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THE EFFECT OF ELECTROMAGNETIC PERTURBATION ON THE OPTICAL PROPERTIES OF $(III, Mn)V$ DILUTED MAGNETIC SEMICONDUCTORS

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ABSTRACT: Nowadays, electronic, magnetic and optical properties of (III, Mn)V diluted magnetic semiconductor (DMS) is a frontline research topic. This emerging research area, inspired us to give a great attention on the study of optical properties of these DMSs. The work can be affirmed by calculating optical conductivity of the aforementioned DMSs, particularly for the prototype, and for its base semiconductor. Hence the focus area of the study is to determine optical, magnetic and electronic properties using quantum concepts and quantum mechanical models such as linear response theory of Kubo formalism, Kohn-Luttinger Hamiltonian (theory), and Green function. In addition to these models, analytical calculations could be treated using perturbation theory of approximation, Fermi golden rules and Kramers-Kronig relations. Having all these models and analytical techniques, optical conductivity will be related as the product of the incorporated magnetic ion concentration and that of the reciprocal of the square root of photon energy in the infrared spectrum. All in all the results obtained fairly fits with others theoretical and experimental reports. Therefore, the findings indicated that there is a promising advancement of optoelectronic device functionalities, laser or spectroscopic experiments to provide background information about the optical linear response functions, high efficiency solar cells and thin film fabrications, magneto-optic materials determinations.

Key Words/phrases: Green's function, Kubo formalism, Magnetic properties, Optical properties.

INTRODUCTION

In the fundamental concept of spintronics study (Lino Miguel da C. P, 2011) data is stored in metal-based magnetic devices, while data processing is carried out in semiconductor-based electronic devices. Integrating the existence of charge and spin functionalities in one system would necessarily increase the speed, energy efficiency and packing densities of electronic devices.

For instance gallium nitride (GaN) are not magnetic but semiconductor. However, ferromagnetic materials used in digital storage devices, such as iron (Fe), cobalt (Co), nickel (Ni) and their alloys are not semiconductors. In addition, the crystal and electronic structures of magnetic materials are usually quite different from that of the semiconductors used in ordinary electronics, making it difficult to combine the two in functional hetero structures. Also, the conductivity mismatch between metals and semiconductors limits the efficiency of spin-injection. The alternative is then to use materials which combine ferromagnetic and semiconducting behavior.

Crystal structure in diluted magnetic semiconductors

The primitive unit cell is the smallest volume which can be periodically translated to reproduce the whole crystal structure (Razeghi M., 2002). There are two common crystal structures among semiconductors with the more common one being zinc-blende (ZB) and the other one is wurtzite. For cubic crystal structure, such as ZB, there is only one distance to be defined: the distance between nearest-neighbor of similar crystal sites, i.e., compounds such as $GaAs$ have the zinc-blende structure. The arrangement of the atoms is the same as in diamond but the two species alternate: Ga occupies the original sites of the face centered cubic lattice, while As occupies the tetrahedral sites, to give a unit cell that contains four atoms of each species.

As already mentioned, isomorphic crystals of$(Ga, Mn)As$ are fabricated by randomly

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substituting a small amount of, typically 2% to 6%, for cation sites of the semiconductor host lattice. Hence, they have a ZB structure similar to that of $GaAs$.

Nowadays, optical properties of diluted magnetic semiconductors are among the best studied phenomena in solid state physics, mainly because of their technological advancement (Dressel M. and Griner G., 2002). Recently, new findings are emerging in the area. Hence advances made to date in photonic devices that have enabled optical communications could not have been achieved without rigorously conducting research in the optical properties of materials and how these properties influence the overall device performance. Consequently, research on optical properties of materials draws in not only physicists, who used to be the usual traditional researchers in this field, but also scientists and engineers from widely different disciplines (Singh J., 2006)

Optical techniques have established themselves as providing key insight into the band structure of materials. In fact, the electronic structure of many semiconductors has been determined by comparing theoretical calculations to optical results (Yu P. and Cardona M., 2010). Furthermore, optical techniques cover a broad range of energy scales in magnetic semiconductors. Thus, optical studies have provided a unique view into the underlying physics governing various correlated electron materials.

