Resonant Pair Exchange and Percolation in the Disordered Hubbard Model

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We show that the effect of disorder on a Mott-Hubbard insulator of interacting electrons produces a quantum phase transition from an antiferromagnetic to a paramagnetic phase governed by a percolation of the singly occupied weakly disordered sites. Near the transition, we propose that a new type of defect, formed by the resonant exchange between a spin singlet and a doubly occupied site with an attractive disorder potential, plays an important role. These resonant pair exchange defects reduce the staggered magnetization but enhance the coupling of the two spins and produce characteristic signatures in the temperature dependent specific heat and the non-Curie spin susceptibility, at temperatures on the order the hopping $t$, higher than the typical exchange scale $J$.

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Recent theoretical and experimental studies show that the interplay of random disorder with correlations in electronic systems can produce striking effects. Such studies have largely focused on the metal insulator transition\textsuperscript{1,2,3}, where the problem still remains open from the theoretical perspective. Also of great interest is the question of magnetism in strongly correlated disordered systems. Besides the destruction of magnetic long range order with increasing disorder, we show in this Letter that disorder induces new types of magnetic coupling, absent in the ordered system.

We address these questions by studying the effect of disorder on the magnetic properties of the Hubbard model. Besides its importance as a theoretical paradigm, this model is also relevant for the high $T_c$ superconductors, where mobile carriers are produced by stoichiometric doping that is also a source of quenched disorder. At half filling, in the absence of disorder, the model describes a Mott insulator with a spin at every site, and antiferromagnetic (AF) coupling of order $J \sim t^2/U$ between spins at adjacent sites. We show that in the presence of disorder, it is also possible to have a novel type of magnetic coupling that we call resonant pair exchange (RPE), which operates between particular pairs of sites. This interaction is the outcome of the resonance to order $t$ between two configurations of spins on adjacent sites: (a) a spin singlet formed by single spins on adjacent sites and (b) a non-magnetic doublon formed by a doubly occupied and empty pair due to a binary potential close to $(U/2)$ and $(-U/2)$ at two nearest neighbor sites. This sort of ‘defect pair’ occurs with a finite probability in a disordered system, and has several important characteristics: (i) Similar to two-level systems in glasses, these defects produce a characteristic maximum in the specific heat. (ii) The staggered spin susceptibility is suppressed because of the mixing of the singlet configurations with a non-magnetic configuration. However, interestingly, the non-Curie behavior persists to temperatures $T \sim t$, which is much higher than the kinetic exchange scale $J \sim t^2/U < t$. (iii) The resonant tunneling produces high kinetic energy on the bond connecting the two sites. This provides a source of noise in RPE defects that should be trackable in conductance noise experiments\textsuperscript{4}.

These RPE defects are most active near the antiferromagnetic (AF) to paramagnetic (PM) transition driven by increasing disorder. Strong disorder generates two types of sites: non-magnetic unoccupied or doubly occupied sites, and magnetic sites with a single spin\textsuperscript{5}. We find that a percolation-based description then becomes possible, as electron hopping results in coupling between neighbouring magnetic sites; increasing the disorder leads to a decreasing number of magnetic sites, and eventually to a transition marking the loss of long range AF order. We supplement this with other calculational approaches, including an inhomogeneous Hartree Fock (HF) calculation whose results are found to agree remarkably well with quantum Monte Carlo simulations\textsuperscript{6,7}.

Model: We start with the repulsive Hubbard model on a two-dimensional square lattice with site disorder:

$$H = -t \sum_{\langle ij \rangle, \sigma} (c_{i\sigma}^\dag c_{j\sigma} + c_{j\sigma}^\dag c_{i\sigma}) + U \sum_i n_{i\uparrow} n_{i\downarrow} + \sum_{i\sigma} (V_i - \mu) n_{i\sigma}$$

(1)

where $c_{i\sigma}^\dag (c_{i\sigma})$ is the electron creation (annihilation) operator with spin $\sigma$ on site $i$ and $n_{i\sigma} = c_{i\sigma}^\dag c_{i\sigma}$ is the corresponding density operator. The first term describes hopping of electrons between nearest neighbors with amplitude $t$. The second term is the repulsive on-site interaction for doubly occupied sites with strength $U$, $\mu$ is the chemical potential that determines the filling and $V_i$ is a random potential chosen from a uniform distribution between $-V$ and $V$. 

