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Original Article Ferromagnetic-antiferromagnetic Competition in Diluted Magnetic Semiconductors

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Abstract: The signature of the magnetic competition in a nearly filled impurity band diluted magnetic semiconductors is addressed. In the formalism of the Kondo lattice model, self-consistent equations determining the Green function of the itinerant carrier in the system are evaluated by use of the dynamical mean-field theory. An analytical solution of the static spin susceptibility of the itinerant carrier to specify the magnetic instability is then established. Once the impurity band is nearly filled, one finds the antiferromagnetic instability against the ferromagnetic state. Phase diagrams of the magnetic structures in the diluted magnetic semiconductor were also discussed.

Keywords: III-V and II-VI semiconductors, Ferromagnetic-antiferromagnetic, Kondo lattice, Saturation moments and magnetic susceptibilities.

1. Introduction

Magnetic instability in diluted magnetic semiconductors (DMSs) is one of the most issues that stimulated much interest because of its potential for applications in future spintronics [1, 2]. By doping a magnetic ion (e.g. Mn^{2+}), a semiconducting host (e.g. GaAs) induces localized magnetic moment and an impurity band is formed [3]. Once the impurity band is partly filled, the ferromagnetic (FM) state is stabilized at a low temperature [1, 2]. However, if the impurity band is filled or nearly filled, the FM configuration might be forbidden due to Pauli's principle. In this situation, instead, the antiferromagnetic (AFM) state is favored [4]. So far, magnetic instability

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in DMSs is considered when the conduction band is not fully filled, and the system stabilizes in FM state [5-10]. The AFM state in DMSs, up to now, has not been considered in detail. Considering the competition of the AFM and FM states and especially their transition features from the paramagnetic (PM) state in a unique theoretical framework is thus extremely fruitful. Feature of the PM-FM transition in DMSs has been proposed in a formation of bound magnetic polarons [7-12]. However, it has not been used to explain a mechanism of the PM-AFM transition. In the present work, both the PM-FM and PM-AFM transitions are discussed in the signature of the static spin susceptibility. Divergence of the susceptibility indicates an instability of the magnetic order state. To treat the problem, we use the highly successful dynamical meanfield theory (DMFT) [13]. The DMFT has been widely used in studying the magnetic properties in DMSs and similar systems [6, 12, 14, 15]. The DMFT is applied to the Kondo lattice model to evaluate the static spin susceptibility function that momentum dependence keeps only on one parameter in the infinite dimensional limit [16]. Considering the influence of that parameter on the divergence of the static spin susceptibility, one might establish stability of the magnetic order state [16]. Note here that, to describe the magnetic stability in DMSs, we have assumed that all active magnetic moments distribute homogeneously in the system, the Kondo lattice formalism is thus applicable [17].

The paper is organized as follows. The next section (namely Section 2) briefly describes the Kondo lattice model applied for DMSs and its DMFT solution that is used to evaluate the static spin susceptibility in Section 3. Section 4 discusses the numerical results of the susceptibility function for a given set of model parameters. The magnetic phase diagram is then addressed. Section 5, finally, concludes our work.

2. Hamiltonian and Dynamical Mean-field Theory

To describe the magnetic signatures in DMSs we examine the following Hamiltonian

$$H = -t \sum_{\langle i,j \rangle \sigma} c^{\dagger}_{i\sigma} c_{j\sigma} + 2J \sum_{i} \alpha_{i} \vec{S}_{i} \vec{s}_{i} - \mu \sum_{i} n_{i}$$
(0)

where $c_{i\sigma}^{\dagger}(c_{i\sigma})$ is the creation (annihilation) operator of the itinerant carrier with spin *s* at lattice site *i*. $\vec{s}_i = \sum_{\sigma\sigma'} c_{i\sigma}^{\dagger} \vec{\sigma}_{\sigma\sigma'} c_{i\sigma'} / 2$ ($\vec{\sigma}_{\sigma\sigma'}$ are the Pauli matrices) and $n_i = \sum_{\sigma} c_{i\sigma}^{\dagger} c_{i\sigma}$, respectively, are the spin and occupation operators of the carrier. \vec{S}_i is the impurity moment at lattice site *i*. The second term in Eq. (1) thus indicates the magnetic coupling between the itinerant carrier and the localized impurity spins at the same site. $\alpha_i = 1(0)$ if site *i* is occupied with probability *x* (unoccupied with probability 1 - *x*) by magnetic ion. In the case of $\alpha_i = 1$ for all *i*, the Hamiltonian recovers the Kondo lattice model without magnetic randomness disorder [6]. *m* is the chemical potential.