To better understand the effects of magnetism on the band structure of $(III-V)$ semiconductors (and vice versa) extensive optical studies have been applied to $(III, TM)V$ferromagnetic DMSs. As detailed throughout in many recent studies, magneto-optical experiments have demonstrated the ferromagnetism originates from a single $(III, TM)V$phase and not from inclusions, such as in a matrix. Furthermore, magneto-optical measurements have provided a determination of the strength of the hybridization between the local moments and holes that mediate the magnetic state (Dietl T., 1994) As far as the evolution of the electronic structure with Mn doping is concerned, optical spectroscopy has uncovered a number of fundamental distinctions with the properties triggered by non-magnetic impurities of diluted magnetic semiconductors in the III-V series.

Theoretical models

In order to solve optical properties of the system, we need to formulate the Hamiltonian. However, it is essentially difficult to treat the Hamiltonian of the systems accurately for small Mn concentrations (Moca etal, 2009). We therefore used the model amiltonian to describe the valence band system in a $(III, Mn)V$ ferromagnet which adds the kinetic exchange field interactions to the six-band $k.p$ Hamiltonian of the host III-V semiconductor. In order to manage the complexity of calculations, in this scheme, we ignored the effect of disorder. In the mean-field virtual crystal approximation, the interactions are replaced by their spatial averages, so that the Coulomb interaction vanishes and holes interact with a homogeneous exchange field. The unperturbed Hamiltonian for the valence band holes reads:

$$H = H_{KL} + \vec{h}s, \qquad (1)$$

Where H_{KL} is the Kohn-Luttinger Hamiltonian of pure $GaAs$, s is the envelope function hole spin operator, and $\vec{h}$ is the effective exchange field that splits the valence bands and estimated as:

$$\vec{h} = N_{Mn} S J_{pd} \hat{z}. \qquad (2)$$

Here $N_{mn} = 4\,x_{mn}/a_{lc}^3$ is the substitutional Mn density in the$Ga_{1-x}Mn_xAs$epilayer with a lattice constant $a_{lc} \approx 0.565\,nm$ and $x_{mn} = 3\%, 4\%, 5\%$ is the Mn-concentration range fixed for this theoretical work, the spin $S = 5/2$is the Mn local moments with a semi-phenomenological local exchange interaction treated at a mean-field level, the strength of the exchange coupling constant is $J_{pd} = 55\,meVnm^{-3}$ and$\hat{z}$indicates the angular momentum quantization direction chosen to be along the z-axis at zero temperature limit (Hankiewicz etal, 2004; Jungwirth et al, 2005).

The host band part of the Hamiltonian is described via the four- or six-band Kohn-Luttinger model. The $k = 0$states at the top valence band have p-like character and can be represented by the$l = 1$ orbital angular momentum eigen states $|m_l>$. For systems with spin-orbit coupling, the KL Hamiltonian, H_{KL}, by using the basis formed by angular momentum eigen states $|j, m_j>$ can be written in the combination form.

The split-off energy for the semiconductor $GaAs$is of the order of few hundred meV. On the other hand, the Fermi energy is of the order of few

meV for typical hole density. Thus, one can safely ignore the split-off band when the Fermi energy is sufficiently smaller than the split-off energy and hence the calculation can be approximated to describe the two upper most valence bands (known as heavy hole and light hole bands). The four-band KL Hamiltonian can be diagonalized analytically and yields a pair of Kramer's doublets with eigen-energies (Chow W. W. and Koch S. W., 1999)