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The Large $U$ limit: In the atomic limit $t = 0$, the competition between repulsive interactions and disorder produces site-dependent integral occupancies $n_i$ in the ground state. Sites with $V_i > U/2$ are unoccupied; and those with $V_i < -U/2$ are doubly occupied; neither of these two types of sites has a free spin. On the other hand, sites with $|V_i| < U/2$ have $n_i = 1$ with a free spin residing on each such site. Thus the fraction of randomly placed singly-occupied (magnetic) sites is $x = U/2V$, while the remaining sites are nonmagnetic.

In the atomic limit, the spin degeneracy of the ground state is $2^N$, where $N$ is the total number of sites. The effect of turning on a small hopping amplitude $t$ is to lift this degeneracy and produce an AF coupling of magnitude $J_{ij}$ between nearest-neighbor (nn) singly-occupied sites $i$ and $j$, by the well-known mechanism of kinetic exchange. To second order in $t$, the coupling $J_0 = 4t^2/U$ in the pure system is modified by the disorder difference $\delta V = V_i - V_j$ on the two sites to

$$J_{ij} = \frac{J_0}{1 - (\delta V)^2/U^2}.$$  (2)

The effective leading-order Hamiltonian is then $\mathcal{H} = \sum_{(ij)} J_{ij} \sigma_i \cdot \sigma_j$, where $J_{ij}$ is nonzero only if both sites are magnetic.

Resonant Pair Exchange: It is evident from the above analysis that there will be some rare regions where the disorder at a pair of neighbouring magnetic sites $V_i$ and $V_j$ is such that the conditions $|V_i - V_j - U| < t \ll U$ and $|V_{ij}| \approx U/2$ hold. In that case, the denominator in Eq. (2) becomes very large, and the perturbative expression is no longer valid. In fact, in this regime the electron hopping couples these pairs of sites to first order in $t$, and we show that it induces a new type of coupling, which we call resonant pair exchange (RPE). The RPE process differs qualitatively from normal kinetic exchange, and has important consequences for the thermodynamic and transport properties of the system.

Consider states with 2 electrons on a pair of sites characterized by disorder parameters $V_1$, $V_2$, when the hopping $t = 0$. Of the total of 6 states, there are 3 singlet ($S = 0$) states $\{|1,1\}_S$, $\{|2,0\}$, $\{|0,2\}$, and 3 triplet ($S = 1$) states $\{|\uparrow,\uparrow\}$, $\{|\downarrow,\downarrow\}$, $\{|1,1\}_t$. Since the Hamiltonian conserves total spin $S$, we examine each subspace separately. Of the three singlet states, two states $\{|2,0\}$ and $\{|0,2\}$ involve unequal charges at each of the two sites, whereas one state $\{|1,1\}_S = 1/\sqrt{2}(|\uparrow,\downarrow) - |\downarrow,\uparrow\rangle)$ involves one electron on each site. We are interested in the case when one of the two unequal-charge states is nearly degenerate with $\{|1,1\}_S$. For specificity, let us take $V_1 = -V_2 = U/2$. Then 5 of the six states (the 3 triplet states and 2 singlet states $\{|1,1\}_S$ and $\{|0,2\}$) are degenerate with energy $-U$, while $\{|2,0\}$ has energy $U$. The primary effect of nonzero but small hopping is to mix the two degenerate singlets, and produce the eigenstates

$$|\psi^\pm\rangle = \frac{1}{\sqrt{2}} \left( |1,1\rangle \pm |0,2\rangle \right),$$  (3)

with eigenvalues $-U - \sqrt{2}t$ and $-U + \sqrt{2}t$ respectively. The resulting pattern of energy levels is as shown in Fig. 1a.

