The Double Exchange Model at Low Densities

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We obtain the phase diagram of the double-exchange model at low electronic densities in the presence of electron-electron interactions. The single particle problem and its extension to low electronic densities, when a Wigner crystal of magnetic polarons is generated due to unscreened Coulomb interactions, is studied. It is argued that the Wigner crystal is the natural alternative to phase separation when the Coulomb interaction is taken into account. We discuss the thermal and quantum stability of the crystalline phase towards a polaronic Fermi liquid and a homogeneous, metallic, ferromagnetic phase. The relevance and application of our results to EuB6 is also considered.

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Introduction. Magnetic polarons are ubiquitous in the physics of double-exchange systems such as mixed valence manganites 1, antiferromagnetic 2 and ferromagnetic semiconductors (such as EuO 3), diluted magnetic semiconductors (DMS) (such as Ga1-xMnxAs) 4,5, and in EuB6 where they have been observed in the optical response 6. Colossal magnetoresistance (CMR) has been attributed in part to the presence of magnetic polarons close to the paramagnetic-ferromagnetic transition 7. In fact, CMR in the Mn pyrochlores has been proposed on the basis of magnetic polarons 8. Given that these different classes of magnetic materials have attracted considerable attention in recent years because of their potential for the development of new magnetoelectronic devices, a number of theoretical approaches to the problem have been developed through the years. In particular, extensive work has been done with emphasis in the physics of magnetic semiconductors 2,4,8,10 and CMR manganites 11, where electron-electron interactions are assumed to be weak.

In this work we will focus our attention on the phase diagram of the double-exchange model (DEM) at low densities, taking into account the effects of Coulomb interactions between the electrons. Our results are summarized in Fig. 1. Studies devoted to the polaronic stability in this particular model and its variations have been performed by several authors, both analytically and numerically 12, but no unified theory has been presented so far. Here we study the extreme case of zero density (that is, the single magnetic polaron), the Wigner crystallization of magnetic polarons due to Coulomb interactions, and its thermal and quantum melting into a polaronic Fermi liquid and a ferromagnetic metal. We argue that the phase separation instability of the non-interacting DEM at low densities is replaced by Wigner crystallization and a polaronic Fermi liquid, stabilized by electrostatic interactions. We also show that the mean-field approach breaks down in the low density regime and that electron-electron interactions have to be taken into account properly.

The Model. In the presence of the Coulomb term, the DEM reads (we use units such that ℏ = 1 = kB):

$$\mathcal{H}_{DE} = - \sum_{(i,j)} t_{ij} a_i^\dagger a_j + \text{h.c.} + e^2 \sum_{i>j} \frac{(n_i-n_e)(n_j-n_e)}{|r_i-r_j|},$$

where $a_i$ ($a_i^\dagger$) is the annihilation (creation) operator for an electron on site $r_i$ with its spin aligned in the direction of a classical localized spin $S_i = S(\sin \theta_i \cos \phi_i, \sin \theta_i \sin \phi_i, \cos \theta_i)$, $\theta_i$ and $\phi_i$ being the local Euler angles. In Eq. (1), $t_{ij} = t|\cos (\theta_i/2) \cos (\theta_j/2) + \sin (\theta_i/2) \sin (\theta_j/2) e^{-i(\phi_i-\phi_j)}|$ is the hopping energy, $e$ the electric charge, $n_i = a_i^\dagger a_i$ the number operator, and $n_e$ the average number of electrons per atom. This model can be obtained from an interacting Kondo lattice Hamiltonian in the limit where the exchange interaction between local and electron spin, $J_H$, satisfies $J_H \gg t$. 13

The presence of the full Coulomb interaction in eq. (1) is to be understood in connection with the case of extremely reduced electron density. In a conventional Fermi
The magnetic polaron level (marked by an arrow) agrees with
for the homogeneous case (recursive method, continuous line).

