Electronic and optical properties of InGaAs quantum wells with Mn-delta-doping GaAs barriers

Udson C. Mendes,1, * M. A. G. Balanta,1 Maria J. S. P. Brasil,1 and José A. Brum1

1 Instituto de Física “Gleb Wataghin”, Universidade Estadual de Campinas, UNICAMP, 13083-859 Campinas, São Paulo, Brazil

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We present here the electronic structure and optical properties of InGaAs quantum wells with barrier doped with Manganese. We calculated the electronic states and optical emission within the envelope function and effective mass approximations using the spin-density functional theory in the presence of an external magnetic field. We observe magneto-oscillations of the Landau levels at low-magnetic fields (B < 5 T) that are dominated by the magnetic interaction between holes spin and Mn spin, while at high magnetic fields the spin-polarization of the hole gas is the dominant effect. Our results also show that a gate voltage alter significantly the magneto-oscillations of the emission energy and may be an external control parameter for the magnetic properties of the system. Finally, we discuss the influence of the Landau Levels oscillations in the emission spectra and compare with available experimental.

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I. INTRODUCTION

The research on magnetic semiconductors has attracted much attention for more than two decades1–3. The most investigated material is the (Ga,Mn)As system, where it has been observed ferromagnetic phase with Curie temperatures (Tc) reaching 190 K for samples with Mn concentration of ~ 10%.4,5 Others (III,Mn)V materials, such as (In,Mn)As, (Ga,Mn)Sb, (In,Mn)Sb, have also shown ferromagnetic phase.6 In these materials, Mn acts as both an acceptor and a magnetic impurity and its ferromagnetism is mediated by the interaction between holes and Mn spins.7,8 As the magnetic interactions are mediated by charged carriers, the control of the magnetic properties can be achieved by electrical and optical means.9–10

Much of the research efforts have been concentrated on (III,Mn)V bulk or in its heterostructures where both Mn and holes are in the same spatial region. These structures allow a high ferromagnetic Curie temperature due to the strong interaction between the hole gas and the Mn ions. However, the hole gas is strongly scattered by the Mn ions, reducing its mobility and the optical quality. To overcome this difficulty, the (Ga,Mn)As layers were grown in the presence of a quantum well, as for example in a GaAs-(In,Ga)As-(Ga,Mn)As sequence. In this situation, the hole gas is located in the quantum well, separated from the Mn ions which need to be controlled in order to maintain a certain level of overlap between the holes and the Mn ions to assure the magnetic properties. Recently, such heterostructures have been investigated by means of transport11,12 and optical experiments.13–17. The results suggest that the interaction between holes and Mn is determinant for these systems properties.

Gazoto et al.14 investigated (In,Ga)As QWs with GaAs barriers δ-doped with both carbon and Mn in alternate sides of the QW. The samples were δ-doped with Mn in order to increase the Mn doping concentration beyond the solubility limit. The presence of the δ-doped C layer in the other side of the QW aimed to increase the hole gas concentration and, with that, to increase the magnetic effects. They observed that the circularly polarized magneto-photoluminescence presents strong oscillations with the magnetic field and they are more pronounced in the samples with higher Mn concentration. These oscillations were attributed to the Landau level filling factor. Magneto-oscillations of the circularly polarized emission were also observed in both two-dimensional electron (2DEG) and hole gas (2DHG)18–20. The origin of these oscillations is in the many-body effects of both the two-dimensional gas21,22 and in the optical recombination process.23–25. The oscillations of the transition energies observed in Ref. 14 are much stronger than those observed in other high quality 2DHG20,26. These oscillations were correlated to the presence of Mn spins in the heterostructure. Several other experiments such as low-magnetic field circularly polarized, photoluminescence excitation and time-resolved photoluminescence13,15–17 and transport measurements27, were performed in similar heterostructures. They all showed significant magnetic interaction correlated to the presence of the hole gas.

Here, we present the results of a calculation for the electronic structure and the emission energy of (In,Ga)As QWs with GaAs barriers δ-doped with Mn and C. The electronic states of the heterostructure are calculated using the spin-density function theory (SDFT)28–31 within the envelope function and effective mass approximations.32. The Mn-hole spin interaction is described by the Zener kinetic exchange theory.27,33. We aim to provide useful information to understand this system and to obtain a microscopic interpretation of the observed effects. We compare our results with the experimental results from Gazoto et al.14 and suggest an interpretation for the observed magneto-oscillations they observe in the emission spectra.

The paper is organized as follows. In Section II we present the model used to obtain the electronic states of the structure and the emission energies. In Sec. III we discuss in details the theoretical results focusing on the samples used by Gazoto et al.14. Finally, in Sec. IV we present our concluding remarks.

II. MODEL

The model heterostructure investigated is illustrated in Fig. 1(a) and it is based on the system studied by Gazoto et
It is composed by a 500 nm GaAs buffer layer, a carbon (C) δ-doping layer, followed by 10 nm GaAs spacer, a 10 nm In$_{0.17}$Ga$_{0.83}$As layer, a GaAs spacer $L_s$, a Mn δ-doping layer with a concentration $x_{Mn}$ given in percentage of monolayers (MLs), and finally a 60 nm GaAs top layer. The distance $L_s$ between the QW and the Mn doping layer and the Mn concentration will be considered as free parameters in our model to study the effects of the Mn interaction. All the other parameters are fixed. As a consequence of the Mn and C δ-doping and the thermodynamic equilibrium, the QW presents a 2DHG. We consider the system under an external magnetic field applied along the growth direction, here the z-direction.

\[ H_{hh}(lh) = H^z_{hh}(lh) + H^x_{xy}(lh), \]

where the first term is the $z$-part of the Hamiltonian and the second term is the in-plane Hamiltonian. The $xy$-part of the Hamiltonian is responsible for the formation of the Landau Levels (LLs). The Hamiltonian in the $z$-direction is

\[ H^z_{hh}(lh) = - \frac{\hbar^2}{2m_{hh}(h)} \frac{d^2}{dz^2} + v^h_{het}(lh)(z) + v(h)(z) + v_{XC}(z) + g^e \mu_B \tau^h_{zh}(lh) B + V_{pd}^{hh}(lh)(z). \]

The first term is the kinetic energy, the second is the heterostructure potential, the third and fourth terms are the Hartree and XC potentials, respectively. The fifth is the Zeeman contribution, and lastly, the hole-Mn (h-Mn) coupling. We will now describe each of the Hamiltonian terms in details.

