Photoluminescence of Double Quantum Wells: Asymmetry and Excitation Laser Wavelength Effects

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Circularly polarized photoluminescence (PL) spectroscopy measured at 19 K on GaAs/AlGaAs symmetric and asymmetric double quantum wells (DQW) is reported. The PL is obtained by exciting the sample with a circularly polarized (left or right) laser in order to create an initial unbalanced distribution of electron spins in the conduction band and, in this way, obtain the electron spin lifetime $\tau_s$. The effects of the excitation laser wavelength are estimated by exciting with laser wavelengths of 701.0, 787.0, 801.5, and 806.5 nm. The increase of $\tau_s$ with the excitation wavelength is attributed to the lower initial quasimomentum $k$ of the excited carriers, which also reduces spin–orbit relaxation processes. $\tau_s$ is found to be higher in asymmetric DQWs: this is attributed to the wider QWs in these samples, which reduces spin relaxation due to the Dresselhaus mechanism. In addition, a smaller contribution from the Rashba mechanism is also detected by comparing samples with built-in electric fields of different orientations defined by doped barrier layers.

1. Introduction

Spin dynamics has become a focus of interest in semiconductor physics in the last years due to the ever closer need of the development of quantum devices. It is important to have an efficient control of spin currents and the manipulation and detection of spin states. For the latter, the value of the spin relaxation time $\tau_s$ is very important. The spin–orbit coupling (SOC) of carriers in a semiconductor plays an important role in the spin dynamics and, in particular, in the spin relaxation time $\tau_s$. SOC induces an energy splitting of the spin states that depends on the quasimomentum $k$ of the carriers. In a quantum well (QW) or a double quantum well (DQW) consisting of two QWs separated by a thin tunneling barrier based on GaAs/AlGaAs structures, the main mechanisms that induce the SOC splitting are the Dresselhaus and the Rashba effects. The relative strength of these mechanisms is quite important and allows the tuning of $\tau_s$. In our case, the thickness of the QWs in the DQW system is different, that is, the DQW is asymmetric. The asymmetry and the built-in electric field in our DQW structures are, in general, very important to establish the relative strengths of the Dresselhaus and the Rashba effects.

Figure 1 shows a simplified energy band diagram corresponding to a QW structure or a DQW. Due to the reduction in symmetry from $T_d$ to $D_{2h}$ relative to bulk GaAs, the heavy- and light-hole levels lift their degeneracy. In the photoluminescence (PL) setup used in this work, the sample is excited with a $\sigma^+$ polarized laser beam with an energy larger than the energy between conduction and light-hole bands. Under this condition, the transitions $-3/2 \rightarrow -1/2$ and $-1/2 \rightarrow +1/2$ are excited with intensities $I \propto 3/4$ and $I \propto 1/4$, respectively. The difference in intensities creates an initial unbalance in the distribution of electron spins in the conduction band. If the recombination lifetime $\tau$ of the carriers is much longer than $\tau_s$ (i.e., if spin relaxation occurs), the intensity of the PL signal will be the same for the $\sigma^+$ and $\sigma^-$ polarizations. In the opposite case, the difference in intensities is associated to the spin distribution of the electrons in the conduction band before recombination.

When an electron–hole pair is optically excited, the electron spin in the conduction band can be depolarized via the Dyakonov–Perel (DP) mechanism in a time $\tau_\Omega$ before its recombination. In the DP mechanism, the electron spin precesses around an effective magnetic field caused by the Dresselhaus and the Rashba effects. The effective magnetic field and the corresponding precession frequency $\Omega$ depend on the quasimomentum $k$ of the electrons. During $\tau_\Omega$, the electron can scatter with other electrons, phonons, and crystalline imperfections, and thus, in general, $\Omega$ changes randomly.

