Itinerant ferromagnetism in a spin-fermion model for diluted spin systems

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We investigate the itinerant ferromagnetism using a diluted spin-fermion model, derived from a repulsive Hubbard model, where itinerant fermions are coupled antiferromagnetically to auxiliary fields in a three-dimensional simple cubic lattice. We focus, in particular, on understanding the spin-dependent transport properties of the itinerant fermions in the impurity band by taking positional disorder of the auxiliary fields into account. For on-site repulsion $U \sim$ bandwidth the density of the itinerant carriers confined to the impurity band, play a key role in determining the kinetic energy of the system and consequently the carrier spin polarization. Our semi-classical Monte Carlo calculations show that the ferromagnetic transition temperature of the carrier spins indeed shows an optimization behavior with the carrier density. We calculate the transport properties in details to establish a one-to-one correspondence between the magnetic and transport properties of the carriers. Our results obtained beyond the perturbative regime are significant for understanding the ferromagnetism in diluted magnetic semiconductors.

I. INTRODUCTION

The history of magnetism is very very old but scientists have begun to understand the concept during the twentieth century. A detailed understanding of magnetism is always necessary to invent and design new magnetic materials. As of now microscopic description of itinerant ferromagnetism still remains a subject of intense research. Due to the technological applications even today the study of itinerant ferromagnetism remains as one of the most interesting as well as challenging avenue for both experimental and theoretical condensed matter physicists.

In recent years, significant progress has been made to understand the magnetic ordering using the microscopic theories of itinerant magnetism. In 1938, Stoner model was first introduced to interpret the itinerant ferromagnetism. In his phenomenal work Stoner pointed out that the ferromagnetic order can arise due to the interaction among the itinerant electrons, which spin split the electronic band structure. For various metals, such as Fe, Co, and Ni, the itinerant electrons exhibit ferromagnetic behavior. In these materials electrons whose spins aligned to form ferromagnetic state are extended and give rise to metallicity. Earlier to Stoner, one of the first justifications of quantum ferromagnetism was put forward by Heisenberg who established that the exchange interactions, arising due to spin-dependent Coulomb repulsion, drives the magnetism between localized moments.

Heisenberg model both in classical and quantum forms remains one of the finest and oldest tools to explain the magnetism and related physical observables in strongly correlated magnetic insulators. Although ferromagnetic Heisenberg model is explored to understand itinerant ferromagnetism but the real difficulty lies in arriving at the effective Hamiltonian of an interacting spin system with negative exchange coupling. Ferromagnetic kinetic exchange between localized spins that arises from an interplay of spin and orbital degrees of freedom is relatively rare. However, probing ferromagnetism in diluted ferromagnetic semiconductors using Heisenberg model is limited. Dual semiconducting and magnetic property of ferromagnetic semiconductors, an diluted spin system, where magnetic impurities are doped in a host semiconductor, is expected to bring technological revolution in spintronics. Theoretical investigation is necessary to understand the physics of these materials that will help the us to push the ferromagnetic transition $T_C$ beyond the room temperature.

In this class of diluted spin systems the magnetic impurities provides both the itinerant carriers and localized moment. The itinerant carriers reside in the shallow acceptor level introduced by magnetic impurity ions in the host semiconductor band gap. It is widely accepted that the (exchange) interaction between the magnetic spins is mediated by the itinerant electrons. This warrants an additional inter-band coupling between the itinerant carriers and the localized moments to study the magnetic and transport properties of charge carriers.

In order to understand the physics of spin-splitting in the carrier impurity band from the perspective of itinerant-exchange mechanism we focus on strong coupling limit. In this limit carriers are firmly localized to the impurity sites and as a result the acceptor levels give rise to distinct impurity band. Although impurity band picture in the most studied GaMnAs semiconductor remains controversial till date, addition of Mn to GaN like large band gap semiconductors give rise to a deep impurity band within the host band gap. Due to disorder (anti-site disorder and interstitial defects) the impurity band remains less than half filled, i.e. carrier density remains smaller than the impurity density and the Fermi energy lies in the impurity band. The position of the Fermi energy, decided by the density of the itinerant carriers plays a key role in determining the

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kinetic energy of the system. Consequently, the gain in kinetic energy is considered a major factor that decides the carrier spin polarization. Detailed study comprising of carrier-spin-dependent transport of localized carriers are limited to date.