**Heterostructure potential** - $V^h_{het}(lh)(z)$ is the structural potential which is built up from the band gap difference and band alignment between Ga$_{0.83}$In$_{0.17}$As and GaAs layers plus the strain effects. The energy gaps, at low-temperature, are 1.264 eV for GaAs and 1.519 eV for Ga$_{0.83}$In$_{0.17}$As and GaAs, respectively. The band alignment is type-I. As the QW is strained, its total band offset contains both the band gap alignment and strain contributions. We first define a band offset without strain. In this case, we assume a VB (CB) offset, $\Delta_V$ ($\Delta_C$) of 15% (85%) of the energy gap difference.

Our system is dominated by the GaAs layers and we assume that the whole structure presents the GaAs lattice parameter. This generates a compressive biaxial strain in the Ga$_{0.83}$In$_{0.17}$As layer, which alters the QW band offset and strain contributions. We first define a band offset without strain. In this case, we assume a VB (CB) offset, $\Delta_V$ ($\Delta_C$) of 15% (85%) of the energy gap difference.

We used the envelope function and effective mass approximations to describe the electronic states of the QW. The alloy is treated in the Virtual Crystal Approximation. In general to describe the valence band (VB) states it is necessary to use the six-band Luttinger-Kohn Hamiltonian that describes the heavy-hole ($hh$), light-hole ($lh$) and split-off ($so$) bands and the coupling among them as well as the spin-orbit effects. However, in our system, the In$_{0.17}$Ga$_{0.83}$As layer is under compressed stress which splits the $hh$ and the $lh$ bands by a value of the order of 50 meV. The $lh$ band is actually a type-II or marginally type-I heterostructure, depending on the parameters chosen to describe the structure. This results in a near parabolic dispersion for the $hh$ QW ground state for the energies of interest to study the optical emission. We therefore simplify our approximation considering a simple parabolic dispersion and tested it against a full Luttinger Hamiltonian calculation at zero magnetic field. The comparison is very good for the ground state responsible for the optical emission. As we go for the excited states, some discrepancies may be present but they do not alter the main results we show here.

The many-body effects of the 2DHG are considered within the SDFT. This allows us to calculate the ground-state properties of the 2DHG including exchange and correlation (XC) effects in the presence of a spin-dependent potential. We employed the Kohn-Sham minimization scheme to obtain the electronic structure. This procedure maps the many-body problem in a set of non-interacting equations, which are solved self-consistently.

We approximate the sample as being homogeneous in the plane. The $z$-direction and the in-plane ($x$, $y$) directions are therefore not coupled. The Hamiltonian can be written as

\[ H_{QW}^{hh/lh} = \Delta_V - \delta E^V_{lh} \pm \delta E_s, \]

where

\[ \delta E^V_{lh} = a_{Vlh}(\epsilon_{xx} + \epsilon_{yy} + \epsilon_{zz}) \]
\[ \delta E_s = b_{V}(\epsilon_{xx} + \epsilon_{yy} - 2\epsilon_{zz}). \]

For compressive biaxial strain, the strain components are given by $\epsilon_{xx} = \epsilon_{yy} = (a_{GaAs} - a_{GaAs})/a_{GaAs} < 0$, and $\epsilon_{zz} = -2C_{21}\epsilon_{xx}/C_{11}$, where $a_{GaAs}$ and $a_{GaAs}$ are the GaAs and Ga$_{0.83}$In$_{0.17}$As lattice parameters, respectively.
\( a_{\text{VBC(B)}} \) and \( b_c \) are the deformation potentials. \( C_{11} \) and \( C_{21} \) are elastic stiffness constants.\(^{32-36}\)

We can now turn our attention to the insertion of the Mn in the GaAs. The Ga\(_{1-x}\)Mn\(_x\)As layer has a larger lattice parameter than GaAs and follows a similar analysis as for the In\(_{0.17}\)Ga\(_{0.83}\)As layer regarding the strain effects. One additional difficulty is that the Mn strongly diffuses towards the surface in the GaAs as it was shown from secondary ion mass spectroscopy (SIMS)\(^{31-33}\). Based on the SIMS results, instead of considering a Mn \( \delta \)-doping layer, we assumed that the total Mn is distributed over many GaAs layers, forming a Ga\(_{1-x}\)Mn\(_x\)Ga\(_x\)As alloy. To take this effect into account, we assume that the system is homogeneous in the \( x-y \) directions and only variations of the Mn concentration in the \( z \)-direction are considered. We construct a distribution function that takes in account the Mn diffusion in the GaAs\(^{31-33}\). The SIMS results show that the Mn diffuses following an approximate gaussian-like function in both directions of the heterostructure. However, it diffuses more strongly in the direction of the surface than towards the QW. We consider therefore a double-gaussian distribution function, as defined below

\[
f(z - L_z) = f_0 \exp \left\{ -\left( \left( z - L_z \right)/\Delta(z) \right)^2 \right\},
\]

where \( L_z \) is the gaussian center, that is, the nominal \( \delta \)-Mn doping position. \( \Delta(z) \) is the average width of the gaussian that describes the Mn diffusion

\[
\Delta(z) = \begin{cases} 
  d & \text{if } z < L_z, \\
  D & \text{if } z \geq L_z,
\end{cases}
\]

and \( f_0 \) is the normalization constant. We considered \( D = 2 \) nm and \( d = 1 \) nm, which are compatible with the SIMS results.\(^{31-33}\) The Ga\(_{1-x}\)Mn\(_x\)Mn\(_x\)Ga\(_x\)As layers also show a different gap than GaAs, and an intrinsic band offset should be present. The main contribution for the Ga\(_{1-x}\)Mn\(_x\)Mn\(_x\)Ga\(_x\)As band offset, however, has origin in the \( sp-d \) interaction, which is discussed below. We therefore will neglect the intrinsic band offset.