An electron in an initial spin state $+1/2$ or $-1/2$ and an initial momentum $k$ just after the optical excitation will suffer collisions losing its energy and reaching the bottom of the conduction band ($k \approx 0$) before recombination. The initial momentum $k$ is determined by the excitation wavelength. During the time $\tau$, the...
the electron follows a path in the k-space, from the initial k value to the bottom of the conduction band. The effective magnetic field is proportional to \( k \cdot k \). If the initial modulus of k in the PL experiment is decreased, the maximum magnetic field and the depolarization of the spins will also be reduced. Thus, for excitation energies close to the bandgap of the DQW, the DP mechanism can be diminished. In this way, by varying the excitation energies, the average degree of polarization of the spins before the recombination can be tuned.

As previously mentioned, a built-in or externally applied electric field to the DQW structure induces a Rashba effect. Considering that one of the main contributions of this work is to demonstrate experimentally the dependence of the degree of circular polarization of the PL induced by the asymmetry of the DQW structure, the contribution of the built-in electric field of the structure must be quantified. To take into account this point, two types of DQW structures were studied. One with an AlGaAs n-type barrier and the other with an AlGaAs p-type barrier.

The DQW structures used in this work are shown in Figure 2. The Al\(_x\)Ga\(_{1-x}\)As layers in the structure have an Al concentration of \( x = 0.15 \). The QWs have thicknesses of 23.7 and 11.9 nm separated by a thin AlGaAs layer of 2.0 nm, constituting an asymmetric coupled system. Two structures are considered, one with the lower AlGaAs barrier composed by an AlGaAs (300 nm) and a n-type (6 \( \times 10^{18} \) cm\(^{-3} \)) AlGaAs (600 nm) layers and the other by an AlGaAs (300 nm) and a p-type (5 \( \times 10^{19} \) cm\(^{-3} \)) AlGaAs (600 nm) layers. The samples were grown by molecular beam epitaxy (MBE) on a GaAs (001) semi-insulating substrate covered with a 200 nm-thick smooth GaAs buffer layer. As the Fermi energy at the sample surface is pinned approximately at the center of the bandgap, the n- and p-doped layers will generate electric fields of opposite directions across the DQW structure. This fact has an impact in the strength of the PL polarization as it will be

Figure 1. a) Electron–hole pairs are generated by exciting with a \( \sigma^+ \) polarized laser. Light is absorbed with strengths of 3/4 and 1/4 for electron-HH transitions [3/2 \(-\) 3/2] and electron-LH [3/2 \(-\) 1/2], respectively. After the absorption, the spin population for \(-\)1/2 and \(+\)1/2 has a ratio of 3:1. b) Electrons scatter and reach the bottom of the conduction band before recombination. The relative intensity of the PL strength for \( \sigma^- \) and \( \sigma^+ \) polarization is associated to the population of \(-\)1/2 and \(+\)1/2 spins in the conduction band and thus can be used to study the spin dynamic during the recombination time \( \tau \).

Figure 2. Structure of the asymmetric DQWs used in this work. Two samples with an AlGaAs layer of n- and p-type are studied. A circular polarized laser (\( \sigma^+ \) or \( \sigma^- \)) of different wavelengths is used to excite the sample. The PL is collected and the spectra for both \( \sigma^- \) and \( \sigma^+ \) polarizations are measured at \( T = 19 \) K. All of the Al\(_x\)Ga\(_{1-x}\)As layers in the structure have an Al concentration of \( x = 0.15 \).
discussed. To support our results, a symmetric DQW was also analyzed. In this symmetric DQW, both QWs have 11.9 nm of thickness and it contains a 600 nm thick n-type (6 \times 10^{18} \text{ cm}^{-3}) layer similar to the n-type asymmetric DQW sample described above. Table 1 summarizes the main parameters of the DQWs structures.

| Sample       | AlGaAs barrier doping level | QWs thicknesses $d_1$, $d_2$         |
|--------------|-----------------------------|-------------------------------------|
| Symmetric    | Si-doped $n = 6 \times 10^{18} \text{ cm}^{-3}$ | 11.9, 11.9 nm                       |
| Asymmetric   | Si-doped $n = 6 \times 10^{18} \text{ cm}^{-3}$ | 23.7, 11.9 nm                       |
| Asymmetric   | Be-doped $p = 5 \times 10^{18} \text{ cm}^{-3}$ | 23.7, 11.9 nm                       |