Figure 2: Top: DOS for the 2-D spin configuration shown on
(bottom) and its underlying lattice spin configuration (bottom).

In a very diluted electron gas the Coulomb interaction
can be disregarded, and the problem reduces to a single magnetic polaron. We con-
sider the electronic density of states (DOS) for a given spin configuration, \( \{ S_i \} \), generated with a Boltzmann
weight that includes the mean-field required to obtain a given average magnetization, \( \mathcal{M} \), of the lattice spins \( \mathcal{N} \). Averaging over several realizations of disorder, we can write the electronic energy as:

\[
E_{el}(\mathcal{M}, n_e) = \int \Theta (E_F(n_e, \mathcal{M}) - \epsilon) e \langle N(\epsilon, \mathcal{M}) \rangle d\epsilon, \tag{2}
\]

where the explicit dependence of both the DOS \( N(\epsilon) \) and Fermi energy \( E_F \) on the magnetization is empha-
sized. For \( n_e \ll 1 \) we approximate eq. \( \text{eq.} \) simply by
\[ E_{el}(\mathcal{M}, n_e) \approx E_b(\mathcal{M}) n_e, \]
with \( E_b \) representing the bottom of the band that, in 3-D, is found to lie at \(-4t\) for the paramagnet \( (\mathcal{M} = 0) \), and at \(-6t\) in the fully polarized ferromagnetic phase \( (\mathcal{M} = 1) \). Let us assume
that the polaron consists of a cubic volume of side \( R \) (in units of the lattice parameter, \( a \)), inside which \( \mathcal{M} = 1 \). Outside the polaron the system remains paramagnetic \( (\mathcal{M} = 0) \). In Fig. \( \text{Fig.} \) we show exact diagonalization results of a two-dimensional ferromagnetic region embed-
ded in a paramagnetic phase, leading to the formation of a bound state, that is, a magnetic polaron \( \mathcal{M} \). The free energy per electron, at a temperature \( T \), required to form a magnetic polaron from the paramagnetic phase is given by:

\[
\Delta F_{pol}(R, T)/n_e = 4t - 6t \cos(\pi/(R + 1)) + TR^2 \log(2S + 1) - TS_{cfg}(n_e, R), \tag{3}
\]

where \( S_{cfg} \) is the configurational entropy. The first three terms in \( \text{eq.} \) reflect the two competing effects at play: the first is the electron's preference for a ferromagnetic back-
ground, accompanied by an energy cost for localization; the second is the reduction of entropy caused by the ap-
ppearance of the (fully polarized) magnetic polarons. For \( n_e \ll 1 \) we find that \( S_{cfg}(n_e, R) \) is always negligible \( \mathcal{M} \). Minimizing eq. \( \text{eq.} \) \( (R \gg 1) \) with respect to \( R \) gives:

\[
R_{eq}(T) \simeq \left[ 2t \pi^2/(T \log(2S + 1)) \right]^{1/5} \tag{4}
\]
for the equilibrium radius of the magnetic polaron, increasing at low temperatures as $T^{-1/5}$. The stability condition for the polaron is $\Delta F_{\text{pol}}(R_{\text{eq}}, T) < 0$, which provides the temperature, $T_m$, below which the polaron first appear:

$$T_m/t = 8\sqrt{2}/(25\sqrt{5}\pi^3 \log (2S + 1)).$$

(5)

Note that at $T = T_m$ the polaron are formed with a size given by $R_{\text{eq}}(T_m)$, and hence, the crossover from the paramagnetic to the polaronic phase is discontinuous.

