Electronic and optical spectra in a diluted magnetic semiconductor multilayer

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(Dated: Friday 2nd May, 2014)

The effects of random distribution of magnetic impurities with concentration \( x \) in a semiconductor alloy multilayer at a paramagnetic temperature are investigated by means of coherent potential approximation and tight-binding model. The change in the electronic states and the optical absorption spectrum with \( x \) is calculated for weak and strong exchange interactions between carrier spins and localized spin moments on magnetic ions. We find that the density of states and optical absorption are strongly layer-dependent due to the quantum size effects. The electronic and optical spectra are broadened due to the spin fluctuations of magnetic ions and in the case of strong exchange interaction, an energy gap appears in both spectra. Furthermore, the interior layers show higher contribution in the optical absorption of the system. The results can be helpful for magneto-optical devices at a paramagnetic temperature.

I. INTRODUCTION

Semiconductor multilayers are materials with attractive electronic and optical properties and possible application in nanodevices [1]. Such unique properties which are caused by their dependence on the number of atoms in a confined direction, have led to a rapid growth on research activities in this field in recent years. For instance, it has been shown that for semiconducting multilayer armchair graphene nanoribbon with more than two layers, semiconductor-to-metallic transition is obtained with increasing electric field, which can be utilized to enhance the efficiency of graphene based transistor devices [2]. In addition, for the thicker systems (i.e. more layers), this transition occurs at a smaller electric field which makes them to enhance the on/off ratio of a field-effect transistor device [2].

Furthermore, magnetic and nonmagnetic impurities in layered structures have received considerable attention [3][11]. Spin-dependent scattering of charge carriers in magnetic multilayers is the origin of magnetotransport effects and has been the subject of intensive interest in recent semiconductor electronics research [6][11]. Doping of magnetic ions in nonmagnetic semiconductors (NMSs) introduces not only magnetic moment but also free carriers in the system. In diluted magnetic semiconductors (DMSs), the host material is an alloy semiconductor of the type II-VI or III-V semiconductor with characteristic of \( AB \), in which the atom \( A \) is substituted by a magnetic atom with concentration of \( x \) [12][13]. For example, Mn ions as magnetic impurities into GaAs and InAs act as acceptors in the III-V compound semiconductors [12][14]. These semiconducting alloys are a novel class of ferromagnetic materials and one of the promising materials for spintronics [15]. The exchange interaction between a carrier spin and localized spin of magnetic ions, plays an important role in magneto-optical effects in DMSs [14][16]. Because this interaction strongly affects on band splitting that is observed in magneto-optical measurements such as magneto-reflectivity and magneto-absorption spectra. Thus, the most powerful tool for studying the exchange interaction between a carrier and localized spins in DMS-based structures is optical measurement. Furthermore, because of the ternary nature of DMSs, one can tune the lattice constants and band parameters by varying the composition of the material. Therefore, most of the magnetic semiconducting alloys are excellent candidates for preparation of quantum wells, superlattices, and other heterostructures that involve band gap engineering [17–20].

In previous studies [21–23], by applying the single-site coherent potential approximation (CPA) [24] to semiconductor alloy multilayers, the effects of chemical (non-magnetic) disorder on optical absorption spectrum were investigated. The results showed that, the photon absorption by the system is layer dependent and strongly changes by variation of the scattering-strength of the chemical impurities [21]. In the present study, we extend our previous approach [21] for the magnetic impurities at a paramagnetic temperature, and investigate the electronic and optical properties of DMS multilayers. To study the optical absorption spectrum in the presence of the magnetic impurities, we use the Onodera-Toyozawa theory [25], which is related to a binary mixed crystal or an alloy consisting of atoms (or molecules) \( A \) and \( B \), in which the transition dipole moments of \( A \) and \( B \) atoms are equal. We should point out that, the application of the Onodera-Toyozawa theory together with the CPA for DMSs was first employed by Takahashi [26] in which the optical band edge was studied for bulk materials. The calculations, however, were performed only for a simple semicircular model density of states.

