

Spin splittings in nanostructures without inversion symmetry

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Abstract: In a crystal lacking inversion symmetry, in the presence of spin-orbit interaction, the spin degeneracy is in general removed at $k \neq 0$ even in the absence of an external magnetic field. Such is the case for bulk III-V semiconductors having the zincblende structure. In nanostructures having a reduced symmetry with respect to the bulk, additional zero-field spin-splitting mechanisms are allowed. The interplay between the different spin-orbit interactions is thoroughly investigated and the relevance of the spin-splittings for excitonic properties is addressed. In particular, the competition between intraexciton exchange and single particle spin flips in the relaxation of the exciton luminescence polarization is analysed.

INTRODUCTION

It is well known that in crystals having a center of inversion the spin degeneracy of the electronic states is not removed by the spin-orbit interaction. In fact, time reversal symmetry guarantees that a state with wavevector \vec{k} and spin up has the same energy of one having wavevector $-\vec{k}$ and spin down (Kramers degeneracy). Then, inversion symmetry guarantees that a state with wavevector $-\vec{k}$ and spin down is degenerate with one having wavevector \vec{k} and spin down. Therefore, in a centrosymmetric crystal, with spin taken into account, even in the presence of spin-orbit coupling, the electronic states at any given \vec{k} are at least doubly degenerate. Of course, the spin degeneracy is removed by the Zeeman interaction in an external magnetic field \vec{B} which breaks time reversal symmetry. In crystals which are not centrosymmetric, however, the spin degeneracy of the electronic states with $\vec{k} \neq 0$ is in general removed by the spin-orbit interaction in the absence of any *external* magnetic field. Such zero-field spin splittings can be thought of as due to the total crystal electric field seen by the spin of a moving electron as an *internal* \vec{k} -dependent magnetic field; such interaction does preserve time reversal symmetry.

The spin splitting in the Γ_6 conduction band of zinc-blende semiconductor compounds, due to inversion asymmetry, has been predicted long ago by Dresselhaus [1], and detected with different experimental techniques [2], one of its characteristic manifestations being a beating pattern in the amplitude of the magneto-oscillations [3, 4]. The corresponding term in the effective mass Hamiltonian is proportional to k^3 and of the form

$$H_D = \gamma \left(\sigma_x k_x (k_y^2 - k_z^2) + \sigma_y k_y (k_z^2 - k_x^2) + \sigma_z k_z (k_x^2 - k_y^2) \right), \quad (1)$$

where σ_i stands for the Pauli spin matrices, γ is a material constant and the coordinate axes are parallel to the crystallographic cubic axes. In the degenerate Γ_8 valence band, instead, the splitting

is linear in k as given by[1]

$$H_V = \frac{C}{\sqrt{3}} (k_x (J_x J_y^2 - J_x J_z^2 + J_y^2 J_x - J_z^2 J_x) + \text{cyclic permutations}) , \quad (2)$$

where J_i are the $j = 3/2$ hole spin matrices, and C is a material constant.

In semiconductor nanostructures such as asymmetric quantum wells or inversion layers, there are additional spin splitting mechanisms due to the lack of mirror symmetry with respect to the plane perpendicular to the growth axis. For the conduction electrons, in particular, Rashba[5] has pointed out the existence of a spin-orbit interaction linear in k of the form

$$H_R = \alpha \vec{\sigma} \cdot (\vec{k} \times \hat{z}), \quad (3)$$

where \hat{z} is the growth axis and α a phenomenological parameter. The Rashba term, in general, depends both on the band bending and the heterojunction discontinuities. In III-V semiconductor asymmetric nanostructures, the total electron zero-field spin splitting is due to both the Dresselhaus and the Rashba contributions.

The spin splittings in the bulk and heterostructures of zincblende semiconductors have been studied in many different contexts[6, 7]. In the following, we report on our recent work on the electron spin-orbit couplings in 2D asymmetric nanostructures and discuss how such spin splittings are germane to exciton spin relaxation.

