Stability and dynamics of free magnetic polarons

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The stability and dynamics of a free magnetic polaron are studied by Monte Carlo simulation of a classical two-dimensional Heisenberg model coupled to a single electron. We compare our results to the earlier mean-field analysis of the stability of the polaron, finding qualitative similarity but quantitative differences. The dynamical simulations give estimates of the temperature dependence of the polaron diffusion, as well as a crossover to a tunnelling regime.

''Colossal'' magnetoresistance (CMR) arises in several different classes of materials where the electrical transport is strongly coupled to the degree of ferromagnetic order, and may arise from different mechanisms. In the perovskite manganites[1] there is evidence that CMR is intimately linked to strong *lattice* polaronic effects in these high carrier density materials. In low carrier density magnets, spin scattering alone can be a dominant contributor to CMR, as in the manganite pyrochlores[2,3] and EuB_6.[4] Strong exchange coupling between the carrier and local moments is then expected to give rise to magnetically self-trapped carriers,[5–7] for which experimental evidence can be gleaned from transport measurements.[3] These entities are free magnetic polarons (FMP) in contrast to the more common bound magnetic polarons (BMP) where a carrier is trapped by an impurity and the local magnetization is a secondary phenomenon. When a magnetic field is applied, the ferromagnetic clusters overlap leading to delocalization of carriers and, consequently, to negative magnetoresistance.

Magnetic polarons have been also found in systems other than CMR materials. BMP have been extensively studied in diluted magnetic semiconductors[8] and rare earth chalcogenides.[9] Golnik *et al.*[10] found experimental evidence of the existence of bound and free magnetic polarons in $Cd_{1-x}Mn_xTe$ and $Pb_{1-x}Mn_xTe$. Magnetic polarons can also explain the temperature-dependent spin splitting seen in magneto-optical experiments.[11] Theoretical models of FMP have been developed within a mean-field approach[12–14] and generalized to a fluctuation-dominated regime.[15] In most of these systems, the underlying (super)-exchange interaction between the localized spins is antiferromagnetic in nature; we shall however be concerned with the ferromagnetic case relevant to manganese pyrochlores and EuB_6. These systems also have very low carrier concentration.

Hence we study the model of a single electron interacting with a spin background that itself is ordering ferromagnetically.[7] We consider the Hamiltonian:

$$H=-t\sum_{\langle i,j\rangle\sigma}c^{\dagger}_{i\sigma}c_{j\sigma}-J'\sum_{i}\vec{\sigma}_i\cdot\vec{S}_i-J\sum_{\langle i,j\rangle}\{(1-\alpha)\times(S^x_i\cdot S^x_j+S^y_i\cdot S^y_j)+S^z_i\cdot S^z_j\}, \quad (1)$$

where $\vec{S}_i$ refer to the spin of the magnetic ions in the system. $c^{\dagger}_{i\sigma}$ creates an electron with spin σ on the site i, $\vec{\sigma}_i=c^{\dagger}_{i\alpha}\vec{\sigma}_{\alpha\beta}c_{i\beta}$ is the conduction spin operator and $\langle i,j\rangle$ denotes sum over the nearest-neighbors pairs. We have added to the Heisenberg term a small Ising anisotropy $\alpha=0.1$ to improve convergence at low temperatures and by enforcing a nonzero transition temperature T_c in bidimensional systems.[16] J' is the coupling between a localized spin and a conduction electron. The qualitative behavior is well understood from previous mean-field analyses.[7,12,14] Below a temperature T_p a ferromagnetic polaron forms by self-trapping in a ferromagnetically aligned cluster of spins. As the temperature is lowered toward the Curie temperature T_c the polaron grows in size and becomes more stable, because the small-q magnetic susceptibility is growing. Near and below T_c the polaron will again become unstable because of the ease of motion in the background ferromagnetic spin alignment. Notice that this is quite different from the case of *anti*-ferromagnetic coupling of spins, where the polaron may remain stable well below the magnetic ordering temperature.

