Spin wave spectrum in disordered Heisenberg spin systems

E.-V. MACOCIAN[*], S. FILIP

Department of Physics, University of Oradea, Oradea, Romania

We calculate the spin wave spectrum for three phenomenological diluted Heisenberg-like spin models that are representative for a qualitative description of the magnetic properties of diluted magnetic semiconductors. We investigate the combined effect of frustration induced by anisotropy and simulated annealing on the spin wave spectrum and temperature dependence of the magnetization. We analyze short-range model characterized by a constant coupling between the spins and a low coordination number, long-range model with a increased connectivity and RKKY model with an oscillating sign in the exchange coupling.

1. Introduction

Recently ferromagnetism was discovered in $Ga_{1-x}Mn_xAs$ semiconductor [1], with a critical temperature as high as $T_c = 110\,K$, and has initiated a tremendous amount of experimental and theoretical work to explain the magnetic properties of this simple diluted magnetic semiconductor (DMS). Until now a large body of experimental work focused on the study of magnetic properties to explain the origin of ferromagnetism of this semiconductor. The consequences of understanding the physics behind these DMS's may leads to the developing of new materials which will combine the advantage of semiconducting devices with the new features due to the possibility of controlling the magnetic state. Measurements of magnetoresistance and concentration effects [2] on T_c strongly suggest that ferromagnetism in $Ga_{1-x}Mn_xAs$ has the origin in Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction between the Mn spins, mediated by the holes from the valence band of the bulk system. The doping dependence of T_c may be understood as arising from the modification of the free hole concentration originally induced by the Mn impurities. The non-equilibrium method used for obtaining these DMS may lead to the formation of As-antisites and Mn-interstitial impurities, which play a minor role by partially compensating the hole concentrations and decreasing T_c. Because of the sensitivity to growing conditions and the strong valence band spin-orbit coupling the simplest magnetic properties such as temperature dependence of magnetization for DMS are completely different from the well known characteristics of itinerant electron ferromagnet. Besides properties of the $T = 0$, magnetization and Curie temperature were found to depend on post-growth annealing protocol [3]. Zaránd and Jankó [4] showed that within the RKKY approximation the spin-orbit coupling leads to anisotropic exchange

between magnetic spins. Other mechanism such as double-exchange mechanism based on d electron hopping [5], double-resonance mechanism [6], the Zener model description [7] have been proposed. In most of the previous theoretical calculations an effective interaction between the magnetic moments is constructed and the magnetic properties of the system are analyzed in the framework of a disordered Heisenberg-like spin system with or without anisotropy included. In this paper we consider three different phenomenological models that describe the interaction between the magnetic spins that are relevant for understanding the physics that governs DMS. The effect of simulated annealing and anisotropy on the spin wave spectrum is studied in detailed. We assume that the magnetic properties of the system can be modeled by an effective Hamiltonian [8] which explicitly includes the anisotropy in the system, but it is not the purpose of the present work to present a microscopic derivation of the effective Hamiltonian. Disorder effects are considered to play a major role in the overall magnetic behavior of DMS. To simulate annealing we introduced a screened Coulomb repulsion between the Mn ions and let them relax for sometime t_{rel} when we use $T=0$ Monte Carlo simulation. After the relaxation process for the positions of magnetic moments we used Metropolis Monte Carlo calculations to generate the ground state at $T=0$ which in most cases is non-collinear. For completion we also do finite temperature Monte Carlo calculations and compute the temperature dependence of the magnetization for each model. We study unrelaxed systems where the initial positions of the Mn ions, which were randomly generated remain fixed, together with the fully relaxed systems where the relaxation time is large. For these systems the Mn ions form a regular bcc lattice with some defects, when periodic boundary conditions are used. We have to emphasize that the use of periodic boundary conditions is absolutely necessary, otherwise the Mn ions move to the sides of the finite lattice in such a way to minimize the

energy (maximize the distances between the ions) and this effect leads to unphysical arrangement of the lattice. The plan of the paper is as follows: In sec II we present the main theoretical approach and approximations used. In sec. III we present the results for the spin wave spectrum together with the temperature dependence magnetization curves for relaxed and unrelaxed systems for isotropic and anisotropic samples. Section IV is dedicated to conclusions.

