Exciton-bound electron-spin relaxation

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Exciton luminescence polarization studies in semiconductor quantum wells have revealed the coexistence of two main mechanisms of exciton-spin relaxation: a well-known direct relaxation with simultaneous electron and hole spin flip due to the electron-hole exchange interaction and an indirect one with sequential spin flips of the single particles. The rate of exciton-spin relaxation in this indirect channel is limited by the slower single-particle spin-flip rate, which is typically the electron one. In this work a theory of exciton-bound electron-spin dynamics driven by the spin-orbit splitting in the conduction band is presented. It is shown that the off-diagonal matrix element between optical active and inactive exciton states that differ only with regard to the electron spin direction represents an effective magnetic field that changes randomly as the exciton is elastically scattered and relaxes its spin. The exchange splitting between the optical active and inactive states acts as a constant external magnetic field, reducing the relaxation. The estimated rate of the bound electron spin flip agrees well with values obtained from previous fittings of the experimental data. Semiconductor heterostructures with real-space indirect excitons, for which the sequential spin-flip relaxation channel becomes the dominant one, are also briefly discussed together with the dependence of the relaxation time on the well width. [S0163-1829(97)08840-1]

The study of the optical properties of semiconductor heterostructures, which are dominated by excitonic effects, has been intense in the last twenty years or so. The physics of the light-matter interaction has been elucidated in terms of exciton formation (absorption), relaxation, and recombination (emission). Many new results have been obtained thanks to recent advances in ultrafast laser spectroscopy. In particular, the measured femtosecond resolved time evolution of the luminescence polarization has revealed a complex exciton and single-particle spin dynamics.

Luminescence polarization was first used in the study of semiconductor heterostructures to determine the symmetry of the states and the complex structure of the valence subbands. More recently, the luminescence polarization decay after excitation with a pulse of circularly polarized light, from both intrinsic and doped samples, has been investigated and exciton and minority-carrier spin relaxation rates have been determined.[1] The angular momentum initially transferred from the pump light to the matter excitations changes during their energy and linear momentum relaxation. Different mechanisms or relaxation channels have been evoked to describe different experiments. The problem of the theory of the spin relaxation in semiconductor quantum wells is, however, complex and far from a complete solution. The experimentally observed dependence on the sample,[2] on the energy,[3] and on the intensity[3,2] of the exciting light, for example, have not been completely understood yet.

Progress has been made though in the understanding of the exciton-spin relaxation in good-quality quantum-well samples. The theory of exciton-spin relaxation in quantum wells given in Ref. 4 has been guiding the research on the problem since then. The spin dynamics of the heavy-hole exciton in quantum wells is described with a set of coupled rate equations for the population of the four exciton-spin states (which are $|\pm 1\rangle$ (optical active) and $|\pm 2\rangle$ (optical inactive)], including all possibilities of spin flip. A simple analysis of the experimental data reveals the coexistence of two relaxation channels for the exciton spin. First, there is a direct spin-relaxation mechanism between the optical active states with simultaneous electron and hole spin flip, with rate W_{ex}, driven mainly by the long-range part of the intraexciton electron-hole exchange interaction. Different experiments by different groups agree that this is the leading exciton-spin relaxation channel in intrinsic GaAs/$Ga_xAl_{1-x}As$ quantum wells. The other relaxation channel, in which the exciton-spin flip occurs via independent electron and hole spin flips, is less known. This second channel is indirect in the sense that transitions between the optical active states in this case involve an intermediate optical inactive state, as is schematically shown in Fig. 1.

While a microscopic theory of the direct exciton-spin re-

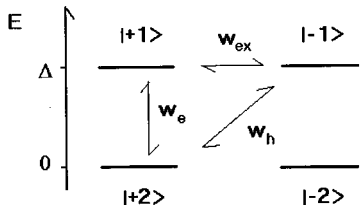

FIG. 1. Set of heavy-hole exciton-spin states with the possible spin-flip processes. W_{ex} stands for the direct exciton-spin-relaxation rate between the optical active states. The other transitions correspond to exciton-bound single-particle spin flips (with rates W_e and W_h for electrons and holes, respectively) that sequentially lead to the indirect exciton-spin relaxation (the similar transition via $|-2\rangle$ have not been drawn for clarity).

laxation via the exchange interaction in good agreement with the experiment was given in Ref. 4, no microscopic theory exists yet for the indirect exciton-spin-relaxation mechanism. Such an indirect channel, besides being always present, dominates the exciton-spin relaxation in the case of real-space indirect excitons, with drastically reduced electron-hole overlap. It is determined by the slower spin-flip rate between the exciton-bound electron (W_e) and hole (W_h). At low carrier concentration the single-particle spin flip is driven by the spin-orbit interaction and in most III-V compounds having a complex valence band is much faster for holes than for electrons. The spin-relaxation rate for the exciton-bound electrons is then expected to determine the indirect exciton-spin-relaxation rate. Following the work in Ref. 4, we then present here a theory of the exciton-bound electron-spin relaxation driven by the k-dependent spin-orbit splitting in the conduction band.