2D ASYMMETRIC NANOSTRUCTURES

A very useful theoretical approach to the study of the electronic properties of semiconductor nanostructures is the envelope function approximation. The simplest possible model to study in a consistent way the Rashba term as due to both interface discontinuities and band bending is the eight band $\vec{k} \cdot \vec{p}$ Kane model. The resulting multiband effective mass Hamiltonian can be projected on the subspace of the conduction electron states giving rise to a Pauli like equation for a two component wavefunction (f_+ , f_-) containing all the effects of the electrostatic potential and material composition variation along \hat{z} on the spin-orbit interaction. With the same notation as in Ref.[7], we finally have

$$\left[-\frac{\hbar^2}{2} \frac{d}{dz} \frac{1}{m(z, \varepsilon_{\pm})} \frac{d}{dz} + \frac{\hbar^2 k^2}{2m(z, \varepsilon_{\pm})} + E_c(z) + V(z) \mp \left(\frac{d\beta}{dz}(z, \varepsilon_{\pm}) k - \varepsilon_{\pm} \right) f_{\pm} \right] = 0, \quad (4)$$

with

$$\frac{1}{m(z, \varepsilon_{\pm})} = \frac{P^2}{\hbar^2} \left(\frac{2}{\varepsilon_{\pm} - V(z) - E_v(z)} + \frac{1}{\varepsilon_{\pm} - V(z) - E_v(z) + \Delta(z)} \right), \quad (5)$$

and

$$\beta(z, \varepsilon_{\pm}) = \frac{P^2}{2} \left(\frac{1}{\varepsilon_{\pm} - V(z) - E_v(z)} - \frac{1}{\varepsilon_{\pm} - V(z) - E_v(z) + \Delta(z)} \right). \quad (6)$$

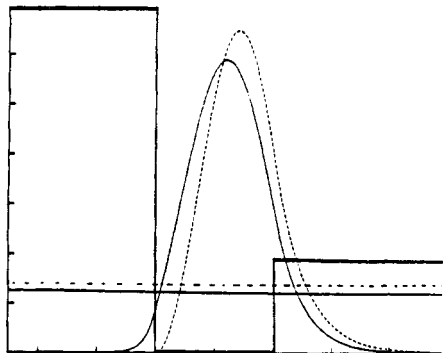


Figure 1: Flat band asymmetric AlAs/GaAs/Al_{0.3}Ga_{0.7}As quantum well (width of 4 nm): solid and dashed lines refer, respectively, to the case with and without wavefunction penetration in the AlAs barrier

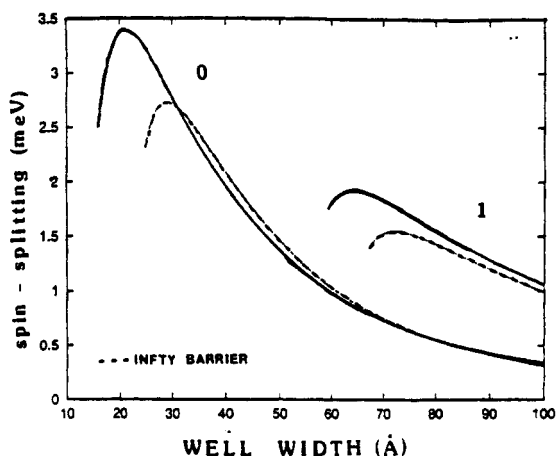


Figure 2: Spin splitting for the quantum well of Fig. 1 as a function of well width: the first and, when present, second confined state splittings are shown for a fixed parallel wavevector $k = 0.03 \text{ \AA}^{-1}$.

