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Softening of spin-wave stiffness near the ferromagnetic phase transition in diluted magnetic semiconductors

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Abstract. Employing the self-consistent Green's function approach, we studied the temperature dependence of the spin-wave stiffness in diluted magnetic semiconductors. Note that the Green's function approach includes the spatial and temperature fluctuations simultaneously which was not possible within conventional Weiss mean-field theory. It is rather interesting that we found the stiffness becomes dramatically softened as the critical temperature is approached, which seems to explain the mysterious sharp drop of magnetization curves in samples within diffusive regime.

PACS. 75.30.Ds Spin waves – 75.40.Gb Dynamic properties (dynamic susceptibility, spin waves, spin diffusion, dynamic scaling, etc.) – 75.50.Dd Nonmetallic ferromagnetic materials

1 introduction

Spintronics [1] brings out an industrial renaissance in the past decades for its powerful usage of the extra spin degrees of freedom in many electronic applications. For instances, giant magneto-resistance (GMR) had been applied to read-write head of computer hard disk and tunneling magneto-resistance (TMR) to the newest nonvolatile memory for MRAM. The huge success kicked off the intense investigations on possible realizations of similar devices in semiconducting materials, which can be directly integrated with the existing industrial techniques. The idea has enjoyed its primary success in the so-called diluted magnetic semiconductors (DMS), where the magnetic ions are doped into the host semiconductors and lead to a ferromagnetic phase.

The key issue at current stage is how to raise the critical temperature so that the ferromagnetic order is robust even at room temperature. Taking the well-known material (Ga, Mn)As as example [2–4], it was demonstrated that the critical temperature can be raised significantly by thermal annealing. However, the highest critical temperature at the time of writing is around 160 K [5], which is still far from the goal for room-temperature DMS. Based on Zener model, Dietl et al. proposed to look for room-temperature DMS in wide bandgap semiconductors and oxides such as GaN, ZnO, TiO_2 [6–8]. While the critical temperatures in these materials are typically higher compared with (Ga, Mn)As, clustering seems to be a serious problem which prevents their potential usage in realistic devices.

While the issue of raising up the critical temperature seems to lie in experimentalists' hands, we believe a better understanding of the ferromagnetic phase would also help. After intense theoretical investigations, the origin of ferromagnetism in DMS is believed to be carrier mediated [9]. Typically, the doped transition metal ions provide the localized impurity spins with small direct exchange (often antiferromagnetic) among themselves. Furthermore, the experimental results show that only $10 \sim 30\%$ of the doped magnetic ions contribute itinerant carriers [10] into the host semiconducting bands. Through double-exchange mechanism, the itinerant carriers mediated the indirect exchange interactions between the localized impurity spins and the ferromagnetic order sets in when the system is cooled below the critical temperature.

While the origin of the ferromagnetic phase is more or less clear, some of its physical properties remain puzzling. To achieve a full understanding, it is crucial to include the exchange coupling between itinerant and localized spin densities, thermal fluctuations, the random locations of the doped ions, the realistic band structure and the repulsive interaction between itinerant carriers. Since it is almost impossible to incorporate all effects in single formalism, one needs to glue up piecewise information from different approaches.

In this paper we employ the self-consistent Green's function method [11], which includes the spatial and thermal fluctuations simultaneously, to study the temperature

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dependence of spin-wave stiffness in DMS. Since our goal is to demonstrate the peculiar temperature dependence, we simplify the realistic band structure by the single-band approximation, at price of sacrificing the *quantitative* description for realistic materials. However, within single-band approximation, we were able to establish the close tie between the softening of stiffness and the sudden drop in magnetization curves [2]. Moreover, our self-consistent Green's function approach also shows the appearance of concave magnetization curve [12] in the regime where itinerant carriers are dilute. This implies a smooth crossover from the diffusive regime to the localized regime with strong disorder. Since previous studies including the full six bands only renormalize various physical parameters and twist the phase boundaries [13], we expect our results to be *qualitatively* robust.

The organization of the paper is the following: in the Section 2 of this paper, we derive the formalism for the self-consistent Green's function approach. In Section 3, we show our numerical results and discuss connections to other approaches in the literature.