Optical conductivity of the III-V diluted magnetic semiconductors

In direct interband transitions, the optical conductivity, $\sigma(\omega)$, is obtained from the Kubo formalism. Hence, we can evaluate the real part of the frequency dependent optical conductivity from the formula expression which relates it to the quasi-particle eigenvectors and eigen values as:

$$Re[\sigma_{xy}(\omega)] = -\frac{e^2\hbar}{m^2}\int\frac{dk}{(2\pi)^3}\sum_{l\neq l'}(f_{l'}-f_l)\frac{Im[<l'|\hat{p}_\alpha|l><l|\hat{p}_\beta|l'>]}{(\hbar\omega-E_l+E_{l'})(E_l-E_{l'})}, \quad (3)$$

where $\alpha,\beta = x,y$; m is the bare electron mass; $\hat{p}_\alpha$, $\hat{p}_\beta$ are components of the momentum operator; $\hat{p}/m$ is the $k.p$ velocity operator obtained by differentiating the $k.p$ Hamiltonian with respect to the wave vector; f_l and $f_{l'}$ are the Fermi distribution functions and the appearance of $(f_{l'}-f_l)$ follows from the Pauli exclusion principle, an interband transition occurs from an occupied state below the Fermi level to an unoccupied state above the Fermi level (i.e., $l = 1\ to\ l' = 0\ at\ T = 0K$); $|l>\ and\ |l'>$ are the eigen vectors (or crystal wave functions), corresponding to the Kohn-Luttingereigen values E_l and $E_{l'}$.
Therefore, the dipole matrix elements in Eq. (3) can be written as:

$$<l'|\hat{p}_\alpha|l>=\frac{m}{\hbar}<z_{l'}|\frac{\partial H}{\partial k_\alpha}|z_l>=\frac{m}{\hbar}(E_l-E_{l'})<\frac{\partial l'}{\partial k_\alpha}|l>. \quad (4)$$

To compute the ac-conductivity, we rewrite Eq. (3) in terms of the spectral function:

$$\sigma_{xy}(\omega) = \int_{-\infty}^{\infty} d\omega'\frac{A_{xy}(\omega)}{\omega-\omega'},$$
with
$$A_{xy}(\omega) = -\frac{e^2\hbar}{m^2V}\sum_{l\neq l'}(f_{l'}-f_l)\frac{Im[<l'|\hat{p}_\alpha|l><l|\hat{p}_\beta|l'>]}{(E_l-E_{l'})}\delta[\hbar\omega-(E_l-E_{l'})], \quad (5)$$

Where $A_{xy}(\omega)$ is given by different expressions in three different energy intervals. In the intermediate energy range transitions at all wave vectors within the Fermi surface are allowed, whereas in the lower and upper energy intervals only transitions at a fraction of the wave vectors within the Fermi surface contribute to $A_{xy}(\omega)$.Hence, considering the upper and lower limits of the heavy-hole and light-hole bands and their respective differences, we can obtain the three energy intervals that contribute to $A_{xy}(\omega)$ and the fraction of the Fermi volume that contributes to it. But for the sake of simplicity we have taken one easily understandable case for the energy intervals. However, for clarity and more completed discussion in case of other intervals, see reference (Sinova etal, 2003)

The spectral function $A_{xy}(\omega)$ is an odd function ofω, and hence we need only spectrum. In line with this, let us consider the transition $hh^+ \rightarrow lh^+$, among the four types of transitions that contribute to $A_{xy}(\omega)$, separately. Therefore, we use:

$$m_{hh}=\frac{m_0}{(\gamma_1-2\gamma_2)},\ m_{lh}=\frac{m_0}{(\gamma_1+2\gamma_2)},\ \mu=\frac{m_{hh}m_{lh}}{m_{hh}+m_{lh}}(E_l-E_{l'}),$$
and $\frac{\hbar k_{lh}^2}{2\mu}=\omega-\widehat{\omega}$,