The Hamiltonian in Eq. (1) is solved by use of DMFT. As a nonperturbative local theory, DMFT solution is exact in the infinite space dimensional limit. The key point of the DMFT is that the local Green function of itinerant carriers

$$G_{\sigma}(i\omega_n) = \int d\varepsilon \rho(\varepsilon) \frac{1}{i\omega_n - \varepsilon + \mu - \Sigma_{\sigma}(i\omega_n)},$$

must coincide with one of the effective single impurity embedded in the dynamical mean-field medium [13]. For the Hamiltonian written in Eq. (1), Green function of the effective single impurity reads.

$$G_{\sigma}(i\omega_n) = \sum_{\alpha s} \frac{W_{\alpha s}}{\mathcal{G}_{\sigma}^{-1}(i\omega_n) - Js\sigma\alpha}$$

 $\mathcal{G}_{\sigma}(i\omega_n)$ here is the Green function of an effective medium. Note here that the Green functions in Eq. (2) and in Eq. (3) depend only on the Matsubara frequency $\omega_n = (2n+1)\pi T$ (*T* is temperature). In the infinite dimensional limitation, the self-energy $\Sigma_{\sigma}(i\omega_n)$ is the momentum independent and $\rho(\epsilon)=\exp(-\epsilon^2)/\sqrt{2}$ for the hypercubic lattice. In Eq. (3), $W_{\alpha s}$ (α =0;1) act as the weight factors, that explicitly read

$$W_{0s} = \frac{(1-x)\exp\sum_{n\sigma}\ln[\mathcal{G}_{\sigma}^{-1}(i\omega_{n})/i\omega_{n}]}{\operatorname{Tr}\exp\sum_{n\sigma}\ln[\mathcal{G}_{\sigma}^{-1}(i\omega_{n})]},$$

and

$$W_{1s} = \frac{x \exp \sum_{n\sigma} \ln\{[\mathcal{G}_{\sigma}^{-1}(i\omega_{n}) - Js\sigma]/i\omega_{n}\}}{\operatorname{Tr} \exp \sum_{n\sigma} \ln[\mathcal{G}_{\sigma}^{-1}(i\omega_{n}) - Js\sigma]}$$

The local Green function is found self-consistently via the Dyson equation

$$G_{\sigma}^{-1}(i\omega_n) = \mathcal{G}_{\sigma}^{-1}(i\omega_n) - \Sigma_{\sigma}(i\omega_n).$$

From Eqs. (2-6) one finds a set of self-consistent equations to evaluate the self-energies $\Sigma_{\sigma}(i\omega_n)$. The Green function of the itinerant carrier thus then can be established.

3. Static Spin Susceptibility

From the result of the Green function in the previous section, one might detect the magnetic phase transition in the systems. In this paper, magnetic properties of the Hamiltonian (1) will be considered in an analyzation of the static magnetic susceptibility of the itinerant carrier, related with its Green function as [18]

$$\chi(\mathbf{q}) = T^2 \sum_{ijn,\sigma\sigma'} e^{-i\mathbf{q}(\mathbf{R}_i - \mathbf{R}_j)} \frac{dG_{ii,\sigma}(i\omega_n)}{dh_{j\sigma'}} \Big|_{h=0} \sigma\sigma',$$

where $h_{j\sigma}$ is the external magnetic field which couple to the magnetization of the itierant carrier into the Hamiltonian. From the identity

$$G_{ii,\sigma}(i\omega_n) = \sum_{lm} G_{il,\sigma}(i\omega_n) G_{lm,\sigma}^{-1}(i\omega_n) G_{mi,\sigma}(i\omega_n),$$

and by noting that $\sum_{m} G_{lm,\sigma}^{-1}(i\omega_n) G_{mi,\sigma}(i\omega_n) = \delta_{li}$, one might obtain

$$\frac{dG_{ii,\sigma}(i\omega_n)}{dh_{j\sigma'}} = -\sum_{lm} G_{il,\sigma}(i\omega_n) \frac{dG_{lm,\sigma}^{-1}(i\omega_n)}{dh_{j\sigma'}} G_{mi,\sigma}(i\omega_n)$$
$$= \sum_{lm} G_{il,\sigma}(i\omega_n) \left[\delta_{lm} \delta_{mj} + \frac{d\Sigma_{lm,\sigma}(i\omega_n)}{dh_{j\sigma'}} \delta_{lm} \right] G_{mi,\sigma}(i\omega_n)$$

Here we have used a representation of the Green function depending on the external magnetic field

