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Full Length Research Paper

Photo - excitation and spin wave scattering effects on specific heat of the (Ga_{1-X}, Mn_X) as diluted magnetic semiconductor

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The effects of photo - excitation and spin wave scattering on magnetic specific heat of the diluted magnetic semiconductor (DMS) (Ga, Mn)As are theoretically studied using Green function formalism and employing a model system Hamiltonian consisting of magnons, photons and an interaction of magnons with photons. At very low temperatures, the DMS is found to have anomalous magnetic specific heat. This could be interpreted as the material is in non - equilibrium situation due to fluctuation effects undergoing heat losses. The specific heat is found to decrease with increase in magnetic impurity concentration and further decreases in the presence of the magnon-photon interaction. On the other hand, in the presence of magnon-scattering it is shown to increase rapidly at relatively higher temperatures.

Key words: (Ga, Mn) As DMS, photo - excitation, spin wave scattering, magnetization, heat capacity.

INTRODUCTION

Recently, the diluted magnetic semiconductor (DMS) (Ga, Mn)As has attracted intense research interest for the purpose of spintronic application due to the special optical property, direct band gap and large carrier density. The material is believed to circumvent difficulty of combining data processing and mass storage facilities in a single crystal, besides solving non-volatility problems ($\breve{Z}$ uti c' et al., 2004; Timm, 2003; Wolf, 2000). The concentration x, of Mn that substitutes for a fraction of Ga in the compound is known to contribute large amount of magnetic moments and holes to the system (Sanvito and Hill, 2002; Ohino et al., 1996). The holes have been understood as mediating ferromagnetic interaction between the localized magnetic moments, in which the interaction could be described as the Ruderman-Kittel-Kasuya-Yosida (RKKY)-type (Munekata et al., 1989; Kasuya, 1956). Though spintronic devices should operate at room temperature the highest ferromagnetic transition temperature T_C obtained to date is nearly 173 K at $x \approx 0.08$ (Wang et al., 2005; Das Sarma et al., 2003). This was an obstacle for application, and initiated for further search of new mechanism of enhancing magne-tization and raise T_C where photo-excitation has been found to be useful (Dalpian and Wei, (2005); Munekata, 2005; Fernandez-Rossier et al., 2004; Oiwa et al., 2002; Piermarocchi et al., 2002; Munekata et al., 1997; Koshihara et al., 1997).

The role of photo - excitation in magnetization of the (Ga, Mn) As DMS thin films was studied by Oiwa et al. (2002), following the pioneer work of Koshihara et al. (1997). They suggested that the generated spin - polarized carriers (holes) could change the orientation of ferromagnetically coupled Mn spins depending on the light ellipticity. This is indicative of the fact that the carrier mediated spin exchange is the novel mechanism to establish a strong tie between light, Mn and hole spins (Munekata, 2005). Moreover, a specific heat of magnetic ions for a self - consistent spin wave approach has been calculated and shown to be proportional to $T^{3/2}$ at low temperature, and found to decrease with increase in impurity concentration (Konig et al., 2000). However, this could not describe the situation in high temperature ferromagnetism, and requires new mechanism of understanding. In the present work we study effects of

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photo-excitation and magnon-scattering on magnon specific heat of the diluted magnetic semiconductor (Ga,Mn)As. Starting with a model Hamiltonian describing the system and using the method of double-time temperature depen-dent Green function (Zubarev, 1960) the average number of magnons is derived, which helps us to determine specific heat, C_{mag}.

FORMULATION OF THE PROBLEM

The Hamiltonian consists of magnon, photon and magnon-photon interaction energies, written as

$$H = H_{mag} + H_{phot} + H_{mag-phot} \tag{1}$$

Where;

$$H_{mag} = \sum_k \omega_k b_k^+ b_k + ZxJ_{nm}\zeta(k_1,k_2,k_3,k_4) \tag{2}$$

$\sum_k \omega_k b_k^+ b_k$ represents free magnon energy, where $\omega_k = 2xJ_{nm}Sa^2k^2 + g\mu_B B$ is the free magnon dispersion in which J_{nm} is the exchange integral between spins localized at sites n and m, S represents localized spins per atom, a is lattice constant, k is magnon wave vector, g is the Lande' g-factor, μ_B is the Bohr magneton, B is magnitude of applied field, $b_k^+(b_k)$ denotes the magnon creation (annihilation) operator, x is impurity concentration given by $x = N_{imp}/N$ in which N_{imp} is number of impurities randomly distributed on lattice of N sites (Bouzerar, 2007; Jungwirth et al., 2005). Using Holstein - Primakoff (HP) bosons (Auerbach, 1994), and coarse graining the spin density S_n (spin at site n) can be written as,

