} = \tilde{\kappa}_0^{-1} - \frac{1}{2D(\mu_c)} \frac{D'(\mu_c)}{1 - \frac{D''(\mu_c)D(\mu_c)}{3D^4(\mu_c)}} \tag{14}
\]

where \( \tilde{\kappa}_0^{-1} = (2D(\mu_c))^{-1} + (2U_c)^{-1}, \) and \( U_c = (\mathcal{D}(\mu_c))^{-1} \) is the critical coupling for the transition to take place. The compressibility is negative if \( D''(\mu_c)D(\mu_c)/(3D^2(\mu_c)) > 1/2 \). In particular, near a saddle point in the 3D dispersion relation, we have \( \mathcal{D}(\epsilon) = D_0 - c\sqrt{\epsilon - c^2D} \). This implies \( \lim_{\epsilon \rightarrow cD} D''(\epsilon)D(\epsilon)/D^2(\epsilon) \rightarrow +\infty \), and the system is always unstable versus PS.

It is interesting to note that, using a different formalism, phase separation has been shown to appear near ferromagnetic phases of the Hubbard model in two dimensions \cite{23}, in good agreement with the picture presented here.

### B. Doped antiferromagnets

It is well known that the Hubbard model (eq. \[8\]) at half filling, in a bipartite lattice and with nearest neighbor hoppings only, has an antiferromagnetic (AF) ground state, except in one dimension. The main physical features of this state, a charge gap, long range magnetic order and low energy spin waves, are well described using standard mean field techniques and the Random Phase Approximation. Recent work shows that, in the presence of a static magnetization, the opening of a charge gap occurs while the quasiparticle residues remain finite, at least in the infinite dimension limit \cite{23}, further supporting the validity of a mean field ansatz. Alternatively, a calculation of the ground state energy in the limit of large dimensions and small values of \( U/t \) can be obtained by standard perturbation theory around a mean field symmetry breaking state \cite{24}. This approach does suggest the existence of PS near half filling \cite{10}. In the following, we analyze the stability of homogeneous mean field solutions in arbitrary dimensions, following a different approach to that in \cite{10}.

The mean field solution at half filling is straightforward to extend to finite fillings. Hartree Fock calculations show that there are self consistent homogeneous doped solutions for a finite range of fillings around half filling, in two and three dimensions \cite{24} \cite{25}. As reported earlier, these solutions acquire a negative contribution to the electronic compressibility near the transition boundaries, making PS possible. In the following, we analyze the stability of homogeneous mean field solutions in arbitrary dimensions, following a different approach to that in \cite{10}.

The AF distortion shifts the mean field levels, \( c \rightarrow \text{sgn}(c)\sqrt{\epsilon^2 + \Delta^2} \), opening a gap and leading to the following staggered magnetization:

\[
\tilde{s} = \sum_{\sigma=\pm 1} \sigma c^\dagger_{i\sigma} c_{i\sigma} = -2 \int_{-W_0}^{\tilde{\mu}} d\epsilon \, \epsilon \text{sgn}(\epsilon) \Delta \tag{15}
\]

where \( \mathcal{D}(\epsilon) = \mathcal{D}(-\epsilon) \) and \( \tilde{\mu} \) are, respectively, the density of states (per spin) and Fermi level in the paramagnetic phase, with filling factor \( n = 2 \int_{-W_0}^{\tilde{\mu}} d\epsilon \mathcal{D}(\epsilon) \), and half-bandwidth \( W_0 \). The energy is given by

\[
\mathcal{E} = 2 \int_{-W_0}^{\tilde{\mu}} d\epsilon \mathcal{D}(\epsilon) \text{sgn}(\epsilon) \sqrt{\epsilon^2 + \Delta^2 + \Delta\tilde{s}} + \frac{U}{4} (n^2 - \tilde{s}^2) \tag{16}
\]

with the order parameter satisfying the following self-consistency requirement:

\[
\frac{1}{U} = - \int_{-W_0}^{\tilde{\mu}} d\epsilon \mathcal{D}(\epsilon) \frac{\text{sgn}(\epsilon)}{\sqrt{\epsilon^2 + \Delta^2}} \tag{17}
\]

Both energy and self-consistency equation can be obtained from the minimization of the following functional of the order parameter \( \mathcal{F}(\Delta) \), which we take as the starting point for the stability analysis:

\[
\mathcal{F}(\Delta) = 2 \int_{-W_0}^{\tilde{\mu}} d\epsilon \mathcal{D}(\epsilon) \text{sgn}(\epsilon) \sqrt{\epsilon^2 + \Delta^2 + \frac{U}{4} n^2 + \frac{\Delta^2}{U}} \tag{18}
\]