2. Theoretical model

In this section we present the main theoretical framework for computing the spin wave spectrum of a disordered system with anon-collinear ground state. Our system consists of a fixed number of randomly arranged spins on a fcc lattice. By using a fcc lattice and randomly placed spins we try to model the bulk system $GaAs$ when Mn ions substitute Ga atoms. The spins are treated as classical variables with unit length. Considering that the Mn ions have the spin $S=5/2$ this is equivalent to rescaling the coupling constants. We'll use for study the following Hamiltonian:

$$H = -\sum_{i,j}\sum_{\alpha,\beta} S_i^\alpha J^{\alpha\beta}\left(\vec{R}_i - \vec{R}_j\right)S_j^\beta - \sum_i \mu_B g\vec{H}\vec{S}_i \qquad (1)$$

In eq. (1) S_i^α is the $\alpha(x,y,z)$ component of the spin at the site index i, $J^{\alpha\beta}\left(\vec{R}_i - \vec{R}_j\right)$ is the exchange coupling, $\vec{H}$ is the magnetic field which in what follows will be considered along the $\hat{z}$ axis, and introduce the notation $h = \mu_B gH_z$. Throughout of our calculation we consider the following parameterization of the exchange coupling:

$$J_{ij} = J\left(\vec{R}_i - \vec{R}_j\right) = J\left(S_i^\Box S_j^\Box + \lambda S_i^\perp S_j^\perp\right)f\left(r\right) \qquad (2)$$

where parameter J incorporates the positional disorder in the model. Parameter λ controls the anisotropy in the system. For $0 < \lambda < 1$ spins prefer a parallel arrangement relative to the axis that join their positions and for $\lambda > 1$ spins prefer a perpendicular arrangement relative to the same axis. In all the models that we consider the angular part of the interaction is the same and is controlled by the anisotropy parameter λ. The only difference between them is reflected in the radial part of the coupling $f\left(r\right)$. We investigate the following interactions: $f\left(r\right) = \theta\left(R_c - r\right)$ (short range model SRM), $f\left(r\right) = 1/r^4$ (long range model LRM) and $f\left(r\right) = \left(\sin\left(2k_F r\right) - 2k_F r\cos\left(2k_F r\right)\right)/\left(2k_F r\right)^4$ (RKKY model). The short range model allow the interaction of a finite number of Mn spins with equal strength while the long range and RKKY model allow the interaction between all the spins in the lattice [8]. The

method that we present for the calculation of the spin wave spectrum (SWS) assumes that the ground state of the system at the mean field level is known. In our calculations the ground state is obtained by performing extended Monte Carlo calculation using Metropolis algorithm at $T = 0$. Finite temperature calculation were also performed for the ground state to obtain the temperature dependence of the magnetization and compute the critical temperature. Recently a different method using Tyablikov approximation was proposed for computing the SWS of DMS but only for a isotropic Hamiltonian [9]. In what follows we describe the method that we used for computing the eigenenergies and the eigenvectors of the SWS. The analysis start by taking into account that the ground state is not fully polarized therefore at each site we rotate the framework in such a way that the z-component of the spin $\vec{S}_i$ points into the $\hat{n}_i$ direction where $\hat{n}_i$ is the direction of the spin $\vec{S}_i$ in the ground state. By doing this, we have the decomposition:

$$\vec{S}_i = \tilde{S}_i^z \hat{n}_i + \tilde{S}_i^x \hat{e}_i^x + \tilde{S}_i^y \hat{e}_i^y$$

where the unit vectors are given in spherical coordinates as

$$\hat{n}\left(\theta,\phi\right) = \begin{pmatrix}\hat{n}_x \\ \hat{n}_y \\ \hat{n}_z\end{pmatrix} = \begin{pmatrix}\sin\theta\cos\phi \\ \sin\theta\sin\phi \\ \cos\theta\end{pmatrix}$$