In this study, owing to the substitutional disorder of the magnetic ions in a DMS multilayer, the effect of carrier scattering due to the presence of both magnetic and nonmagnetic atoms will be studied within the single-site CPA. Due to the quantum size effect in the multilayer systems the electronic states at each layer strongly depend on the location of that layer relative to the surface layer. Hence, each layer has a different response to the carrier scattering process in the layer and should be
II. THE MODEL

In this study, we consider a semiconductor multilayer described by the single-band tight-binding model with the nearest-neighbor hopping and the on-site delta-function-like potential on a simple cubic lattice in which one of the dimensions (the $z$ direction) is confined. The number of layers along the confined direction is $N_z$ and the label of each layer is denoted by $n$, hence $1 \leq n \leq N_z$. The multilayer is a semiconducting alloy of the form $A_1-xM_xB$, where the sites of $A$ atoms such as Ga can be occupied by $M$ atoms such as Mn with concentration $x$ and $B$ atoms such as As remain unchanged. Due to the low carrier density in the system, the interaction between carriers is ignored in the calculations. Accordingly, the one-electron or a Frenkel exciton Hamiltonian in this system is given by:

\begin{equation}
H = \sum_{r,n} u^A_{r,n} - \sum_{r,n,r',n'} \sum_{\mu} t_{rn,r'n'} |\langle r, n, \mu | r', n', \mu \rangle| (1)
\end{equation}

where, $|\langle r, n, \mu | r', n', \mu \rangle|$ is an atomic orbital with spin $\mu$ (=↑ or ↓) at site $|r, n\rangle$. Here, $r$ denotes the positional vector in the $x - y$ plane of the layer $n$. The hopping integral $t_{rn,r'n'}$ is equal to $t$ for the nearest neighbor sites and zero otherwise. We assume that the hopping integral only depends on the relative position of the lattice sites. Therefore, the type of disorder in our model is considered to be diagonal and accordingly, the off-diagonal disorder is outside of the scope of the present study. The random site energy $u^A_{r,n}$ is assumed to be $u^A_{r,n}$ and $u^M_{r,n}$ with probabilities $1 - x$ and $x$ when the lattice site $|r, n\rangle$ is occupied by the $A$ and $M$ atoms, respectively [21, 26, 27]. For $A$ and $M$ sites we have:

\begin{equation}
u^A_{r,n} = \{\langle r, n, \mu | r, n, \mu \rangle | (2)
\end{equation}

\begin{equation}
u^M_{r,n} = \{\langle r, n, \mu | \delta_{\mu \nu} - IS_{rn} \cdot \sigma_{\mu \nu} | r, n, \nu \rangle | (3)
\end{equation}

where $IS_{rn} \cdot \sigma_{\mu \nu}$ is the $k$-independent exchange interaction in which $|S_{rn}| = S$ is the local spin operator of the $M$ atom and $\sigma$ is the Pauli matrix for carrier spin. In this study, instead of taking the quantum fluctuation of the localized spin of the magnetic ions into account, the classical spin approximation (i.e. $S \rightarrow \infty$) is used, while IS is a constant parameter [26, 27].

According to the CPA, a disordered alloy can be replaced by an effective periodic medium in which the potential of all sites is replaced by an energy-dependent coherent potential, except one site which is denoted by impurity [24]. Therefore, the multilayer Hamiltonian $H$ is replaced by the following effective medium Hamiltonian:

\begin{equation}
H_{\text{eff}} = \sum_{r,n} \sum_{r',n'} \sum_{\mu} \Sigma_n(\omega)\delta_{r,r'}\delta_{n,n'} - t |\langle r, n, \mu | r', n', \mu \rangle| (4)
\end{equation}

where $\Sigma_n(\omega)$ is an energy-dependent self-energy and called the coherent potential. Because of the absence of translational symmetry of the system along the $z$-direction, this self-energy depends on the layer number $n$. Furthermore, the self-energy is spin independent, because the system is at a paramagnetic temperature ($T = T_c$).