There are several deficiencies of the mean field treatment. The most pronounced is a continuum treatment of the spin background where fluctuations are neglected. This approximation is such that the paramagnetic state leads to a vanishing exchange coupling, so that the electron is bound in a potential of depth $J'\overline{S}$, with $\overline{S}$ the average magnetization inside the polaron. As $J'/t\to\infty$, the potential well becomes arbitrarily deep. This is undoubtedly a severe overestimate.

In the paramagnet there will always be low energy states localized in the band tail[17] with energies $O(t)$ above the ferromagnetic ground state, even in the strong coupling limit. Such low energy states are produced by random fluctuations of a few neighboring spins into near-alignment. But now one must distinguish between a self-trapped polaron and a localized band-tail state, if indeed such a distinction is appropriate.

In this paper we address the topic by a dynamical simulation of the Hamiltonian of Eq. (1). We show that polarons may be distinguished (when they exist) by a spectroscopic gap to bandlike states, and that they move diffusively. As temperature is raised, the polaron level moves toward the band edge, and begins to resonate with states in the band tail, leading to a crossover to hopping conductivity.

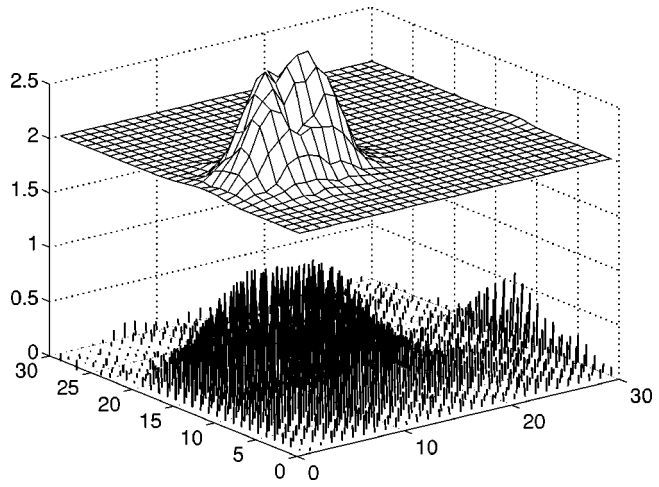

FIG. 1. $|\psi(x,y)|^2$ and the averaged magnetization $\vec{m}_i = \langle \vec{S}_i \rangle / |\vec{S}_i|$ for $J'/t = 5$ and $T = 1.1T_c$ are plotted.

We perform a classical Monte Carlo simulation (MC) in two dimensions on a square lattice of localized spins $\vec{S}_i$ which are treated as classical rotors characterized by the angles θ_i and ϕ_i. We use periodic boundary conditions on a two dimensional lattice of size up to 30×30.

We place a single electron in the system in the lowest energy eigenstate of the Hamiltonian consisting of the first and third terms of Eq. (1), using the instantaneous spin configuration for the classical spins $\vec{S}_i$. The resulting wave function leads to a local magnetic field proportional to $|\psi(x,y)|^2$ (see, for instance, Ref. 8), used in the next step of the MC spin simulation. Hence $\psi(x,y)$ is calculated self-consistently.[18]

The standard Metropolis algorithm is used. Randomly chosen sites suffer a random change of spin orientation. Changes are allowed if the increment in energy ΔE is such that the quantity $\exp(-\Delta E/KT)$ is smaller than a random number between 0 and 1. 4000 reorientations per spin were made for an initial equilibration and 3000 to calculate averages after each diagonalization. Each diagonalization defines our time step. Changing the number of spin reorientations between each diagonalization led to no significant change in either the magnetization or binding energy. All the quantities are given in units of J, the Heisenberg parameter. The hopping parameter t is fixed to 100 (estimated with the mean field relation $T_c \sim zJS^2$ and the values for the parameters expected for the pyrochlores[7]) as we are interested in the behavior versus J'/t and the temperature T. $T_c = 1.8$ in these units. Note that we have a T_c in our two-dimensional system due to the anisotropy in the Heisenberg parameter.[16] This help us to find a quick convergence but does not change our conclusions.