In this work we focus on the spin-dependent transport properties of the carriers confined to the impurity band. In our effective spin-fermion model Hamiltonian, derived from repulsive Hubbard model, we assign the $U \sim BW$ (band width) on few percentage of sites in a simple cubic lattice and set it to zero for rest of the sites. We take the carrier density with respect to the impurity concentration which is concomitant with experimental measurements. We organize this paper as follows: In section II we introduce the effective spin-fermion model derived from the Hubbard Hamiltonian and outline our method. We frame the impurity band scenario in Section III. In Section IV we present our numerical results comprising of spin-dependent transport of carriers for $U \sim BW$. Section V is dedicated to compare the magnetic transition temperature which is a major factor that decides the antiferromagnetic order persists beyond the classical percolation threshold in the diluted one-band Hubbard model at absolute half filling in three dimensions.

For diluted spin systems, we have considered finite $U \sim BW$ at randomly chosen sites $k$ (with concentration $x$) and put $U = 0$ for rest of the sites (with concentration $1 - x$). Our diluted Hamiltonian is of the form:

$$H_{sf} = -t \sum_{<i,j>,\sigma} (c_{i\sigma}^\dagger c_{j\sigma} + h.c.) + U/2 \sum_k (<n_k>_k - n_k - \bar{m}_k \cdot \sigma_k)$$

$$+ (U/4) \sum_k (m_k^2 - <n_k>_k^2) - \frac{U}{2} \sum_i n_i$$

The overall electron density $n$ is controlled through the chemical potential ($\mu$) given in the last term. $\mu$ is chosen self-consistently during the thermalization process to get the desired electron density $n$ at each temperature. In quantum Monte Carlo method $m_k$ variables are often used as Ising-like auxiliary fields were introduced by Hirsch through a discrete Hubbard-Stratonovich transformation. So, we use Ising-like auxiliary fields in our calculations. For this case the carriers are either point towards up or down direction in strong coupling limit.

We use semi-classical Monte Carlo (s-MC) method to anneal the system from high temperature consisting of randomly oriented auxiliary fields to obtain the ground state for a fixed carrier density. First we chose a set of random auxiliary fields $m_k$ at desired number of sites and set $<n_k>$ to be uniform for a system size of $N = L^3 = 10^3$ and calculate the internal energy of the carriers by exact diagonalization scheme. Then we update the auxiliary field at an impurity site say $k$ and recalculate the internal energy of the carrier using the new auxiliary field configuration. We employ Metropolis algorithm to accept or reject the above update. At every 10th Monte Carlo step, using the resulting $m_k$ configuration, we update $<n_k>$ self-consistently. This new set of $<n_k>$ is used to perform the further Monte Carlo steps. In order to access large system size we adopt a Monte Carlo update technique based on the traveling cluster approximation (TCA). For impurity concentration $x$ we assign finite $U$ for $10^3x$ sites (for a system size $10^3$) randomly and set $U = 0$ for rest of the sites. We define carrier density as the electrons (or holes) per impurity site. Direct exchange interaction between the impurity spin sites is not taken in to account, which is a valid approximation in the diluted limit, by avoiding the nearest-neighbor impurity pairing. All physical quantities such as carrier magnetization and conductivity are averaged over ten different positional disorder configurations of auxiliary fields in addition to the quantum and thermal averages taken during the Monte Carlo simulations. In this work we consider mainly $x = 0.25$, but compare our main results between $x = 0.25$ and $0.125$ at the end. All parameters such as on-site repulsion ($U$) and temperature ($T$) are scaled that matches well with DQMC results. Recently, we have used s-MC approach to show that the antiferromagnetic order persists beyond the classical percolation threshold in the diluted one-band Hubbard model at absolute half filling in three dimensions.

II. MODEL HAMILTONIAN AND METHOD

We consider one band electron-hole symmetric Hubbard Hamiltonian

$$H = -t \sum_{<i,j>,\sigma} (c_{i\sigma}^\dagger c_{j\sigma} + h.c.) + U \sum_i (n_i^\uparrow - \frac{1}{2}) (n_i^\downarrow - \frac{1}{2})$$

where first term is the kinetic energy [$t$ is the nearest neighbor hopping parameter and $c_{i\sigma}^\dagger$ ($c_{i\sigma}$) are the fermion creation (annihilation) operators at site $i$ with spin $\sigma$] and the second term presents the repulsive Hubbard interaction ($U > 0$).

