Enhancement of $T_C$ of the (Ga,Mn) As diluted magnetic semiconductor by photoinduced exchange coupling of the Mn$^{2+}$ spins

Chernet Amente and P. Singh

Department of Physics, Addis Ababa University, P. O. Box 1176, Addis Ababa, Ethiopia.

Accepted 30 April, 2010

Effect of photo-excitation on the ferromagnetic transition temperature $T_C$ of the diluted magnetic semiconductor (DMS) (Ga, Mn)As is theoretically studied. The model Hamiltonian used to describe the system consists of interaction of localized Mn$^{2+}$ spins with radiation field as appropriate to ferromagnetic and/or spin wave resonance. Using second order perturbation theory, an expression for photoinduced exchange coupling energy between the Mn spins is obtained. It is argued that the coupling energy could be significantly large to provide very large $T_C$ at/near spin wave resonance. The intensity and energy of the light can further assist enhancement of $T_C$ useful for high temperature spintronic devices. An analogy with carrier mediated Ruderman-Kittel-Kasuya-Yosida (RKKY) type interaction with magnetic ions is also established.

Key words: Spintronics, (Ga,Mn)As, exchange coupling, photo-excitation, resonance, ferromagnetism, DMS.

INTRODUCTION

Photoinduced ferromagnetism has been observed recently in (In,Mn)As (Koshihara et al., 1997; Munekata et al., 1997). The mechanism is believed to be due to photo-excitation generated extra carriers/holes which mediate ferromagnetic order. This was achieved by impinging light on magnetic impurity doped semiconductor, (In,Mn)As, where the concentration of holes was kept lower than required for ferromagnetism. Upon light irradiation, induced photo-generated holes mediate exchange interaction to producing ferromagnetic order (Kaminski and Das Sarma, 2003) resulting in the hysteresis loop confirming ferromagnetism and magnetization (Koshihara et al., 1997).

Among the III-V DMSs, (Ga,Mn)As is the most studied, due to its good optical property and application in which high speed is required (Neamen, 1992). The band gap of GaAs is direct (Bouzerar, 2007) closely below the energy range of visible radiation, making (Ga,Mn)As suitable for the fabrication of efficient optical devices where the phenomena is controlled by spin interactions in the system (Oiwa et al., 2002). In such diluted magnetic semiconductors, hole-spin and Mn spin subsystems are thought to couple through p-d exchange, hence, alteration of either of these two subsystems cause significant influence on the entire system. This can be achieved by the carrier spin injection through the adequate inter-band optical excitation where in (Ga,Mn)As optical-absorption is known to occur in the visible range. However, to what extent the injected carrier spins modify the strength of p-d exchange interaction is the most inspiring question currently (Jungwirth et al., 2006; Dietl et al., 2002).

Intrinsically, Mn is known to contribute holes and localized magnetic moments to the system that could produce ferromagnetic order below transition temperature as high as 173 K at concentration $x$ -0.08 (Wang et al., 2005; Jungwirth et al., 2005; Das et al., 2003). The success has been made possible by the technological progress in controlling crystallographic quality of the materials, such as, reducing the number of unintentional charge and moment compensating defects through optimized growth and post-growth annealing procedures (Jungwirth et al., 2005). On the other hand, the impurity correlations are known to have only small effects on $T_c$ with the neutrally correlated random disorder producing the nominally highest $T_c$ (Priour et al., 2006). It is also

*Corresponding author. E-mail: chernetamente@yahoo.com.
suggested that the general picture of ferromagnetism that applies to these metallic (Ga,Mn)As systems is the one in which magnetic coupling between local Mn moments has its origin in the RKKY interaction mediated by delocalized holes in the (Ga,Mn)As valence band (Matsukura et al., 1998). The density of these excited carriers/holes, thus, determines the enhancement of the magnetic coupling energy. This could be done by impinging light of nearly resonance frequency of the DMS to significantly enhance the exchange coupling energy.

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).