In the case $V_1 = V_2 = 0$, the states $\{|2,0\}$ and $\{|0,2\}$ have a high energy $U$, while the hopping lowers the energy of $\{|1,1\}_S$ with respect to the triplet state by an energy of order $t^2/U$ (Fig. 1b). This is the mechanism of kinetic exchange, which produces AF correlations in the dimer ground state that persist up to $T \sim t^2/U$. With resonant pair exchange, by contrast, the AF correlations in the ground state are reduced (as the non-magnetic state $\{|0,2\}$ is mixed in); but these correlations persist to much higher $T \sim t$ than in the case of kinetic exchange. As a consequence, both the uniform susceptibility $\chi(q = 0)$ and the ordering susceptibility $\chi(q = \pi)$ show deviations from Curie (free moment) behaviour for $T < t$, signaling the onset of AF correlations due to resonant pair exchange, as shown in Fig. 1b. Similar magnetic effects are expected whenever the conditions $|U + V_i - V_j| \leq t$ and $|V_{ij}| \approx U/2$ are satisfied for neighboring pairs of sites $\{ij\}$. The fraction of such pairs in the disordered Hubbard model is of the order of $(t/2V)^2$. As the contribution to energy lowering from each pair is $t$, the overall contribution to the ground state energy is of order $t^3/V^2$, which is of higher order in $t$ than from the majority of pairs, which are coupled by normal kinetic exchange.

![Fig. 1: Eigenstates for two sites. (a) Resonant pair exchange: With $V_1 = -V_2 = U/2$, the difference in energy between the RPE singlet ground state and triplet excited state is $\Delta \approx t$. (b) Kinetic exchange: With $V_1 = V_2 = 0$; the energy difference between the magnetic singlet ground state and the triplet excited state is $\sim J \sim t^2/U$.](image-url)
splitting. In this regime the total specific heat is from RPE sites, due to their large energy for this two level system is obtained by substituting \(0\) and \(V\) for \(\Delta\) in Eq. 4, since \(\Delta \propto t^2/U\). In the low-\(T\) regime, \(C_v\) decays exponentially as \(T \to 0\); this form of the decay found within the pair approximation would change if the system supports extended (spin wave like) states.

Specific Heat and Spin Susceptibility: Since sites coupled by resonant pair exchange are relatively rare and have a very different level structure (see Fig. 1) from the majority of pairs, they act as localized centers and give rise to a distinctive signature in the specific heat \(C_v\) and \(\chi_{avg}\), much as two level centers do in glasses. For an arbitrary pair, the energy splitting \(\Delta\) between the singlet ground state and the triplet states is

\[
\Delta = \sqrt{2t^2 + \left(\frac{U - \delta V}{2}\right)^2 - \frac{U - \delta V}{2}} \tag{4}
\]

where \(\delta V = |V_1 - V_2|\). Here we have assumed the state with energy \(U + \delta V\) decouples from other two levels. If \(V_1\) and \(V_2\) are chosen from a uniform distribution between \(-V\) to \(V\) and we regard each pair as isolated from the others, the probability distribution for the splitting \(\Delta\) is given by

\[
P(\Delta) = \frac{1}{2V} \left[\frac{\Delta}{V} \left(\frac{4t^4}{\Delta^2} - 1\right) + \frac{2V - U}{V} \left(\frac{2t^2}{\Delta^2} + 1\right)\right]. \tag{5}
\]

The total specific heat from such pairs is \(C_v = \int_{\Delta_{min}}^{\Delta_{max}} d\Delta P(\Delta)c_v(\Delta)\) where \(\Delta_{min}\) and \(\Delta_{max}\) can be obtained by substituting \(0\) and \(U\) for \(\Delta\) in Eq. 4 since the integration is only over singly occupied sites. Noting that the average energy for this two level system is \(E(\Delta) = -\Delta \exp(\Delta/T)/[\exp(\Delta/T) + 3]\), the corresponding specific heat is \(c_v(\Delta) \equiv \partial E/\partial T\), shown in Fig. 3.

We see that at \(T \geq t\) most of the contribution to the total specific heat is from RPE sites, due to their large splitting. In this regime \(C_v\) varies as \(1/T^2\). On lowering \(T\), \(C_v\) has a peak at \(T = 2t^2/\lambda U\) with \(\lambda \approx 2.85\). Most of the peak weight comes from pairs with small splitting \((\Delta \propto t^2/U)\). In the low-\(T\) regime, \(C_v\) decays exponentially as \(T \to 0\); this form of the decay found within the pair approximation would change if the system supports extended (spin wave like) states.