$$S_n^+ = (\sqrt{2xS - a_n^+ a_n})\, a_n, \tag{3}$$

$$S_n^- = (a_n^+\sqrt{2xS - a_n^+ a_n}), \tag{4}$$

and

$$S_n^z = xS - a_n^+ a_n, \tag{5}$$

Since the spin deviations are not localized to a particular lattice sites but propagate throughout, it is necessary to use Fourier variables, which are given by

$$a_n = \frac{1}{\sqrt{N}}\sum_k e^{-ik\cdot r_n} b_k;\ a_n^+ = \frac{1}{\sqrt{N}}\sum_k e^{ik\cdot r_n} b_k^+; \tag{6}$$

Where the Fourier transformation variables also satisfy the bosonic relations $[b_k, b_{k'}^+] = \delta_{kk'}$. On the other hand, $ZxJ_{nm}\zeta(k_1,k_2,k_3,k_4)$ represents magnon scattering part of the Hamiltonian, where

$$ZxJ_{nm}\zeta(k_1,k_2,k_3,k_4) = \frac{1}{4N}\sum_{k_1,k_2,k_3,k_4}\Delta(k_1+k_2-k_3-k_4) \times b_{k_1}^+ b_{k_2}^+ b_{k_3} b_{k_4}(\gamma_{1+2-3} + \gamma_{2-3-4} + \gamma_{-4} - 4\gamma_{1-4}), \tag{7}$$

K_1, k_2, k_3 and k_4 represents magnon wave vectors, Z the number of nearest neighbour atoms in which $\gamma_k = \frac{1}{Z}\sum_\delta e^{k\cdot\delta}$ with $\gamma_1 = \gamma_{k_1} = (1 - \frac{k_1^2\delta^2}{6})$, $\gamma_{1-4} = \gamma_{k_1}\gamma_{-k_4}$ and so on. This is based on the assumption that $k\delta << 1$, and δ is supposed to join the central atom to its nearest neighbours. The second term in the right hand side of Equation (1),

$$H_{phot} = \sum_k \lambda_k d_k^+ d_k \tag{8}$$

Represents free photon energy, where $d_k^+(d_k)$ is photon creation (annihilation) operator, $\lambda_k = ck$ is dispersion of photon and c is speed of light in free space. The third term in the right hand side of Equation (1),

$$H_{mag-phot} = \sum \chi_k(d_{-k}^+ b_k + d_{-k} b_k^+) \tag{9}$$

Represents the magnon-photon interaction energy, in which $\chi_k = g\mu_B\left(\frac{S\mu_0 ck}{4V}\right)^{\frac{1}{2}}$ denotes magnon - photon coupling strength, μ_0 permeability of free space, and V volume of the radiation field cavity (Scully et al., 1997). Therefore, making use of the model Hamiltonian magnetization and ferromagnetic transition temperature of the system under consideration are examined.

MAGNON HEAT CAPACITY

In order to estimate the C_{mag} of the diluted magnetic semiconductor, it is essential to find the average number of magnons and dispersion using Green function formalism (Zubarev, 1960). The equation of motion for the Green function is, thus, written as

$$\in_k << b_k ; b_{k'}^{+} >>= \frac{< [b_k , b_{k'}^{+}] >}{2\pi} + << [b_k , H]; b_{k'}^{+} >> \quad (10)$$

Where; $\in_k$ is total dispersion of the system. Computing the commutation relation by substituting Equation (1) in (10) and applying the random phase decoupling approximation,

$$\left(\in_k - \omega'_k - \frac{\chi_k^{\ 2}}{\in_k - \lambda_{-k}} \right) << b_k ; b_{k'}^{+} >>= \frac{\delta_{kk'}}{2\pi}, \quad (11)$$

Where;

$$\omega_k = xZJ_{nm}Sa^2 \left(\frac{1}{6} - \frac{1}{SN} \sum_{k_1} < n_1 > \frac{k^2{}_1\delta^2}{36} - \sum_{k_2} < n_2 > \frac{k^2{}_2\delta^2}{36} \right) + g\mu_B B \quad (12)$$

is magnon dispersion containing the spin wave scattering factor with out inclusion of the light - matter coupling in which $< n_1 >$ and $< n_2 >$ are average number of magnons with wave vector k_1 and k_2 respectively. Since $\sum_{k_1} < n_1 > k_1^{\ 2} = \sum_{k_1} < n_2 > k_2^{\ 2}$, Equation (12) reduces to

$$\omega'_k = xZJ_{nm}Sa^2 \left(\frac{1}{6} - \frac{1}{18SN} \sum_{k_1} < n_1 > k^2 \delta^2 \right) + g\mu_B B. \quad (13)$$