The physical properties of the real system can be obtained from the configurationally averaged Green’s function, $\langle G \rangle_{\text{av}} = (\langle \omega - H \rangle)^{-1}_{\text{av}}$ which is replaced in the CPA with an effective medium Green’s function $G = (\omega - H_{\text{eff}})^{-1}$. A direct consequence of this replacement is the fact that the effective scattering of a carrier at the impurity site is zero, on average [24]. The matrix elements of the effective Green’s function are calculated by the following Dyson equation [27]:

\begin{equation}
G_{n,n'}^{\text{0}}(r,r') = G_{n,n'}^{\text{0}}(r,r') + \sum_{n''=1}^{N_z} \sum_{r''}G_{n,n''}^{0}(r,r'') \times \Sigma_{n''} G_{n'',n'}^{0}(r',r') , (5)
\end{equation}

where the variable $\omega$ has been suppressed for simplicity. Here, $G^{0}(\omega)$ is the clean system Green’s function and its matrix element is given by [27]:

\begin{equation}
G_{n,n'}^{0}(r,r') = \frac{\alpha^2}{4\pi^2} \sum_{k_\parallel} N_z \int_{1BZ} \text{d}k_{\parallel} G_{n,n'}^{0}(\ell, k_{\parallel}) e^{i k_{\parallel}(r-r')} , (6)
\end{equation}

In this equation, $\ell$ denotes the subband number (mode), $k_{\parallel}$ is a wave vector parallel to the layer, and the integral is taken over all the wave vectors in the first Brillouin zone (1BZ) of the two-dimensional lattice [21]. $G_{n,n'}^{0}(\ell, k_{\parallel})$ is the mixed Bloch-Wannier representation of $G^{0}$ which can be expressed as

\begin{equation}
G_{n,n'}^{0}(\ell, k_{\parallel}; \omega) = \frac{h_{n,n'}(\ell)}{\omega + i\eta - \varepsilon_{\ell}(k_{\parallel})} , (7)
\end{equation}