Where $k_{lh}$is the light-hole band Fermi wave vector of Fermi energy (E_F) in zero exchange field. For $hh^+ \rightarrow lh^{\pm}$ transitions, we have

$$A_{xy}(w; hh^+ \rightarrow lh^{\pm}) = -\frac{he^2}{(2\pi)^2m^2}\int_{-1}^{1}d(cos\theta)\frac{\mu k}{\hbar^2}\frac{Im[<k,hh+|\hat{p}_\alpha|k,lh\pm><k,lh\pm|\hat{p}_\beta|k,hh+>]}{E_{hl}^{\pm}-E_{hh}^{+}}|_q, \quad (6)$$

where the subscript q in the Eq. (6) is

$$q = k = \sqrt{\frac{2\mu k}{\hbar}}(1\mp\frac{\hbar cos\theta}{12\hbar\omega\ cos2\theta'}+\frac{\overrightarrow{\hbar}}{4\hbar}\ cos\theta). \quad (7)$$

For the $hh^-\ to\ lh^{\pm}$ transition we obtain the same result, therefore with in this range we have the special function:

$$A_{xy}(\omega)=\left(\frac{5e^2\sqrt{2\mu}}{48\pi^2\hbar^2}\right)\left(\frac{4[x_{mn}]SJ_{pd}}{a_{lc}^3}\hat{z}\right)(\hbar\omega)^{-1/2}\approx K.[x_{mn}]\left(\frac{1}{\sqrt{\hbar\omega}}\right), \quad (8)$$

Where $K=\left(\frac{5e^2\sqrt{2\mu}}{48\pi^2\hbar^2}\right)\left(\frac{4SJ_{pd}}{a_{lc}^3}\right)\hat{z}$ is constant.

Optical conductivity of the bulk GaAs

The transition probability from an initial state$|l>$ to a final state$|l^{'}>$ of higher energy can be written:

$$p_{l\to l^{'}}(t)=\frac{|<l^{'}|H_p|l>|^2 sin^2(1/2)(\omega_{ll^{'}}-\omega)t}{\hbar^2(\omega_{ll^{'}}-\omega)^2}. \quad (9)$$

We note from this equation that the transition probability between two states depends sinusoidally on the time. This probability is proportional to the square of the matrix element of the perturbation. ReplacingH_p by its expression we obtain the following expression for the absorption transition probability, which we shall use below:

$$p_{l\to l^{'}}(t)=\frac{4|<l^{'}|c^{+}|l>|^2 sin^2(1/2)(\omega_{ll^{'}}-\omega)t}{\hbar^2(\omega_{ll^{'}}-\omega)^2}. \quad (10)$$

Now we shall consider the Fermi's golden rule: since the transition probability is proportional to the time the perturbation acts, it is therefore useful to deal with a quantity called the transition probability per unit time.

The probability, for a given initial state$|l>$, of finding an electron in one or another of a set of final states of energy close to$E_{l^{'}}$. If$n(E_{l^{'}})$ is the density of these states the total probability of finding the electron in any one of these states is:

$$p(t)=\frac{4}{\hbar}\int\frac{|<l^{'}|c^{+}|l>|^2\, n(E_{l^{'}})\, sin^2(1/2)(\omega_{ll^{'}}-\omega)t}{(\omega_{ll^{'}}-\omega)^2}d\omega_{ll^{'}}. \quad (11)$$

In this integral, the only non-negligible contribution comes from the region where ω is very close to $\omega_{ll^{'}}$because of the denominator $(\omega_{ll^{'}}-\omega)^2$.We see that only a narrow frequency bandwidth of order $1/t$contributes to the integral. We can therefore extend the limits of the integral to$\pm\infty$and use the fact that both $n(E_{l^{'}})=n(E_{l^{'}}+\hbar\omega_{ll^{'}})$ and the matrix element are approximately constant over the interval $\hbar/t$to write:

$$p(t)=\frac{4}{\hbar}|<l^{'}|c^{+}|l>|^2\, n(E_{l^{'}})\int_{-\infty}^{\infty}\frac{sin^2[(1/2)(\omega_{ll^{'}}-\omega)t]}{(\omega_{ll^{'}}-\omega)^2}d\omega_{ll^{'}}, \quad (12)$$

or setting

$$x=\frac{1}{2}(\omega_{ll^{'}}-\omega)t,$$

$$p(t)=\frac{4}{\hbar}|<l^{'}|c^{+}|l>|^2\, n(E_{l^{'}})2\pi t, \quad (13)$$

where we have used the fact that:

$$\int_{-\infty}^{\infty}(sin^2 x/x^2)dx=\pi. \quad (14)$$

The total probability of transition from a given initial state to one or another of a set of final states which are very close to each other is proportional to the time. We may therefore define a transition probability per unit time W:

$$W=\frac{p(t)}{t}=\frac{2\pi}{\hbar}|<l^{'}|c^{+}|l>|^2 n(E_{l^{'}}). \quad (15)$$

Which is often called Fermi's golden rule.

The fact that the only significant contributions to the integral Eq.(11) come from energies such that$\omega_{if}\cong\frac{t^2}{4}$is an expression of conservation of energy for states which were eigenstates of the unperturbed system. This conservation is exact in the limit of weak perturbations and long-time scales. In this limit, we can write Eq. (15) as:

$$W=\frac{2\pi}{\hbar}|<l^{'}|c^{+}|l>|^2\delta(E_{l^{'}}-E_l-\hbar\omega). \quad (16)$$

After some substitution of common expressions, we obtain:

$$W_T=\frac{e^2A^2Sdx}{\hbar^4m^24\pi}(2m_\Gamma)^{3/2}p^2(\hbar\omega-E_g)^{1/2}. \quad (17)$$

The average incident photon flux is $\frac{\bar{\pi}}{\hbar}$per unit area and by definition the fraction of incident photons absorbed over a depthdx is:

$$w_T=\alpha\frac{\bar{\pi}}{\hbar\omega}Sdx. \quad (18)$$

Therefore, expression for absorption coefficient (α) would be:

$$\alpha(\omega)=\frac{(2m_\Gamma)^{3/2}e^2p^2}{2\pi\epsilon_0cn\, m^2\hbar^3\omega}(\hbar\omega-E_g)^{1/2}, \quad (19)$$

Where $\bar{\pi}=\frac{1}{2}\epsilon_0\omega^2A^2cn$ and $\frac{1}{m_\Gamma}=\frac{1}{m_e}+\frac{1}{m_h}$.

Substituting all the constants in the Eq. (19), we get:

Now, let us define the relationship between absorption coefficient and optical conductivity:

$$\alpha(\omega)=\frac{8\pi Re[\sigma(\omega)]}{c(1+n)}. \quad (20)$$

Equation (20) provides that:

$$Re[\sigma(\omega)] = \frac{c(1+n)\times 10^4}{2\pi}\left(\hbar\omega - E_g\right)^{1/2}. \quad (21)$$

RESULTS AND DISCUSSION

In this paper we dealt with optical absorption coefficient ($\alpha(\omega)$) and then conductivity ($\sigma(\omega)$) of$GaAs$. In vicinity of these and other approximation techniques, we analytically obtained that $\alpha(\omega)$ can be estemeated as the product of some constant and square root of $(\hbar\omega - E_g)$ as given in Eq. (19). By using this result as an input we found that the real part of the frequency dependent optical conductivity is also approximated using Eq. (21) in the infrared region. These dependence of conductivity on photon energy ($\hbar\omega$) with in the given limit is depicted in Fig. 1. Therefore, as the frequency of the electromagnetic radiation (photon energy) increases, strong interactions can occur and modify the optical conductivity substantially. From the same figure (or Fig. 1) one can also read that at lower photon energy less than 1.43 eV (i.e., $\hbar\omega < E_g$), no any effect on optical conductivity or absorption is observed. Therefore, the result agrees with the previous studies (Nag B.R. , 2002).