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$$G_{lm,\sigma}(i\omega_n) = \frac{1}{\delta_{lm}[i\omega_n + \mu - h_{l\sigma} - \Sigma_{lm,\sigma}(i\omega_n)] - t_{kl}^* / 2\sqrt{D}}$$

with an effective hopping t^* in *D* dimensional hypercubic lattice [13]. Assuming that $\Sigma_{\sigma}(i\omega_n)$ is a functional of the Green function, i.e., one can write

$$\frac{d\Sigma_{ll,\sigma}(i\omega_n)}{dh_{j\sigma'}} = \sum_{\nu\sigma''} \frac{d\Sigma_{ll,\sigma}(i\omega_n)}{dG_{ll,\sigma''}(i\omega_\nu)} \frac{dG_{ll,\sigma'}(i\omega_\nu)}{dh_{j,\sigma'}}.$$

Substituting Eq. (9) into Eq. (7) with the help of Eq. (11) one might arrive

$$\chi(\mathbf{q}) = -T^2 \sum_{n\sigma\sigma'} \chi_0(\mathbf{q}, i\omega_n) \sigma\sigma' + \sum_{n\sigma\sigma'} \chi_0(\mathbf{q}, i\omega_n) \sigma\sigma' \sum_{\mu\sigma'} \frac{d\Sigma_{\sigma}(i\omega_n)}{dG_{\sigma'}(i\omega_v)} \chi_{\sigma'\sigma'}(\mathbf{q}, i\omega_n).$$

Here, we have defined

$$\chi_{\sigma\sigma'}(\mathbf{q},i\omega_n) = -T^2 \sum_{ij,\sigma\sigma'} e^{-i\mathbf{q}(\mathbf{R}_i - \mathbf{R}_j)} \frac{dG_{ii,\sigma}(i\omega_n)}{dh_{j\sigma'}} \Big|_{h=0} \sigma\sigma',$$

and the bare particle-hole susceptibility

,

$$\chi_0(\mathbf{q},i\omega_n) = \sum_{\mathbf{k},\sigma} G_{\sigma}(\mathbf{k}+\mathbf{q},i\omega_n) G_{\sigma}(\mathbf{k},i\omega_n).$$

From the set of self-consistent equations addressed above in the previous section, one might consider that the self-energy $\Sigma_{\sigma}(i\omega_n)$ is a functional of the Green function $G_{\sigma}(i\omega_n)$ and the weight factors $W_{\alpha s}$. In this sense, we have

$$\frac{d\Sigma_{\sigma}(i\omega_{n})}{dG_{\sigma''}(i\omega_{\nu})} = \frac{\partial\Sigma_{\sigma}(i\omega_{n})}{\partial G_{\sigma''}(i\omega_{\nu})} \delta_{\sigma\sigma''} \delta_{n\nu} + \sum_{s\alpha} \frac{\partial\Sigma_{\sigma}(i\omega_{n})}{\partial W_{\alpha s}} \frac{dW_{\alpha s}}{dG_{\sigma''}(i\omega_{\nu})}.$$

The partial derivatives can easily be evaluated and finally one arrives a representation of the static spin susceptibility function

$$\chi(\mathbf{q}) = -2T^{2} \sum_{n} \frac{1}{\left[\chi_{0}(\mathbf{q}, i\omega_{n})\right]^{-1} + \sum_{\sigma} \left[G_{\sigma}^{-2}(i\omega_{n}) - S_{n\sigma}^{-1}\right]/2} + T^{2} \sum_{\alpha s \sigma} \frac{\sigma \gamma_{\alpha s}(\mathbf{q})}{S_{n\sigma} Z_{\sigma}^{\alpha s}(i\omega_{n}) \left[\chi_{0}(\mathbf{q}, i\omega_{n})\right]^{-1} + \sum_{\sigma} \left[G_{\sigma}^{-2}(i\omega_{n}) - S_{n\sigma}^{-1}\right]/2} \right],$$

where

$$S_{n\sigma} = \sum_{\alpha s} \frac{W_{\alpha s}}{\left[Z_{\sigma}^{\alpha s}(i\omega_{n})\right]^{2}}$$

and the $\gamma_{as}(\mathbf{q})$ term in Eq. (16) can be determined from a matrix identity

$$\sum_{\alpha's'}\Pi_{\alpha s,\alpha's'}(\mathbf{q})\gamma_{\alpha's'}(\mathbf{q})=\Omega_{\alpha s}(\mathbf{q}),$$

with

$$\Pi_{\alpha s,\alpha' s'}(\mathbf{q}) = \delta_{\alpha \alpha'} \delta_{ss'} - \sum_{n\sigma} \frac{W_{\alpha s} \Gamma_{\sigma}^{\alpha s}(i\omega_n)}{S_{n\sigma} Z_{\sigma}^{\alpha' s'}(i\omega_n)} - \frac{1}{2} \sum_{n\sigma\sigma'} \frac{R_n(\mathbf{q}) W_{\alpha s} \Gamma_{\sigma'}^{\alpha s}(i\omega_n)}{S_{n\sigma} S_{n\sigma'} Z_{\sigma}^{\alpha' s'}(i\omega_n)} \sigma\sigma',$$