Finally, we consider the \( \delta-C \) layer. C act as an acceptor and its growth is well controlled and does not significantly diffuse. We assume an homogeneously distributed 5 Å doped region in a continuous approximation. The heterostructure potential finally can be written as

\[
v_{\text{het}}^{h/h/\ell}(z) = V_{\text{QW}}^{h/h/\ell} \Theta(z^2 - L_{\text{QW}}^2/4) - \left( \delta E_{h_{\text{min}}}^{\text{VB}} + \delta E_{h_{\text{min}}}^{\text{Max}} \right) a_{\text{GaAs}} f(z - L_z)
\]

with \( \Theta(x) \) being the Heaviside function, and \( L_{\text{QW}} \) the QW width, that is, the In\(_{0.17}\)Ga\(_{0.83}\)As layer.

**Hartree potential -** The third term of \( H_z \) is the Hartree potential, which is obtained by solving Poisson’s equation

\[
\frac{d^2 \psi(z)}{dz^2} = -\frac{e^2}{4\pi\epsilon} \left[ p(z) - N_{\text{e}} \Theta(z - z_c') \Theta(z_c - z) \right] - p_{\text{Mn}} f(z - L_z),
\]

where \( \epsilon \) is the GaAs dielectric constant. \( p(z) \) is the total hole density, which at zero temperature \((T = 0 \text{ K})\) is given by

\[
p(z) = \sum_{b, n} \frac{m^2}{2\pi\hbar^2} |\psi_{b,n}(z)|^2 \Delta_{b,n} \Theta(\Delta_{b,n}) \quad \text{if } B = 0 \text{T},
\]

\[
p(z) = \sum_{b, i, \tau} \frac{m^2}{2\pi\hbar^2} |\psi_{b,i,\tau}(z)|^2 \int_{-\infty}^{E_F} g_{b,i,\tau}^{b,n}(\epsilon) d\epsilon \quad \text{if } B \neq 0 \text{T}.
\]

The first (second) line describes the hole density in absence (presence) of an external magnetic field. \( \psi_{b,n}(z) \) are the VB envelope functions, and \( \Delta_{b,n} = E_F - E_{b,n}^0 \), where \( E_F \) is the Fermi level, and \( E_{b,n}^0 \) are the eigenvalues of \( H_z^0 \)(\( b = \text{hh}, \text{lh} \)). \( E_{b,n}^0 = E_{b,n}^0 + \Gamma \) is the total subband energy of the \( b \)-hole with spin \( \tau \), in the \( n \)-th LL of the \( i \)-th subband. \( \Gamma = \Gamma_0 \sqrt{B} \) is the LL broadening related to the 2DHG mobility.\(^{34,44}\) In our calculations, we considered \( \Gamma_0 \) as a parameter.

In the absence of a gate voltage, we assume that the Fermi level, \( E_F \), is pinned at the surface states, that is, in the middle of the gap at the surface, \( E_F = E_{\text{GaAs}}^{\text{GaAs}} / 2 \). We will also consider the case of an applied gate voltage, \( V_g \), which allows us to change the Fermi level position. In this case we have \( E_F = E_{\text{GaAs}}^{\text{GaAs}} / 2 - V_g \).

**Exchange-correlation potential -** The fourth term of Eq. (2) is the XC potential. Here we use the Vosko, Wilk, Nusair (VWN) parametrization\(^{46,47}\) for the local-spin-density approximation. \( v_{\text{XC}}(z) \) depends on both the hole density \( p(z) = p_{\uparrow}(z) + p_{\downarrow}(z) \) and the hole gas magnetization \( \xi(z) = p_{\uparrow}(z) - p_{\downarrow}(z) \). Again, we consider that the density in the plane is homogeneous, and hence, the hole density depends only on the \( z \)-coordinate.

**Zeeman potential -** The fifth term in \( H_z \) is the Zeeman interaction between hole spins and the external magnetic field. \( g^* \) is the effective g-factor and \( \mu_B \) the Bohr magneton. The \( \text{hh} \) and \( \text{lh} \) spins are \( \tau_{\text{hh}}^z = \pm \frac{3}{2} \) and \( \tau_{\text{lh}}^z = \pm \frac{1}{2} \), respectively.

**p-d potential -** The last term of the Hamiltonian is the p-d interaction between holes and Mn spins. This term has its origin in the interaction between VB states with the \( d \)-orbitals of the Mn impurity.\(^{48,49}\) The presence of the hole gas is described via Zener kinetic-exchange model.\(^{2,33}\)

We assumed the magnetization vector aligned along the \( z \)-direction. This is a valid approximation since even a small magnetic field aligns the magnetization in its direction\(^{50}\) against the easy-axis.\(^{31,32,50}\)

The final expression is written as

\[
V_{pd}^{h/h}(z) = -\frac{1}{3} N_0 \beta x_{eff} M_{z}^{h/h} B_M(y) a_{Mn} f(z - L_z),
\]

where \( N_0 \beta \) is the p-d exchange constant of the spin interaction between Mn’s and holes. \( x_{eff} \) is the effective concentration
of Mn spins (see below). $M = 5/2$ is the Mn spin. $B_M(y)$ is the Brillouin function. Its argument is given by

$$y = \frac{g_M \mu_B M B}{k_B T} + \frac{J_{pd} M}{2k_B T} \int \xi(z) f(z - L_s) dz,$$  \hspace{1cm} (10)

the first term is due to the interaction of Mn spin with the external magnetic field, where $g_M$ is the Mn g-factor, $k_B$ is the Boltzmann constant, and $T$ is the temperature. The second term is the antiferromagnetic interaction between holes spins with Mn spins, which is responsible for the ferromagnetic interaction of Mn’s spins.\textsuperscript{2,27,33} $\tilde{J}_{pd} = \beta/N_0$ is the p-d exchange constant, $N_0$ is the cation concentration.