We reduce the quartic fermion problem present in repulsive Hubbard model into a quadratic one by introducing Hubbard-Stratonovich field and extract the following effective spin-fermion type Hamiltonian by suppressing the imaginary-time dependence from the Hubbard-Stratonovich fields (for details please see Refs. [45,46]). Then above Hamiltonian transfers to

$$H_{sf} = -t \sum_{<i,j>,\sigma} (c_{i\sigma}^\dagger c_{j\sigma} + h.c.) + U/2 \sum_i (<n_i>_i n_i - m_k \cdot \sigma_i)$$

$$+ (U/4) \sum_i (m_i^2 - <n_i>_i^2) - \frac{U}{2} \sum_i n_i$$

where fermions are coupled to the classical auxiliary fields ($m_k$).

The non-monotonic $U$ dependence of the antiferromagnetic transition temperature $T_N$ is established for undiluted system ($U$ at all sites, i.e. $x = 1$ limit) at half filling using a semi-classical Monte Carlo (s-MC) approach. Recently, we have used s-MC approach to show that the antiferromagnetic order persists beyond the classical percolation threshold in the diluted one-band Hubbard model at absolute half filling in three dimensions.
by assuming an impurity band picture

For half-filling case (one carrier per each impurity site) we find that the carriers orient randomly due to intervening non-impurity site at half filling. None out of (a) and (b) are found to be the ground state. Bottom Panel: Our calculations shows ferromagnetic state is favored due to gain in kinetic energy via the non-impurity site as shown in (c) when compared to the scenario drawn in (d).

with hopping parameter \( t \). We use \( U \sim BW \) and vary the carrier density with respect to \( x \) from 0 to 1.

**III. IMPURITY BAND PICTURE**

In the impurity band picture, which is relevant to ferromagnetic semiconductors, carriers reside in the shallow acceptor level separated from the valence band due to the strong coupling between the impurity ions and the carriers. The width of the impurity band and gap between impurity band and valence band depend upon the coupling strength. The location of the Fermi energy inside the impurity band plays a vital role in determining the transport and magnetic properties of the system. The optimum \( T_C \) is expected for which Fermi energy is at the center of the band to gain maximum kinetic energy from the delocalization of the carriers and supposed to decrease towards the edge of the impurity band.

Using our semi-classical Monte Carlo calculations we identify that the itinerant ferromagnetic order obtained in the impurity band picture in spin-fermion model is due to the following scenario of event in which one type of carrier (say up) is more mobile than the other one (down) and drives the magnetism. A schematic figure is shown in Fig. 1 by assuming an impurity band picture where carriers are antiferromagnetically aligned to auxiliary fields at the impurity sites (sites \( i \) and \( j \)). Intervening non-impurity site \( k \) is also shown for completeness. For half-filling case (one carrier per each impurity site)

one would naively expect an AFM ground state mediated by carrier via the non-impurity sites. But it is also apparent the magnetic ground state is strongly depends on the magnetic impurity concentration. For \( x = 0.25 \), which is below the classical percolation limit \( (x_P^{sc} \sim 0.31) \), we do not find any magnetic ordering. This shows that the antiferromagnetic coupling between the auxiliary fields and localized carriers favors the paramagnetism at half filling. Beyond half filling case extra electron added to the system is now relatively more mobile as the first electron is already anti-aligned with auxiliary field and give rise to the ferromagnetic ground state by maximizing the kinetic energy via the non-impurity site(s).

We start our calculation for \( x = 0.25 \) and use \( U = 12 \) \(( \sim BW) \) to manifest the formation of an impurity band that imposes the carrier localization. A well separated impurity band for hole density \( p = 0.2 \) is clear from density of states (DOS) shown in Fig. 2(a) for a relatively high temperature \((T = 0.07)\). The DOS at each \( \omega \) is obtained by implementing the Lorentzian representation of the \( \delta \) function: 

\[
N(\omega) = \sum_{\omega_k} \delta(w - \omega_k),
\]

where \( \omega_k \) are the eigenvalues of the fermionic sector and the summation runs up to total number of eigenvalues \((2L^3)\) of the system. The valence band is very much symmetric but the narrow impurity band is asymmetric. This asymmetric character of the impurity band picture remains intact for all the carrier densities. It is important to mention here that ferromagnetism in Hubbard model is attributed to an asymmetric density of states with large spectral weight near one of the band edges.

For high band gap systems the ferromagnetism along with the nature of the charge carriers is controversial till date. Holes (electrons) are accounted to be the charge carriers for p-type conduction \cite{27,28} (n-type...
Uto the formation of the impurity band whereas $x = 12$ (distribution for $U = 12$ sites and $U = 0$ sites are plotted separately). (b) Quantum $s(0)$ and classical $S_m(0)$ ferromagnetic structure factor vs temperature show the same transition. The spin resolved density of states are plotted for (c) $T = 0.07$ (above $T_C$) (d) $T = 0.01$ (below $T_C$). The equal contribution from up or down spin sector in (c) implies that the bands are unpolarized. For $T = 0.01$ the impurity band is completely spin polarized. The Fermi energy is set at zero.