Differently from previous work on the electron-spin relaxation in quantum wells,[5,6] we here look at the spin relaxation of the electron bound to a heavy hole in a free-exciton state and not in a free-electron state. We add to the usual exciton Hamiltonian the following spin-dependent term that acts only on the electron's coordinates and represents the spin-orbit interaction in the conduction band:

$$H_{k^3}=\gamma(\sigma_x k_x(k_y^2-k_z^2)+\sigma_y k_y(k_z^2-k_x^2)+\sigma_z k_z(k_x^2-k_y^2)), \tag{1}$$

and study its effects on the exciton-spin dynamics. In the above expression γ is a band parameter, σ_x, σ_y, and σ_z are the Pauli matrices and, in the effective-mass or envelope-function approximation, the electron's wave vector becomes the operator $\mathbf{k}=-i[(d/dx_e),(d/dy_e),(d/dz_e)]$, which acts only on the electron's coordinate. This is the so called k^3 spin-orbit term in the conduction band of the III-V semiconductor compounds that was first obtained by Dresselhaus[7] and the x, y, and z correspond to the crystal cubic axis.[8]

Such spin-orbit term causes a very small spin splitting in the GaAs conduction band and its effects on the quantum-well exciton state will be treated here within first-order perturbation theory. The unperturbed $1s$-exciton state we start with is written as

$$|\mathbf{K},s\rangle=\phi_{1s}(\rho)\,\frac{e^{i\mathbf{K}\cdot\mathbf{R}}}{\sqrt{A}}\,f_e(z_e)f_h(z_h)|s\rangle. \tag{2}$$

The motion along the growth direction (z) is described by the subband envelope functions $f_e(z_e)$ and $f_h(z_h)$; and in the plane (xy) we have the free motion of the center of mass with wave vector $\mathbf{K}$ and the bound $1s$ state,

$$\phi_{1s}(\rho)=\sqrt{\frac{2}{\pi}}\,\frac{1}{a_{2\mathrm{D}}^*}\,e^{-\rho/a_{2\mathrm{D}}^*}. \tag{3}$$

The single-particle coordinates are written as $\mathbf{r}_i=(\boldsymbol{\rho}_i,z_i)$ with $i=e$ for the electron and h for the heavy hole; $\boldsymbol{\rho}=\boldsymbol{\rho}_e-\boldsymbol{\rho}_h$ and $\mathbf{R}=(m_e\boldsymbol{\rho}_e+m_h\boldsymbol{\rho}_h)/(m_e+m_h)$ are the relative and center of mass coordinates, respectively (m_i stands for the band-edge effective mass and $a_{2\mathrm{D}}^*$ for the effective two-dimensional Bohr radius). Finally, $|s\rangle$ represents the product of the zone-center electron and heavy-hole Bloch states with total spin (or angular momentum) component s along the growth direction; the four heavy-hole exciton spin states mentioned before are written as

$$|\pm 1\rangle=u_{\mp 1/2}(\mathbf{r}_e)v_{\pm 3/2}(\mathbf{r}_h), \tag{4}$$

$$|\pm 2\rangle=u_{\pm 1/2}(\mathbf{r}_e)v_{\pm 3/2}(\mathbf{r}_h). \tag{5}$$

The spin-orbit term in Eq. (1) mixes the optical active $|\pm 1\rangle$ and the inactive $|\pm 2\rangle$ states. Transitions between those states due to this term correspond to the exciton-bound electron spin flip. The interaction H_{k^3} mixes the above unperturbed exciton states and in the basis $\{|\mathbf{K},-2\rangle,|\mathbf{K},-1\rangle,|\mathbf{K},1\rangle,|\mathbf{K},2\rangle\}$ is represented by a block-diagonal matrix,[9]

$$H_{\mathrm{so}}=\begin{pmatrix} h_{\mathrm{so}} & 0 \\ 0 & h_{\mathrm{so}} \end{pmatrix}, \tag{6}$$

with two identical 2×2 blocks h_{so} corresponding to the two equivalent exciton (or heavy-hole) spin orientations. A straightforward calculation of the matrix elements gives

$$h_{\mathrm{so}}=\gamma\,\frac{m_e}{M}\left[K_x\left(\frac{m_e^2}{M^2}\,K_y^2+\frac{1}{2a_{2\mathrm{D}}^{*\,2}}-q_z^2\right)\sigma_x - K_y\left(\frac{m_e^2}{M^2}\,K_x^2+\frac{1}{2a_{2\mathrm{D}}^{*\,2}}-q_z^2\right)\sigma_y\right], \tag{7}$$

where $q_z^2=\langle f_e(z_e)|-(d^2/dz_e^2)|f_e(z_e)\rangle$ is the expectation value of the square of the electron's wave vector along the growth direction and $M=m_e+m_h$ is the exciton's total mass.