$$\hat{e}^x\left(\theta,\phi\right) = \frac{1}{\sqrt{1-n_z^2}}\begin{pmatrix}\hat{n}_x\hat{n}_z \\ \hat{n}_y\hat{n}_z \\ \hat{n}_z^2 - 1\end{pmatrix} = \begin{pmatrix}\cos\theta\cos\phi \\ \cos\theta\sin\phi \\ -\sin\theta\end{pmatrix}$$

$$\hat{e}^y\left(\theta,\phi\right) = \frac{1}{\sqrt{1-n_z^2}}\begin{pmatrix}-\hat{n}_y \\ \hat{n}_x \\ 0\end{pmatrix} = \begin{pmatrix}-\sin\phi \\ \cos\phi \\ 0\end{pmatrix}$$

and $\tilde{S}_i^\alpha$ are the components of the $\vec{S}_i$ in the rotated frame. We are interested in computing the spin wave spectrum in the linear approximation therefore we neglect non-linear effects [10]. By using Holstein-Primakoff [11] transformation and the linear spin wave theory [12], retaining only terms up to the second quadratic order in the boson operators we have:

$$\tilde{S}_i^z = S - b_i^\dagger b_i$$
$$\tilde{S}^\dagger = \sqrt{2S}b_i \qquad (3)$$
$$\tilde{S}^- = \sqrt{2S}b_i^\dagger$$

Each spin is characterized by a unit vector that describes its direction in the local frame. We use $\left(\hat{e}^+, \hat{e}^-, \hat{n}\right)$ as the basis in the local frame with:

$$\hat{e}^\pm = \frac{1}{\sqrt{2}}\left(\hat{e}^x \pm \hat{e}^y\right)$$

By combining these expressions we can express the spin operator in terms of creation and annihilation operators as:

$$\vec{S}_i = \left(S - b_i^\dagger b_i\right)\hat{n}_i + \sqrt{S}\left(\hat{e}_i^+ b_i^\dagger + \hat{e}_i^- b_i\right)$$

In the linear approximation we need to expand the Hamiltonian up to quadratic order in the creation and annihilation operators for the spin waves $b_i^\dagger$ and b_i. By doing that we end-up with the following expression for the Hamiltonian:

$$
\begin{aligned}
H = H\left(b\dagger, b = 0\right) + h\sum_i \hat{n}_i \hat{z} b_i^\dagger b_i + \\
+ S\sum_{i,j} \hat{n}_i J_{ij} \hat{n}_j \left(b_i^\dagger b_i + b_j^\dagger b_j\right) - \\
- S\sum_{i,j}\left(\hat{e}_i^+ J_{ij}\hat{e}_j^- b_i^\dagger b_j + \hat{e}_i^- J_{ij}\hat{e}_j^+ b_i b_j^\dagger + \right. \\
\left. + \hat{e}_i^+ J_{ij}\hat{e}_j^+ b_i^\dagger b_j^\dagger + \hat{e}_i^- J_{ij}\hat{e}_j^- b_i b_j\right)
\end{aligned}
\tag{4}
$$

The time evolution for the creation operator is: $-i\dot{b}_i^\dagger = \left[H, b_i^\dagger\right]$. By introducing the following notations

$$
\begin{aligned}
f_i &= h\hat{n}_i\hat{z} + 2S\sum_{j\neq i}\hat{n}_i J_{ij}\hat{n}_j \\
g_{ij} &= 2S\hat{e}_j^+ J_{ij}\hat{e}_i^- \\
r_{ij} &= 2S\hat{e}_j^- J_{ij}\hat{e}_i^-
\end{aligned}
\tag{5}
$$

with g hermitian matrix, $\left(g = g^+\right)$, r symmetric matrix $\left(r = r^T\right)$ and f_i real $\left(f_i = f_i^*\right)$ the commutator is:

$$\left[H, b_i^\dagger\right] = f_i b_i^\dagger - \sum_j\left(g_{ji}b_j^\dagger + r_{ji}b_j\right) \tag{6}$$