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$$\Omega_{\alpha s}(\mathbf{q}) = -2\sum_{n\sigma} \frac{R_n(\mathbf{q})W_{\alpha s}\Gamma_{\sigma}^{\alpha s}(i\omega_n)}{S_{n\sigma}}\sigma.$$

Here definitions

$$\Gamma_{\sigma}^{\alpha s}(i\omega_n) = Z_{\sigma}^{\alpha s}(i\omega_n)^{-1} - G_{\sigma}(i\omega_n)$$

and

$$R_{n}(\mathbf{q}) = \frac{1}{[\chi_{0}(\mathbf{q}, i\omega_{n})]^{-1} + \sum_{\sigma} [G_{\sigma}^{-2}(i\omega_{n}) - S_{n\sigma}^{-1}]/2}$$

have been used. Note here that the momentum dependence of $\chi(\mathbf{q})$ sets only on the bare susceptibility $\chi_0(\mathbf{q},i\omega_n)$. In the infinite dimension limit, the \mathbf{q} dependence of $\chi_0(\mathbf{q},i\omega_n)$ is summarized in a single parameter $X = \sum i \cos qi/D$ [13, 16]

$$\chi_0(\mathbf{q},i\omega_n) \equiv \chi_0(X,i\omega_n) = \frac{-1}{\sqrt{1-X^2}} \int \frac{d\epsilon\,\rho(\epsilon)}{z-\epsilon} F\left(\frac{z-X\epsilon}{\sqrt{1-X^2}}\right),$$

where $F(z) = \int d\epsilon \rho(\epsilon)/(z-\epsilon)$ is the Hilbert transform of the noninteracting density of states, with $z=i\omega_n+\mu-\sum(i\omega_n)$. Instability of the PM state is addressed depending on a certain value of X the susceptibility $\chi(X)$ diverges. If $\chi(X)$ diverges at X = -1 (or the zone-boundary point $\mathbf{q} = (\pi, \pi, ..., \pi)$) the PM-AFM transition happens, whereas a divergence of $\chi(X)$ at X = 1 (or uniform zone center point $\mathbf{q} = 0$) indicates the FM stability against the PM state.

4. Numerical Results

To examine the instability of the PM state in DMSs, firstly, we inspect signatures of the static spin susceptibility as a function of X for some values of different temperatures T at a given carrier density n. For $n \approx x$, the impurity band reaches a nearly filled situation. The static spin susceptibility is evaluated from Eq. (16) once we have arrived results of the Green function evaluated by DMFT. In the numerical calculation below, we fix the magnetic coupling J=3 and dopping density x = 0.1. μ is varied to satisfy a given carrier density.



Figure 1. Static spin susceptibility χ depending on X at different temperatures T for J = 3 and x = 0.1 at n = 0.097.

Figure 1 shows χ as a function of X at some temperatures T with J = 3, and x = 0.1, at carrier density n = 0.097. Once the impurity band is not fully filled, the static spin susceptibility comes to diverge at X = 1 by lowering the temperature. The tendency indicates that the PM state becomes

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instability and the system might stabilize at the FM if the temperature is smaller than a critical value.

Next, in Figure 2, we address the signature of the static spin susceptibility as a function of X for the same set of parameters in the Figure 1, but with a larger carrier density, n = 0.099. In the contrast to the case of n = 0.097, the static spin susceptibility for n = 0.099 has a tendency to diverge at X=-1 by lowering the temperature. The system thus refers to stabilize in the AFM state if the impurity band becomes completely filled.



Figure 2. Static spin susceptibility χ depending on X at different temperatures T for J = 3 and x = 0.1 at n = 0.099.