This antiferromagnetic interaction depends on the 2DHG magnetization and the overlap between hole and Mn ions. If Mn ions and holes are equally homogeneously distribute, the distribution function $a_{Mn} f(z - L_s)$ is replaced by a unitary constant and we recover the well-known results from (Ga,Mn)As bulk.\textsuperscript{7} Here, they are non-homogeneous and the second term in the function $y$ depends strongly on the structural parameters, namely, the Mn position in the GaAs and the holes states.

Equations 2, 6, and 7 are solved self-consistently. The Schrödinger equation is solved via split-operator method.\textsuperscript{52}

**Conduction band states** - The CB states are calculated within the same approximations. The in-plane Hamiltonian gives the LLs for the CB. The electron Hamiltonian in the z-direction is

$$H_z^e = -\frac{\hbar^2}{2m_e} \frac{d^2}{dz^2} + v_{he}^e(z) - v_H(z) + g_e \mu_B \sigma_z B$$

$$+ V_{sd}^e(z) + v_C(z)$$  \hspace{1cm} (11)

where $m_e$ is the electron effective mass in the CB. The second term is the CB heterostructure potential, which can be written as

$$v_{he}(z) = (\Delta_C - \delta E_{CB}^h) \Theta(z - L_{QW}/2) \Theta(L_{QW}/2 - z) - \delta E_{CB}^0 \alpha_{GaMnAs} f(z - L_s).$$

The strain contribution in the CB offset is limited to the hydrostatic term ($\delta E_{CB}^h$). The third term is the Hartree potential defined in Eq. 6. The fourth term is the Zeeman interaction, where $g_e$ is the electron g-factor and $\sigma_z = \pm 1/2$ is the electron spin. In the fifth term we have the d-d interaction between the electron’s and Mn’s spins, which is written as

$$V_{sd}^e(z) = N_{0d} \nu_{sd} f M \sigma_z B_M(y) a_{Mn} f(z - L_s),$$  \hspace{1cm} (13)

where $N_{0d}$ is the s-d exchange constant between electron and Mn spin. The last term is the effect of the correlation potential on the conduction band due to the presence of the hole gas. Since electrons and holes are treated as different particles, there is no exchange contribution for the CB. This potential was parametrized for the case of a spin-unpolarized 2DHG and gives an important contribution for the band-gap renormalization observed in the optical spectrum of modulated-doped QWs.\textsuperscript{43} From our knowledge there is no parametrization for the spin-dependent electron-hole correlation energy. We will neglect this term in our calculations. Within our description the electron-hole correlation potential should not present a dependence with the spin-polarization of the hole gas, but only on the total hole gas density. Therefore, the main effect of this contribution will depend on the total charge transfer between the hole reservoirs of the heterostructure which plays a minor effect in most of our results (see next Section).

**Optical transitions** - The transition energy is calculated as the energy difference between the electron and hole eigenstates. Our focus is in the circularly polarized emission. The right circularly polarized ($\sigma_+$) light is given by the recombination of a spin-down electron with a spin-up $hh$, while the left circularly polarized ($\sigma_-$) light is the recombination of a spin-up electron with a spin-down $hh$. The recombination energies are given by

$$E_{T_{tot}}^{\sigma_+} + E_{T_{tot}}^{\sigma_-} = E_{T_{tot}}^{\sigma_+}(\sigma_+) + E_{T_{tot}}^{\sigma_-}(\sigma_-).$$

The emission is allowed only if the electron and hole states are in the same LL ($n = n_m$). To enhance the magnetic field effects on the transition energies, we subtract the transition energy at zero magnetic field from $E_{T_{tot}}^{\sigma_+} + E_{T_{tot}}^{\sigma_-}$, redefining the transition energy as

$$E_{T_{tot}}^{\sigma_+} + E_{T_{tot}}^{\sigma_-} = E_{T_{tot}}^{\sigma_+} - E_{T_{tot}}^{\sigma_-}(0).$$  \hspace{1cm} (14)

We define the non-linear energy shift as

$$\Delta E_{T_{tot}}^{\sigma_+} + E_{T_{tot}}^{\sigma_-} = E_{T_{tot}}^{\sigma_+} - E_{T_{tot}}^{\sigma_-}(0).$$

where we subtracted all the linear terms in $B$ in order to magnify the non-linear effects in the transition energy.

