Spin Dynamics Driven by the Electron-Hole Exchange Interaction in Quantum Wells

M. Z. Maialle

Universidade São Francisco, 13251-900 Itatiba-SP, Brazil

Received on April 23, 2001

We present a theoretical study of the spin dynamics of site-localized excitons in semiconductor quantum wells driven by the electron-hole exchange interaction. Using a simple model for the distribution of sites we observed spin dephasing leading to a strong spin polarization decay.

I Introduction

In general, control over the degree of spin orientation of photocreated charge carriers in semiconductor structures is achieved by polarized-light excitation of optical transitions in conformity with the angular momentum selection rules. In semiconductor quantum wells (QWs) the spin dynamics results from the individual dynamics of the electron spin (components $\pm 1/2$, in units of $\hbar$) or heavy hole spin (angular momentum projections $\pm 3/2$), or from the spin dynamics of the bound electron-hole pair (exciton). Individual electron and hole spinflips follow from band spin mixing in addition to momentum scattering, and they have little efficiency on cold carriers at the band edges of narrow QWs due to vanishing spin mixing and reduced available phase space for scattering.

In intrinsic QWs the band-edge optical properties are dominated by the exciton state for which the correlation between the electron and hole *spins* is set by the electron-hole (e-h) *exchange* Coulomb interaction. Although small, the exchange contribution to the spin dynamics becomes important in the aforementioned situation of strongly confined carriers at the band edges. The long-range part of the e-h exchange interaction (LRX) depends on the exciton center-of-mass (c.m.) momentum, such that scattering leads to an exciton spin relaxation according to a motional narrowing process, for which shorter scattering time τ^* gives longer exciton spin relaxation time τ_s (which is valid for $\tau^* \ll \tau_s$). However, such mechanism cannot be directly applied for samples that exhibit lateral localization of exciton in sites created by QW interface imperfections. Localized excitons do not scatter as frequently as free excitons, such that the motional narrowing process is inappropriate if $\tau^*_{\rm loc} \approx \tau_s$. Also, the localization of the exciton in large sites quantizes the c.m. motion and as a result the LRX coupling for exciton spin states $|+1\rangle \longleftrightarrow |-1\rangle$ vanishes in symmetric sites.[1]

The inhomogeneity of sites and it effects on the exciton spin dynamics via the e-h exchange have been addressed experimentally and theoretically by Nickolaus *et al.*[2] for (Zn,Cd)Se/ZnSe QWs. They have found that the dominant mechanism in the spin decay was not a true relaxation process, but instead a dephasing of the macroscopic spin polarization resulting from the LRX matrix elements, which are subjected to the same inhomogeneity of the sites. Further evidences of the combined role of the exciton localization and e-h exchange interaction are provided by experiments in QWs with transverse magnetic field (Voigt configuration) where Larmor precessions of the electron spin are observed in the case that the exchange interaction lacks strength to correlate the electron and hole spins. Otherwise, the precessing electron spin would have to drag the heavy-hole spin that, in first approximation, is pinned along the growth axis. In GaAs/AlAsGa QWs[3] electron spin precessions were observed, however, in (Zn,Cd)Se/ZnSe QWs,[4] the precessions were seen only at high temperatures (~100 K).

In this paper, we present a brief account of the theoretical study[5] of the spin dynamics in ZnSe/(Zn,Cd)Se QWs including the effects of exciton localization, e-h exchange interaction and spin precessions about a transverse magnetic field.

II Spin dynamics by the e-h exchange interaction

The exchange interaction matrix for QWs is calculated in the effective-mass approximation using an exciton ground-state wavefunction written as

$$\Phi_{1s,\mathbf{K}}(\mathbf{r}_e,\mathbf{r}_h) = \xi(z_e)\zeta(z_h)\varphi_{1s}(\rho)\frac{e^{i\mathbf{K}\cdot\mathbf{R}}}{\sqrt{A}}, \qquad (1)$$

where ρ and $\mathbf{R}$ are the relative and c.m. e-h coordinates in the QW plane of area A, respectively. $\hbar\mathbf{K}$ is the in-plane c.m. momentum. The envelope wavefunctions for the QW ground states for electron and heavy hole are respectively $\xi(z)$ and $\zeta(z)$. Spin mixing between heavy holes and light holes is neglected.