2. Theory

In 2D systems, such as the asymmetric DQWs used in this work, the spin relaxation $\tau_s$ of the electrons in the conduction band is dominated by the Dyakonov–Perel (DP) mechanism.\(^{5,9,10}\) The spin relaxation rate is given by\(^{5,9,10}\)

\[
\frac{1}{\tau_s} = \langle \Omega \cdot \Omega \rangle \tau_p
\]  

(1)

where $\langle \Omega \cdot \Omega \rangle$ is the mean of the precession vector's square magnitude in the QW plane; $\tau_p$ and $\tau_s$ are the spin and momentum relaxation times, respectively. The precession vector $\Omega$ is associated to the built-in effective magnetic field induced, in general, by the Dresselhaus and the Rashba effects. The Dresselhaus effect\(^{2,4,7}\) is associated to the lack of inversion symmetry in the zincblende symmetry semiconductors ($T_d$ symmetry), while the Rashba effect is further reduced by extrinsic effects. This occurs if an electric field is applied perpendicular to the (001) surface or for asymmetric QWs.\(^{11-13}\) In the latter case, an asymmetry may appear in DQWs with barriers of different heights,\(^{11-13}\) with different chemical composition\(^{16}\) or by triangular barriers\(^{11}\) for instance.

In principle, the relative values for the Dresselhaus and the Rashba effects (and, consequently, the value of the precession vector) can be controlled by: 1) the built-in electric field of the DQW structure and 2) the relative thicknesses of the QWs. The electric field induces a Rashba effect that depends linearly with the electric field and can then be reversed if the AlGaAs barrier is n- or p-type. If the DQW system is symmetric (and no electric field is applied), only the Dresselhaus effect is important and the effective magnetic field will have no preferential in-plane orientation, leading to an efficient spin depolarization. In the case of the asymmetric DQW system, if both the Rashba and the Dresselhaus mechanisms have similar strengths, the direction of the effective magnetic field becomes uniaxially and parallel to the DQW plane. This case leads to the formation of helical spin density waves, the so-called persistent spin helix (PSH).\(^{17-21}\) In this case, $\Omega$ is oriented only along either the [110] or the [1$ar{1}$0] direction.

The degree of circular polarization of the PL is defined as

\[
P = \frac{P^- - P^+}{P^- + P^+}
\]  

(2)

where $P^+$ and $P^-$ are the PL intensity for $\sigma^+$ and $\sigma^-$ polarizations. The spin relaxation time $\tau_s$ is related to the degree of circular polarization of the PL according with\(^{9}\)

\[
P = \frac{P_o}{1 + \tau/\tau_s}
\]  

(3)

where $\tau$ is the recombination lifetime of the electrons and $P_o$ is the polarization degree if no relaxation of the spin occurs. In our experiments, the polarization degree of PL is expected to have a maximum value of $P_o = 0.5$. In the limits $\tau_s << \tau$ and $\tau_s >> \tau$, $P$ tends to zero and $P_o$, respectively.