Now we can diagonalize the Hamiltonian by a canonical transformation:

$$A(E) = \sum_i\left(\phi_{i,-}(E)b_i + \phi_{i,+}(E)b_i^\dagger\right)$$

by requiring that $\left[H, A(E)\right] = -EA(E)$. We end-up with the following eigenvalue problem:

$$
\begin{aligned}
-E\sum_{i,\mu}\phi_{i,\mu}(E) = -\sum_i f_i\left(\phi_{i,-}(E)b_i + \phi_{i,+}(E)b_i^\dagger\right) \\
+ \sum_j\left(g_{ji}^*\phi_{i,-}(E) - r_{ji}\phi_{i,+}(E)\right)b_j \\
- \sum_j\left(g_{ji}\phi_{i,+}(E) - r_{ji}^*\phi_{i,-}(E)\right)b_j^\dagger
\end{aligned}
$$

or in matrix form:

$$
\begin{pmatrix} E - \left(f - g^*\right) & -r \\ r^* & E + \left(f - g\right) \end{pmatrix}\begin{pmatrix} \phi_{i,-} \\ \phi_{i,+} \end{pmatrix} = 0
\tag{7}
$$

Next we want to find the orthogonality relations satisfied by the canonical operators $A(E)$ (E=energy) and wave functions $\phi_{i,\mu}(E)$. For the particular case of positive energy a compact expression for the annihilation operator is:

$$A(E > 0) = \sum_{i,\mu}\phi_{i,\mu}(E)b_i^\mu \tag{8}$$

where $\mu = \pm$, and we also introduced the notation $b_i^- = b_i$ and $b_i^+ = b_i^\dagger$. By calculating the commutator we have:

$$
\begin{aligned}
\left[A(E'), A(E)\right] &= \sum_{i,\mu,i',\mu'}\phi_{i,\mu}(E)\phi_{i',\mu'}(E')\left[b_i^\mu, b_{i'}^{-\mu'}\right] = \\
&= \sum_{i,\mu}\mu\phi_{i,\mu}(E')\phi_{i,\mu}(E) = \begin{cases} 1, & \text{if } E = E' > 0 \\ 0 & \text{otherwise} \end{cases}
\end{aligned}
\tag{9}
$$

and

$$\sum_i \left|\phi_{i,-}(E)\right|^2 - \left|\phi_{i,+}(E)\right|^2 = 1$$

In the case of negative energies (E<0) we can construct the wave function in a similar way by making the observation that in this case

$$\phi_{i,\mu}(E < 0) = \phi_{i,-\mu}(E > 0)$$

The normalization condition in this case is:

$$\sum_i \left|\phi_{i,-}(E)\right|^2 - \left|\phi_{i,+}(E)\right|^2 = -1$$

Combining negative and positive energy results we can write a compact expression for the orthogonality relations satisfied by the wave function as

$$\sum_{i,\mu}\phi_{i,\mu}^*(E')\phi_{i,\mu}(E) = sgn(E)\delta_{E,E'}$$

and for the completitude relation

$$\sum_E \phi_{i,\mu}(E)\phi_{i',\mu'}^*(E)sgn(E) = -\mu\delta_{\mu,\mu'}\delta_{i,i'}$$

The local spin wave operators b_i^μ can be expressed in term of canonical operators also:

$$b_i^\mu = -\sum_E \mu\, sgn(E)\phi_{i,\mu}^*(E)A(E) \tag{10}$$

Our task is to solve the eigenvalue equation (10) that gives us the spin wave energies for a given spin configuration. Through of our calculations we consider that the magnetic field is zero. As can be observed the spin wave spectrum is symmetric in the sense that both positive and negative energy $\pm E$ belong to the spectrum.