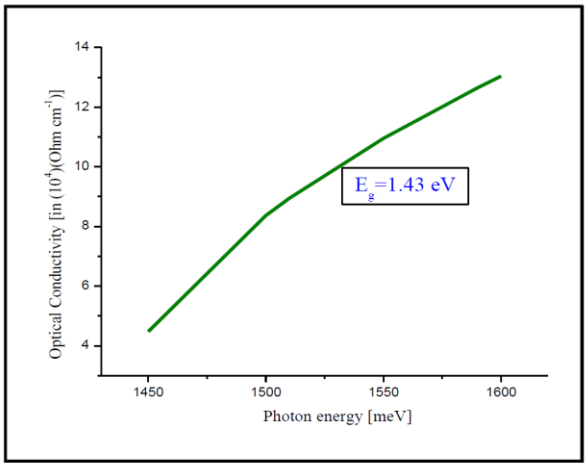

Figure 1. A schematic of the optical conductivity versus photon energy of *GaAs* in the infrared spectral region.

Spectral function of GaMnAs with respect to the infrared photon energy

In this work, we ideally handled an intrinsic optically active base (or host); intentionally doped with a few amount of magnetic impurity; allowed this doped semiconductors to respond towards the externally applied electromagnetic radiation in the infrared limit; and treated the nature of the response with the help of quantum mechanically sophisticated models listed. In running these calculations, we followed a long chain of physical and analytical steps as we have discussed before. When we express these comprehensive ideas into more technical and text wised terms again, because of the technological importance and practical applications, we focused on electronic nature rather than lattice vibrational (absorption and creation of phonons) techniques of study. Fairly to execute these tasks, conduction band for a few cases, and valence band (for almost 90% of the work) were mainly chosen to see the contributions of degenerate hh, lh and split-off energy to the spectral function. For this matter, initially we started from concepts of electrodynamics (Ohm's law) and then passed over into fully quantum mechanical approaches. This method was straightforwardly directed to the problems at hand with the help of linear response Kubo formalism. Then 6-band Kohn Luttinger Hamiltonian (theory) was used and also 4-band spherical model was dominantly applied in the diagonalization of the matrix and eigen values or states determination. Finally, Kramers-Kronig relations were applied and spectral function was calculated.

On the other hand, by relating Kramers-Kronig relations and discussions given in the above sections, we can see that spectral function (an imaginary part of the optical response function or conductivity) conveys important information about the triggered response in terms of the electron excitation.

Therefore, these organized way of calculation nourished us with lastly simplified equation $(A_{xy}(\omega) \approx K.[x_{mn}][\frac{1}{\sqrt{(\hbar\omega)}}])$, where K stands for some multiple of constants resulted from Eq. (8). This dependency of optical conductivity on the frequency of photon energy is shown in Fig. 2. Thus, one can easily observe that the imaginary part of the optical conductivity exponentially decreases with increasing the infrared photon energy.

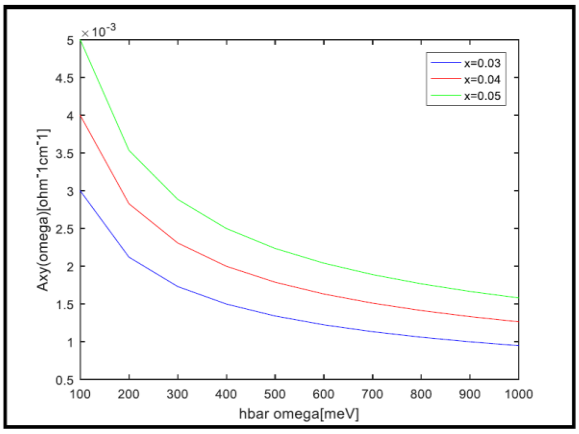

Figure 2.The spectral function versus photon energy of $GaMnAs$ at three different impurity (Mn) concentrations.