The term $\sum_{k_1} < n_1 > k_1^{\ 2}$ can be evaluated as in reference (Kittel, 1987), leading to $\omega'_k = Ak^2$ for $B = 0$ where;

$A = 2xJ_{nm} Sa^2 \left(1 - \Omega_0 T^{5/2}\right)$ in which $\Omega_0 = \frac{0.15 k_B^{\ 5/2}}{SNa^3 \pi^2 (2xSJ_{nm})^{5/2}}$ is a term appeared as coefficient of $T^{5/2}$ in the part contributing to the scattering and, hence, $\Omega_0 T^{5/2}$ is referred to as scattering factor for clarity as it results from the magnon scattering where $k^2\delta^2 = k^2 a^2$. For $k' = k$, Equation (11) can be written in a simplified form as,

$$<< b_k ; b_{k'}^{+} >>= \frac{\delta_{kk'}}{2\pi \left(\in_k - \omega'_k - \frac{\chi_k^{\ 2}}{\in_k - \lambda_{-k}} \right)}, \quad (14)$$

From which poles of the Green function can be found and expressed as $\in_k = \omega'_k + \frac{\chi_k^{\ 2}}{\in_k - \lambda_{-k}}$. Applying straight forward mathematics, $\in_k$ reduces to

$$\in_k \cong \frac{1}{2} (\omega'_k + \frac{2\alpha^2}{c}) \cdot \quad (15)$$

$\alpha^2 = \frac{c\mu_0 Sg^2 \mu_B^{\ 2}}{4V}$ is the photon - magnon coupling constant that depends on the volume V of the optical resonator (Scully et al., 1997).

Employing the double - time Green function formalism it is possible to calculate the number of magnons excited at temperature T. This can be done starting with writing the correlation function, and making use of Equation (14). Hence,

$$< b_{k'}^{+} b_k >= \lim_{\varepsilon \to 0} \int_{-\infty}^{\infty} \frac{e^{-iE(t-t')}}{e^{\beta E} - 1} \left(<< b_k ; b_{k'}^{+} >>_{E+i\varepsilon} - << b_k ; b_{k'}^{+} >>_{E-i\varepsilon} \right) dE \quad (16)$$

Where the Dirac identity gives

$$<< b_k ; b_k^{+} >>_{E+i\varepsilon} = \frac{1}{2\pi [(E + i\varepsilon) - \in_k]} = \frac{1}{2\pi} \left[\mathrm{P} \frac{1}{E + \in_k} - i\pi\delta (E - \in_k) \right] \quad (17)$$

and

$$<< b_k ; b_k^{+} >>_{E-i\varepsilon} = \frac{1}{2\pi [(E - i\varepsilon) - \in_k]} = \frac{1}{2\pi} \left[\mathrm{P} \frac{1}{E - \in_k} + i\pi\delta (E - \in_k) \right] \quad (18)$$

in which P is the principal part of the integral. Substitution of Equations (17) and (18) in to (16), and using equal time correlation, $t' = t$, gives the number;

$$< n_k >= \frac{1}{e^{\beta \in_k} - 1} \quad (19)$$

Where; $\beta = \frac{1}{k_B T}$.

The expression for magnon specific heat C_{mag} of the diluted magnetic semiconductor system would be obtained from internal energy of magnon U,