Signatures of the static spin susceptibility discussed above might help us establish the magnetic phase competition in the system. A divergent point of the susceptibility function is exactly the critical point of the phase transition. However, note here that, for a given set of parameters, the susceptibility might diverge at a different value of X as long as a temperature is small enough. To specify which state is more stabilized, one has to find the critical temperature as a function of X. The maximum point of the critical temperature versus X would determine which state should be stabilized. Figure 3 shows us the critical temperature T_c as a function of X for different values of n with J = 3 and x = 0.1. At n = 0.096, the critical temperature is nonzero and monotonically increases for $0 < X \le 1$ [see Figure 3(a)]. Maximum of the critical temperature is sufficiently small. The behavior of $T_c(X)$ is slightly changed if the carrier density is further increased to n = 0.097 or n = 0.098 [see Figure 3(b-c)]. In these cases, one finds a nonzero values of T_c even at X < 0.



Figure 3. The critical temperture of the magnetic instability Tc as a function of X for some values of carrier density n at J = 3 and x = 0.1.

However, $T_c(X = -1)$ is always smaller than that at X = 1, the FM thus is still stabilized in the range of the carrier density. By increasing the carrier density the critical temperture for X < 0 becomes comparable with that for X > 0, i.e., the AFM state has a tendency to stabilize against the FM state. If the carrier density is sufficiently close to x, i.e., once the impurity band is close to be completely filled, the critical temperature is nonzero only for X < 0 and it gets maximum at X = -1 [see Figure 2(d)]. In the contrast to the previous situations, in this case, the system stabilizes in the AFM state



Figure 4. Magnetic phase diagram in the T-n plane for different values of J at x = 0.1. The FM state is indicated by the green, the AFM state is indicated by the red, while PM state is expressed in white.

To discuss in more detail the magnetic competition in DMSs, in Figure 4 we analyze a magnetic phase diagram in the T -n plane for some values of J at x = 0.1. The FM state is specified if T_c gets maximum at X = 1 and vise versa, one finds the AFM state if T_c gets maximum at X = -1. Note here that, for a sufficiently large magnetic coupling J > 2, the impurity band is completely separated from the main band [6,12]. For small *n*, the impurity band is partially filled and the FM phase is formed due to the delocalization of the itinerant carriers. Increasing n leads the impurity band further to be filled, the low energy hopping processes for the FM state are suppressed, whereas, that of the AFM state is developed. The FM regime thus is collapsed and instead the AFM state becomes stabilize. The AFM state is found for $n \approx x$, i.e., once the impurity band is completely filled [see Figure 4(b&c)]. Otherwise, a small magnetic coupling is not sufficient to open an impurity gap. The impurity band thus merges to the main band and one finds only the FM state at low temperature in the whole range of the carrier density [see Figure 4(a)]. Note here that the AFM state becomes dominant at the intermediate magnetic coupling. Increasing the magnetic coupling from a small value, the AFM appears and then is suppressed if the magnetic coupling is large enough. It makes sense when the magnetic coupling between the carrier spin and local magnetic moment in our consideration is in ferromagnetism.



Figure 5. Magnetic phase diagram in the *T*-*n* plane for J = 3 with three different values of *x*. The FM state is indicated by the green, the AFM state is indicated by the red, while PM state is expressed in white.

The influence of the magnetic impurity doping on the magnetic instabilities in DMSs is also discussed in Figure 5 analyzing the *T*-*n* magnetic phase diagram evaluated in our approach for J = 3

with three different values of x. Here, we consider only in the case that the carrier density is always smaller than the magnetic impurity density, i.e., $n \le x$. For a slight change of the impurity density, one finds an abrupt change of the magnetic stability. Indeed, at x=0.1, the AFM state is stabilized at $n\sim0.1$ instead of the FM state [Figure 5(a)]. The FM state is expanded by a small increase of the impurity density and at x=0.105 [Figure 5(c)], the AFM is completely disappeared and one finds only the FM state. Note here that at x=0.101 [Figure 5(b)] or x=0.105 [Figure 5(c)], the AFM is completely disappeared and one finds only the FM state. Note here that at x=0.101 [Figure 5(b)] or x=0.105 [Figure 5(c)], the arrive fully filled impurity band so the AFM state becomes less stabilized in the range of the carrier density.

5. Conclusion

To conclude, we have considered the phase structure analyzing the magnetic competition in DMSs when the impurity band is nearly filled. By assuming that the carrier in DMSs satisfies the Kondo lattice formalism, the Green function of the carrier is evaluated by the use of the dynamical mean-field theory. In this sense, an analytical expression of the static spin susceptibility is found and it might be calculated once the self-consistent solution of the Green function is established. Numerical results indicate that when the impurity band is filled or nearly filled, the antiferromagnetic state becomes stabilized against the ferromagnetic state. Detailed phase structure of the magnetic competition in the nearly filled impurity band DMSs then has been addressed.

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