**Parameters** - The holes in the heterostructure are provided by both the C and the Mn doping. We will consider the effective Mn concentrations parameters that better agree with those from [14]. The C concentration $N_C$ is fixed for all systems we investigate here. We consider $N_C = 13.35 \times 10^{18}$ cm$^{-3}$ which is the value obtained by fitting the measured hole concentration in the QW for a sample without Mn and comparing with our calculations.\textsuperscript{14} The nominal concentration of Mn, $x_{Mn}$, is known from the growth process. However, it does not provide the real hole density, since Mn can be either a substitutional impurity or an interstitial one.\textsuperscript{7} In the first case Mn replaces Ga, and provides one hole to the system, while at the interstitial position it is a double donor, and gives two electrons. Therefore, there is a self-compensation of holes by the electrons, and the total density of holes provided by the Mn is given by $p_{Mn} = x_S - 2x_I$, where $x_S$ and $x_I$ are the concentration of substitutional and interstitial Mn, respectively.\textsuperscript{7} Furthermore, because of the attractive Coulomb interaction, the interstitial Mn ions tends to be near to the substitutional ones presenting an antiferromagnetic coupling, which reduces the net Mn spins.\textsuperscript{7} The effective Mn spin concentration is given by $x_{eff} = x_S - x_I$.\textsuperscript{6,7} In our model, we only describe the uncompensated substitutional Mn, $p_{Mn}$, with the effective spin concentration, $x_{eff}$. We do not have direct access to $x_S$ and $x_I$. These values are strongly dependent on sample growing conditions. We extract $p_{Mn}$ from our calculations by fitting
the theoretical value \( p_{QW} \) with its experimental value, which was estimated from Shubnikov-de-Hass and Stoke shift measurements. This allows to determine \( p_{Mn} \) for each sample which is used as fixed parameters for the remaining calculations. The other parameters are described in Table I.

TABLE I. Parameters used in the self-consistent calculation. The In\(_{0.17}\)Ga\(_{0.83}\)As strain parameters are linear interpolation between GaAs and InAs parameters. The parameters were extracted from the refs. 7, 37–39, 55–57.

| Parameters | GaAs | InAs |
|------------|------|------|
| \( \gamma_1 \) | 6.98 | 20.0 |
| \( \gamma_2 \) | 2.06 | 8.5 |
| \( C_{11} (10^{10} \, \text{Pa}) \) | 12.21 | 8.329 |
| \( C_{21} (10^{10} \, \text{Pa}) \) | 5.66 | 4.526 |
| \( a_L (\text{Å}) \) | 5.65325 | 6.0583 |
| \( a_{ob} (\text{eV}) \) | -7.17 | -5.08 |
| \( b_v (\text{eV}) \) | -1.16 | -1.0 |
| \( b_o (\text{eV}) \) | -2.0 | -1.8 |

\[ m_{hh}^{Mn} 0.11 \]

\[ g_{c} 2.9 \]

\[ g_{a} 2.3 \]

\[ E_{B}(x_{1n}) (\text{eV}) = 1.519 - 1.583 x_{1n} + 475 x_{1n}^2 \]

\[ E_{B}(x_{1n}) (\text{eV}) = 0.15 E_{B}(x_{1n}) \]

\[ N_{s} \beta (\text{eV}) 1.2 \]

\[ J_{pd} (\text{meV nm}^{-1}) \]

\[ x_{Mn} 0.4 \]

\[ x_{eff} 0.13 \]

\[ p_{Mn} (10^{11} \, \text{cm}^{-2}) \]

\[ p_{QW}^{1st} (10^{11} \, \text{cm}^{-2}) \]

\[ 9.92 \]

\[ 5.2 \]

III. RESULTS

We consider a sample with \( x_{Mn} = 0.4 \) ML and \( L_w = 1 \) nm as our case study. The LLs broadening (\( \Gamma_0 \)) is fixed for all investigated heterostructures independently of \( x_{Mn} \). \( \Gamma_0 \) is related to the 2DHG mobility, which is approximately \( 2 \times 10^{3} \) cm\(^2\!/\text{Vs} \) at 77 K in the QW for the samples investigated in Ref. 14. This implies in \( \Gamma_0 \approx 1.8 \, \text{meV B}^{-1/2} \). The experimental photoluminescence is performed at 2 K. At lower temperatures the mobility increases and this leads to smaller values for the LLs broadening. In our calculation we considered \( \Gamma_0 = 0.25 \, \text{meV B}^{-1/2} \). Fig. 1(b) illustrates the self-consistent profile potential for the hh and lh VB. The wavefunctions in the VB represent the occupied hh-subbands. We note that there is a 2DHG in the carbon doped layer. In the Mn layer there is no occupied state. Therefore, the only hole gas interacting with the Mn ions has its origin in the QW states. We assume that the Fermi level is pinned at the surface states which creates a high electric field between the surface and the QW as it can be observed in Fig. 1(b). As a consequence of this strong potential anisotropy and the different doping at the barriers, the QW CB ground state (not shown here) is pushed towards the Mn doped barrier. These features are common for most of the case discussed here.

First we examine the evolution of the CB LLs as a function of the magnetic field. In Fig. 2(a) we plot the LLs associated to the CB QW ground state, \( x_{n \sigma}^{1} \). At low-magnetic fields, more precisely at 1.5 T, there is a crossing of the \( n=0 \) CB LLs. This crossing originates from the interplay between the Zeeman energy and the s-d interaction. It is, however, a too small effect to be observed experimentally. At high-magnetic fields, \( B > 5 \) T, we observe oscillations in the CB LLs, which are associated to the crossing of the QW hole LLs with the Fermi energy. These oscillations did not change the CB LLs dependence with the magnetic field and they show the same behavior for both spins. This is expected because the electron spin does not interact with the hole spin via exchange interaction, and also the s-d exchange coupling cause only a rigid energy shift at large magnetic field. Therefore, the observed oscillations have their origin in the charge transfer between the C layer and the QW, as it will be discussed below. We remind that the electron-hole correlation was not taken into account in our model. However, we do not expect a qualitative influence from this term on the spin-properties of the QW hole gas. It would certainly lower the energy levels and enhance the oscillations since correlation potential should be dependent on the total hole density. Its effect is associated to the oscillations in the total hole concentration. As we observed, this is a minor effect [see Fig. 3(a)]. The energy transitions calculated within this approximation should give a reliable behavior with the magnetic field.