In the present work we study the effect of photo-excitation on the $T_C$ of the diluted magnetic semiconductor (Ga,Mn)As. The model Hamiltonian used in describing the system is the standard Zeeman energy semiconductor (Ga,Mn)As. The model Hamiltonian used in describing the system is the standard Zeeman energy

$$H = g \mu_B H x S_z,$$

(1)

where $g$ is the $g$-factor, $\mu_B$ is the Bohr magneton and $H_x$ is the magnetic field, expressed by:

$$H_x =\sum_{n} \left( \frac{\hbar}{2\epsilon_0 \omega_0} \right)^{1/2} \left[ \hat{d}_q(t) e^{i q R_n} - \hat{d}^+_q(t) e^{-i q R_n} \right] \frac{q \times \hat{e}_\rho}{V},$$

(2)

in which $\hat{d}_q$ ($\hat{d}_q^+$) is the time dependent annihilation (creation) operator for photon, $q$ is the wave vector of photon, $R_n$ is position of a spin at site $n$, $V$ is the volume in which the radiation field is confined, $\epsilon_0$ is permittivity of free space, $\hat{e}_\rho$ is a unit polarization vector, and $i q \times \hat{e}_\rho$ = $q$ due to transversality condition (Scully et al., 1997). $S_z$ is the $z$-component of spin of the localized Mn $d$-shell electrons (magnetic spins) given by:

$$S_z = \frac{1}{2} (S^+ + S^-)$$

(3)

The model assumed no direct interaction between the localized moments as it is weak and no free carriers without the incident light, so that the system is paramagnetic in the absence of light and can be ferromagnetic only via the indirect interaction with the photoinduced carriers.

Substituting Equation (2) and (3) into Equation (1) gives:

$$H_I = i \sum_{n,q} \eta_q \left[ \hat{d}_q(t) e^{i q R_n} - \hat{d}^+_q(t) e^{-i q R_n} \right] \left[ \frac{S^+_n + S^-_n}{2} \right]$$

(4)

where $\eta_q = g \mu_B \omega_0 / \hbar$ is magnon-photon coupling constant, $\omega_0 = c q$ is frequency of the electromagnetic radiation, $c$ is speed of light in free space, and $\mu_B$ is magnetic permeability of vacuum.

By substituting Equation (4) into an expression for the second order energy correction, the photon-induced exchange coupling energy would be obtained. This enables one to find the indirect exchange interaction between localized spins (Kasuya, 1956) mediated by carriers/holes. The correction to energy of the second order perturbation can be determined as follows:

$$E^{(2)} = \sum_{n,m} \sum_{\sigma,\sigma'} \left| \langle n_{\sigma} | \sigma H_I | n_{\sigma'} \rangle \right|^2 \delta_{\omega - \omega_{\sigma}},$$

(5)

where $\omega_{\sigma} = (E_{\sigma} - E_{\sigma'}) / \hbar$ is transition frequency due to Zeeman splitting that might be from $-\hbar/2$ (down) to $+\hbar/2$ (up) spin state and/or dictated by the selection rule. Using (4) in (5) lead to an energy operator that can be expressed in the form of:

$$\hat{H} = \sum_{nm} J_{nm} S^+_n S^-_m$$

(6)

This last expression has resemblance of the Heisenberg type of Hamiltonian. The term $S^+_n S^-_m$ appears from the commutation relation

$$[S^+_n, S^-_m] = -2i S^Z \delta_{nm}$$

for spin state $S$ in which the delta function vanishes for the reason that the spin wave is propagation between different arbitrary sites, $n$ and $m$, and not a spin flip at a site. $J_{nm}$ is the photoinduced exchange coupling energy, for $n_q >> 1$, given by:

$$J_{nm} = \sum_q g^2 \mu_B^2 \mu_0 \frac{n_q}{2 V} \left( \frac{\omega_0}{\omega_q - \omega_{\sigma}} \right) \cos q R_{nm}$$

(7)

where $\cos q R_{nm}$ indicates oscillatory character of the energy; and $n_q$ is number of photons (intensity).

Introducing a small imaginary factor $i \mathcal{E}$, in which $\mathcal{E}$ can be
associated with the resonance line broadening, and using the Dirac identity:

\[
\frac{1}{\omega_q - \omega_{\sigma\sigma'} + i\varepsilon} = \text{Sp} \left( \frac{1}{\omega_q - \omega_{\sigma\sigma'}} \right) - i\pi\delta (\omega_q - \omega_{\sigma\sigma'}) \tag{8}
\]

where \(\text{Sp}\) denotes the principal value. Substituting Equation (8) in (7) takes the form of:

\[
J_{nm} = \chi + i\gamma \tag{9}
\]

\(\chi\) denotes the shift in energy as a result of photo-excitation, and \(\gamma\) represents the line broadening that will be a delta function at resonance.

RESULT AND DISCUSSION

Figure 1 shows that \(J_{nm}\) is an oscillating parameter with constant amplitude. The figure illustrates the possible appearance of ferromagnetic and antiferromagnetic interaction of the impurity ions depending on their separation.