The LRX Hamiltonian[6] can be written as[7]

$$H^{\rm LR}(\mathbf{K}) = F(K) \begin{bmatrix} 0 & 0 & 0 & 0 \\ 0 & K & Ke^{-2i\phi} & 0 \\ 0 & Ke^{2i\phi} & K & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix}, \quad (2)$$

where the matrix elements are arranged for the exciton spin basis in the order $|+2\rangle$, $|+1\rangle$, $|-1\rangle$, $|-2\rangle$. Also, $\mathbf{K}=(K,\phi)$ in polar coordinates and $F(K)$ gives the quasi–two-dimensional exchange strength. Assuming no mixing with the light-hole states, the short-range part of the exchange interaction is diagonal in the exciton spin basis and can written as[7]

$$H_{i,j}^{\rm SR} = \Delta_{\rm SR}\delta_{i,\pm1}\delta_{j,\pm1}, \quad (3)$$

with $\Delta_{\rm SR}$ setting the splitting between the dark $|\pm2\rangle$ and optically active $|\pm1\rangle$ excitons.

II.1 Localized excitons

We treat only the localization of excitons in sites larger than the exciton Bohr radius. In this way, the exciton c.m. motion in the QW plane is assumed to be bound into the minima of the site potential. We describe such bound state in the harmonic approximation

$$\Psi_{\mathbf{b}}(\mathbf{K}) = \left(\frac{b_x b_y}{\pi^2\hbar^2}\right)^{\frac{1}{4}} \exp\left[-\frac{1}{2}(b_x K_x^2 + b_y K_y^2)\right], \quad (4)$$

where $\mathbf{b}=(b_x,b_y)$ specifies the shape of the site, with $b_x=\hbar/\sqrt{MV_{xx}}$ given by the second derivative V_{xx} of the site potential along the site principal axis x at the minimum (and similarly for y). The exciton mass M is for the dispersion on the QW plane.

A spin Hamiltonian for the LRX can be obtained by first-order perturbation theory for the localized exciton if we neglect the K dependence on $F(K)$ and substitute K in the matrix Eq. (2) by

$$\overline{K_{\mathbf{b}}}^{(d/o)} = \int d\mathbf{K}\,|\Psi_{\mathbf{b}}(\mathbf{K})|^2 \,\frac{K_x^2 \pm K_y^2}{\sqrt{K_x^2+K_y^2}}, \quad (5)$$

with the + and − signs holding for the diagonal (d) and off-diagonal (o) terms, respectively. The short-range exchange term for localized states remains as in Eq. (3) since it does not depend on $\mathbf{K}$ and because of the approximation used that the relative e-h motion is not modified by c.m. localization.

We model the distribution of sites as done by Wilkinson *et al.* [8] using a Gaussian random function to simulate the in-plane potential $V(\mathbf{r})$ created by the QW imperfections. The distribution of potential is characterized by two parameters: the variance σ_V^2 of the site potential and the correlation length l such that $\langle V(\mathbf{0})V(\mathbf{r})\rangle=\sigma_V^2\exp(-r^2/2l^2)$, where the brackets denote either ensemble average or spatial average. The absorption optical density in this model is a Gaussian function of variance σ_V^2. The density probability of minima $N(V)$ of this potential gives a narrower distribution centered at a lower energy when compared to the absorption Gaussian curve. This model was used to interpret the Stokes shift and Nickolaus *et al.*[2] have used it to derived the density of minima $N(V,V_{xx},V_{yy})$, now also as a function of the second derivatives, that gives the distribution of sites with different shapes. The inhomogeneity in the values of the exchange matrices for the sites is calculated using the distribution of minima $N(V,V_{xx},V_{yy})$.