Away from half-filling, a critical value \( U_c(n) \) is required for the instability to take place, with \( U_c(n) = -1/\int_{-W_0}^{\tilde{\mu}} d\epsilon \mathcal{D}(\epsilon) \). Close to the transition line \( U_c(n) \), a Ginzburg-Landau analysis is straightforward, leading to:

\[
\mathcal{F}(\Delta) = f_0 + f_2 \Delta^2 + f_4 \Delta^4 \tag{19}
\]

with

\[
f_0 = 2 \int_{-W_0}^{\tilde{\mu}} d\epsilon \, \epsilon \, \mathcal{D}(\epsilon) + \frac{U}{4} n^2 \tag{20}
\]

\[
f_2 = 1/U - 1/U_c(n) \tag{21}
\]

\[
f_4 = -\frac{1}{4} \int_{-W_0}^{\tilde{\mu}} d\epsilon \, \frac{\mathcal{D}(\epsilon)}{\epsilon^3} \tag{22}
\]

The scaled inverse compressibility satisfies:

\[
\tilde{\kappa}^{-1} = \frac{\partial^2 \mathcal{E}}{\partial n^2} = \left\{ \begin{array}{ll}
\frac{1}{2D(n)} + \frac{U}{2} & \text{for } \mathcal{D}(\epsilon) \geq 1/U_c^-
\frac{1}{2D(n)} + \frac{U}{2} - \frac{(\partial f_2)^2}{2f_4} & \text{for } \mathcal{D}(\epsilon) \geq 1/U_c^+
\end{array} \right. \tag{23}
\]

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Following the transition line on the AF side to the limit of small doping, one has \( \frac{(\partial_{\mu} f)^2}{2f} \rightarrow 1/D(\mu) \), leading to \( \kappa^{-1} \rightarrow \frac{1}{2D(\mu)} \). Therefore we have proven the existence of PS in the vicinity of \( U_c(n) \), at least close to half filling.

In fact, PS is not restricted to the previous region. It is also present around half-filling, \( n \rightarrow 1^\pm \), for all values of \( U \), as we now show. Close to half-filling and at finite \( U \), the compressibility satisfies

\[
\kappa^{-1} = \frac{U}{2} - \left[ 2\Delta^2 \int_{-W_0}^0 \frac{D(\epsilon)}{(\epsilon^2 + \Delta^2)^{3/2}} \right]^{-1}
\]

and, given the following inequality (see eq. [17]),

\[
2\Delta^2 \int_{-W_0}^{\tilde{\mu}} \frac{D(\epsilon)}{(\epsilon^2 + \Delta^2)^{3/2}} < 2 \int_{-W_0}^{\tilde{\mu}} \frac{D(\epsilon)}{\sqrt{\epsilon^2 + \Delta^2}} = \frac{2}{U}
\]

one obtains

\[
\kappa^{-1} < 0
\]

proving PS around half-filling for all values of \( U \), as previously stated. This analysis indicates that PS is a very robust feature of the AF instability at the mean field level. In fact, for a flat model density of states, numerical results suggests that the entire AF region has negative compressibility.

The study of this section can be extented to the case of a diverging density of states at half-filling: \( D(\epsilon) \sim |\epsilon|^\alpha \), \( 0 \leq \alpha < 1 \). The existence of PS is also proved in this case, at least around the transition line \( U_c(n \rightarrow 1) \). In particular, this includes the mean field solution of the Hubbard model in the square lattice, where the density of states has a van Hove logarithmic divergence. The conclusions of our analysis are consistent with the tendency towards inhomogeneous solutions found in mean field studies of the Hubbard model [24,22].

IV. PYROCHLORES

The pyrochlores are metallic oxides, \( \text{Mn}_2\text{Ti}_2\text{O}_7 \), which show various anomalous transport and magnetic properties, including colossal magnetoresistance. The magnetism is mostly due to the spins of Mn ions. The electronic carriers are assumed to come from a wide s band from Ti orbitals, with a very low occupation. These electrons are coupled ferromagnetically to the Mn spins [23,24]. We can describe the coupled system by the hamiltonian \( \mathcal{H} \):

\[
\mathcal{H} = \sum_{k,s} \epsilon_k c_{k,s}^\dagger c_{k,s} - J \sum_{ij} \bar{s}_i \bar{s}_j - J' \sum_i c_{i,s}^\dagger \bar{\sigma}_{ss'} c_{i,s'} \bar{s}_i
\]

where \( J \) and \( J' \) are positive (ferromagnetic). The model undergoes a transition to an ordered where the local spins have a finite magnetization, \( s = |\langle \bar{s} \rangle| \neq 0 \), and the electron gas is polarized, \( \bar{s} = |\langle c_{i,s}^\dagger \bar{\sigma}_{ss'} c_{i,s'} \rangle| \neq 0 \).

In principle, the entire issue of PS in the pyrochlores could be settled using results of Majumdar-Littlewood (ML) [25]. These authors have shown that magnetic polarons (i.e. self-trapped, polarized electrons in a magnetic bubble of core spins embedded in a paramagnetic environment) are the stable configuration of carriers in the paramagnetic phase at low densities. Now we will show that the existence of polarons in the paramagnetic phase is a sufficient condition for the thermodynamic instability of the uniform phase of the standard mean field analysis.