This approach allows us to calculate in a self-consistent way the wave function and the magnetic polarization over a large range of temperature. In particular, we can explore the region around T_c where the mean-field treatment fails.

In Fig. 1 we plot $|\psi(x,y)|^2$ and the averaged local magnetization close to T_c. Visually, the existence of a magnetic polaron is clear, and there is substantial alignment of the moments in the vicinity of the carrier. Note that far from the influence of the wave function the average magnetization is close to 0, so a large part of the spin configurations are explored, while for the spins close to the center of the wave function there are few accepted spin flips.

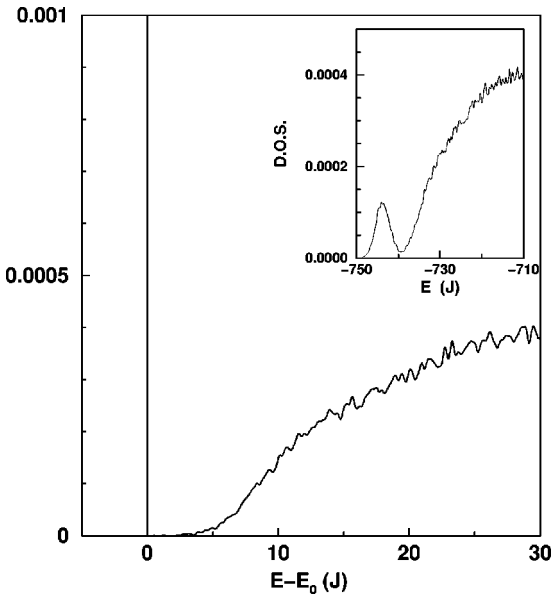

FIG. 2. The density of states (inset) and excitation spectrum at low energies for $J'/t = 5$, $T = 1.1T_c$, and $|\vec{S}| = 3/2$ is plotted.

Pictorial evidence is purely qualitative, and does not allow one to extract reliable estimates for the polaron size or binding energy, especially at higher temperatures and lower J'/t, when the polarons are smaller and fluctuating in time. More reliable evidence comes from the time-averaged electronic density of states (DOS), and of the excitation spectrum shown in Fig. 2. In the density of states (inset) a sharp low energy feature is pulled from the bottom of the band (only the lowest 1% of the spectrum is shown) that contains exactly one state. This is the bound polaron level. The level width comes from thermal fluctuations in the energy of the bound state, and the stability of the bound polaron is seen more clearly in the excitation spectrum (main figure) that demonstrates a clear gap corresponding to the electronic part of the binding energy E_p of the polaron.

The continuum of excited states can be characterized as the band tail formed by the fluctuating paramagnetic background; the lowest energy states are produced by rare fluctuations of nearby spins into near-alignment. Consequently, the ''gap'' in the excitation spectrum is soft, and indeed statistically very rare states may occur at energies *below* the bound state of the polaron. We will discuss this below.

We estimate the electronic binding energy E_p by the configurationally averaged gap $\Delta = E_1 - E_0$ to the lowest excited state in our simulations (we have checked that the separation between excited states scales as $1/N^2$ so that these are true continuum states). In Fig. 3 we show the dependence of Δ and the absolute value of the local magnetization M (weighted with the wave function) on T and J'/t. M is defined as

$$M = \left\langle \left| \sum_i \vec{S}_i' \right| \right\rangle,$$

where $\vec{S}_i' = |\psi(i)|^2 (\vec{S}_i / |\vec{S}_i|)$.