2 Theory

The sd model is proper to describe the DMS systems including a strong exchange interaction between local spins, which come from electrons in d orbits of transition atom, and itinerant spins around whole system, which come from impurity doping donation. The Hamiltonian of the sd model can be expressed by

$$H = H_0 + J\int d^3r S(r)\cdot\sigma(r), \tag{1}$$

the first term H_0 is the kinetic energy of the itinerant carriers and the second term is the exchange interaction between itinerant carrier spins and the localized spin moments, where the spin density of the localized moments is $S(r) = \sum_I \delta^3(r - R_I)S_I$ and the itinerant spin density is $\sigma(r) = \psi^\dagger(r)(\tau/2)\psi(r)$. In momentum representation the Hamiltonian is expressed as

$$\begin{aligned} H &= H_k + H_J \\ &= \sum_{k,\sigma}\varepsilon_k c^\dagger_{k,\sigma}c_{k,\sigma} - c\frac{J}{2}\sum_k S^+_k\sigma^-_k \\ &\quad -c\frac{J}{2}\sum_k S^-_k\sigma^+_k - cJ\sum_k S^z_k\sigma^z_{-k}, \end{aligned} \tag{2}$$

where the constant c is the density of the magnetic ions in DMS and J is the magnetic coupling integral in the unit of $eVnm^3$. Since the RKKY interaction dominates the magnetism in DMS, to investigate the magnon dispersion is necessary for studying in microscopic. In order to obtain the magnon dispersion of local spin in DMS we have to calculate the retarded local spin Green's function defined by

$$\begin{aligned} G_{i,j}(t) &= \langle\langle S^+_i(t); S^-_j(0)y\rangle\rangle \\ &= -i\theta(t)\langle[S^+_i(t), S^-_j(0)]\rangle, \end{aligned} \tag{3}$$

where $\theta(t)$ is a step function of time t, $\langle\cdots\rangle$ representing the expectation value and $[\cdots]$ is the commutator. It is more convenient to derive this Green's function in momentum space. Through the Fourier transformation to obtain the spin Green's function in momentum representation, we get

$$G(q,t) = \langle\langle S^+_q(t); S^-(0,0)\rangle\rangle. \tag{4}$$

Employing the equation of motion to equation (4) makes the spin Green's function calculation reduced to

$$i\frac{d}{dt}G(q,t) = \varphi + \langle\langle[S^+_q(t), H]; S^-(0,0)\rangle\rangle, \tag{5}$$

where function φ comes from the derivative of time for step function. Following commutation rules in RPA (random phase approximation) are we used in this paper, there are

$$\begin{aligned} [S^+_q, S^-_k] &= \frac{2}{N}\sum_\ell e^{i(q-k)\cdot R_\ell}S^z_\ell \\ &= 2\langle S^z\rangle\delta_{q,k}, \end{aligned} \tag{6}$$

$$[S^+_q, S^z_k] = -\frac{1}{N}S^+_{q+k}, \tag{7}$$

$$[\sigma^+_k, \sigma^-_{k'}] = \sum_p (c^\dagger_{p+k,\uparrow}c_{p-k',\uparrow} - c^\dagger_{p+k'+k,\downarrow}c_{p,\downarrow}) \tag{8}$$

and

$$[\sigma^+_k, \sigma^z_{k'}] = -\frac{1}{N}\sigma^+_{k+k'}, \tag{9}$$

where $\langle S^z\rangle$ is the expectation value of magnetization for local spin. Employing these commutation rules to equation (5) results to the equation

$$\begin{aligned} \omega G(q,\omega) &= \varphi - cJ\langle S^z\rangle\langle\langle\sigma^+_q; S^-(0,0)\rangle\rangle \\ &\quad + cJ\langle\sigma^z\rangle\langle\langle S^+_q; S^-(0,0)\rangle\rangle, \end{aligned} \tag{10}$$

where $\langle\sigma^z\rangle$ is the expectation value of the magnetization for itinerant carriers. We found the derivation process for local spin Green's function resulting to another new Green's function $\zeta(k,\omega) = \langle\langle\sigma^+_k; S^-(0,0)\rangle\rangle$ in the meanwhile. It is clearly the new resulted Green's function reveals the physics that the exchange interaction between local spins needs the itinerant carriers' mediation. Therefore we need to calculate the new Green's function by the same way,

$$i\frac{d}{dt}\langle\langle\sigma^+_q; S^-(0,0)\rangle\rangle = \langle\langle[\sigma^+_q, H]; S^-(0,0)\rangle\rangle. \tag{11}$$