Let us consider a low concentration \( (n) \) of carriers in the polaronic configuration. The free energy at fixed temperature is proportional to the number of carriers (polarons):

\[
\mathcal{F}_{\text{pol}}(T, n) = \delta f_{\text{pol}}(T) \ n
\]

where \( \delta f_{\text{pol}}(T) < 0 \) is the free energy reduction per carrier due to its polaronic bubble. The preference for polarons implies that the homogeneous (paramagnetic) phase \( \mathcal{F}_{\text{hom}} \) is above \( \mathcal{F}_{\text{pol}} \) in the limit \( n \rightarrow 0 \). Consider now that the carrier density reaches the value \( \tilde{n} \) at which all volume is filled with polarons, each one with its confined carrier. It is obvious that, allowing carriers to delocalize (keeping core magnetization and temperature constant), their kinetic energy diminishes. This implies the existence of a homogeneous phase (with finite core magnetization and total carrier polarization) with energy below the polaronic phase at finite carrier concentration. A variational reasoning guarantees that the true homogeneous solution of the mean-field treatment is even below in free energy. In conclusion, if the single polaron is stable, the free energy of the homogeneous phase must satisfy

\[
\mathcal{F}_{\text{pol}}(T, n \rightarrow 0) = \delta f_{\text{pol}}(T) \ n < \mathcal{F}_{\text{hom}}(T, n \rightarrow 0)
\]

\[
\mathcal{F}_{\text{pol}}(T, \tilde{n}) = \delta f_{\text{pol}}(T) \ \tilde{n} > \mathcal{F}_{\text{hom}}(T, \tilde{n})
\]

Remenber that \( \mathcal{F}_{\text{pol}} \) is linear in \( n \), and both \( \mathcal{F}_{\text{pol}} \) and \( \mathcal{F}_{\text{hom}} \) start from the same point. Then, the previous inequalities can only happen if the curvature of \( \mathcal{F}_{\text{pol}} \) with concentration changes sign, leading to a negative compressibility and PS. Therefore we have proven our original assertion: stable polarons imply thermodynamic instability towards PS. Notice that this is a sufficient condition, but not necessary: PS can exists (see below) even if the ML polarons are not the stable configuration in the paramagnetic phase. A graphical description of the previous argument comparing polaronic and homogeneous phase free energies for \( kT = 0.115 \ t \) is shown in fig. [1] (two upper curves). In addition, we also show the free energy of the correct treatment, including carriers entropy (see below).
FIG. 1. Free energy versus electron concentration for the exact mean field treatment (continuous line), the polaronic ansatz (dashed straight line), and the degenerate approximation for electrons (dashed-dotted line), for $kT = 0.125\ t$, and parameters explained in the text. The arrow marks the onset of negative curvature.

There is, however, an important source of concern with the previous conclusion. In the ML analysis, carriers in the homogeneous phase enter with energy but no entropy. This would be correct for the usual case of degenerate fermions. But the temperature is finite and the interesting region corresponds to very low carrier concentration, making the degenerate assumption questionable. For the parameters expected to apply in the pyrochlores, the classical limit would be a more appropriate starting point. In fact, in the limit of zero concentration at finite temperature, the chemical potential ($\mu = \partial_n F_{\text{hom}}$) corresponds to classical particles, and diverges: $\mu(n \to 0) \sim \log(n)$. This implies that, in that limit, the homogeneous paramagnetic phase (including carrier’s entropy) is always more stable that the ML polaronic ansatz. Notice that our original statement remains true: the existence of stable polarons would imply thermodynamic instability of the homogeneous phase. It only happens that the carrier’s entropy makes the paramagnetic homogeneous phase to be preferred over the polaronic one at low doping, making the very existence of polarons at finite doping uncertain.

Given the previous situation, we study the existence of PS in the pyrochlores performing the usual mean field calculation without simplifying hypothesis for the carriers. The Helmholtz free energy $F$ contains the core $F_s$, carrier $F_s$, and core-carrier contributions $F_{ss}$, with expressions:

$$F_s = \sum_{\sigma = \uparrow \downarrow} \int d\epsilon \ D(\epsilon) \left\{ \epsilon - kT \log \left( e^{\epsilon/kT} + 1 \right) \right\}$$

$$F_{ss} = -J'ss$$

where $s$ is the number of nearest neighbors, $D(\epsilon)$ represents the paramagnetic density of states (per spin) and $h$ describes the effective field characterizing the mean field distribution of core spins, assumed to be classical vectors of unit length, therefore, $s = \tanh(h) - h^{-1}$.