3. Results and discussions

In this section we present the results for the spin wave spectrum and for the temperature dependence of the magnetization for different strength of the anisotropy and
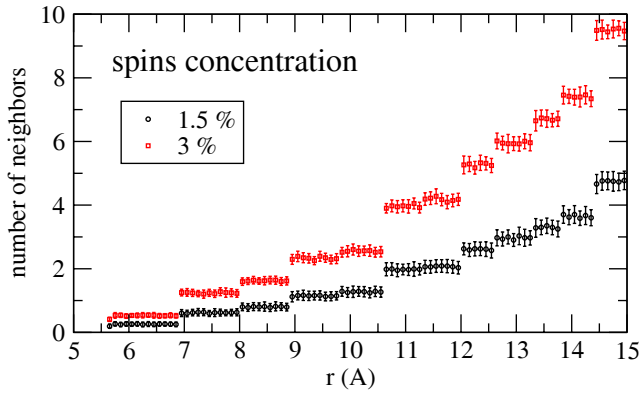
Fig. 1. Average number of neighbors inside a sphere of radius r for two spin concentrations.
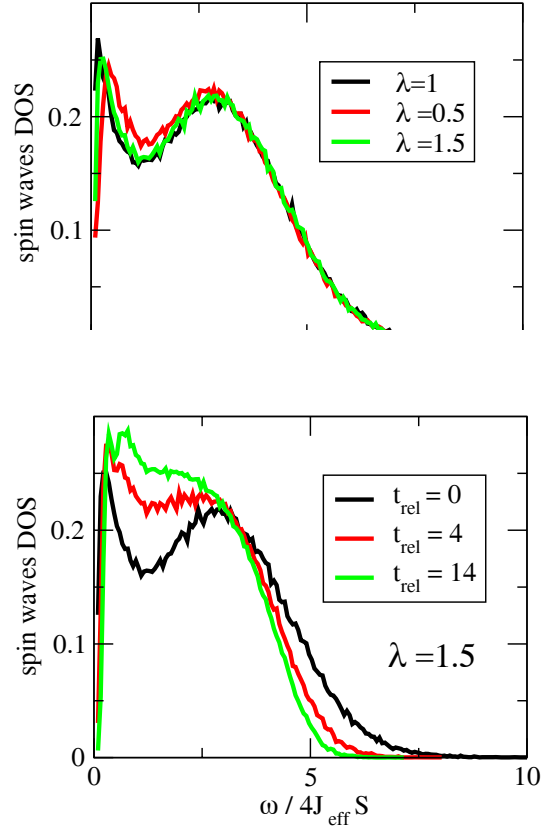

Fig. 2. Spin waves density of states for the short range model. Top: the effect of anisotropy is presented for a non-relaxed lattice with $t_{rel} = 0$. Bottom: The effect of annealing in a anisotropic system with preferred perpendicular alignment.

We did calculation for the isotropic case $(\lambda = 1)$ and for the anisotropic case with preferred parallel alignment $(\lambda = 0.5)$ and perpendicular alignment $(\lambda = 1.5)$. Unless otherwise specified our results are for 3% *Mn* concentration. We did calculation for 1.5 and 5% concentrations with similar results, so are not presented here.

Short range model

This model is characterized by a constant coupling between the spins and a cut-off range that characterize the extension of interaction. In our simulation we have fixed the value for the short range cut-off to 12 Å. However, the anisotropy coefficient rescales the coupling between the spins [8] and introduce an effective coupling which depends on the anisotropy parameter λ, and this has to be taken into account in the numerical calculations in order to get consistent results for the critical temperature. The critical temperature depends also on the number of neighbors in the system. In Fig. 1 we present the number of neighbors inside a sphere of a given radius for two different spin concentrations. This result was obtained as an average over a 500 samples.

The effect of low coordination number is felt also in the SWS. In Fig. 2 we present SWS and magnetization curves for isotropic and anisotropic systems for an unrelaxed sample. Frustration has a small effect on both temperature dependence of magnetization and on spin wave spectrum. The effect of simulated annealing time is more drastic. As we increase the relaxation time, the spin wave spectrum starts to shrink and extends up to the maximum energy that is allowed in the system for a spin wave in a perfectly ordered lattice. In this case an expression for the dispersion relation as function of the momentum $\vec{k}$ can be written:

$$E_k = 2J_{eff} Sz \left(1 - 1/z \sum_{\delta} exp \left(i \vec{k} \vec{\delta} \right) \right) \qquad (11)$$