We calculate the magnetic moments $P_q(M)$ for $U = 12$ (distribution for $U = 12$ sites and $U = 0$ sites are plotted separately). (b) Quantum $s(0)$ and classical $S_m(0)$ ferromagnetic structure factor vs temperature show the same transition. The spin resolved density of states are plotted for (c) $T = 0.07$ (above $T_C$) (d) $T = 0.01$ (below $T_C$). The equal contribution from up or down spin sector in (c) implies that the bands are unpolarized. For $T = 0.01$ the impurity band is completely spin polarized. The Fermi energy is set at zero.

We present the spin resolved DOS for both high and low temperature cases in Fig. 3(c) and (d). For $T = 0.07$ (which is above $T_C$) the impurity band remain unpolarized. Both the valence band and the impurity band are completely symmetric for both up or down spin sectors. The impurity band is completely spin polarized for $T = 0.01$ which depicts the complete ferromagnetic ordering of the carriers that reside within the impurity band. This agrees well with experiment. In addition we plot the classical structure factor for the auxiliary fields $S_m(0)$, where $S_m(q) = \frac{1}{2N_\uparrow N_\downarrow} \sum_{ij} \mathbf{m}_i \cdot \mathbf{m}_j e^{i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)}$.
moments at the interacting sites (see Fig. 2a) and decreases sharply beyond \( n = 0.5 \). We also calculated the system averaged uniform magnetization \( m_f \) vs electron density at \( T = 0.01 \) show one to one correspondence with \( s(0) \) as it is proportional to the square root of \( s(0) \). (b) Ferromagnetic window with respect to the electron density \( (n) \). The ferromagnetic transition temperature \( (T_c) \) shows optimization behavior. The inset shows the FM window for both electron and hole densities. (c) dc conductivity calculated at \( T = 0.01 \) show metallic at middle of the ferromagnetic window and depicts an insulator-metal-insulator (IMI) transition with respect to \( n \). (d) Participation ratio around Fermi energy \( \text{PR}(E_F) \) shows that the states at the middle of ferromagnetic windows are more extended and agrees well with conductivity results. Corresponding quantities for two system sizes are compared in the inset of (c) and (d).

In order to figure out the correspondence between the ferromagnetism and the metallicity we plot the temperature dependence of the resistivity for \( n = 0.2 \) in Fig. 4a). We calculate the dc limit of the optical conductivity by using the Kubo-Greenwood formula. At low temperature the system shows metallic behavior. The insulator-metal transition coincides with the onset of ferromagnetism (see Fig. 4a). To check for finite size effect which remains a concern in the small system size based Monte Carlo calculations, we show \( s(0) \) with temperature for two system sizes \( L = 10 \) and 12 in Fig. 4b). Our results indicate that the curves are pretty similar to each other.

The moment distributions \( P_q(M) \) for the different densities using only \( U = 12 \) sites are shown in Fig. 5a) at \( T = 0.01 \). The moment distributions get steeper and the peak value (defined as \( P_q(M^*) \)) increases up to \( n = 0.3 \) and decreases thereafter. This shows that the moment distribution curve gets broadened with a reduction in \( P_q(M^*) \) value on both sides of \( n = 0.3 \). We plot the peak value of moment distribution curves \( P_q(M^*) \) vs \( n \) for \( T = 0.01 \) and 0.07 in Fig. 5b). Next we will show that the optimization of \( P_q(M^*) \) is very similar to the optimized ferromagnetic, conductivity and participation-ratio windows [see Fig. 5]. This emphasize the fact that the optimum ferromagnetic \( T_c \) is obtained for which the moment fluctuation at \( U = 12 \) sites is minimal.