We are interested in the exciton-spin dynamics driven by such a spin-orbit term. The study of such spin dynamics starts from an effective spin Hamiltonian. The optical active exciton states have a small center-of-mass momentum and in the well-width range of two-dimensional confinement we typically have

$$K^2\ll\frac{1}{2a_{2\mathrm{D}}^{*\,2}},q_z^2,\left|\frac{1}{2a_{2\mathrm{D}}^{*\,2}}-q_z^2\right|.$$

In first approximation, one can then neglect the above cubic-in-K terms and add the exchange splitting Δ between the optical active and inactive exciton states[4,10] to obtain the following effective spin Hamiltonian (with an analogous expression for the $\{|-1\rangle,|-2\rangle\}$ block)

$$H_{\mathrm{spin}}=\begin{pmatrix} \Delta/2 & \alpha K_+ \\ \alpha K_- & -\Delta/2 \end{pmatrix}, \tag{8}$$

where $K_\pm=K_x\pm iK_y$ and

$$\alpha=\gamma\,\frac{m_e}{M}\left(\frac{1}{2a_{2\mathrm{D}}^{*\,2}}-q_z^2\right). \tag{9}$$

Similarly to the case of the $\mathbf{K}$-dependent long-range part of the exchange interaction between the optical active states,[4] we see here that the conduction-band spin-orbit interaction leads to an effective spin Hamiltonian in the $\{|1\rangle,|2\rangle\}$ basis corresponding to that of a pseudospin 1/2 in the presence of a $\mathbf{K}$-dependent effective magnetic field. This means that the exciton-bound electron-spin dynamics presents a motional narrowing type of relaxation analogous to the D'yakonov-

Perel free-electron spin relaxation[11] and to the direct exciton-spin relaxation. A random variation of the center-of-mass momentum **K**, e.g., due to elastic scattering by lattice imperfections, will correspond to a randomly varying effective magnetic field. This effective internal magnetic field will cause the relaxation of the pseudospin 1/2, which in this case corresponds to the exciton-bound electron spin. Before proceeding in the calculation of the corresponding relaxation rate, we would like to point out the twofold qualitative difference of the above effective spin Hamiltonian, with respect to that describing the exchange exciton-spin relaxation (i.e., mixing the $|+1\rangle$ and $|-1\rangle$ states[4]). We first note that the off-diagonal element (or the effective transverse magnetic field) is in this case linear (and not quadratic) in K, and second, that the exchange splitting Δ here works as an additional longitudinal component of the effective magnetic field, which is constant.

A general solution to the time evolution equation for the **K**-dependent density matrix of a pseudo spin 1/2 in the presence of an effective magnetic field $\mathbf{B}(\mathbf{K})$ considering elastic scattering of the center-of-mass momentum was presented in Ref. 4. The same approach, appropriate to explain resonant excitation experiments,[12] can be applied to the exciton-bound electron spin relaxation. For the spin Hamiltonian above, the spin-relaxation rate is finally obtained as

$$W_e=\frac{1}{t_e}=\frac{4\alpha^2K^2}{\hbar^2}\,\frac{t_p}{1+(\Delta t_p/\hbar)^2}, \tag{10}$$

where t_p is the usual momentum scattering time. The above expression for W_e can be reduced to the one appropriate for a free electron[13] of in-plane wave vector $k_{\rm el}$ (with $k_{\rm el}^2 \ll q_z^2$) by simply letting $(1/a_{\rm 2D}^*)\rightarrow 0$, $\Delta\rightarrow 0$ and $(m_e/M)K\rightarrow k_{\rm el}$.