Here, the sum is over the neighbors and z is the number of neighbors in the lattice. So the energy range allowed for the spin waves is $E \in [0.4\, J_{eff}\, S_z]$. For the particular case that we consider the number of neighbors for our fixed cut-off $R_c = 12\, \overset{\circ}{A}$ is $z \cong 4 \div 5$ when *Mn* concentration is fixed to 3%. This behavior can be understand by considering that as the relaxation time t_{rel} increase, the lattice becomes a bcc lattice with point defects and the upper limit of the spectrum moves to the

maximum allowed value for a perfect lattice. For unrelaxed samples the disordered effects broaden the spin wave spectrum by shifting it up to higher energies. The temperature dependence of magnetization is presented in Fig. 3. The anisotropy parameter λ does not have a strong influence on these curves also in agreement with the results of Ref.[8].

The relaxation time t_{rel} has a much stronger influence on the magnetization curves also. Two major effects are observed when increasing the relaxation time in both isotropic and anisotropic cases. First, an increase of magnetization at $T=0$ is observed so the ground state

Long range model

In this model a single spin is interacting with all the other spins in the lattice. However the coupling is decaying fast with the distance and this lead to a smaller criti

Fig. 4. Spin waves density of states for long range model. Top: The anisotropy effect is presented for a unrelaxed sample. Bottom: The effect of relaxation on the SWS for an anisotropic system with $\lambda = 1.5$.
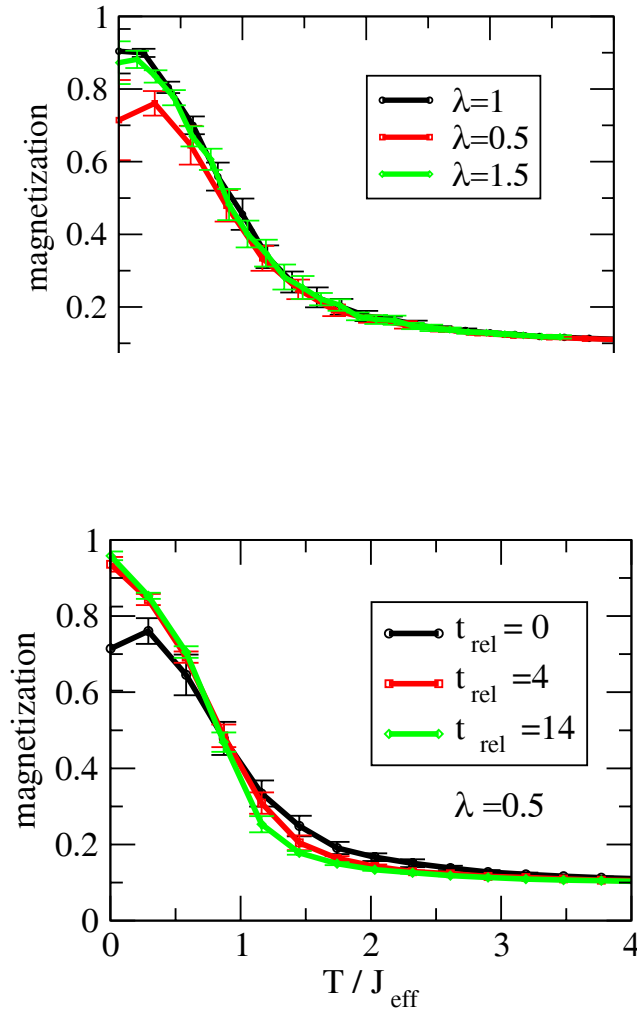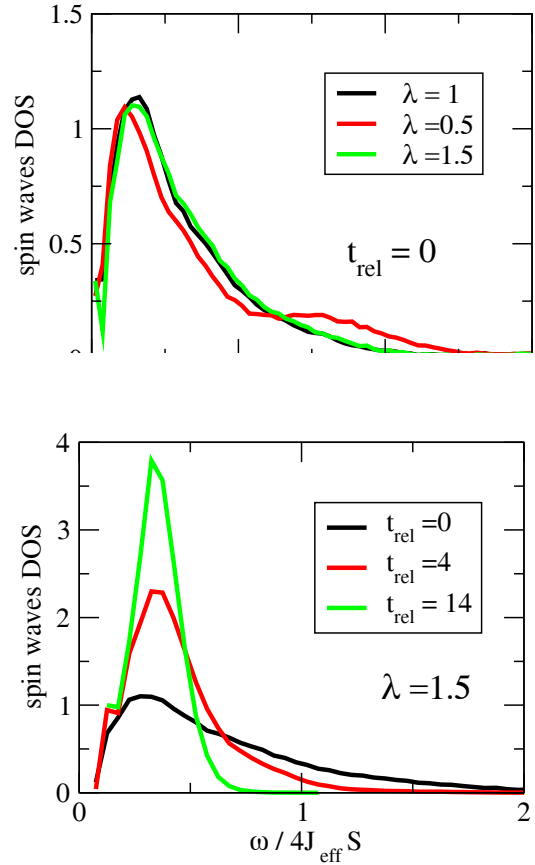