Next we examine the VB electronic structure. Fig. 2(b) shows the LLs fan diagram. In order of increasing energy we have at zero magnetic field \( hh_{1n,\tau}^{1} \) and \( hh_{2n,\tau}^{1} \) subbands, where the upper index, \( n \) and \( \tau \), refers to the subband order, LL and hh-spin, respectively. The first and second subbands are located in the QW and in the carbon layer, respectively, as depicted in Fig. 1(b). There is also a marginally occupied light-hole subband in the carbon layer, which is not shown in Fig. 1(b) and does not play any significant role in the subsequent analysis. We observe a low- and high-magnetic field regimes for the Landau Levels magnetic-field dependence.

We first consider the low-magnetic field regime, i.e., \( B < 5 \) T. Let us concentrate on \( hh_{1n,\tau}^{1} \) and \( hh_{2n,\tau}^{1} \) states. They are the more important states for the emission spectra. In particular, for \( B \leq 2 \) T, \( hh_{1n,\tau}^{1} \) state shows an almost flat dependence with the magnetic field while \( hh_{2n,\tau}^{1} \) state shows a stronger one. The origin of this behavior is three fold: the Zeeman effect, p-d interaction, and the spin-polarization of the hole gas, which is enhanced by the hole-hole exchange energy. For low magnetic fields the spin-up and spin-down hole densities are nearly the same, and hence, the 2DHG spin-polarization does not influence its magnetic field dependence (see discussion below for high-magnetic fields, where this contribution is important). Also, in this regime, the Zeeman energy is small and its effect is to split the hh-spin states by the same amount of energy. Therefore, the hole-Mn spin interaction is the dominant effect at low magnetic fields. As the magnetic field in-
creases, the Mn spins are aligned and the Brillouin function saturates. The net result is a sizable spin-splitting originated by the $p$-$d$ coupling that remains at higher magnetic fields. The flat behavior of $hh_{0,↑}^1$ originates from the competition between $p$-$d$ interaction and diamagnetic shift of the LLs, which tends to cancel out, while for the $hh_{0,↓}^1$ they add up. This was verified by turning off $p$-$d$ interaction (not shown here) in our model.

We turn our attention now to the high-magnetic field regime, i.e., $B > 5$ T [see Fig. 2(b)]. We observe that all LLs and the Fermi level oscillate as a function of the magnetic field. These oscillations have origin in both the crossing of the LLs with $E_F$, and the charge transfer between the hole gas in the QW and the carbon layer. However, we observe that $hh_{0,↑}^1$ and $hh_{0,↓}^1$ oscillate in the same or opposite directions, depending on which spin hole level is crossing $E_F$. On the other way, the $hh_{0,↑}^2$ and $hh_{0,↓}^2$ levels roughly following the $E_F$ oscillations. Fig. 3(a) shows the QW and the C 2DHG concentration as a function of the magnetic field. It gives us a measure of the charge transfer between the QW and the C layer as a function of the LL filling factor. Fig. 3(b) shows the LLs occupation as a function of the magnetic field. Most of the LLs show a similar behavior. Their hole concentration increases linearly with the magnetic field, as it is expected from the LL degeneracy. As one LL crosses the Fermi level, it starts to be depopulated. If there was no broadening, this should be an abrupt decrease. In our case, the broadening makes the depopulation of the LL to last a finite range of magnetic field, but with a nearly linear decrease. This behavior is consistent for all LLs associated to QW subbands. The C layer LLs show a different behavior. As they start to be depopulated, they do not follow a linear behavior. Actually, this behavior is associated with the C layer LL states that roughly follow in energy the Fermi level, as if they were partially pinned on it.

Taking that in consideration let us look now in more detail in the QW LLs oscillations and their correlation with the LL filling factor. Here again we focus on the analyses of the lowest energy QW LLs, i.e., $hh_{0,↑}^1$ and $hh_{0,↓}^1$ since they are responsible for the optical emission. At $B \sim 5$ T the states at the QW that are fully occupied are $hh_{1,↑}^i, i = 0, 1$ and $\tau_s = ↑, ↓$. We have in this situation a spin-unpolarized hole gas in the QW. Other LLs from states at the C layer are also occupied but they do not affect the results we discuss here. At $B \sim 5$ T, as depicted in Fig. 3(b), $hh_{1,↑}^1$ starts to be depopulated, spin-polarizing the QW hole gas. It depopulates entirely at $B \sim 7$ T when the hole gas in the QW becomes partially spin-polarized. In this interval, 5-7 T, $hh_{0,↑}^1$ and $hh_{0,↓}^1$ have a significant difference in their magnetic field dependence, with $hh_{0,↑}^1$ energy increasing strongly with the magnetic field while $hh_{0,↓}^1$ shows a weak dependence with it. At $B \sim 7$ T $hh_{1,↑}^1$ starts to be depopulated and the QW hole gas starts to decrease its spin-polarization until $B \sim 11$ T when $hh_{1,↓}^1$ is completely emptied and the QW hole gas is spin-unpolarized. In this interval of magnetic field, both $hh_{0,↑}^1$ and $hh_{0,↓}^1$ show similar weak magnetic field dependence. At $B \sim 11$ T $hh_{0,↓}^1$ starts to be depopulated and the hole gas is again spin-polarized. The magnetic field dependence of $hh_{0,↑}^1$ and $hh_{0,↓}^1$ states again differ significantly, repeating the previous pattern. The spin-polarization and spin-unpolarization manifests itself by an oscillation in the LL dependence with the magnetic field. These oscillations take place each time a QW LL is emptied and a new one starts to be depopulated.

It should be observed that during this range of magnetic fields 5-11 T, $hh$ and $lh$ LLs associated to the C layer are also changing their occupation in relation to their maximum occupation but that does not affect the $hh_{1,↑}^1$ and $hh_{1,↓}^1$ magnetic field dependence. Actually, this picture is confirmed by the charge transfer between the C layer and the QW. Fig. 3(a) shows that the charge transfer oscillates following the QW LL filling factor. This charge transfer has its origin in the thermo-
dynamic equilibrium. However, it is not significant due to the weak overlap between \( hh^1 \) and \( hh^2 \) wave-functions. Clearly, the most important effect is the spin-polarization of the hole gas in the QW, a consequence of the charge transfer between LLs within the QW.