Minimization of $F$ leads to the following equations:

$$\mu_\uparrow - J's - \mu = 0$$

$$\mu_\downarrow + J's - \mu = 0$$

$$-Jzs + kTh - J's = 0$$

with the occupation constraint:

$$n = \sum_{\sigma = \uparrow \downarrow} \int d\epsilon \ \frac{D(\epsilon)}{\exp(\epsilon/\sigma kT) + 1}$$

We have solved the previous equations for parameters expected to apply in the case of pyrochlores: $kT_0 = 0.4 = 0.1\ t$, and $J' = t$, where $t$ a measure the electronic energy scale, is taken to be the kinetic energy of an electron confined to a unit cell volume. Searching for negative values of $\mu = \partial_n F$, we arrive at the phase diagram plotted in fig. 2 for small carrier concentration. We see that, beyond a carrier concentration, a region of negative compressibility opens around the ferro-para transition. Therefore, we confirm the existence of PS as suggested by the general arguments of section II. Notice that in fig. 2 we show the area of intrinsic instability: the standard Maxwell construction would increase this PS region even into the paramagnetic phase. We have plotted (fig. 2) the free energy of the exact solution (continuous line), compared to the ML polaron ansatz (dashed, straight line) and the solution ignoring the electron entropy (dashed-dotted line) for $kT = 0.115\ t$. As explained at the beginning of this section, the correct homogeneous phase (including entropy) has the lowest free energy even at low concentration. In contrast, having ignored the electronic entropy would have led to the prediction that polarons are the preferred configuration (see two upper curves of fig. 2). Nevertheless, PS does exists, and the onset of negative curvature (hardly visible to the bare eye) is marked with an arrow in that figure.

For the parameters corresponding to fig. 2, electrons are strongly non degenerate, and the numerical results almost coincide with the classical limit for the carriers, whose Ginsburg-Landau analytic treatment we now present for completeness. Assuming the electrons to be a classical ideal gas and performing an expansion in the core $s$, and electron magnetization $\bar{s}$, one obtains the following expressions for the contributions to the free energy:

$$F_s = \left( \frac{3}{2} kT - \frac{Jz^2}{2} \right) s^2 + \frac{9}{20} kT^2 s^4$$

$$F_{ss} = -J'ss$$

$$F_{s\bar{s}} = -J's\bar{s}$$

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\[ F_s = -nkT + nkT \log \left( \frac{n}{n_Q} \right) + \frac{kT}{2n} s^2 + \frac{kT}{12n^3} s^4 \]  
\[ F_{s \bar{s}} = -J's\bar{s} \]  
(35)

where \( n_Q \) is a reference density marking the onset of the quantum regime.

Minimization of the free energy leads to the following transition temperature:

\[ kT_c = \frac{kT_0}{2} + \sqrt{\left( \frac{kT_0}{2} \right)^2 + \frac{J^2 n}{3}} \]  
(37)

In the vicinity of \( T_c \), core and electron magnetization are related by

\[ \bar{s} = \frac{sJn}{kT} \left[ 1 - \frac{J^2 s^2}{3(kT)^2} \right] \]  
(38)

and the scaled inverse compressibility at \( T \to T_c^- \) is given by:

\[ \tilde{\kappa}^{-1} = \frac{\partial^2 F_{MF}}{\partial n^2} = \tilde{\kappa}_0^{-1} - \frac{15 kT_c J^4}{54(kT)^4} + 10J^4 n \]  
(39)

where \( \tilde{\kappa}_0^{-1} = kT_c/n \), is the inverse compressibility of the paramagnetic phase. \( \tilde{\kappa} \) becomes negative beyond \( n_{PS} = 54T_c/(5J^4) = 1.266 \times 10^{-3} \), for the parameters used here, very close to the exact results of fig. 3.

As a final remark, it is important to realize that the precise configuration of the coexisting phases in the real material will be complicated by effects beyond the present treatment (disorder, domain boundary contributions ...). Nevertheless, the associated spatial inhomogeneity will certainly affect the physics of these compounds in an important way. This strongly suggests that, even if the original polaron argument is doubtful, the associated physics explored by ML might well apply to the pyrochlores.

V. DOUBLE EXCHANGE SYSTEMS.

The double exchange (DE) model describes systems with local spins and itinerant electrons, in the limit where the Hund coupling between the electrons and the spins is much larger than other scales \([7] \). The electrons are always polarized in the direction of the local spins, and hopping \( (t) \) to neighboring ions is modulated by the relative orientation of these spins, leading to the following Hamiltonian

\[ H = -t \sum_{\langle ij \rangle, \sigma} \left[ z_{i\sigma} z_{j\sigma} c_{i}^\dagger c_{j} + \text{H.c.} \right]. \]  
(40)

where \( z_{i\sigma} \) is the spinor describing the orientation of the core at site \( i \): \( z_{i\uparrow} = \cos(\theta_i), z_{i\downarrow} = \sin(\theta_i) \exp(-i\phi_i) \).