The commutator in equation (11) results to

$$[\sigma^+_q, H_k] = \sum_p(\varepsilon_p - \varepsilon_{p+q})c^\dagger_{p+q,\uparrow}c_{p,\downarrow} \tag{12}$$

$$[\sigma^+_q, H_J] = -\frac{cJ}{2}\sum_k[\sigma^+_q, \sigma^-_k]S^+_k - cJ\sum_k[\sigma^+_q, \sigma^z_k]S^z_k, \tag{13}$$

where ε_k is the electrons kinetic energy with momentum k. Apply the Fourier transformation and RPA for carriers density to the new Green's function ζ resulting to

$$\omega\zeta(q,\omega) = \sum_p(\varepsilon_p - \varepsilon_{p+q})\langle\langle c^\dagger_{p+q,\uparrow}c_{p,\downarrow}; S^-(0,0)\rangle\rangle - \frac{cJ}{2}\sum_p(\langle c^\dagger_{p+q,\uparrow}c_{p+q,\uparrow}\rangle - \langle c^\dagger_{p,\downarrow}c_{p,\downarrow}\rangle) \times G(q,\omega) + cJ\langle S^z\rangle\zeta(q,\omega). \quad (14)$$

Abstracting momentum p from $\zeta(q,\omega)$ in equation (14) results to a relation

$$\langle\langle c^\dagger_{p+q,\uparrow}c_{p,\downarrow}; S^-(0,0)\rangle\rangle = \frac{cJ}{2}\frac{\langle c^\dagger_{p,\downarrow}c_{p,\downarrow}\rangle - \langle c^\dagger_{p+q,\uparrow}c_{p+q,\uparrow}\rangle}{\omega - \varepsilon_p + \varepsilon_{p+q} - cJ\langle S^z\rangle} \times G(q,\omega). \quad (15)$$

Combining equations (10) and (14) results to a closed form of Green's function equation $G(q,\omega)$,

$$(\omega - cJ\langle\sigma^z\rangle + \frac{cJ^2}{2}\langle S^z\rangle\sum_p\frac{\langle c^\dagger_{p,\downarrow}c_{p,\downarrow}\rangle - \langle c^\dagger_{p+q,\uparrow}c_{p+q,\uparrow}\rangle}{\omega - \varepsilon_p + \varepsilon_{p+q} - cJ\langle S^z\rangle}) \times G(q,\omega) = \varphi, \quad (16)$$

where the $\langle c^\dagger_{p,\sigma}c_{p,\sigma}\rangle = f_{p,\sigma} = (\beta\varepsilon_{p,\sigma}+1)^{-1}$ is the carrier density for spin σ and $\beta = 1/K_BT$. The poles in equation (16) represent magnon excitations. In the diluted limitation, the minority is the itinerant carriers which could be seen as a free carrier gas with effective mass $m^* = 0.5m_e$ [9], where m_e is the free electron mass, and in the ferromagnetic state the majority of local magnetic ions produce an effective magnetic field causing both spins to split with Zeeman energy $cJ\langle S^z\rangle$. The kinetic energies of different spins are $\varepsilon_{k,\sigma} = \hbar^2k^2/2m^* \mp \sigma 1/2cJ\langle S^z\rangle$ respectively. Therefore the magnon excitation energy for each momentum q is

$$\omega_q = J\langle\sigma^z\rangle - \frac{cJ^2}{8\pi^2}\langle S^z\rangle \times \left\{\int_0^\infty \frac{km^*f_{k\uparrow}}{\hbar^2q} \times \ln\left(\frac{\omega_q - cJ\langle S^z\rangle - \frac{\hbar^2q^2}{2m^*} - \frac{\hbar^2kq}{m^*}}{\omega_q - cJ\langle S^z\rangle - \frac{\hbar^2q^2}{2m^*} + \frac{\hbar^2kq}{m^*}}\right)dk + \int_0^\infty \frac{km^*f_{k\downarrow}}{\hbar^2q}\ln\left(\frac{\omega_q - cJ\langle S^z\rangle + \frac{\hbar^2q^2}{2m^*} - \frac{\hbar^2kq}{m^*}}{\omega_q - cJ\langle S^z\rangle + \frac{\hbar^2q^2}{2m^*} + \frac{\hbar^2kq}{m^*}}\right) \times dk\right\}. \quad (17)$$