We now turn our attention to the consequences of these effects on the optical emission. Fig. 2(c) illustrates the fundamental energy transition shift, \( E_T^{\sigma_+ (\sigma_-)}(B) \), and the non-linear energy shift \( \Delta E_T^{\sigma_+ (\sigma_-)} \) as a function of the magnetic field. We focus our attention on the non-linear energy shifts where the magnetic effects are more clearly displayed. We first observe at low magnetic fields a strong non-linear splitting between the two polarized emissions. This is a consequence of the \( sp-d \) interaction. At higher magnetic-fields this splitting is superimposed by opposed oscillations originated from the QW holes LLs oscillations. We observe that the non-linear behavior for the \( \sigma_- \) transition increases as the QW hole gas starts to be polarized reaching a maximum value when the hole gas is spin-polarized. As this polarization starts to decrease, the non-linear behavior for \( \sigma_- \) decreases. The opposite behavior is observed for the \( \sigma_+ \) transition. It shows a negative non-linear behavior which roughly follows the same dependence. As a consequence, if we look at the transition energy shift, \( E_T^{\sigma_+} (E_T^{\sigma_-}) \) shows a maximum oscillation at odd (even) filling factor. Combining all the effects, the non-linear splitting oscillates between 1.2 meV \( (B = 3.8 \, T) \) and a maximum of 2 meV \( (B = 15 \, T) \). These values are in qualitative agreement and of the same order of the non-linear splitting observed experimentally.

Our results suggest that the oscillations observed experimentally by Gazoto et al. [Ref. 14] are a consequence of a combined effect of the spin polarized hole gas, the Coulomb exchange interaction and the \( p-d \) exchange interaction. However, in order to observe similar Mn dependence in the effect we consider the Mn ions closer to the QW as compared to their samples.

### A. Role of interactions and Mn position on the electronic structure

We shall now investigate how the spin-dependent interactions and Mn position affects the electronic properties of the 2DHG. We start our discussion considering the same sample parameters as discussed above, but turning off the \( sp-d \) interaction, with Mn ions acting as non-magnetic acceptor impurity. This gives us a clear picture of the effects induced by Coulomb exchange interaction on the hole gas, and consequently, by comparison with the previous section, we extract the effects of Mn on the hole gas. In the sequence, we change the Mn-doping position to 3 nm and turn-off the Coulomb exchange interaction. This allows us to infer about the effects Mn position on the electronic structure. Here we will focus on the hole gas LLs and transition energy behavior with the magnetic field. The CB electronic states have the same quantitative behavior as in the case described in the previous section.

Figure 4(a) illustrates the \( hh \)-LLs fan diagram in absence of the \( p-d \) exchange coupling. We first observe that the LLs always increase with the magnetic field independent of the \( hh \)-spin, differently of what was observed with the full Hamiltonian for the same heterostructure, but in the presence of the hole–Mn spin interaction. The transition energies, shown in Fig. 4(b), for both polarizations, increases linearly with the magnetic field, as expected, and show oscillations that are directly related with the LLs filling factor. A clear picture of the oscillations is given in the non-linear energy shift. For low-magnetic fields \( (B < 2 \, T) \) the non-linear energy shift presents a week dependence with \( B \). For higher-magnetic fields it oscillates with LLs filling factor, with the maximum (minimum) shift at odd (even) filling factors. This result indicates that the dominant effect of the \( sp-d \) interactions at low magnetic fields with the consequent energy splitting between the different polarizations while at higher magnetic fields the spin-polarized hole gas occupation is the dominant effect being responsible for the alternate oscillations observed.

Following, we consider the Mn layer located at 3 nm from the QW/barrier interface. This is the nominal position of the Mn layer in the samples investigated in Ref. [14]. All the other parameters are the same as discussed before. We set now \( \delta_{xc} = 0 \) in the Hamiltonian, Eq. (2), to focus on the remaining \( sp-d \) interactions. Fig. 5(a) shows the holes LLs fan diagram. We observe the non-linear energy splitting between the two polarizations at low magnetic fields. This splitting, however, is considerable diminished as a consequence of the weaker overlap of the hole wave-functions with the Mn layer. We also observe an oscillatory behavior in the \( hh^1 \) LLs with the magnetic field which is associated to the QW LL filling factor. These oscillations, however, have a completely different qualitative and quantitative behavior in comparison with the results shown in the previously section. First of all,
we do not observe a qualitative dependence with the hole-spin in the LLs oscillations. Both fundamental states, $hh_{1,\uparrow}$ and $hh_{1,\downarrow}$, oscillate following the same pattern with the magnetic field. Second, the value of these oscillations is significantly diminished. This can be better visualized in Fig. 5(b) where we plot the energy transition and the non-linear energy shift as a function of the magnetic field. The non-linear behavior is the same for both circularly polarized transitions, as expected from the $hh$ LLs behavior. The tiny split observed in the total energy shift is of the order of 0.1 meV. These results confirm the combined origin of the oscillations in the Coulomb exchange interaction and the $p$-$d$ exchange interaction in the presence of the hole gas. They also demonstrate that the Mn ions have to be close to the QW states to observe a sizeable $p$-$d$ coupling.

![Figure 5](image-url)

**FIG. 5.** (Color online) (a) LL fan diagram for heavy-holes. The energy states are, in order of increasing energy, labelled as $h_{1,\uparrow}$ (solid), $h_{1,\downarrow}$ (dash-dot), $h_{1,\uparrow}$ (dash-three-dots), $h_{1,\downarrow}$ (dash two-dots) and $E_{F}$ (dash-line), respectively. (b) Transition energy (right $y$-axis) and non-linear energy shift (left $y$-axis) as function of the magnetic field. We considered $L_{a} = 3$ nm and $v_{XC} = 0$.