Fig. 3. Temperature dependence of magnetization for short range model. Top: the effect of anisotropy is presented on the magnetization curves for a un-relaxed lattice with $t_{rel} = 0$. Bottom: The effect of relaxation for a anisotropic lattice with parallel preferential alignment.

In Figs. 4, we present the result for the SWS in the case of long range model. Here the spectrum is much narrow as compared with the previous case and this is due entirely to the fast decay of the coupling. Interaction between spins separated by large distances generates a tail in the SWS which extends up to very large energies. The effect of anisotropy on SWS is small also. A much stronger effect is felt by the magnetization curves of a unrelaxed systems. For isotropic case the ground state is fully polarized at T=0 for both relaxed and unrelaxed samples. For unrelaxed anisotropic systems the ground state is no longer collinear and this effect is enhanced in the systems with parallel preferential alignment. For large enough relaxation times the ground state is becoming collinear also. Here an increase in t_{rel} leads to a decrease of T_c similar with the SR model.
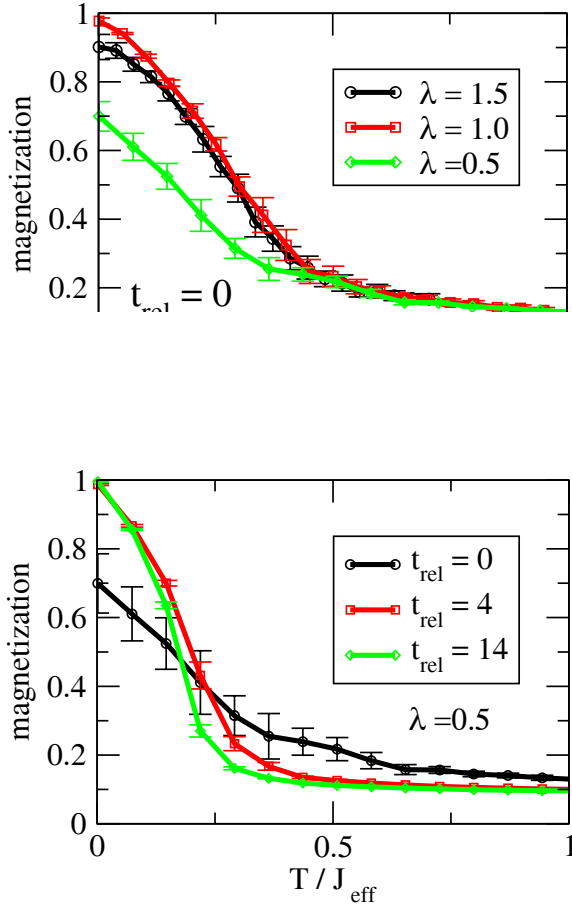
Fig. 5. Temperature dependence of magnetization, for the long range model. Top: Anisotropy effect for an un-relaxed sample. Bottom: Magnetization for different annealing times for a system with.
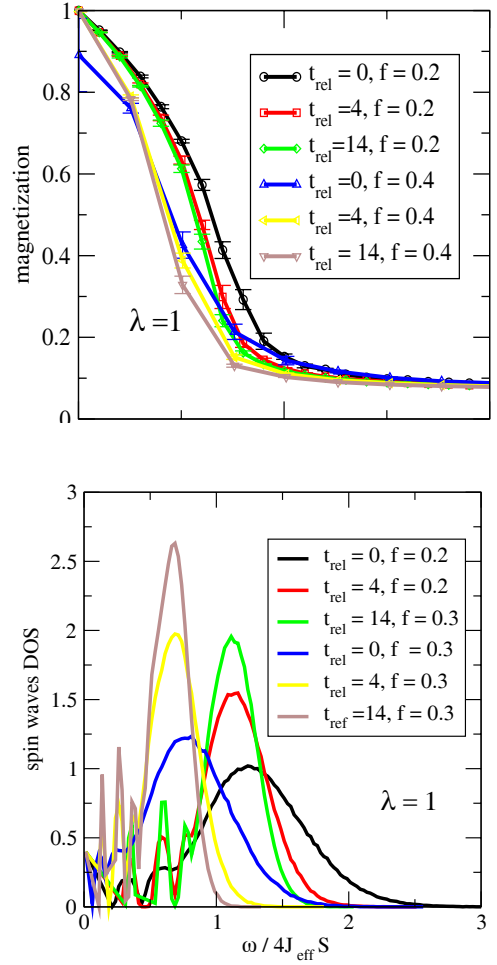
Fig. 6. Top: Temperature dependence of the magnetization for a RKKY model for different relaxation times and for different hole concentrations. Bottom: The effect of simulated annealing on the spin wave spectrum. Results are presented for hole fractions $f = 0.2$ and