where $h_{n,n'}(\ell) = \frac{2}{N_z+1} \sin(\frac{\ell}{N_z+1} n) \sin(\frac{\ell}{N_z+1} n')$, $\eta$ is a positive infinitesimal and $\varepsilon_{\ell}(k_{\parallel})$ is the electronic band structure for mode $\ell$ [21].
To obtain the CPA condition, we define a perturbation potential energy as
\[ V = H - H_{\text{eff}} = \sum_{\mathbf{r}, \mathbf{n}} v_{\mathbf{r}, \mathbf{n}}, \]  
where \( v_{\mathbf{r}, \mathbf{n}} = v_{\mathbf{r}, \mathbf{n}}^A \) for the A site and \( v_{\mathbf{r}, \mathbf{n}} = v_{\mathbf{r}, \mathbf{n}}^M \) for the M site are given by the following equations [27]:
\[ v_{\mathbf{r}, \mathbf{n}}^A = \sum_{\mu} |\mathbf{r}, \mathbf{n}, \mu| \{ \epsilon_A - \Delta_n \}(\mathbf{r}, \mathbf{n}, \mu), \]  
\[ v_{\mathbf{r}, \mathbf{n}}^M = \sum_{\mu\nu} |\mathbf{r}, \mathbf{n}, \mu| ((\epsilon_A - \epsilon_M)\delta_{\mu\nu} - IS_{\mathbf{r}, \mathbf{n}, \mu\nu})|\mathbf{r}, \mathbf{n}, \nu|. \]
Substitution of a fraction \( x \) of the element A by magnetic impurity atom M will include both substitutional disorder and spin scattering, hence we can expect a semi-conductor with magnetic properties. In such a case, the CPA equation for the system is expressed as:
\[ \langle t_{\mathbf{r}, \mathbf{n}} \rangle_{\text{av}} = (1 - x)t_{\mathbf{r}, \mathbf{n}}^A + x(t_{\mathbf{r}, \mathbf{n}}^M)_{\text{spin}} = 0, \]
where \( t_{\mathbf{r}, \mathbf{n}}^A (t_{\mathbf{r}, \mathbf{n}}^M) \) represents the complete scattering associated with the isolated potential \( v_{\mathbf{r}, \mathbf{n}}^A (v_{\mathbf{r}, \mathbf{n}}^M) \) due to the A(M) atom in the effective medium and \( \langle \cdots \rangle_{\text{spin}} \) denotes average over the spin scattering at the M site [27]. At the paramagnetic temperature the orientation of localized spin is completely random; hence the probability of each state is \( 1/2 \). Therefore, the associated \( t \)-matrices for an arbitrary site \( \mathbf{r} \) in the \( n \)th atomic layer are given by:
\[ t_{\mathbf{r}, \mathbf{n}}^A = \frac{\epsilon_A - \Delta_n(\omega)}{1 - (\epsilon_A - \epsilon_M(\omega))F_n(\omega)} \]
\[ t_{\mathbf{r}, \mathbf{n}}^M = \frac{1}{2} \left[ \frac{\epsilon_M - IS - \Delta_n(\omega)}{1 - (\epsilon_M - IS - \epsilon_n(\omega))F_n(\omega)} \right] + \frac{1}{2} \left[ \frac{\epsilon_M + IS - \Delta_n(\omega)}{1 - (\epsilon_M + IS - \epsilon_n(\omega))F_n(\omega)} \right], \]
where \( F_n(\omega) = \tilde{G}_{n, n}(\mathbf{r}, \mathbf{r}; \omega) \) is the diagonal element of the effective Green’s function matrix [24, 27]. From the CPA condition, Eq. (11), one can derive an equation for the self-energy of \( n \)th layer. Then, using such an equation and also Eq. (5), which gives a system of linear equations, one can obtain self-consistently the self-energy \( \Delta_n(\omega) \) and the Green’s function \( F_n(\omega) \) in each layer. Then, the LDOS per site in the \( n \)th layer, \( g_n(\omega) \), is calculated by
\[ g_n(\omega) = -\frac{1}{\pi} \text{Im} F_n(\omega). \]
On the other hand, to calculate the optical absorption spectrum, we assume that both the magnetic and non-magnetic atoms have equal transition dipole moments. Therefore, when the \( \ell \)th subband is optically excited, the layer-dependent optical absorption is given by the contribution of \( k_\parallel = 0 \) component of the electronic states that is related to the \( \Gamma \) point in the BZ [27]. Accordingly, the optical absorption of the \( n \)th layer, due to the creation of an exciton in the system, can be defined as
\[ A_n(\omega) = -\frac{1}{\pi} \text{Im} \sum_{\ell=1}^{N_\ell} G_{n, n}(\ell, k_\parallel = 0; \omega). \]
Note that, the calculation of \( A_n(\omega) \) is not restricted to the lowest \( \ell = 1 \) subband, because the numerical results showed that the contribution of \( k_\parallel = 0 \) component in the other subbands, i.e. \( \ell = 2, 3, 4 \), is finite and hence, all the subband contributions must be included.

III. RESULTS AND DISCUSSION

Based on the above formalism we study the electronic states and the optical absorption spectrum in a DMS alloy multilayer consisting of five atomic layers, i.e. \( N_x = 5 \). All the energies are measured in units of \( t \) and we set \( \epsilon_A = 0 \) as an origin of the energy. We perform the calculations for two sets of weak (\( \epsilon_M = 0.75 t \), \( IS = -1.5 t \)) and strong (\( \epsilon_M = -4.0 t \), \( IS = -3.5 t \)) interactions. It has been shown that, the band gap opening in DMSs depends on the value of magnetic ion chemical (or spin independent) potential and the strength of exchange interaction [28]. Therefore, by choosing the above values for \( \epsilon_M \) and \( IS \) we can model the system to show gap opening in the electronic states due to the doping of M atoms into the AB structure. Since the system consists of five atomic layers, one can expect five energy subbands in the band structure of the system [27]. Each subband is attributed to one of the atomic layers. In addition, due to the geometrical symmetry of the system in the \( z \) direction, the electronic states and hence the optical properties in the layers \( n = 1 \) and 5 and also in the layers \( n = 2 \) and 4 are the same. Therefore, we only present the LDOS and the optical absorption spectrum for the layers \( n = 1, 2 \) and 3.