$$U = \sum_k \in_k < n_k >_T \quad (20)$$

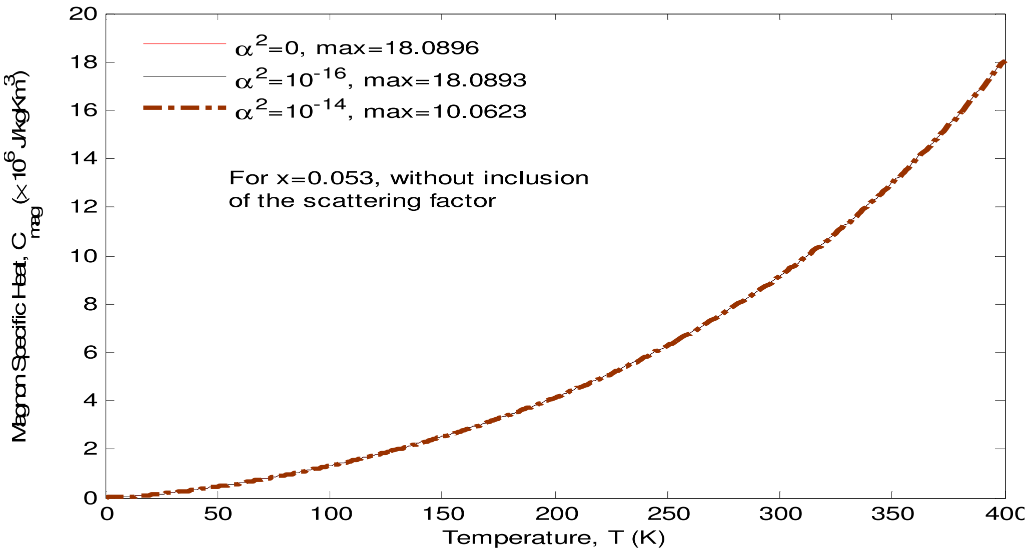

Figure 1. Magnon heat capacity vs. temperature for $x = 0.053$.

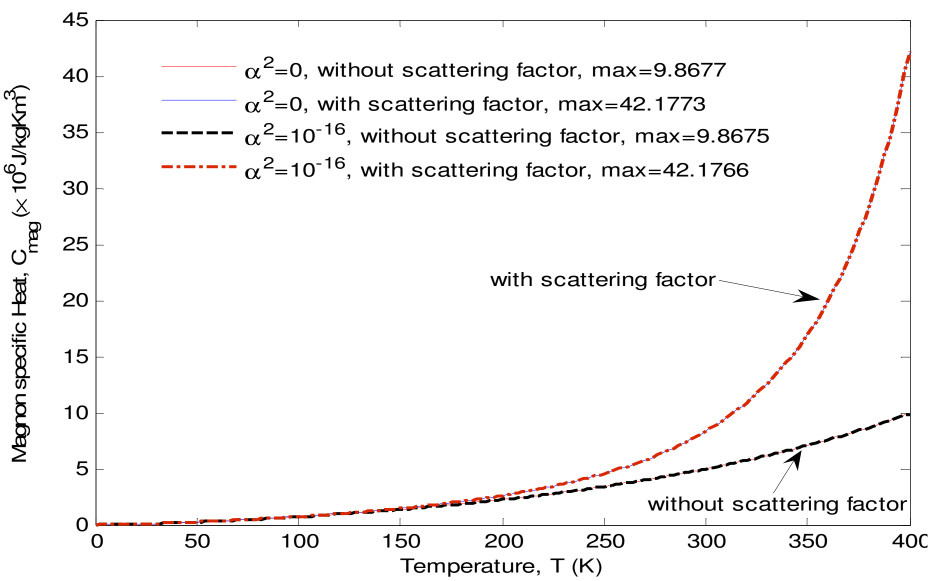

Figure 2. Magnon heat capacity vs. temperature for x = 0.08.

Where n_k is given by Equation (19). Hence,

$$C_{mag} = \frac{1}{(2xJ_{nm}Sa^2)^{3/2}(1-\Omega_0 T^{5/2})^{3/2}}\left(0.325k_B^{5/2}T^{3/2} - 0.0621\frac{\alpha^2 k_B^{3/2}}{c}T^{1/2}\right)$$
$$+\frac{1}{(2xJ_{nm}Sa^2)^{3/2}(1-\Omega_0 T^{5/2})^{5/2}}\left(0.325k_B^{5/2}T^4 - 0.0621\frac{\alpha^2 k_B^{3/2}}{c}T^3\right) \qquad (21)$$

According to Equation (21), the magnetic specific heat would change with change in the optical resonator cavity and spin wave scattering effects. This is explained by varying the photon-magnon coupling constant and inclusion of the scattering factor, as shown in Figures 1 and 2, where the maximum values are indicated for temperature of 400K. These demonstrate that, the presence of magnon - photon coupling constant would decrease the magnon specific heat resulting in enhanced magnetization. Further scrutiny of these graphs shows that there could be an unusual, negative specific heat for larger coupling strength at very low temperatures. This can be shown for $\alpha^2 = 10^{-13}$ and $\alpha^2 = 10^{-12}$.