The model, put forward by Zener \([8] \), has received great attention in recent times as the building block needed to explain the physics of Mn perovskites. We will restrict ourselves to its simplest one-band version, leaving aside further features such as orbital degeneracy, antiferromagnetic couplings or Jahn-Teller distortion, probably required for a more realistic description of manganites.

PS in double exchange models has been extensively studied \([9,10,11,12] \). It was originally observed in numerical studies of the DE model with AF couplings between core spins, raising doubts about its existence in the bare version \([13] \). However, we have recently shown that PS tendency is an intrinsic feature of the simplest DE model without additional terms \([10] \). For completeness, we include here an analysis of the compressibility using the Ginzburg-Landau expansion outlined in the preceding sections. The approach is a direct extension of that used in \([15] \). We leave out the study of PS at zero temperature due to the competition between direct antiferromagnetic interactions and the double exchange mechanism \([16] \), which also can be cast in terms of a Ginzburg-Landau expansion.

The model is determined by the polarization of the localized spins, \( s \), the density of states of the itinerant electrons in the absence of spin disorder, \( D_0(\epsilon) \), and the temperature \( T \). The free energy of the spins comes from the entropy due to their thermal fluctuations only, and coincides with eq. \([16] \), except for the absence of the direct exchange term. We assume that the electron gas is degenerate, an excellent approximation for temperatures in the range of \( T_c \), and replace its free energy by the ground state energy in a background of fluctuating spins. The bandwidth of the electrons is reduced by a

\[ \text{FIG. 2. Phase diagrams for the mean field theory of eqns. (41, 42, 43) for the parameters explained in the text. The solid line is the (scaled) critical temperature versus carrier concentration. The region of negative compressibility is bound by the dashed line and the critical temperature.} \]
factor \( f = \langle \cos(\vartheta_{ij}/2) \rangle \), where \( \vartheta_{ij} \) is the angle between neighboring spins. Thus, the electronic energy can be written as:

\[
E = f K_0 = f \int_{-W_0}^{\mu_0} \epsilon \, D_0(\epsilon)d\epsilon
\]

where \( \mu_0 \) denotes the chemical potential in the absence of spin disorder, and \( W_0 \) is the lower band edge (note that \( -W_0 \leq \mu_0 \leq W_0 \)). We can now expand \( f \) in terms of the magnetization \( s \), to obtain:

\[
E = \frac{2}{3} K_0 + \frac{2}{5} K_0 s^2 - \frac{6}{175} K_0 s^4
\]

which completes the required Ginzburg-Landau expansion. The Curie temperature of the model is given by

\[
T_c = -\frac{4}{15} K_0 \quad (K_0 \text{ is negative})
\]

The compressibility below \( T_c \) is negative when:

\[
\frac{|K_0|}{D_0(\mu_0)} \leq \frac{7}{9} \mu_0^2
\]

This result is independent of the initial bandwidth, \( 2W_0 \).

If \( \mu_0 \to -W_0 \) while \( \frac{K_0}{D_0(\mu_0)} \to 0 \). At half filling, \( \mu_0 \to 0 \), and there is no PS.

Finally, the scaled inverse compressibility is:

\[
\tilde{K}^{-1} = \begin{cases} 
\frac{2}{3D_0(\mu_0)} & T \to T_c + 0 \\
\frac{14\mu_0^2}{27|K_0|} & T \to T_c - 0
\end{cases}
\]

The magnetization is:

\[
\langle \vartheta_{ij} \rangle = \frac{1}{2} \left( \frac{W_0 + W_0}{W_0} \right) \tanh \left( \frac{\alpha + f \gamma}{2 \Theta} \right)
\]

where \( s = \text{ctnh}(Q) - Q^{-1} \) is the magnetization, \( x \) describes the doping level (electron/hole), and \( f = \sqrt{\frac{1}{2} \left( 1 + s^2 \right)} = \langle \cos^2(\vartheta_{ij}) \rangle^{1/2} \). The energy, temperature and chemical potential are scaled in units of the half-bandwidth in the absence of spin disorder by \( \gamma \equiv \epsilon/W_0 \), \( \alpha \equiv \mu/W_0 \), and \( \Theta \equiv kT/W_0 \).

If \( D_0 \) is taken to be the density of states of the original crystal lattice, neglect of the Berry phase \( \text{fig. 3} \) collected by the hopping electron is implicit. More appropriately, we can assume that spin disorder cancellations favor retraced paths \( \text{fig. 4} \), leading to a Bethe lattice density of states, with eq. (17) as the infinite coordination limit. In fig. 3 (upper panel) we show the phase diagram (scaled temperature versus hole concentration \( x \)) for the density of eq. (17). Notice that the PS region corresponds to the intrinsic instability (negative compressibility). Global stability (Maxwell construction) would increase this region.