For \(qR_{nm} \ll 1\) limiting case of Equation (7), Figure 2 is plotted and shows dependence of exchange coupling \(J_{nm}\) on the impinged light energy \(\omega_q\). This coupling energy rises to maximum near/at resonance.

Broadening of the line width is also thought to decrease the exchange energy revealing that the light frequency far from the resonance value would create fewer amounts of carriers/holes required for mediation. Moreover, increase in intensity \(n_q\) of the irradiated photon would augment the mode of excitation and hence, enhance ferromagnetism, as also shown by Mishra et al. (2008). The density of excited holes, thus, increase with this intensity as experiments can reveal, which in turn would increase the exchange coupling energy. For Mn of concentration \(x\) and band broadening of half line width \(\varepsilon\), \(T_C\) can be verified using expression \(T_C = 2\varepsilon S + 0.2J_{nm}/3\). Equation (9) describes interaction of the system in analogy with the RKKY expression (Priour et al., 2006; Matsukura et al., 1998).

\[
J_{nm} = J_0 F(2k_F R_{nm}) \tag{10}
\]

where the term \(F(y) = (y \cos y - \sin y) y^{-1}\), for \(y = 2k_F R_{nm}\) is oscillatory \(k_F\) being the Fermi wave vector, \(J_0 = 8\pi n_{pd}^2/3N^2\varepsilon_F^{-1}\) in which \(J_{pd}\) is an exchange coupling between the Mn local moments and hole spins, \(\varepsilon_F\) is the Fermi energy, \(n = k_F^3/6\pi^2\) (Priour et al., 2006; Kasuya, 1956) is the number of ground state Fermi electrons, and \(N\) is the total number of lattice points. Inline with this, if the spin-orbit interaction and band warping are neglected, the top of the valence band is

![Figure 1](image1.png)

**Figure 1.** \(J_{nm}\) vs. \(qR_{nm}\) and \(\omega_q\) for \(\varepsilon = 0.1\omega_{\sigma\sigma'}\).

![Figure 2](image2.png)

**Figure 2.** The exchange coupling energy, \(J_{nm}\), vs. energy of the radiation field \(\omega_q\) for different half line width, \(\varepsilon\), is plotted.

![Figure 3](image3.png)

**Figure 3.** The RKKY exchange energy, \(J_{nm}\), and \(F(2k_F R_{nm})\), vs. \(2k_F R_{nm}\).
Acknowledged. and partially supported by Sida/SAREC, are gratefully acknowledged.

Figure 3 is plotted using Equation (10), from which the maximum value of \( J_{nm} \) could be estimated. Accordingly, the exchange coupling energy is shown to increase and become strong as the factor \( 2k_F R_{nm} \), described in terms of the separation between localized impurity spins, decreases until the ferromagnetism becomes maximum. This energy is also found to increase with the hole density, \( N_s \), that might be expressed via \( k_F \) and decrease with increase in the Fermi energy \( \varepsilon_F \). The later indicates that the exchange interaction could take place more by valence band subsystem where \( \varepsilon_F \) may shift towards the top of the valence band approaching the acceptor level, hence, the localized spins interact with holes. Comparison of Figures 2 and 3 indicates that excitation by photon energy of frequency at/near resonance would increase density of carriers/holes to maximum. On the basis of inference to the exchange couplings, maximum value of the ferromagnetic transition temperature \( T_C \) for a known impurity concentration, \( x \), can be calculated.

**Conclusion**

In this work the effect of photon irradiation on \( T_C \) of the diluted magnetic semiconductor (Ga,Mn)As is studied. It is shown that photoinduced exchange coupling energy would greatly enhance ferromagnetic transition temperature at/near resonance. It will be influenced by magnon-photon coupling constant \( \eta_p \), energy of the irradiated light \( \omega_p \), number (intensity) of photons \( n_p \), and volume of the radiation field cavity with \( J_{nm} \approx V^{-1} \), inferring that the enhancement of the magnetization could be more pronounced in miniaturized size crystals. The exchange energy at/near resonance is analogous to that of the hole mediated RKKY indirect exchange for a particular value of \( k_F R_{nm} \).

The difference is, here the mediation is done by photo created holes in which with larger photon intensity, greater number of holes can be created so as to further increase ferromagnetic order and critical temperature \( T_C \).

**REFERENCES**


ACKNOWLEDGEMENTS

The work is sponsored by Nekemte College of teacher education under Oromiya regional state education bureau and partially supported by Sida/SAREC, are gratefully acknowledged.