The appearance of such unphysical result is perhaps

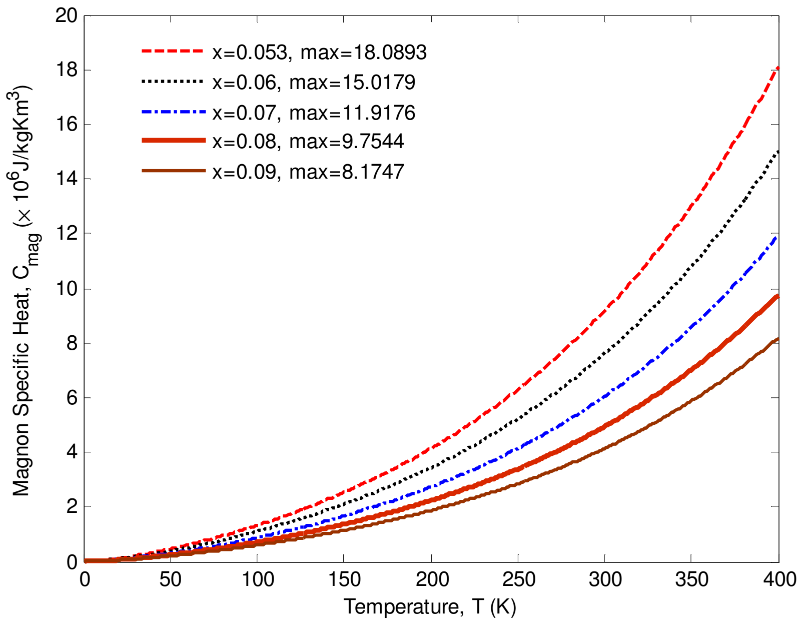

Figure 3. Magnon heat capacity vs. temperature for $\alpha^2 = 10^{-16}$ without inclusion of spin wave scattering factor.

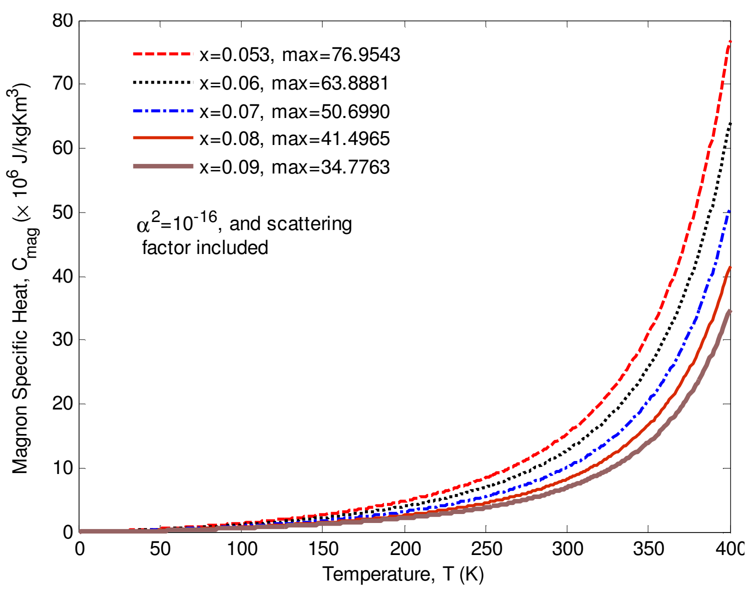

Figure 4. Magnon heat capacity vs. temperature for $\alpha^2 = 10^{-16}$ with inclusion of spin wave scattering factor.

due to confinement, non -equilibrium situation, and fluctuation effects in the radiation field cavity. Such anomaly was also observed in experimental study of the heat capacity of $Zn_{0.98}Co_{0.02}O$ and tentatively attributed to a change in the magnetic properties of the substance (Gavrichev et al., 2009). In Figures 1 and 2 presence and absence of spin wave scattering is compared, in which the specific heat is found to increase with inclusion of the factor contributing to the scattering and decrease with increase in magnon - photon coupling parameter. Figures 3 and 4 show the decrease in magnon specific heat with increase in Mn concentration in the DMS, indicating an interaction between the impurity ions in which, when the exchange interaction is sufficiently strong, magnetic impurity bands could split from the host band. Carriers in the magnetic impurity band mainly stay at magnetic impurity sites and coupling between the carrier spin and the localized spins is very strong (Takahashi and Kubo, 2003). The result is in agreement with what Konig and MacDonald, (2000) and Twardowski et al., (1991) have obtained for diluted magnetic semiconductors ferromagnetism at lower temperatures and the situation for $Zn_{1-x}Fe_xS$ DMS, respectively.

CONCLUSION

The radiation and spin wave scattering are shown to affect ferromagnetism of the diluted magnetic semiconductor (Ga, Mn)As. In inclusion of these factors, especially the magnon - photon coupling energy, we observed anomalous magnetic specific heat at very low temperatures. The phenomenon of negative heat capacity is also evolved in other compounds (Schmidt et al., 2001). These effects look pronounced in the absence of spin wave scattering, and might be due to immediate loss of absorbed heat in which the system is probably in non - equilibrium. With inclusion of spin wave scattering the heat capacity seems relatively to decrease with increase in temperature, compared to that with both photon and scattering absent.

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