=bf II.2 Equations of motion – Coherent spin dynamics

The equation of motion for the density matrix $\rho(\mathbf{K})$ (with elements $\rho_{i,j}=|i\rangle\langle j|$, where $|i\rangle=|\pm2\rangle, |\pm1\rangle$ are the exciton spin states) is given by

$$\frac{d\rho(\mathbf{K})}{dt} = \frac{i}{\hbar}[\rho(\mathbf{K}),H(\mathbf{K})] + \left(\frac{\partial\rho}{\partial t}\right)^{\rm incoh} + G, \quad (6)$$

where G is the generation rate, $\left(\frac{\partial\rho}{\partial t}\right)^{\rm incoh}$ contains incoherent contributions such as the recombination rates for the optically active excitons with spins $|\pm1\rangle$ and spin relaxation for electrons and hole due to spin-orbit interaction. An applied magnetic field $\mathbf{B}$ introduces a Zeeman term in H

$$H_{\rm e}^{\rm B} = \mu_B g_e \frac{1}{2}\sigma^{\rm e}\cdot\mathbf{B} \quad \text{and} \quad H_{\rm h}^{\rm B} = \mu_B g_h \frac{3}{2}\sigma_z^{\rm h} B_z, \quad (7)$$

where μ_B is Bohr magneton, $g_{e(h)}$ is the electron (hole) g factor and σ are Pauli matrices used for the electron ($s=\pm\frac{1}{2}$) and heavy hole ($m=\pm\frac{3}{2}$) spins. Notice that only the z component of $\mathbf{B}$ acts on the heavy holes (the other components would involve smaller contributions due to mixing with the light holes).[9] The equation of motion Eq. (6), with the contributions Eqs. (2), (3) and (7), is a system of first-order linear differential equations that couples all the 16 matrix elements $\rho_{i,j}$. In the most general cases, no physically simple solution emerges from the equations of motion and in the following they are solved numerically. The time dependent solutions are then average out using the distribution of minima $N(V,V_{xx},V_{yy})$ as weight to account for the distribution of sites.

III Results and discussion

We have solved the problem of a free exciton in a $Zn_{0.8}Cd_{0.2}Se$/ZnSe QW using a trial wavefunction with two variational parameters.[10] The exchange induced splittings we have obtained were compared to the available experimental data $\Delta E_{\rm exch}$=0.25 meV [4] and $\Delta E_{\rm exch}$=0.5 meV.[11] Reasonable values were found when using the *bulk* exchange splittings $\Delta E_{\rm SR}$=0.2 meV and $\Delta E_{\rm LR}$=0.8 meV. In what follows we consider a 5nm-QW for which $\Delta_{\rm SR}$=0.3 meV in Eq. (3) and F= 14 meVÅ in Eq. (2). In addition, we have used[11] g_e=1.1 and looked at excitons with energy $V=-1.5\sigma_V$ (luminescence tail)[2] with σ_V=6 meV and site correlation length l=200 Å. The exciting pulse is a Lorentzian of width 1 ps centered in t=0 s with circular polarization σ^+ exciting excitons $|+1\rangle$.

In Fig.1(a) we show the time evolution of the electron and hole spin components along the z direction (growth axis) for zero magnetic field. We have set zero decaying rates $\left(\frac{\partial\rho}{\partial t}\right)^{\rm incoh}$=0 in Eq. (6), such that the decay observed in Fig. 1(a) for the spin components are actually due to spin dephasing. The physical reason for such dephasing is that the LRX, through the off-diagonal terms in Eq. (2), induces precessions of the hole and electron spins about each other. The distribution of sites yields a similar distribution on the exchange strength and consequently on the precession frequency resulting in dephasing. In Fig. 1(b) a transverse magnetic field is applied B_x=2 T. Besides the site-exchange induced dephasing, now the electron spin goes through Larmor precession about the field.

In Fig. 1(c) we have used different correlation lengths along the x and y directions, that is, there is still a distribution of inhomogeneity of sites as in the results of the previous figures, but now elongated sites are more likely. In this case, the LRX assumes a distribution about nonzero values, which set a finite frequency for the exchange induced e-h spin precession. This is seen by the long period oscillation on the time evolution of the electron and hole spin components.