The simple DE model offers a clear example of the general mechanism for PS in the presence of an ordering process. In fact, the mean field treatment is intrinsically unstable versus PS in the limit of low carrier density. To see this, notice that the energy scale of the magnetic order is the electronic energy, proportional to the carrier concentration in the dilute limit. Doubling, for instance, the carrier concentration close to the Curie temperature produces a finite increase of the bandwidth. The chemical potential instead remains tied to the band edge and follows its fate: it decreases with increasing electron density (thermodynamic instability). In our electron-hole symmetric model, the same would apply to the dilute
hole limit.

Although we do not expect the mean field approximation to be a serious drawback, certain limitations are clear. For instance, the inability to distinguish between short-range correlations and long-range order confines the negative compressibility region to the ferromagnetic phase, for only there does the bandwidth change with magnetic order. In what follows, we will show that PS is a robust feature that survives several improvements of this basic mean field approach.

A. Schwinger bosons.

An improved mean field theory can be implemented using the Schwinger boson method. A nondynamical field \( \lambda \) enforces the constraint \( z_{i\sigma}^*, z_{i\sigma} = 1 \) at every site. The core spins are quantized according to \([z_{i\sigma}, z_{j\sigma'}] = \delta_{ij} \delta_{\sigma\sigma'}/2S\). The mean field Hamiltonian is obtained through a Hartree decoupling of the bosonic \( z_{i\sigma}^*, z_{i\sigma} \) and fermionic \( c_i^\dagger c_j \) hopping terms:

\[
\mathcal{H}_{\text{MF}} = N (ztfK - \mu) - \mu \sum_i c_i^\dagger c_i + \lambda \sum_{i,\sigma} \bar{z}_{i\sigma} z_{i\sigma}
\]

\[
- t f \sum_{\langle ij \rangle} (c_i^\dagger c_j + c_j^\dagger c_i) - tK \sum_{\langle ij \rangle, \sigma} (\bar{z}_{i\sigma} z_{j\sigma} + \bar{z}_{j\sigma} z_{i\sigma}),
\]

where \( K = \langle c_i^\dagger c_j \rangle \) and \( f = \langle z_{i\sigma}, \bar{z}_{j\sigma} \rangle \). Such a model was introduced by Sarker, who identified a Curie transition and found that the \( e_g \) fermion band becomes incoherent above \( T_c \). Accounting for the possibility of condensation of Schwinger bosons, we write \( \Psi_{k\sigma} \equiv \langle z_{k\sigma} \rangle \). Assuming condensation only at \( \mathbf{k} = 0 \), we define \( \rho \equiv |\Psi_{0,\sigma}|^2 \).

We have solved the mean field equations for a semi-elliptic density of states and \( S = 3/2 \). Several features of the solution are shown in fig. \( \mathcal{F} \) (lower panel). While the critical temperature is hardly affected, the region of negative compressibility enters now into the paramagnetic phase, an expected physical feature not present in the previous approximation. In this approach, the fermion bandwidth collapses to zero at high temperature. Although this is likely an artifact of the approximations, it takes place well above the Curie temperatures to be a source of concern.

B. Critical spin fluctuations

The rapid change of spin correlations ties the PS region to the magnetic ordering transition. One can worry about the importance of critical spin fluctuations, neglected in the molecular field approximation. Assuming a nearest-neighbors, tight-binding lattice with hopping amplitude \( t_{ij} = -t \cos(\theta_{ij}/2) \), and expanding the half-angle between spins in the usual manner: \( \cos(\theta/2) \approx a_0 + a_1 \cos(\theta) \), the standard virtual crystal decoupling of the electron-spin system allows integration of charge degrees of freedom. This produces an effective Heisenberg model for the core spins (assumed classical for simplicity) and the total free energy (charge and core spins) is given by

\[
\mathcal{F} = K_\infty(x) + \mathcal{F}_H(J, T)
\]

where \( K_\infty \) is the electron energy (degenerate limit assumed) of the fully disordered limit \( t_{ij} = a_0 t \). \( \mathcal{F}_H \) describes the free energy of the classical Heisenberg model, with coupling constant \( J = 2a_1 |\langle c_i^\dagger c_j \rangle|t \), where tilded quantities are electron properties scaled to unit half-bandwidth. A lengthy but simple calculation permits de following connection between the stability criterium and thermal properties of the Heisenberg model (cubic lattice):

\[
\frac{\partial^2 \mathcal{F}}{\partial \eta^2} = \frac{1}{\mathcal{D}(\mu)} \left[ 1 - g(x) f_H(T) \right]
\]

with

\[
g(x) = \frac{2a_1 \tilde{\mu}^2 \mathcal{D}(\tilde{\mu})}{6 |\langle c_i^\dagger c_j \rangle|}
\]

and

\[
f_H(T) = \frac{T C(T)}{\left[ a_0 + a_1 \langle \cos(\theta_{ij}) \rangle \right]}
\]