The \pm sign refers to spin up and down along the y direction, the 2D wave vector being set along x ($\vec{k} = k \hat{x}$). The Kane momentum matrix element, taken to be independent of z [8], is denoted by P , E_c , E_v , V , Δ are, respectively, the conduction band edge, the valence band edge, the band bending electrostatic potential energy and the spin-orbit splitting in the valence band. For a given k , the solutions $\varepsilon_{\pm}(k)$ are the spin dependent eigenenergies; $|\varepsilon_+ - \varepsilon_-|$ being the zero-field spin splitting. In a lowest order perturbation treatment, the β term corresponds to the Rashba term (proportional to $\vec{\sigma} \cdot \vec{k} \times \hat{z}$). When considering the dependence of β on z in equation (4) for a generic asymmetric heterostructure, two contributions of the Rashba spin-orbit splitting can be identified: the first is proportional to the space-charge and/or external electrostatic field (V), the second is related to the discontinuities of the band profiles (E_v , Δ). The former gives a spin-dependent term in the effective mass Hamiltonian proportional to the long range band bending, the latter corresponds to an interface short range term that, in the case of ideal abrupt junctions, can be dealt with using the following spin-dependent boundary conditions:

$$f_{\pm} \text{ continuous and } -\frac{\hbar^2}{2m} \frac{df_{\pm}}{dz} \mp \beta k f_{\pm} \text{ continuous.} \quad (7)$$

The two contributions to the Rashba term are in general comparable; under flat band conditions only the short range term survives, whereas in an inversion layer the interface contribution is negligible when the conduction band offset (the confining barrier) is much larger than the valence band offset. In the latter case, the Rashba term coupling constant can be expressed as:

$$\alpha = \frac{\hbar^2}{2m^*} \frac{\Delta}{E_g} \frac{2E_g + \Delta}{(E_g + \Delta)(3E_g + 2\Delta)} eE, \quad (8)$$

being E the electric field and m^* the band edge effective mass. We stress that the above approach is equivalent to the solution of the multiband Kane model (nonparabolicity included) and that $E_c(z)$ and $V(z)$ clearly do not play the same role, which is consistent with other treatments [10, 11]. As

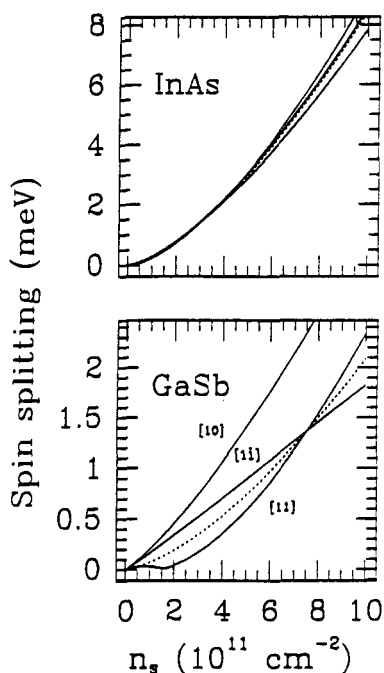


Figure 3: Zero-field spin splitting at three points of the Fermi "surface" along different directions in k -space (indicated in the case of GaSb), as a function of the carrier density n_s in inversion layers. The dashed line gives the isotropic contribution of the Rashba term alone.

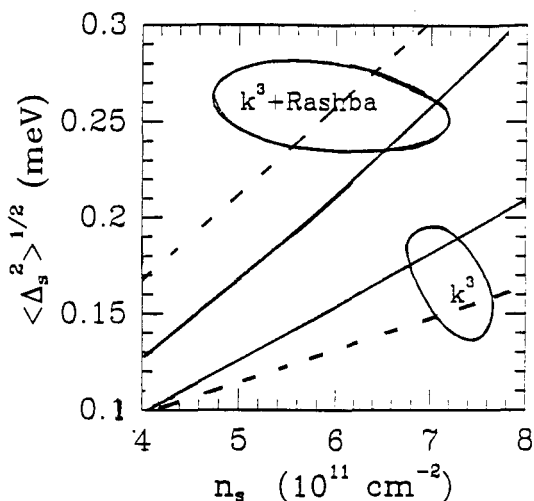


Figure 4: Comparison of the computed total spin splitting averaged over the Fermi surface with that obtained in Ref. 10, shown by dashed lines. Two theoretical cases are considered: one with both contributions to the spin splitting and one with only the Dresselhaus term.

an example, we consider the problem of the stationary states for electrons confined in asymmetric semiconductor quantum wells of the type $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}/\text{Al}_y\text{Ga}_{1-y}\text{As}$. Sizable splittings are obtained with a pure AlAs barrier on one side of the GaAs quantum well. The conduction band edge profile and the first bound state are as shown in Fig. 1. In Fig. 2 we plot the splitting at a fixed parallel wave vector $k = 0.03 \text{ \AA}^{-1}$ as a function of the well width. It is also interesting to compare the results with those obtained in the infinite barrier approximation, when no wavefunction penetration is allowed in the AlAs barrier, as shown by the dashed lines; in this case, the difference is small as the splitting is dominated by the penetration in the lower barrier junction.