3. Experimental Results and Discussion

Figure 3 shows the PL spectra for the n-type sample for excitation wavelengths of 701.0, 787.0, 801.5, and 806.5 nm. The excitation energy range lies above the electron-light-hole (LH) and the electron-heavy-hole (HH) transitions. The excitation laser has a $\sigma^+$ circular polarization state and the $\sigma^+$ and $\sigma^-$ PL curves are displayed as blue and red spectra, respectively. For the HH transitions (HH in Figure 3), the PL intensity of the $\sigma^-$ polarization is stronger than the one for the $\sigma^+$, while for the LH transitions (LH in Figure 3) this relation is the opposite. This fact is expected considering that HH transitions $-1/2 \rightarrow -3/2$ and $+1/2 \rightarrow +3/2$ produce PL with polarizations $\sigma^-$ and $\sigma^+$, respectively, while for the LH transitions $-1/2 \rightarrow +1/2$ and $+1/2 \rightarrow -1/2$ produce PL with polarizations $\sigma^+$ and $\sigma^-$, respectively. Thus, the degree of circular polarization has the opposite sign for the HH and LH transitions. Note that the degree of circular polarization becomes larger (in particular, for the HH transition) when the excitation energy approaches the HH transition. This can be interpreted in the frame of the reduction of the initial quasimomentum $k$. With this reduction, the average magnitude of $k$ must be smaller and, consequently, the average precession vector's magnitude $\langle \Omega \cdot \Omega \rangle$, leading to an increase in the relaxation time $\tau_s$. To demonstrate that the degree of circular polarization is directly related to the asymmetry of the DQW structure, the inset of Figure 3 shows the PL spectra for the symmetric DQW for an excitation of 787.0 nm. In this case, $P \approx 0.001$ is more than one order of magnitude smaller than the ones obtained for the asymmetric DQW structure at the same excitation wavelength. It is important to note that for any excitation wavelength used in this work, $P$ does not change significantly for the symmetric DQW.

It is important to note that the different laser wavelengths will also excite the barriers and the sub-bands of the DQW structure. In the case of the barrier (excited only with the 701 nm laser), a fraction of the excited electrons may diffuse to the QW suffering also spin depolarization (in the same way than the electrons excited in the QW) reaching the bottom of the QW conduction band with some degree of polarization and have some contribution to the bottom PL spectra of Figure 3 and 4. This contribution can be quantified comparing the PL intensities for 787 and...
701 nm. We found that the intensities of both PL are comparable, concluding that there is no significant contribution of the barriers to the spin polarization. In the case of the sub-bands, the polarization of the spins for the HH and LH transitions (in the ratio 3:1) only applies for excitation in resonance with these transitions in a QW. The excited spin polarization changes for laser wavelengths away from these transitions because the excited sub-bands will no longer have a pure HH or LH character (and are also expected to be modified by the DQW structure).

![Figure 3](image-url)

Figure 3. PL spectra for the asymmetric DQW with a n-type AlGaAs barrier. The excitation wavelength for each spectrum is a) 701 nm, b) 787 nm, c) 801.5 nm, and d) 806.5 nm. Blue and red spectra correspond to the intensity of the PL for $\sigma^+$ and $\sigma^-$, respectively. As can be seen, the degree of polarization increases with the excitation wavelength. Inset shows the spectra for the symmetric DQW excited with 787.0 nm. Note that in this case $\sigma^+$ and $\sigma^-$ spectra do not differ significantly.

In addition, the excited spins can flip as it relaxes to the bottom of the electron-HH band before recombination, thereby reducing the measured spin polarization. The laser excitation wavelength of 806.5 nm only the sub-band ($E_1$) is expected to be excited considering that the energy of the second sub-band ($E_2$) is located around 1.536 eV (806.9 nm). Thus, only the laser wavelengths of 801.5, 787 and 701 nm excite HH, LH and their higher-lying sub-bands. The sub-bands with energy higher than $E_1$ are expected to have approximately the same relative weight of spins.
The electrons of these sub-bands will contribute to the PL in a region below the wavelength shown in Figure 3 and 4. From the above discussion, the initial spin state distribution in the level $E_1$ induced by the exciting laser may be considered independent on the laser wavelength.