The above arguments hold also for the averaged susceptibility. For the two level system mentioned above spin susceptibility is \(\chi_{avg}(\Delta, T) = 2/T[\exp(\Delta/T) + 3]\). The susceptibility averaged over pairs shows Curie behavior \((\propto 1/T)\) at high \(T\) and a peak at \(T \approx 2t^2/U\) (see Fig. 3): below this temperature, triplet states make a very small contribution to the susceptibility. As \(T\) tends to zero \(\chi_{avg}\) falls as \(\exp(-\Delta/T)\). Therefore the high temperature \((T \geq t)\) behavior of the specific heat and susceptibility is determined by the RPE sites while the low temperature behavior is governed by pairs of sites with small energy splittings.

Percolation of Magnetic Sites: Recall that in the atomic limit, a fraction \(x = U/2V\) of sites is magnetic with \(n_i = 1\), while the remaining sites with \(n_i = 0\) or 2, are nonmagnetic. Since only nearest neighbour magnetic sites are coupled if \(t\) is small, the system maps to a dilute magnet. A necessary condition for antiferromagnetic long range order (AFLRO) is that there be an infinite connected cluster of singly occupied sites, i.e. \(x > x_c\) where \(x_c\) is the site percolation threshold for a given lattice geometry. Thus in the limit \(t/U < 1\), we can rigorously rule out LRO for \(x < x_c\). Further, in this limit, the model reduces...
FIG. 4: Solid line is AF order parameter $m^\dagger$ vs $1 - x = 1 - U/2V$ using HF approximation for a 2D square lattice of the model mentioned in Eq. (1) of a system size $28 \times 28$, $U = 4$ and $T = 0$. The dashed and dotted lines are staggered magnetization vs dilution in the 2D quantum Heisenberg model with spin-1/2 using quantum Monte Carlo and spin wave theory. The square symbols are experimental data points using neutron scattering. The percolation threshold is $1 - x_c \simeq 0.41$.

to a dilute Heisenberg AF with random couplings. The question is then whether there is AFLRO for $x > x_c$. Since $J_{ij}$ in Eq. 2 is bounded below by $J_0$, and a dilute AF with uniform coupling between magnetic sites on a square lattice seems to show AFLRO, we expect AFLRO in the disordered Hubbard model with $t/U \ll 1$, provided that $V < U/2x_c$. The occurrence of a small number of resonant pairs in the infinite clusters would result in a slight loss of AF order in the ground state. However, if the temperature is much larger than $t^2/U$ but still of the order of $t$, only the resonant pairs would make a non-Curie contribution to magnetic properties.

Results of the Hartree-Fock approximation: The HF approximation, allowing for inhomogeneity in spin and densities on all the sites, sheds light on both magnetic and conducting properties of the disordered Hubbard model. Earlier work using this approximation focused on 3D and the effects of moving away from half filling. Here we focus on the case of half filling in 2D. We find that adding site disorder leads to closing of the Mott gap in the density of states (DOS), while AFLRO persists up to the percolation threshold of magnetic sites, $2V_{cr} \simeq U/0.59$ (Fig. 1). At an intermediate regime of disorder and interaction the system has metallic behavior. Paramagnetic sites start appearing only when $V > U/2$. For disorder strength $V > V_{cr}$ the system breaks into clusters of AF sites with no long range order, and displays a glassy behavior; the final state of self-consistent iterations depends on the initial inputs of the variational parameters of the trial HF Hamiltonian. With increasing disorder the size of AF clusters shrinks further, and at the limit of very large disorder the system is a paramagnetic Anderson insulator.

Figure 1 shows a good consistency of the staggered magnetization obtained within the HF approximation with results of other theoretical and experimental studies of the diluted Heisenberg model. The HF approximation captures rather subtle effects as well. In particular the next nn coupling, coming from a fourth order expansion in $t$ can compete with the nn coupling to produce occasional mismatches in the alignments of particular spins/clusters — and these are reproduced by HF treatment. Further for a honeycomb lattice at half filling the AFLRO vanishes at a disorder strength, predicted by the percolation picture.

Conclusion: There are two significant results in this paper: Firstly, we have identified a new type of disorder induced defect that involves pairs of binary potentials and produces a magnetic state with a reduced staggered magnetization, but with an enhanced coupling. Secondly, we have shown that the disordered Hubbard model at large $U$ can be mapped to a disordered diluted AF spin-1/2 quantum Heisenberg model. This allows us to connect the existence of AFLRO with a percolating infinite cluster of magnetic sites with $|V_c| < U/2$. We find a remarkable consistency between the percolation picture and HF results even for intermediate $U$.