As mentioned in the introduction, the Dresselhaus term proportional to k^3 also contributes to the zero-field electron spin splitting in III-V semiconductor nanostructures (symmetric or not). This splitting is much smaller than the quantized subband energy ε of the confined electrons and its effects can be treated within first order perturbation theory. In this case, it is sufficient to take the expectation value of H_D on the unperturbed electron subband wavefunction without modifying the boundary conditions[12]. The lowest subband of the triangular well corresponding to an inversion layer can be well described with the Fang-Howard trial function,

$$f_{\pm}(z) = \sqrt{\frac{b^3}{2}} z e^{-bz/2}, \quad (9)$$

where b is a variational parameter determined by minimizing the total energy. When the Dresselhaus term is comparable to the Rashba one, the spin splitting exhibits a typical anisotropy with respect

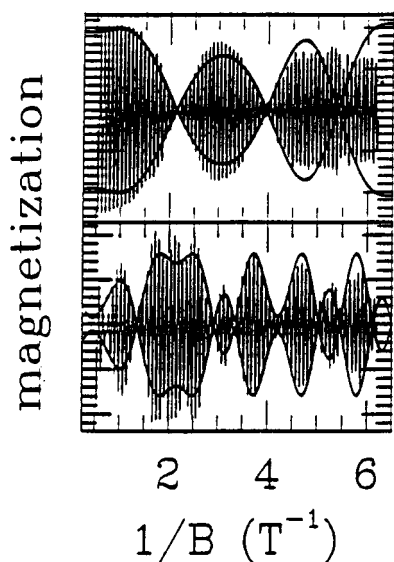


Figure 5: Anomalous magneto oscillations for a $n_s = 6.0 \times 10^{11} \text{ cm}^{-2}$ GaSb heterojunction under zero and 50 kV/cm of applied electric fields. We have traced with envelope curves the semiclassical magnetic breakdown fit.

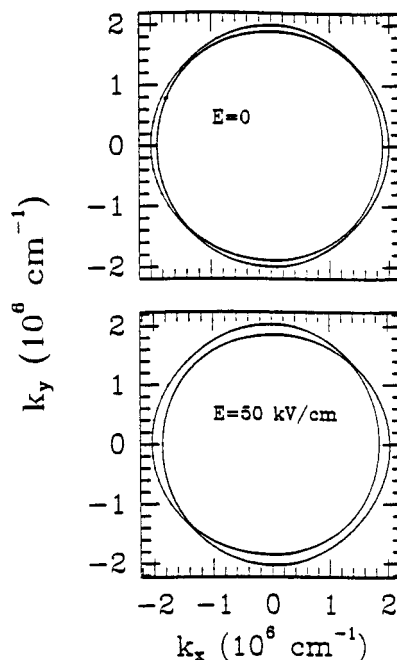


Figure 6: The 2D Fermi "surfaces" for the GaSb heterojunctions considered in Figure 5. The two curves correspond to the two spin split subbands and give the two sheets of the Fermi "surface".