Finally, we could utilize the Callen's arbitrary spin formula,

$$\langle S^z\rangle = \frac{[S-\Phi(S)][1+\Phi(S)]^{2S+1} + [S+1+\Phi(S)][\Phi(S)]^{2S+1}}{[1+\Phi(S)]^{2S+1} - [\Phi(S)]^{2S+1}} \quad (18)$$

to obtain the magnetization values, where $\Phi(S) = \frac{1}{N}\sum_q(\exp(\beta\omega_q)-1)^{-1}$ is the magnon number.

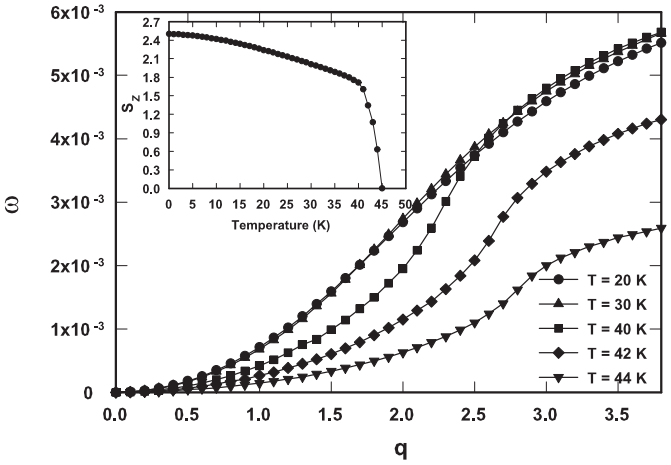

Fig. 1. We take the exchange coupling and the effective mass are fixed at typical values $J = 0.15$ eV nm^3, $m^* = 0.5m_e$ and the ratio of itinerant and localized spin densities fixed at $c^*/c = 0.1$ to calculation resulting to $T_C = 45$ K. In the insert of the figure shows a sharp drop of magnetization in the vicinity of T_C. In the sharp drop region the magnon shows a softening effect.

3 Results and discussions

The anomalous temperature dependence of magnetization of sharp drop in the vicinity of T_C represents in our theoretical result as shown in the insert of Figure 1. is consistent with experimental result implying an important interaction existing in the magnetization collapse region. According to the RKKY mechanism the magnetism is established by itinerant carriers mediation. The sharp drop of magnetization reveals a possibility of carrier-magnon decoupling in the vicinity of T_C. This decoupling effect reduces the effective magnetic interaction and the magnetization disappears in decoupling completely.

The dispersion of spin wave derived from the conventional spin wave theories [14] is temperature independent, which is an intrinsic characteristic for many kinds of magnets. The Figure 1 exhibits the dispersion of magnon from our theoretical calculation showing a temperature independent dispersion at far from T_Cs, which reveals a result hat the temperature independence of normal spin wave existing at robust magnetism region, meanwhile it reveals an obvious magnon softening effect in the vicinity of T_C leading to the magnetization falling down sharply. Interestingly, this softening effect starts from small magnon momentum qs then extending to whole dispersion region eventually. From the conventional spin wave theory as the $q \ll$, the magnon dispersion relation has $\omega(q) = Dq^2$, where the stiffness constant $D \propto J^{'}$ and $J^{'}$ is the magnetic coupling integral between two separated spins. From the linear response theory we have derived [15] before, the coupling $J^{'}$ is $\propto J^2$, where the J is the coupling between itinerant spins and local spins. Therefore this softening effect results to D decreasing, which reveals a fact that the effective coupling J reduces and gives an implication with carrier-magnon decouple in system.

In the conclusion, by our theoretical study the anomalous magnetization sharp drop in the vicinity of T_C comes from the magnon softening effect, and this softening effect possibly comes from the carrier-magnon decoupling.

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