To quantify the effect of the built-in electric field on the degree of polarization of the PL, a DQW structure with p-type AlGaAs barrier was measured. Figure 4 shows the equivalent experiments of Figure 3, but now performed on the p-type structure. For the PL excited with the 701.0 nm laser wavelength, the degree of circular polarization for the HH transition increases a factor of 2, while for 806.5 nm the factor diminishes to 1.2. Considering that the electric field has opposite direction for the n- and p-type DQWs, the effect on the polarization degree is not so large. This fact suggests that the Rashba effect induced by the electric field has a small contribution in the spin depolarization. However, a Rashba effect induced by the asymmetry could have a contribution. If only the Dresselhaus effect is considered, a possible explanation to the increase of $P$ in the asymmetric DQW is that the PL from the asymmetric DQW arises from the wider QW, which has a smaller bandgap. In QWs, the

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**Figure 4.** PL spectra for the asymmetric DQW with a p-type AlGaAs barrier. The excitation wavelength for each spectrum is a) 701 nm, b) 787 nm, c) 801.5 nm, and d) 806.5 nm. Blue and red spectra correspond to the intensity of the PL for $\sigma^+$ and $\sigma^-$, respectively. As can be seen, the degree of polarization increases with the excitation wavelength.
The Dresselhaus effect reduces with the inverse square of thickness, increasing $\tau_s$. However, to clarify this point more experiments and calculations must be performed on these DQW systems.

The dependence of the degree of polarization $P$ on the excitation wavelength is shown in Figure 5a,b for the HH and LH transitions, respectively. As we mentioned at 701 nm, $P$ has its maximum difference for the n- and p-type DQW structures. When the wavelength increases, $P$ tends to increase rapidly in both cases. This fact is consistent with the fact that when the excitation approaches to the gap of the structure (814.7 nm), the electron in the initial $k$ has its minimum possible value and in the same way the average precession vector's magnitude $\Omega$ reaches its minimum. This condition implies that electrons with lower initial momentum have a higher probability of conserving their spin after photoexcitation. By using Equation (1), the experimental values of $P$ (for HH transition) and a recombination lifetime for the electrons of the order of $\tau = 1.0$ ns,[22] the relaxation time can be estimated. For the n- and p-type samples, $\tau_s$ takes values in the range from 0.07 to 0.3 ns and 0.17 to 0.5 ns, respectively.

4. Conclusion

The results reported in the present work demonstrate that the asymmetry in a DQW system increases the value of the degree of circular polarization in PL measurements. The experiments also show the dependence of the spin relaxation time of electrons in the conduction band excited by different laser wavelengths in PL measurements. By measuring the degree of circular polarization of the PL, it was demonstrated that the spin relaxation time increases for wavelengths closer to energy in the fundamental gap of the DQW system. The degree of PL circular polarization was found to be higher for asymmetric than for symmetric DQW structures. The latter is attributed to the fact the electrons are stored in a wider QW in the case of the asymmetric samples. Furthermore, we compared the spin lifetimes of asymmetric samples with a n- and a p-type AlGaAs lower DQW barriers and, thus, with built-in electric fields of opposite amplitudes. $\tau_s$ was found to be slightly larger for samples with a p-doped barrier, thus demonstrating the presence of a small Rashba effect. We believe that the asymmetric DQWs constitute an excellent system to characterize and study the spin dynamics in 2D semiconductor structures.

5. Experimental Section

Experiments were carried out by using a low-vibration, helium closed-cycle cryocooler (19 K). The excitation of the samples was performed by using semiconductor lasers with wavelengths of $\lambda = 701$ and 787 nm and a Ti:Sph tunable laser for the wavelength of $\lambda = 801.5$ and 806.5 nm. The power density in any case was $I = 0.6$ mW mm$^{-2}$. By using a $\lambda/4$ plate and a liquid crystal half-wave variable retarder in tandem, the polarization of the laser can be switched between left- and right-circular polarization. The laser incidence was normal to the surface of the sample and focused within a spot size of 1.0 mm diameter. The PL was directed to a linear polarizer prism to select the left- and right-circular polarizations, by rotating the prism. The PL signal was collected and detected by using an imaging spectrometer with a resolution of 0.025 nm equipped with a thermoelectric-cooled camera as detector.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding upon reasonable request.