to the 2D wavevector \vec{k} [7]. The total spin splitting Δ_s , for a triangular well is given by:

$$\Delta_s(k, \theta) = 2[(\gamma^2 \frac{b^4}{16} + \alpha^2)k^2 + \gamma\alpha(k^2 - \frac{b^2}{2})k^2 \sin 2\theta + \gamma^2(k^2 - b^2) \frac{k^4}{4} \sin^2 2\theta]^{1/2}, \quad (10)$$

where θ is the angle between the parallel wave vector \vec{k} and the x cubic axis. The total (Rashba + Dresselhaus) spin splitting in inversion layers of InAs and GaSb is shown in Fig. 3 by the solid lines, the anisotropy being particularly strong for the case of GaSb (notice the difference between [11] and $\bar{1}\bar{1}$ directions). This anisotropic pattern has been recently confirmed directly by spin flip Raman scattering[9]. Our results for the spin splitting in a GaAs inversion layer averaged over the Fermi surface are shown in Fig. 4 together with the corresponding results from a more sophisticated calculation[10]: in view of the transparency of our approach as well as of the uncertainty in the material parameters the agreement is quite satisfactory.

A very sensitive probe of the electronic structure are the magneto-oscillatory phenomena like the de Haas-van Alphen effect: the magnetization exhibits a periodicity in $1/B$ with frequency given by $\hbar c S_F/e$ (Onsager relation) where S_F represents the section of the Fermi surface (area in reciprocal space) perpendicular to the magnetic field \vec{B} . In the presence of zero-field spin splittings, the Fermi surface has two sheets and, thus, the magneto-oscillations show a beating frequency proportional to $S_F^+ - S_F^-$ [3, 4]. In a two dimensional system such frequencies are simply proportional to the electron

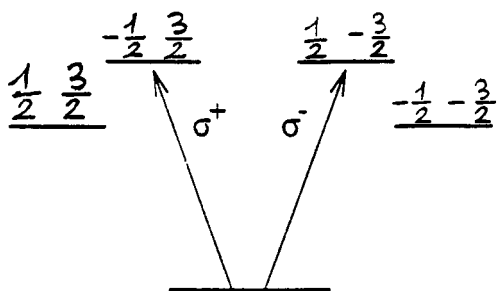


Figure 7: Schematic diagram of the HH exciton spectroscopy, the states are labelled with the electron and hole J_z quantum numbers. The two optically active states can be selectively excited with circularly polarized light, the two “dark” states are also shown.

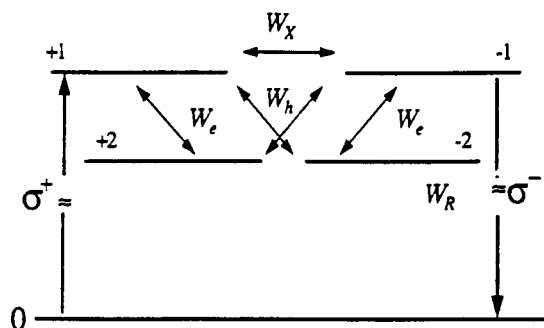


Figure 8: Schematic diagram of the heavy hole exciton spin relaxation: the two optically active states can transform into one another either through a single step exchange spin flip or through a double step single particle spin relaxation.

densities in the split bands:

$$n_{\pm} = \frac{1}{(2\pi)^2} \int d\vec{k} \Theta(\epsilon_F - \epsilon_{\pm}(\vec{k})), \quad (11)$$

$\Theta(x)$ being the Heaviside function and ϵ_F the Fermi energy. For strongly anisotropic splittings, the possibility of magnetic breakdown leads to a complex beating pattern from the measurement of which it is possible to extract very detailed information on the various spin splitting mechanisms[3, 4, 7, 13]. As an example, we show in Fig. 5 the simulated de Haas-van Alphen effect for an inversion layer of GaSb with $n_s = 6.0 \cdot 10^{11} \text{ cm}^{-2}$; the lower panel shows the effect of an additional bias field which increases the spin splitting anisotropy (see Fig. 6) leading to a very complex beating pattern. The heavy solid lines are the envelopes obtained from a semiclassical fit[7] based on the Fermi surface sections shown in Fig. 6.