**B. Gate Voltage**

It is interesting to consider now the possibility to control the magnetic effects in the structure by the application of an electric field. This can be achieved through a gate voltage modifying the Fermi level in the structure and, therefore, the carrier distribution. Essentially, we will simple change the Fermi level at the surface by the expression $E_{F} \rightarrow E_{g}/2 - V_{g}$ and will consider the value $V_{g} = 0.71$ eV which leads to an almost flat band condition near the surface. We consider a value of 1 nm for the Mn layer spacing. All the other parameters are the same.

Figure 6(a) shows the $hh$ potential profile and the wave-functions of the occupied $hh$ levels at (a) $B=0$ T and for (c) $B=10$ T, (b) the $hh$ LLs fan diagram and (d) the $hh$-LLs charge concentration as a function of the magnetic field. We first observe that there are three occupied $hh$ states at $B = 0$ T. As the magnetic field increases, this picture changes completely. The Mn rich region becomes attractive for the spin up $hh$s and repulsive for spin down $hh$s, and the QW spin up levels are pushed towards this region. As a consequence, following the previous terminology, $hh_{1,\uparrow}$ strongly overlaps with the Mn region. The same happens for $hh_{1,\downarrow}$ although it is less pronounced. $hh_{2}$ also presents a significant overlap with the Mn region, while $hh_{1}$ is mainly confined in the QW. The states $hh_{2}$ and $hh_{3}$ become almost fully confined in the C layer. All this evolution in the wave-functions produces a complex structure in the LLs fan diagram.

![Figure 6](image-url)

**FIG. 6.** (Color online) (a) and (c) Self-consistent profile potential and heavy-hole wave-functions for $B = 0$ T and $B = 10$ T, respectively. (b) Heavy-hole LLs and (d) two-dimensional density of the LLs. The LLs and its density are, in order of increasing energy, labelled as $h_{1,\uparrow}$ (solid), $h_{1,\downarrow}$ (dash-dot), $h_{2,\uparrow}$ (dash-three-dots), $h_{2,\downarrow}$ (dash two-dots) and $E_{F}$ (dash-line), respectively. For $x_{Mn} = 0.4$ MLs, $V_{g} = 0.71$ eV and $L_{a} = 1$ nm.

We focus now our analysis in the $hh$ ground-state LLs, that is, $hh_{0,\uparrow}$ and $hh_{0,\downarrow}$. At low magnetic fields, the two states show a strong non-linear behavior, with $hh_{0,\uparrow}$ showing a convex curvature while $hh_{0,\downarrow}$ shows a concave curvature with the magnetic field. This regime is entirely dominated by the $p$-$d$ interaction. At higher magnetic fields, for $B > 6$ T, the oscillations with the LLs filling factors become more pronounced and dominate the features. However, in this case, we do not observe the difference in the magnetic field dependence from spin up and spin down with the odd and even filling factors which was originated from the hole gas exchange energy in the presence of the spin-polarization. Here, both states oscillate in a similar way. The main reason for this behavior is in the way that the LLs associated to the QW are depopulated. This can be observed in Fig. 6(d). In particular, for $B \sim 13$ T, both LLs associated to spin up and down become depopulated almost at the same magnetic field. This prevents the spin-polarization of the QW hole gas. At the same time, the participation of the states $hh_{1}$ in the polarization of the
QW hole gas prevents a clear oscillation in this polarization. On the other way, the states $\hbar \omega_{1,\uparrow}$ have a strong influence of the Mn ions which dominates the magnetic field dependence. The oscillations are therefore mainly dominated by the charge transfer among the QW levels but not in the polarization of the hole gas.

FIG. 7. (Color online) (a) CB LL fan diagram. (b) Transition energy (right y-axis) and non-linear energy shift (left y-axis) as function of the magnetic field. $x_{\text{Mn}} = 0.4$ MLs, $V_0 = 0.71$ eV and $L_z = 1$ nm.

Figure 7 shows (a) the CB LLs fan diagram and (b) the transition energies and the non-linear energy shift for this case. The CB LLs show the usual oscillation associated to the charge transfer between different hole gas reservoirs. The non-linear shift observes a rapid split at low magnetic fields due to the increasing of the $s-p$ interaction with the magnetic field. For $B > 3$ T this splitting saturates and the oscillatory behavior dominates its features. As it was already mentioned for the $hh$ LLs, we observe symmetric oscillations for the two circularly polarized transitions associated to the LLs filling factor. These results show that by applying a gate voltage we are able to qualitatively change the behaviour of the optical emission.

IV. CONCLUDING REMARKS

We presented here the results of calculations of the electronic structure and optical emission energy for (In,Ga)As QWs with barriers doped with Mn and C acceptors with a focus on the influence of the Mn impurities. Our results clearly show that for a significant magnetic effect on the optical emission the Mn layer has to be near the QW interface, allowing a strong overlap between QW wave functions and Mn spin. We can separate two regimes for the optical emission effects. At low-magnetic fields, $B < 5$ T, the $p-d$ exchange interaction dominates the effects while at higher magnetic fields the spin-polarization of the hole gas becomes the dominant effect. We also demonstrated that the non-linear effects can be controlled applying an external electric field.

Our results explain the experimental observations by Gazoto et al. [Ref. 14]. The oscillatory magneto-shift observed in the magneto-photoluminescence can be interpreted as a combined effect of the exchange hole interaction in the presence of the spin-polarized hole gas enhanced by the $p-d$ interaction. However, to observe a clear dependence with the Mn it is necessary to consider a Mn layer closer to the QW than the nominal value.

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