Now we analyze the quantum ferromagnetic structure factor for different carrier densities \( n \) at \( T = 0.01 \) using interacting \( U = 12 \) sites as mentioned in Fig. 3a) and Fig. 4a). For \( T = 0.01 \) the quantum ferromagnetic structure factor as shown in Fig. 3a) increases with carrier density as expected due to the enhancement of the

\( \mathbf{q} \) are the wave vectors along with quantum ferromagnetic structure factor \( s(0) \) in Fig. 3b) and it shows the same transition. Due to strong coupling between itinerant carriers and auxiliary fields the carriers are always align anti-parallel to the fields. For this reason classical ferromagnetic structure factor \( S(0) \) and quantum fermionic structure factor \( s(0) \) behaves very similar to each other.
that the kinetic energy is minimum at the edge of the ferromagnetic window. As a result, one expects a metallic system at the center of the band and an insulating state at the edge for low temperatures. In fact, conductivity calculations for different $n$ plotted at $T = 0.01$ (see Fig. 7(c)) shows the same and depicts an insulator-metal-insulator (IMI) transition with carrier density. This also establishes the fact that the mobility is minimum near the edge of the ferromagnetic window. The participation ratio (PR) which is a measure of delocalization also signifies that the states corresponding to FM order are extended, though for higher $U$ values PR($E_F$) decreases. Inset in (b) shows that peak of local moment distribution shift towards the carrier density value (for $n = 0.2$) for higher $U$ and indicate the electrons are more and more localized for larger $U$.

V. COMPARISON OF FERROMAGNETIC WINDOWS FOR DIFFERENT $U$

Next, we explore the ferromagnetic window for four different $U$ values. The ferromagnetic window in Fig. 7(a) shifts to the right with increasing $U$. This is because the carriers are more localized for larger $U$ values [see in inset of Fig. 7(b)] and enhances the carriers mobility among the interacting sites due to availability of more interacting lattice sites beyond $n = 0.5$. This analysis is true for higher density edge of the impurity band. In fact, in large $U$ ($>> BW$) limit ferromagnetic window is expected to span up to $n = 1.0$ like undiluted Kondo lattice model. At the same time ferromagnetism in lower part of the FM window gets depleted for stronger localization of carriers that remain localized far apart on impurity sites. The quantum magnetic structure factor for different $U$ values (see Fig. 7(b)) at low temperature ($T = 0.01$) also shows the non-monotonic behavior similar to $U = 12$ case. Here, for all $U$, the maximum $\sigma(0)$ is found for the particular carrier density beyond which the ground state is paramagnetic. In Fig. 7(c) the conductivity results show that the insulator-metal-insulator (IMI) pattern also remains intact for all $U$. With increasing $U$ the conductivity decreases, mainly at the middle of FM window, as carriers are more localized at larger $U$ values. In addition PR($E_F$) in Fig. 7(d) supports the localization-delocalization-localization pattern with the electron density which is also obtained from the ferromagnetic and conductivity calculations.

VI. COMPARISON OF FERROMAGNETIC WINDOWS BETWEEN $x = 0.25$ AND $x = 0.125$

In addition to $x = 0.25$, we performed systematic calculations to unveil the ferromagnetic window for $x = 0.125$ case. In Fig. 8(a) we compare the ferromagnetic...
windows for $x = 0.25$ and $0.125$ using $U = 12$. Here the FM window with respect to $n$ shifts to the right, but $T_C$ reduces considerably. The average local moments on $U = 12$ sites remain same for both $x$ [see Fig. 8b)]. This indicate that the induced moment at $U = 0$ sites would decrease for smaller $x$. In fact as we decrease the $x$ the resulted moments at $U = 0$ sites decreases [see the inset of Fig. 8b)]. Although moments for $U = 12$ sites are the same, the density of states in Fig. 8c) shows that the carriers are more localized as the impurity band gets narrower for $x = 0.125$. This is due the increase of the distance between interacting sites in $x = 0.125$ as compared to $x = 0.25$. This is one of the reason for the drop in $T_C$ value. In Fig. 8d) the reduction of conductivity with $x$ agrees with the fact that the mobility of carriers decline for $x = 0.125$.

VII. CONCLUSIONS

In summary, based on spin-fermion model, we show that the ferromagnetism is favored for low density of carriers, which is concomitant to the experimental results. In a non-perturbative limit ($U \sim BW$) our analysis shows that the density of the itinerant carriers, confined to the impurity band, that decides the kinetic energy of the system, plays an important role in determining the carrier spin polarization. The ferromagnetic ordering temperature shows an optimization behavior with the carrier density. We have provided a systematic study of carrier spin dependent transport properties of the carriers over the whole carrier density range. A insulator-metal-insulator transition is observed across the ferromagnetic window. Due to strong coupling between itinerant carriers and auxiliary fields the polarization of auxiliary fields follows the property of carrier spin polarization very closely. Thus our results are significant for the understanding of ferromagnetism in diluted magnetic semiconductors.

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