In conclusion, we have modeled the spin dephasing induced by the e-h exchange interaction for excitons localized in sites created by imperfections on the QW interfaces. The spin dephasing yielded quite short spin decay times ~30 ps when using typical parameters for (Zn,Cd)Se/ZnSe QWs, which shows the relevance of such process in the study of spin dynamics in these systems. In GaAs QWs the role of spin dephasing by the e-h exchange for localized excitons is not so clear because in these systems excitons exhibit weaker localization and the exchange strength is considerably smaller ($\Delta E_{\rm exch}\sim$ 0.05 meV). Electron-hole spin precession that leads to dephasing is then less likely to happen.

Acknowledgments: This work was supported by FAPESP and PROPEP-USF (Brazil).

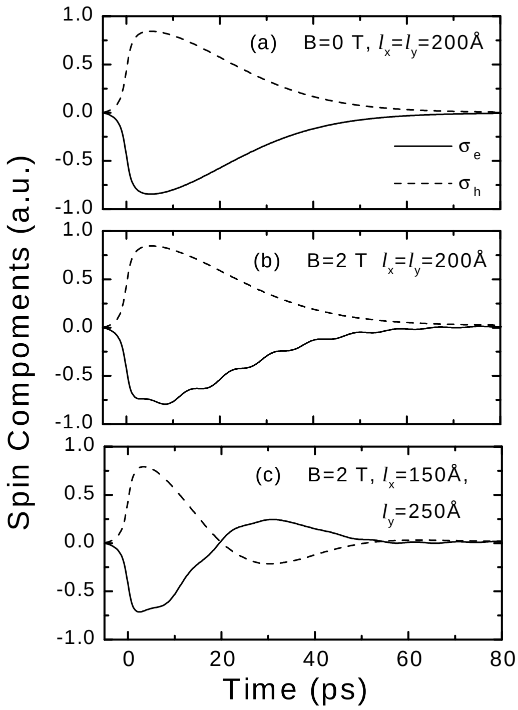

Figure 1. Time evolution of the spin components of electron $\sigma_z^{\rm e}$, in full lines, and heavy hole $\sigma_z^{\rm h}$, in dashed lines. (a) No applied field B=0 T, correlation length l=200 Å for the x and y directions. (b) Same as before but for B=2 T. (c) Time evolutions calculated for B=2 T, l_x=150 Å and l_y=250 Å, simulating elongated sites.

References

[1] S. V. Goupalov, E. L. Ivchenko, A. V. Kavokin, J. Exp. Theor. Phys. **86**, 388 (1998) [Zh. Eksp. Teor. Fiz. **113**, 703 (1998)].

[2] H. Nickolaus, H. -J. Wünsche, and F. Henneberger, Phys. Rev. Lett. **81**, 2586 (1998).

[3] T. Amand, X. Marie, P. Le Jeune, M. Brousseau, D. Robart, and J. Barrau, Phys. Rev. Lett. **78**, 1355 (1997).

[4] S. A. Crooker, D. D. Awschalom, J. J. Baumberg, F. Flack, and N. Samarth, Phys. Rev. B **56**, 7576 (1997).

[5] M. Z. Maialle, Phys. Rev. B **61,** 10877 (2000).

[6] G. E. Pikus and G. L. Bir, Zh. Eksp. Teor. Fiz. **60**, 195 (1971). [Sov. Phys.-JETP **33**, 108 (1971)].

[7] M. Z. Maialle, E. A. de Andrada e Silva, and L. J. Sham, Phys. Rev. B **47**, 15776 (1993).

[8] M. Wilkinson, Fang Yang, E. J. Austin, and K. P. O'Donnel, J. Phys.: Condens. Matter **4**, 8863 (1992).

[9] X. Marie, T. Amand, P. Le Jeune, M. Paillard, P. Renucci, L. E. Golub, V. D. Dymnikov, and E. L. Ivchenko, Phys. Rev. B **60**, 5811 (1999).

[10] To be published elsewhere.

[11] J. Puls and F. Henneberger, Phys. Stat. Sol. (a) **164**, 499 (1997).