Fig. 1 shows the dependence of electronic states on the carrier energy \( \omega \) and the alloy concentration \( x \) in the case of weak interaction, i.e., \( \epsilon_M = -0.75 t \), \( IS = -1.5 t \). Because of the 2D nature of the atomic layers of the system, one can see the van Hove singularities and steplike features in the LDOS which strongly depend on the magnetic ion concentration. The energy position of these features is different in various layers due to the quantum size effects which cause different electronic states in different layers. In the case of \( x = 0 \) the system is in completely nonmagnetic case, i.e., the original AB system, while in the case of \( x = 1 \), the AB system changes to the MB
system that is a clean magnetic system. When the alloy concentration is increased, the bottom of the bands shifts towards lower energies while the top of the bands remains approximately at a fixed energy. The shift of the bands is due to the enhancement in the spin fluctuation of $M$ ions with increasing $x$. In the limit of $x \rightarrow 1$, a dip appears close to the center of the band that for $x = 1$, its energy position is at $\varepsilon = -0.75 t$, i.e. at the on-site energy of the $M$ ions. The optical absorption spectrum, associated with the above electronic states, is shown in Fig. 2. The results show sharp features in the optical spectra corresponding to some of the features seen in the LDOS at low energies. These features are remarkable in the case of the clean system, i.e. in the $AB$ system. When the magnetic ions are introduced, that is in the case of alloy system, the absorption peaks are broadened and the height of sharp features decreases. Because of
the dependence of $A_n(\omega)$ to the electronic states through the imaginary part of Green’s function, one can see a shift in the optical spectra toward lower energies, similar to that of the electronic states. An important feature in Fig. 2 is the dependence of optical absorption spectrum on the layer number. This means that the photon absorption in low dimensional systems is different in various layers. In addition, one can find that in the case of weak interaction no energy gap appears in the optical spectrum. Accordingly, the density of states and the absorption spectrum in the weak interaction indicate that the $A$ and $M$ atomic states are amalgamated into a single band when the exchange coupling is small.

To study the effect of strong exchange interaction on the electronic and optical properties of the system, we have shown in Fig. 3 and 4 the layer-dependent of LDOS and optical absorption for $\varepsilon = -4.0 t$ and $IS = -3.5 t$. The influence of magnetic impurity causes an impurity band at the bottom of the host band, whose bandwidth depends on the $M$ concentration $x$. With increasing $x$,
the contribution of spin scattering by the localized spins in the scattering process of the carriers increases and the impurity band center and hence, the whole electronic spectrum is broadened. This broadening which does not appear in LDOS of the NMS alloys is a consequence of the localized spin fluctuation of magnetic impurities. It is evident that the strong exchange interaction splits the host band into two subbands. We see that, the behavior of the electronic states of each impurity band completely depends on the magnetic ion concentration. For \(x=1\), the two bands are symmetric and correspond to the parallel coupling and antiparallel coupling between the carrier spin and the localized spin, in agreement with the results of the CPA for ordinary magnetic semiconductors at a paramagnetic temperature. The energy position of the CPA for ordinary magnetic semiconductors at a spin and the localized spin, in agreement with the results of the CPA for ordinary magnetic semiconductors at a paramagnetic temperature. 28, 29. The energy position of the electronic band center is located at the energy \(\varepsilon = -4.0t\), i.e. at the on-site energy of the \(M\) ions, which is a consequence of the classical spin treatment of the localized spins at a paramagnetic temperature. Note that, the quantum effect of the localized spins introduces some further complications in the results and the band symmetry is broken.