Figure 5. Polarization degree ($P$) of the HH and LH transitions for the asymmetric QW with a) n-type and b) p-type barriers versus the excitation wavelength. The lines are guides to the eye.
Keywords
photoluminescence, quantum wells, spin dynamics

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[1] A. Hirohata, K. Yamada, Y. Nakatani, I.-L. Prejbeanu, B. Diény, P. Pirro, B. Hillebrands, J. Magn. Magn. Mater. 2020, 509, 166711.
[2] G. Dresselhaus, Phys. Rev. 1955, 100, 580.
[3] S. D. Ganichev, V. V. Bel’kov, L. E. Golub, E. L. Ivchenko, P. Schneider, S. Giglberger, J. Eroms, J. De Boeck, G. Borghs, W. Wegscheider, D. Weiss, W. Prettl, Phys. Rev. Lett. 2004, 92, 256601.
[4] J.-W. Luo, G. Bester, A. Zunger, Phys. Rev. Lett. 2009, 102, 056405.
[5] P. S. Eldridge, J. Hübner, S. Oertel, R. T. Harley, M. Henini, M. Oestreich, Phys. Rev. B 2011, 83, 041301.
[6] A. Violante, R. Hey, P. V. Santos, Phys. Rev. B 2015, 91, 125302.
[7] M. Schlipf, F. Giustino, Phys. Rev. Lett. 2021, 127, 237601.
[8] M. I. Dyakonov, V. I. Perel, in Optical Orientation, Vol. 8 (Eds: F. Meier, B. P. Zakharchenya), North-Holland, Amsterdam 1984, Ch. 2.
[9] M. I. Dyakonov, in Spin Physics in Semiconductors, Springer Series in Solid-State Sciences, Vol. 157 (Ed: M. I. Dyakonov), Springer-Verlag, Berlin, Germany 2008, Ch. 1.
[10] R. T. Harley, in Spin Physics in Semiconductors, Springer Series in Solid-State Sciences, Vol. 157 (Ed: M. I. Dyakonov), Springer-Verlag, Berlin, Germany 2008, Ch. 2.
[11] N. S. Averkiev, L. E. Golub, A. S. Gurevich, V. P. Evtikhiev, V. P. Kochereshko, A. V. Platonov, A. S. Shkolnik, Yu. P. Efimov, Phys. Rev. B 2006, 74, 033305.
[12] D. J. English, J. Hübner, P. S. Eldridge, D. Taylor, M. Henini, R. T. Harley, M. Oestreich, Phys. Rev. B 2013, 87, 075304.
[13] Y. F. Hao, Y. H. Chen, Y. Liu, Z. G. Wang, EPL 2009, 85, 37003.
[14] Y. F. Hao, Phys. Lett. A 2015, 379, 2859.
[15] L. Han, Y. Zhu, X. Zhang, P. Tan, H. Ni, Z. Niu, Nanoscale Res. Lett. 2011, 6, 1.
[16] D. Richards, B. Jusserand, H. Peric, B. Etienne, Phys. Rev. B 1993, 47, 16028.
[17] J. D. Koralek, C. P. Weber, J. Orenstein, B. A. Bernevig, S.-C. Zhang, S. Mack, D. D. Awschalom, Nature 2009, 458, 610.
[18] M. P. Walser, C. Reichl, W. Wegscheider, G. Salis, Nat. Phys. 2012, 757.
[19] B. A. Bernevig, J. Orenstein, S.-C. Zhang, Phys. Rev. Lett. 2006, 97, 236601.
[20] F. Dettwiler, J. Fu, S. Mack, P. J. Weigele, J. C. Egues, D. D. Awschalom, D. M. Zumbühl, Phys. Rev. X 2017, 7, 031010.
[21] S. Karimi, C. A. Ullrich, I. D’Amico, F. Perez, Sci. Rep. 2018, 8, 1.
[22] E. O. Göbel, H. Jung, J. Kuhl, K. Ploog, Phys. Rev. Lett. 1983, 51, 1588.