Effect of impurity (magnetic ion) concentration on optical conductivity

In this study, based on the explanation given on sections above, we used three different manganese ion (Mn^{+2}) concentrations that are 3%, 4% and 5% as depicted in Fig. 3. Accordingly, one can easily see that optical conductivity increases as concentration increases within the given limit. The physics behind this may be due to the fact that dopants provide the substrate with both a local moment and a hole. In other words, by keeping other factors constant, increasing concentration to the material agitates electron excitation and hole creation via the valence and conduction bands. This in turn, derives the relative elevation of the response function.

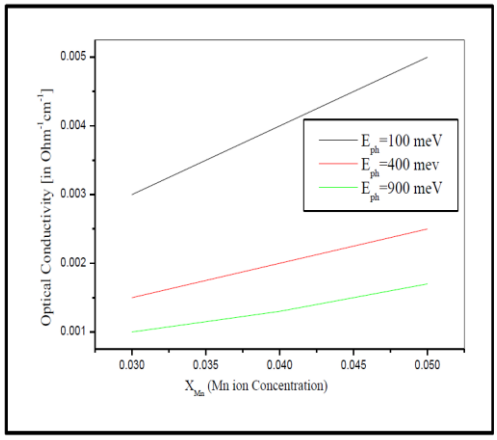

Figure 3.The effect of $Mn-$ ions concentration on optical conductivity of $GaMnAs$ and E_{ph} stands for infrared photon energy.

Comparison of our results with other experimental or theoretical works

We obtained that for bulk $GaAs$ within a small fraction of an electron-volt at energy nearly equal to the energy gap (E_g) of the material. The increased absorption or conductivity is caused by transitions of electrons from the valence band to the conduction band and this concept is available in any fundamental semiconductor books.

Other important result of this work is: optical conductivity of our system exponentially decreases with increasing photon energy in the infrared region and optical conductivity of diluted magnetic semiconductor that we considered is directly related to the concentration the dopant magnetic ion. In general, previous studies (both experimentally and theoretically) using one band V-J model have been pointed out similar results with this study (Charles P. Poole Jr., 2004 ; Oudovenko etal, 2004 ; Eric Yang, etal, 2003 ; . Hartmut H. and Stephan W. K., 2004).

CONCLUSIONS

The desire to integrate storage capabilities and information processing in single semiconductor based devices has fueled the development of ferromagnetic semiconductors based on $(III, Mn)V$ materials. The motivation behind the choice of this specific group of semiconductors is very clear: these semiconductors have enough potential for new device applications enriched with optoelectronic behavior. This is due to the fact that their magnetic, optical, and electronic effects are all interconnected so that they are likely to have a major technological impact if systems with Curie temperatures or above room temperature can be created and better control of disorder effects can be achieved.

We know that scientific study have its own sequence and nature, without losing these, we have also proposed to use quantum mechanically sophisticated approaches like linear response theory of Kubo formalism, Kohn-Luttinger Hamiltonian ($k.p$theory), and Green function models. These models are not chosen without any reason. Instead, those very recent nobilities like spintronics and optoelectronics features are quantum concepts, and any problems targeted to have important contribution towards or against these concepts should be treated quantum mechanically, otherwise classical models may fail.

Due to these quantum mechanical advantages, our problem (i.e., optical conductivity of the chosen diluted magnetic semiconductors and their base semiconductors) are treated systematically. In order to manage some challenges and complexities during analytical calculations, we used some approximations, rules or relations like perturbation theory of approximation, Fermi golden rule, and Kramers-Kronig relations.

In conclusion, we obtained somewhat interesting relationships in which the magnetic, optical, and electronic behaviors of our system coexisted. The conductivity of $GaMnAs$is the product of the incorporated magnetic ion concentration and the reciprocal of the square root of photon energy in the infrared regime. In comparison, this result agrees with the others theoretical and experimental findings.

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