Experimental investigations of the exciton-spin dynamics in high-quality GaAs/Al_xGa_{1-x}As multiple quantum wells ($x=0.3$ and $L=150$ Å) have determined through comprehensive fittings that the exciton-bound electron-spin relaxation rate lays in the range $3\times10^8<W_e<3\times10^9$ s^{-1},[12] and is much smaller than the direct exchange exciton-spin relaxation rate. In order to estimate W_e using the present theory we take the average center-of-mass motion energy of the optically active excitons as given by the homogeneous line width, i.e., we set $\hbar^2K^2/2M=\Gamma_h\simeq 0.22$ meV;[14] we set $q_z^2=\pi^2/L_{\rm eff}^2$, where $L_{\rm eff}$ is the effective well width[15] (in our case, $L_{\rm eff}=L+35$ Å$=185$ Å) and use $\gamma=17$ eV Å^3, $a_{\rm 2D}^*=100$ Å, $m_e=0.067$, $m_h=0.17$, $\Delta=0.05$ meV (Ref. 10) and $t_p=6$ ps, to obtain $W_e=8.2\times10^8$ s^{-1}. Even allowing for the uncertainties in the parameters and in the model used to fit the data, the agreement is very good. We therefore conclude that the exciton-bound electron-spin relaxation here considered is responsible for the observed indirect exciton-spin relaxation. In Fig. 2 we show how this indirect spin-relaxation time varies with the well width L and the momentum relaxation time t_p. In the well-width dependence we have included that of the two-dimensional Bohr radius, obtained from a simple variational calculation, that of q_z^2 and also that of the exchange energy splitting Δ.[10] The spin relaxation time t_e increases with well width due to the corresponding decrease in the average spin-orbit splitting in the conduction subband that the bound electron feels. Except in the narrow well limit, we observe the usual motional narrowing behavior with the exciton-bound spin-relaxation time inversally proportional to the momentum scattering time. As the well width decreases Δ increases almost exponentially and the opposite behavior is observed, i.e., larger relaxation times with larger scattering times.

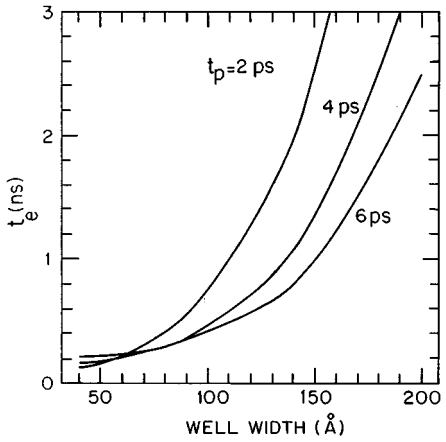

FIG. 2. Well-width variation of the exciton-bound electron-spin relaxation time t_e for different values of the elastic momentum scattering time t_p, as given by expression (10).

In the case of a type-I quantum well,[12] however, the sensitivity of the exciton luminescence polarization decay to W_e is low because of the much larger value of $W_{\rm ex}$ ($W_{\rm ex}\sim 1.5\times10^{10}$ s^{-1}). Such a situation can be drastically changed diminishing the direct exchange spin-flip rate by reducing the electron-hole overlap; in fact, $W_{\rm ex}$ is proportional to the fourth power of the overlap.[4] A suitable experimental configuration can be realized, for instance, employing AlSb/GaSb/AlSb/InAs/AlSb polytype heterostructures[16] having space indirect excitons as sketched in Fig. 3. Without reducing too much the exciton oscillator strength, a reduction of $W_{\rm ex}$ by a factor of 100 can be easily achieved. Furthermore, in InAs the value of the spin-orbit coupling parameter γ is expected to be larger than in GaAs: $\gamma_{\rm InAs}\sim 130$ eV Å^3.[17] In this case, the regime $W_h>W_e>W_{\rm ex}$ is realized and the exciton-spin relaxation is dominated by W_e. Therefore, a more sensitive dependence of the time-resolved polarized luminescence spectra on the zero-field conduction subband splittings due to H_{k^3} would occur.

We have presented above a microscopic theory of the spin-orbit-driven exciton-bound electron-spin relaxation. The relaxation rate W_e was calculated assuming random elastic

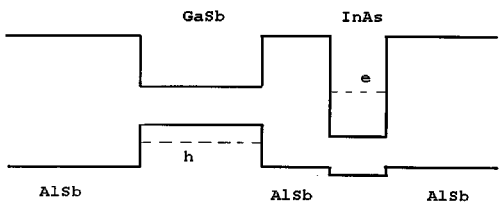

FIG. 3. Sketch of the band-edge modulation in a suggested polytype heterostructure with space indirect exciton.

scattering of the exciton center of mass. Comparison with the available experimental data shows that it explains the observed indirect exciton-spin relaxation. More measurements, however, are necessary in order to test further the theory, looking, for example, at the well-width dependence of W_e. We have also considered the case of real-space indirect excitons for which the indirect spin-relaxation rate dominates over the direct one due to the exchange interaction. In such a situation, the exciton luminescence polarization decay is determined mainly by the spin-orbit exciton-bound electron-spin-relaxation mechanism here studied.

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