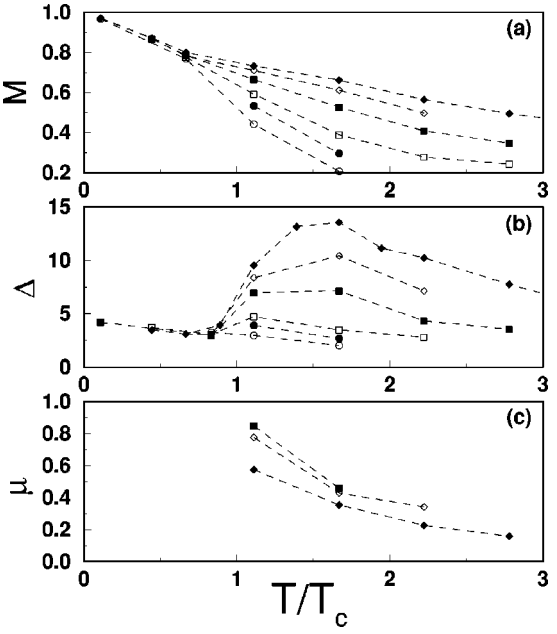

FIG. 3. We plot here the dependence on J'/t and temperature of the local magnetization in the lattice produced by the polaron (a), its binding energy (b), and the mobility $\mu = D/T$ (c) for $|\vec{S}| = 3/2$. The different curves correspond to different J'/t such that its value is 1 for open circles, 1.5 for closed circles, 2 for open squares, 3 for closed squares, 4 for open diamonds, and 5 for closed diamonds.

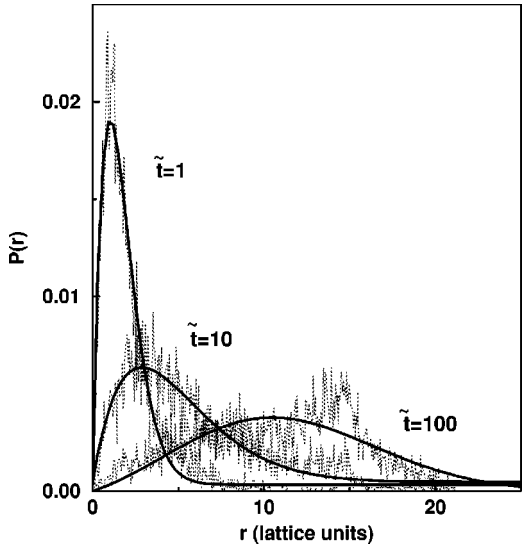

FIG. 4. The probability of moving a distance r for different times, $\tilde{t} = 1$, 10, and 100, taken from a single run, is shown. Solid lines show fits to a 2D Gaussian, plus an offset. This background is due to the rare appearance of FM clusters away from the polaron location, where the polaron hops a large distance. The curves for $\tilde{t} = 10$ and $\tilde{t} = 100$ are the result of iterations of the $\tilde{t} = 1$ curve; thus the rare long-distance hops eventually dominate the distribution, as can be seen in the trace at the longest times.

We are not taking into account thermal excitations of the quasiparticle so our results are valid only when Δ is bigger than T. This condition is fulfilled for all the values of Δ shown in Fig. 3. As expected from previous analyses, the polaron binding energy increases as temperature is lowered from high temperatures, as the thermal spin fluctuations are reduced. For large J'/t we find a new behavior on Δ not found within mean-field theory, namely, that it has a maximum at some temperature above T_c. The existence of a maximum can be understood in terms of the correlation length ξ. This quantity increases as we decrease the temperature above T_c. For very small ξ (large T) is very difficult to have a FM cluster for the spin-polaron to sit in and for large ξ the electron would rather spread out. This will lead to an intermediate optimum ξ for the existence of the polaron that would happen close to T_c but not necessarily at T_c.