2D EXCITON SPIN RELAXATION

With the development of fast time resolved spectroscopy it has been possible to address the question of the exciton spin relaxation in quantum wells as revealed by the decay of the degree of polarization of the luminescence excited with circularly polarized light. In a zincblende direct gap semiconductor quantum well (e.g., GaAs/AlGaAs), the fundamental optical absorption is dominated by the heavy hole exciton resonance. The heavy holes (HH) having a lower confinement energy with respect to the light holes (LH) possess an angular momentum projection along the growth axis $J_z = \pm 3/2$. Therefore, the HH exciton may have an angular momentum projection $J_z = \pm 2$ or $J_z = \pm 1$ as given by the sum of the electron and hole J_z (see Fig. 7). The two latter states are optically active, while the former are “dark”; the active states are split from the dark ones by the short range electron-hole exchange interaction. The electric dipole selection rules relevant to optical orientation of the HH exciton are depicted in Fig. 7. The exciton spin dynamics following the excitation with circularly

polarized light rules the decay of the time resolved polarized luminescence[14]. The depolarization is either due to a single step process driven by the long range exchange interaction in which electron and hole flip their spins simultaneously[15] or to a two step process driven by single particle spin relaxations in which an optically active state first changes into a dark one and then into the oppositely polarized active one (see Fig. 8). It is therefore clear that if the rate of either one of the single particle spin flips (the electron spin relaxation rate W_e or the hole one W_h) is lower than the rate of the direct exciton exchange process (W_X), the latter dominates. This is the situation observed in Ref. 14 in a high quality 150 Å AlGaAs/GaAs symmetric quantum well, for which an accurate fit of the time resolved polarized luminescence gives $W_X = 1.5 \cdot 10^{10} \text{ s}^{-1}$, $W_h = 0.7 - 1.0 \cdot 10^{10} \text{ s}^{-1}$, $W_e = 0.3 - 3.0 \cdot 10^9 \text{ s}^{-1}$. The hole spin relaxation rate is dominated by effects related to the HH-LH valence band mixing, and is fast compared to W_e . The electron spin relaxation is dominated by the (motional narrowing) D'yakonov-Perel mechanism[16] related to the zero-field spin splitting Δ_s , by:

$$W_e = \left(\frac{\Delta_s}{\hbar} \right)^2 \tau_p, \quad (12)$$

being τ_p the momentum scattering time (assumed to be related to the homogeneous linewidth). In this case, we estimate Δ_s taking the expectation value of the Dresselhaus term on the exciton bound electron wavefunction obtaining:

$$\Delta_s \approx \frac{2 m_e Q_{\parallel} \gamma}{m_e + m_h} \sqrt{\frac{\pi^4}{L^4} - \frac{\pi^2}{L^2} \frac{1}{a_B^{*2}} + \frac{1}{4} \frac{1}{a_B^{*4}}}, \quad (13)$$

where L is the effective well width (allowing for barrier penetration), Q_{\parallel} the 2D exciton center of mass wavevector (from the kinetic energy related to the homogeneous linewidth) and a_B^* the 2D exciton effective Bohr radius (given by a variational calculation). From $\Delta_s \approx 10 \mu\text{eV}$ and $\tau_p \approx 6 \text{ ps}$, the spin relaxation rate turns out to be $W_e \approx 1.4 \cdot 10^9 \text{ s}^{-1}$, i.e. of the right order of magnitude. However, such an agreement should be checked examining the expected trends of W_e with variations of well width and linewidth. In the conditions of Ref. 14, however, the sensitivity to W_e is very low because of the large value of W_X . Such a situation can be changed diminishing W_X by a drastic reduction of the electron-hole overlap[14, 15]. This can be easily achieved by employing a system with *space indirect* excitons, as in biased resonant couple wells[17] in which the radiative recombination rate (which scales with the electron-hole overlap like W_X) can be reduced by a couple of orders of magnitude. A still more flexible experimental configuration can be realized with AlSb/InAs/(AlSb/GaSb)/AlSb polytype heterostructures[18] taking full advantage of the type-II band alignment and the large spin splitting of the constituent materials. In the regime $W_h > W_e > W_X$, the exciton spin relaxation is dominated by the electron spin relaxation and a more sensitive dependence of the time resolved polarized luminescence spectra on the zero-field conduction subband splittings would occur.

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