The previous results can also provide an estimate of the temperature below which a homogeneous ferromagnetic state is established. Assume that each electron forms a cluster, which evolves continuously from the percolation threshold. This percolation threshold is defined from the criterion $n_c R_{\text{eq}}^3 \approx p_c$ ($p_c \approx 0.31$ for a cubic lattice), yielding from (4) a Curie temperature,

$$T_C^p/t \approx 2\pi^2/[\log (2S + 1)] (n_c/p_c)^{5/3}.$$

(6)

This temperature can be compared with the Curie temperature obtained by a mean-field approach: $T_{C}^{MF}/t = 5(1 + S)/(3S)n_c$. Notice that, for $n_c \to 0$, we have $T_C^p \ll T_C^{MF}$, indicating that the mean-field calculation overestimates the Curie temperature in the low density regime. For $T_C < T < T_m$, an anomaly in the paramagnetic susceptibility is expected to signal the polaron presence, through an enhanced effective moment. Naively, the transition from the polaronic phase to the ferromagnetic phase would be first order, for the magnetization jumps at $T_C$. Nevertheless, because this transition is percolative in nature, one would expect a second order phase transition, in the sense that the average magnetization should be weighted by the “mass of the infinite cluster”, which evolves continuously from the percolation threshold.

**Polaronic Wigner Crystal.** The previous estimate of the Curie temperature for a small but finite density of electrons has a fundamental problem: the non-interacting DEM is unstable towards phase separation at low densities [13, 15, 20, 21, 22, 23]. In the presence of Coulomb interactions, however, the phase separation is frustrated by the large electrostatic energy price of confining charge to a given region of the system. It is found that, for reasonable values of the dielectric constant, the tendency for charge neutrality, which favors small radius of electron rich regions, and the kinetic energy of localization, lead to a strong suppression of the phase separation region, in temperature and electron concentration, and its replacement by a polaronic phase [17].

At the lowest densities ($r_s \gg 1$) the Coulomb interaction is unscreened and larger than the kinetic energy in (1). In order to minimize the Coulomb energy, the electrons form a Wigner crystal and undergo zero point motion around their equilibrium position. Delocalization of the electron wavefunction due to quantum fluctuations leads to the polarization of the local spins around the crystalline positions. The local polarization, exactly as in the case of the single polaron discussed previously, produces an extra confining potential to the electron. Following the Wigner-Seitz approximation [13], the Wigner crystal unit cell (much larger than the original lattice spacing $a$) is approximated by an electrically neutral spherical volume, inside which the atomic charge density is homogeneous. The electrostatic potential energy then depends only upon $r$: the distance of the electron from the center of the cell. The Hamiltonian for an electron in this uniform charge and spin background is then:

$$H_0 = -6t - 3\frac{e^2}{2r_o} + \frac{p^2}{2m} + \frac{1}{2}m(\omega_e^2 + \omega_s^2)r^2, \quad (7)$$

where $p$ is the electron momentum, $m = 1/(2a^2t)$ the effective electron mass, and $\omega_e^2 = \omega_s^2 = e^2/mr_o^2$, where $\omega_s$ is the plasma frequency. In Eq. (7) $\omega$ is the frequency of the confining potential due to the DEM mechanism, and is a variational parameter. The radius of the magnetic polaron in the Wigner crystal relates to $\omega$ by:

$$R = \sqrt{3t/\Omega},$$

(8)

where $\Omega = \sqrt{\omega_e^2 + \omega_s^2}$ is the total frequency of oscillation of the electron. Notice that the ground state energy of (7) is $E_0 = -6t - 3e^2/(2r_o) + 3\Omega/2$ and hence the relative gain in free energy is:

$$\Delta F_{W} = -2t + 3(\Omega - \omega_c)/2 + 4\pi R^3T \log (2S + 1)/3.$$ (9)

Minimization of eq. (9) with respect to $\omega$ gives:

$$R(T) = \left\{ \begin{array}{ll} R_S & (T/T^*)^{1/5}, \quad T > T^* \\ R_S & , T \leq T^* \end{array} \right. \quad (10)$$

where $R_S = [3t/(\omega_0)]^{1/5}$ is the saturation radius, and $T^*/t = 9/[4\pi R_S^3 \log (2S + 1)]$ is the temperature below which the polaron radius saturates due to the interplay between the DEM and the Coulomb interaction.