The size of the polaron may be estimated from the separation between the first eigenvalue E_o and the bottom of the band of the uniform ferromagnet. The bottom of the band in this case is given by $-\frac{3}{4}J' - 4t$ and the separation should go roughly as $1/L_p^2$ being L_p the size of the polaron if we assume saturation in the local magnetization. The general trend is that the size decreases as T (above T_c) or J' increases. From Fig. 3 we can also deduce that the ''window'' above T_c where the spin polaron is stable increases with J'/t. These two results are consistent with previous mean-field calculations.[7]

Although qualitative comparison is satisfactory there are large quantitative differences that point to a great decreasing in the stability of the spin polarons when fluctuations are taken into account. To be precise we compare the binding energy at $T = T_c$. From mean-field calculations on Ref. 7 the maximum possible value for Δ is $\sim J'S$ but it is not reached due to the kinetic energy that is lost with the formation of a polaron. In the present work $\Delta/J \sim 1$ while $J'S/J \sim 100$. So binding energies are reduced by two orders of magnitude compared with the mean-field results because the loss of kinetic energy is not well taken into account in the latter.

The study of the stability conditions for a free magnetic polaron is interesting by itself but the MC simulation also opens us the possibility of learning about its dynamics in a spin-fluctuating landscape. In Fig. 4 the probability of moving a distance r (defined as the change in the expectation value of the electron position) for different MC times is shown. For time $\tilde{t} = 1$ one observes dominant short distance motion with occasional rare hops over long distances. For longer times, the peak of the distribution moves out approximately with $\sqrt{\tilde{t}}$ as expected. This is the expected behavior from a diffusing object. The long-distance hops occur when unoccupied band tail states (which may be localized anywhere in the system) temporarily drop below the bound polaron level. In our algorithm—which automatically populates the lowest energy level—the electron moves to occupy this new state and restabilises the polaron there. These rare events eventually dominate the long-time behavior in our simulations. Of course, very long range hops are unphysical because the tunnelling probability will be exponentially small with distance, and the band-tail states survive in one place for only a short time. Hops to band-tail states will then be limited to some finite range. As temperature is raised, and E_p is reduced, hops to band tail states become more frequent; we cross over to a regime of ''passive advection'' of the wave function in the fluctuating spin background.[19]

Our results are fitted to a Gaussian in two dimensions plus a constant (to approximately take account of hops to band-tail states). The Gaussian dominates for the parameters of interest when a spin-polaron is well formed. The distribution

scales with $\sqrt{\tilde{t}}$ as expected for diffusive motion. Hence we calculate the diffusion constant as $D=\Sigma_i P(r,\tilde{t}=1)r^2$ and the mobility ($\mu=D/T$) of the spin polaron for different couplings and temperatures (see Fig. 3). The mobility decreases with temperature, and also with J'/t. The latter is reasonable, because larger polarons should diffuse more slowly. The temperature-dependence is more surprising, and arises because D itself is weakly T dependent. Although the polaron size is decreasing with temperature (tending to increase D), this is counterbalanced by a reduced probability of favorable FM spin configurations near its boundary as T/T_c is increased.

The Heisenberg term has been considered ferromagnetic to compare with the pyrochlores. The change to antiferromagnetic coupling is straightforward and in fact the more common case in manganite perovskites,[1] rare earth chalcogenides,[9] or magnetic semiconductors.[8] In the case of an antiferromagnetic background, we find that the stability of a free magnetic polaron is enhanced. These results will be reported elsewhere. These results are consistent with Ref. 20 where a pseudogap in the DOS is associated with phase separation, that is the large scale effect corresponding to spin polarons.

In conclusion, our dynamical simulations have revealed a picture of the FMP in a ferromagnet above T_c which is considerably more complex than given by the mean field pictures. Provided the exchange coupling is large enough, FMP's are stable above T_c, but considerably more weakly bound than found by mean field calculations. This by itself raises some doubts about the interpretation given earlier for the Mn pyrochlores, because we require an exchange coupling comparable to the bandwidth for a well-formed polaron with nearly saturated magnetization, whereas in the Mn pyrochlores this coupling is expected to be not large.[7] We find that the motion of the polaron is diffusive, but as temperature is raised the electron fluctuates out of the self-trapped configuration into band-tail states formed by opportunistic fluctuations of the moments.

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