$$f = 0.4 .$$

The effect of t_{rel} on SWS is to shrink the energy range of the spectrum as t_{rel} increases. This is due to the fact that Mn spins tries to maximize the distance between them, when relaxing the lattice. LRM has a large coordination number therefore the effects of frustration due to anisotropy are observed in $T = 0$ magnetization value. The effect of frustration on spin wave collective modes is less significant.

RKKY model

The final results are for RKKY model which allows the interaction between all the spin in the lattice with oscillating sign depending on the Fermi wave-vector k_F. This quantity is different for different hole fraction. In Fig. 6 we present the results for the temperature dependence of magnetization and for SWS corresponding to this model with different hole fractions. For 5% Mn impurities concentration the ground state at $T = 0$ is fully polarized as can be seen from the magnetization curves. As we decrease the concentration the ground state becomes non-collinear.

The overall behavior for SWS compared with the previous models is completely different due to the oscillating sign in the coupling. The same oscillating sign is responsible for removing the extended tail in the spectrum at large energies as compared with the long range model, and for the oscillating part of the spectrum at small energies. The upper energy limit moves to lower energies as we increase t_{rel}. Doping plays an important role in the sense that doubling the hole fraction from 0.2 to 0.4, the spectrum shrinks by a factor of 2 also, and this effect is entirely due to the modification of the Fermi wave vector amplitude k_F with doping.

4. Conclusions

We have study three phenomenological models which are representative for understanding the magnetic behavior of magnetic semiconductors. We used a anisotropic disordered Heisenberg model with different radial

interaction behaviors to model the magnetic properties of DMS.A comparison between the short-range, long-range and RKKY models may clarify the role played by the range of the interaction and also the importance of the exchange interaction sign. We have calculated the spin wave spectrum of these phenomenological models, and study the effect of anisotropy and of annealing on SWS. Our simulations demonstrate that annealing strongly affects the spin wave spectrum, and the magnetization curves. The increases of relaxation time leads to an arrangement of the localized *Mn* moments in a periodic lattice with defects, and the spin wave spectrum evolves toward the spectrum of a perfectly ordered lattice. On the other side anisotropy is less important for the SWS, a much stronger effect is observed on temperature dependence of magnetization for the unrelaxed samples. The unrelaxed samples have a non-collinear ground state and therefore the anisotropy plays an important role. Relaxing the lattice the ground state becomes practically collinear (see magnetization curves) and the anisotropy role gets weaker.

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[*]Corresponding author: macocian@uoradea.ro