**Wigner Crystal Melting.** It is clear that the previous results are valid for temperatures so low as not to melt the Wigner solid. The calculations for the independent polaron model reveal that the temperatures for polaron stability are already typically small for reasonable values of $t$, but the electronic solid is much more sensitive to the temperature. The Wigner crystal melting temperature, $T_M$, can be estimated from the Lindemann’s criteria [24]: $T_M \approx 0.01(e^2/\mu a)^{1/3}$. It is known from several numerical calculations [24, 27, 28] on the stability of the one component plasma, that the maximum densities and temperatures at which the Wigner crystal can exist correspond to $r_s \approx 50-100$, and $T \approx 10$ K. Values of $r_s \approx 50-100$ correspond to $n_e \sim 10^{-6} - 10^{-5}$ for $t = 1$ eV and $a = 4$ Å. Because of the absence of magnetic interactions between different polarons, the Wigner crystal is a superparamagnet. In the presence of other long-range...
interactions (such as dipole-dipole) the polaronic Wigner crystal can exhibit long range magnetic order. Increasing the electron density at $T = 0$ causes the Wigner crystal to quantum melt at some critical density with two possible outcomes: a paramagnetic polaronic Fermi liquid or a fully polarized ferromagnet. In both cases the carriers are mobile and can screen the long-range part of the Coulomb interaction leading to a Fermi liquid state. At finite temperatures, where the electron state cannot be described by the zero point motions implicit in eq. (7) alone, the crystal should follow the features of the phase diagram for the electron gas [6]. The characterization of the system in the neighborhood of the melting point, where the presence of a polaron liquid is plausible (Fig. 1), is restrictively hard, even for the simple electron gas. Far from this region, where the electron density is high enough to make the screening process effective, one expects to retrieve the behavior obtained before within the independent polaron model discussed previously. Our results are summarized in the phase diagram presented in Fig. 1.

**Application to EuB$_6$.** While we have produced the general phase diagram for the DEM model at low densities, it is interesting to apply our results to a real system. EuB$_6$ is a good magnetic metal with extremely reduced electron density [27], exhibiting all the characteristic signatures of a polaronic phase in Raman scattering. Such experiments reveal that the FM transition at $T_{m}^C \approx 15$ K is preceded by an interval of temperatures ($T < T_{exp} \approx 30$ K) where the system is dominated by the presence of magnetic polarons. We have recently proposed that EuB$_6$ is a DEM material in the low density regime [16], characterized by a hopping integral $t = 0.55$ eV, and a carrier density per unit cell $n_c \sim 10^{-3}$ (which, according to Fig. 1, puts EuB$_6$ far away from the WS). If we incorporate such values in our previous results (see Fig. 1) we find $T_m \approx 43$ K and $T_{m}^{P} \approx 15$ K, in accordance with the experimental data (notice that the mean-field theory predicts much higher values for $T_{m}^{MF}$, in complete disagreement with the experiments). These results show that our description captures the polaron physics in EuB$_6$.

**Conclusions.** We have studied the DEM at low densities in the presence of Coulomb interactions and shown that its phase diagram is very rich. We find that at extreme low densities the system does not phase separate but, instead, forms a Wigner crystal of magnetic polarons. Due to thermal or quantum effects, this crystal melts into a polaronic Fermi liquid or a metallic ferromagnet. We find that the polaronic liquid is stable in an intermediate temperature range $T_{C}^{P} < T < T_{m}$. Above $T_{m}$ the polaronic liquid evolves into a paramagnetic metal while at low temperatures, $T < T_{C}^{P}$, it percolates into a metallic ferromagnet. Our theory has only two free parameters, the number of electrons per site, $n_e$, and the hopping energy $t$, and should apply to a broad class of materials that are described by the diluted DEM. In particular, our results provide a good description of the physics observed experimentally in EuB$_6$.

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