\( \mu \) is the chemical potential and \( x \) the carrier density. \( C(T) \) represents the specific heat of the Heisenberg model. The expected increase of \( C(T) \) near the critical temperature \( T_c \) links the tendency towards instability with magnetic ordering. In any case, for a fixed value of
$T/T_c$, the system is bound to exhibit PS, $(\partial \mu/\partial x) < 0$, owing to the diverging behavior of $g(x \to 0)$. Extracting the temperature dependence of $f_\mu(T)$ from published Monte Carlo data \cite{10} for the cubic lattice, we estimate PS accompanying the ordering transition up to $x \simeq 0.11$. The resulting phase diagram and intrinsic instability region are shown in fig. 4 (upper panel). Notice that no approximate treatment is assumed for the criticality of the effective magnetic Hamiltonian, the only (major) surviving approximation being the virtual crystal (mean field) decoupling of mobile carriers and core spins.

**C. Infinite dimension**

The mean field decoupling of charge and core spins fluctuations remains in all the previous analysis. This manifests itself in a mere rescaling of the electronic density of states with magnetic order. As the PS instability can be seen as a manifestation of correlation between charge and spin at a macroscopic level (the only possibility left in a mean field approach) concern might arise about its fate when charge-spin coupling is properly considered. Although in the general case a numerical approach would be necessary to answer this question, there is a limit in which the problem is solvable but the charge-spin coupling remains non trivial: infinite coordination (dimension). The relevance of this limit for the understanding of correlated systems in 3-D has been recognized in recent years \cite{11,12}.

In the DE case, this problem has been considered by Furukawa \cite{43}, and indeed PS has been observed \cite{34}. In those works, the DE model included a large though finite Hund coupling, leading to AF correlations between core spins that certainly increase the tendency towards PS. This fact, together with accumulated experience from numerical studies of doped antiferromagnets, has led to the extended belief that antiferromagnetism is required for PS to exist. We now show that, in spite of recent suggestions on the contrary, the simple DE model without AF additions indeed exhibits PS close to the ordering temperature, according to the general explained in this paper.

The average density of states for electrons moving in an infinitely coordinated Bethe lattice according to the DE Hamiltonian is given by:

$$\langle D(\epsilon) \rangle_{\bar{\Omega}} = -(1/\pi) \Im \langle g(\epsilon, \bar{\Omega}) \rangle_{\bar{\Omega}}$$

where the local Green's function satisfies:

$$g(\epsilon, \bar{\Omega}) = \left[ \epsilon - \Sigma(\epsilon, \bar{\Omega}) \right]^{-1}$$

and the self-energy is determined by:

$$\Sigma(\epsilon, \bar{\Omega}) = (W_0/2)^2 \langle \frac{1 + \bar{\Omega} \cdot \bar{\Omega}'}{2} \left[ \epsilon - \Sigma(\epsilon, \bar{\Omega}') \right]^{-1} \rangle_{\bar{\Omega}'}$$

where

![FIG. 4. Phase diagrams (upper panel) for the cubic lattice in the virtual crystal approach, including MonteCarlo data for the Heisenbeg model (subsection B) and (lower panel) for the Bethe lattice of infinite coordination. The solid line is the (scaled) critical temperature versus hole concentration. The dashed line marks the boundary of the region of negative compressibility.](image)

$\bar{\Omega}$ is the unit vector describing the local core (classical) spin, $W_0$ represents the half-bandwidth for the fully aligned case, and angular averages are taken with the probability distribution $P(\bar{\Omega})$. Notice that, though the probability distribution for spins in different sites factorizes (confining PS the the ferromagnetic phase), charge and spin fluctuations remain coupled at the same site, and the density of states is not merely rescaled by magnetic order (eq. \cite{23}).

The complete thermodynamic problem is solved by finding the probability distribution that minimizes the total free energy.

$$F = F_{el} - TS$$

where $F_{el}$ is the free energy of fermions for the average density of states and $S$ is the core spin entropy, both depending on $P(\bar{\Omega})$.

For computational convenience, we have solved the problem with two additional features: degenerate limit for carriers and a core spin distribution parametrized by an effective magnetic field (detailed analysis of the exact solution for a few temperatures shows the previous simplifications to introduce only minute corrections). The generic results of previous treatments are reproduced here. This is shown in fig. 5 (lower panel), where the phase diagram presents the expected region of intrinsic instability (negative compressibility) at low carrier density. The inclusion of a finite Hund coupling will certainly increase the PS region, as observed in previous...
works [5,6]. All these studies support the view that the general scheme for PS presented in this paper applies to the simple DE model with features which are robust and not an artifact of approximations.