One of the main differences between the electronic states in the magnetic and nonmagnetic semiconductor alloys is the variation of \(g_n(\omega)\) with increasing \(x\). In the CPA method and in the absence of magnetic impurities, a NMS alloy for \(x = 1\) is converted to the same original NMS but with an energy shift in the electronic states. This means that, the behavior of LDOS with energy variation at \(x = 1\) is the same behavior of the LDOS at \(x = 0\). By doping a NMS with magnetic impurities, however, the behavior of LDOS might be completely different with the electronic states of the original undoped system, specially at \(x = 1\). This difference appears because of the multiple scattering effect due to the exchange interaction between the carrier spin and the localized spin of magnetic ion, which is most remarkable in both cases of \(x = 0\) and \(x = 1\). Because of the dependence of optical spectrum on the electronic states, this remarkable difference for the cases of \(x = 0\) and \(x = 1\) can be clearly seen in this study. It is important to note that, if we set \(IS=0.0\), the present formalism will give the same results for a NMS multilayer in which there is no band broadening in the electronic spectra.

The electronic band splitting in the strong exchange interaction regime creates the two subbands in the optical absorption spectrum. For \(0 < x < 1\), the lower energy band corresponds to the case that the carrier spin is parallel with the localized spin, while the higher band belongs to the \(A\) atom and the case in which the carrier spin is antiparallel with the localized spin. Therefore, such an optical absorption spectrum might correspond to the persistence type. For \(x = 1\), the spectrum completely belongs to the \(M\) atoms, i.e. the clean magnetic system. Optical band broadening in the all layers can be clearly seen in the spectrum due to the chemical and magnetic disorder. Furthermore, the bands show a sharp feature at the optical band edge, depending on the value of \(x\). The strength of this feature increases for the interior layers and indicates that most of the photon energy can be absorbed by the interior layers of the system. Roughly, for \(x \leq 0.5\), the sharp features appears at the bottom of the higher band, while for \(x > 0.5\) the respective features appear at the bottom of the lower band. Note that, the band broadening, which is accompanied by energy shift of the bottom of the optical bands, demonstrates the operation of a magnetic semiconductor multilayer as an optical device which can be tuned at desired wavelength.

Another set of the results that we present here is the bulk (total) optical absorption for the weak and strong exchange interactions. Due to the fact that the optical absorption of each layer is not experimentally measurable yet, we have calculated the total magnitude of this quantity here as well (see Fig. 5). For this purpose, we make a summation over all five layers to obtain the total optical absorption of the system, i.e. \(A(\omega) = \sum_{n=1}^{N_z} A_n(\omega)\). All features that we see in Figs. 5(a) and (b) are compatible with those of the individual layers. It is clear that the
sharp features of the amalgamation type (Fig. 5(a)) at the bottom of the optical band are much stronger than those of the persistence type (Fig. 5(b)). The results also show that the optical absorption of a DMS multilayer has an obvious broadening in comparison with the NMS multilayers [21], and in the case of larger $IS$ the amount of broadening is higher and a gap opening occurs. Therefore, the transition from the amalgamation type to the persistence type in the DMS multilayers depends on the strength of the exchange coupling $IS$, and the magnetic ion concentration $x$.

IV. CONCLUSION

We have studied the effects of magnetic impurity on the electronic and optical properties of a semiconductor multilayer under the assumption that the transition dipole moments of the magnetic and nonmagnetic ions are the same. Using the single-site CPA for a random distribution of the impurity atoms, we investigated the influence of exchange interaction strength and the impurity concentration on the LDOS and the optical absorption spectra of the system. In such multilayers, the magnetic impurities shift the spectra towards lower energy-side, broaden the bands, and strongly affect on the sharp features of the spectra, even in the case of weak exchange coupling. In the case of strong exchange coupling, an energy gap opens in the electronic and optical bands. The interior layers show a higher contribution in the optical absorption process of the system relative to the surface layer, together with the fact that the amalgamation type alloys in comparison with the persistence type alloys need a higher amount of photons energy to make an excitation. These results might be helpful for the development of DMS multilayers in magneto-optical devices at a para-magnetic temperature.

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