As mentioned before, PS can be thought of as a long range instability caused by the coupling between carriers and core spins. Such mechanism is operative even in the region where the system does not phase separate, and the carriers compressibility can be much enhanced close to the Curie temperature. This statement does not contradict the fact that the density of states at the Fermi level is featureless and shows no major change around the Curie temperature [44]. Remember that, if the bandwidth were not affected by the spin order, the compressibility would be merely proportional to the density of states at the Fermi level, that means featureless. It is the bandwidth change with magnetic order what produces this enhancement in the region close to the onset of magnetic order. This is shown in Fig. 5, where the compressibility of the infinitely coordinated Bethe lattice is measured in units of the compressibility of a free fermion system (that is, no carrier-spin coupling) with the same density of states. This behavior is not a peculiarity of the infinitely dimension limit, and remains the same in all previous approaches. The possible connection between this enhancement and the unusual properties of the manganites remains an open and intriguing question [6].

VI. INFLUENCE OF A MAGNETIC FIELD

So far, we have considered the existence of phase separation near a ferromagnetic (or antiferromagnetic) - paramagnetic transition, at zero magnetic field. When a field is applied, the features associated to the transition are smoothed, and, at sufficiently large fields, the magnetic moments are aligned, and the magnetization has a weak temperature dependence.

The increase in the compressibility which leads to phase separation is associated with the strong coupling between magnetic and charge fluctuations, as measured by the dependence of the critical temperature on electronic concentration. The magnetic field suppresses magnetic fluctuations leading to a smaller increase in the compressibility near $T_c$. Phase separation should disappear at sufficiently large fields, when the temperature no longer induces significant magnetic fluctuations.

A generic case which shows the dependence of the region where phase separation occurs on magnetic field, at fixed nominal electronic concentration, is shown in fig. (6). The calculations have been done for the double exchange model of the previous sections, using the mean field equations (42, 43), plus an applied field. The region of negative compressibility is shown. As in previous examples, a Maxwell construction gives a somewhat larger region.

In a small applied field, phase separation takes place above the Curie temperature. This is due to the rounding of the discontinuity at $T_c$ of the compressibility induced by the field. The region of phase separation lies between a high magnetization and a low magnetization phase, in close analogy with the phase diagram of an ordinary liquid-vapor transition. At high fields, phase separation is completely suppressed. At the highest possible field we find a critical point.

VII. PHASE SEPARATION AND DOMAIN FORMATION

The analysis in the previous sections suggests that PS should be a frequent feature of magnetic transitions. PS on a macroscopic scale, however, cannot occur, as it requires an infinite amount of electrostatic energy. We can...
extend the mean field analysis used in the previous section to study the role of the Coulomb interaction. If we first neglect it, the charge susceptibility obeys:

$$\lim_{q \to 0} \chi(q) = -\partial n \over \partial \rho = -\chi_0(q)$$

where $\chi_0(q)$ is the self-energy of the unit cell. A charge fluctuation $\rho(q)$ induces an electrostatic potential, $(4\pi e^2/q^3)\rho(q)$, which induces more charge polarization. The RPA selfconsistent equation for the charge polarization becomes:

$$\lim_{q \to 0} \chi(q) = \chi_0(q) \to -\chi_0(q)$$

when, in the absence of Coulomb repulsion the system exhibits the instability, $\kappa < 0$, the denominator has a pole at:

$$q_* = \sqrt{-\frac{4\pi e^2}{\chi_0(q)}}$$

and charge fluctuations of shorter wavelengths remain unstable, whereas the long range nature of the Coulomb term prevents the formation of larger charge inhomogeneities. Thus, we expect the formation of domains at length scales comparable to $q_*^{-1}$. As mentioned previously, this analysis does not take into account neither the cost in magnetic energy associated with the formation of domain walls nor the effect of impurities. In addition to macroscopic charge neutrality, these are likely to be among the major factors affecting the spatial coexistence pattern in real materials.

VIII. CONCLUSIONS

We have discussed a general framework which shows that PS is likely to occur near magnetic phase transitions. The dependence of the critical temperature, or critical couplings, on electronic density leads to a reduction of the compressibility in the ordered phase. This reduction tends to be of the same order as the value of the compressibility in the disordered phase. The existence of PS typically depends on numerical constants of order unity, related to the electronic structure. The coupling constants do not require a special fine tuning for PS to take place. The analysis presented here is probably relevant to understand a variety of experimental findings in the manganites, the pyrochlores and doped antiferromagnets.

We have studied PS mostly within a conventional mean field framework. Corrections due to critical fluctuations will usually enhance the effects discussed here. The compressibility is associated with the second derivative of the free energy with respect to electronic density, in a similar way to the specific heat. Critical fluctuations tend to increase the divergences of the derivatives of the free energy at the transition, so that the estimates reported here probably underestimate the tendency towards PS. The analysis also shows that, even in the absence of phase separation, an enhanced coupling between charge and magnetic fluctuations is expected, due to the reduction in the charge compressibility.

Finally, we have discussed the way in which electrostatic effects frustrate the formation of macroscopic domains, and we give a scheme to calculate the scales at which domain formation is expected.

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