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Hole Relaxation and Luminescence Polarization in Doped and Undoped Quantum Wells

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Valence-hole relaxation by acoustic-phonon emission yields spin relaxation through the spin mixing of the hole-band states. In quantum wells, the hole-spin relaxation is incomplete. This leads to a unified theory of the polarization spectra for undoped and doped quantum wells, which explains the diverse features in the spectra.

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Polarization measurements[1,2] of luminescence processes between the conduction and valence bands in semiconductor quantum wells provide information on the electronic states beyond the energy spacings,[2,3] and their interpretation requires inclusion of the many-body effects.[2,4] We report a study of the hole relaxation processes after photoexcitation. The results contradict the common assumption of complete spin relaxation of the photoexcited holes, which is at the base of the previous theories of luminescence in quantum wells.[3-5] Hole-spin populations are calculated as the consequence of acoustic-phonon scattering of spin-orbit-mixed photoexcited hole states. In a symmetric well, the hole relaxation processes are naturally classified in terms of the change or preservation of the parity of the hole state. A theory of hole-spin populations before recombination follows. Calculated polarization spectra for the undoped, *n*-, and *p*-doped wells are in good agreement with experiment, whose major features can also be qualitatively explained in terms of the theory. This work highlights the importance of carrier relaxation in luminescence in quantum wells and points to the possibility of determining the electron- and hole-spin relaxation rates through more detailed polarization measurement and theoretical analysis.

Consider, for example, the polarization, defined as the fractional difference of the luminescence intensities of two circular polarizations σ^+ and σ^- at a fixed energy, as a function of the excitation energy with polarization σ^+. The assumption of complete hole-spin relaxation leads to a qualitative explanation[3] of the polarization spectrum in the undoped quantum wells in terms of the valence-band structure. The same assumption in an *n*-doped quantum well, in which a Fermi sea of electrons of both spins is present, leads to a featureless polarization spectrum, in contradiction to experiment. The spin polarization of the electron Fermi sea by the photoexcited valence hole through an exchange interaction was invoked to explain the polarization reversal in the *n*-doped wells.[4] The qualitative explanation is limited to only one feature of the polarization spectrum. Yet to be explained is the strength of the exchange interaction required.

We examine the hole relaxation by emission of acoustic phonons in both undoped and doped wells and by shakeup[2] of the electron Fermi sea in the *n*-doped wells. While these processes provide *per se* only momentum and energy relaxation, the different mixings of spin components in different hole states through the $k\cdot p$ terms lead to hole-spin relaxation. Since the intrinsic hole-spin relaxation is much slower than the momentum relaxation, the hole-spin relaxation is incomplete prior to radiative recombination. Quantum-well confinement appears to play the same role as the uniaxial stress in bulk GaAs in reducing hole-spin relaxation.[6]

The mixing of the spin-$\frac{3}{2}$ states of the valence bands following from the band structure is important to the determination of the polarization spectrum. The subbands of a quantum well are determined in the effective-mass approximation around Γ, the zone center. The conduction-subband state is given by an amplitude modulation of the spin-$\frac{1}{2}$ Bloch state of the conduction valley. The amplitude modulation is composed of a plane wave along the interface plane and the function $\phi_{ns}(z)$ along the normal or z direction. The subscript n $(=1,2,\ldots)$ denotes the band index and the wave function is independent of the spin index s $(=\pm\frac{1}{2})$. For a symmetric well, the parity of a conduction state under the mirror operation $z\rightarrow -z$ is the same as $n-1$. The valence-subband state is represented by four components of the spin-$\frac{3}{2}$ states. The interface-plane dependence is again like a plane wave. The z dependence of the four components is given by $\zeta_{\nu p_m m}(z)$, where ν includes both the hole-band index and the in-plane wave vector, m $(=\frac{3}{2},\frac{1}{2},-\frac{1}{2},-\frac{3}{2})$ denotes the z component of the $\frac{3}{2}$ spin, and p_m $(=\pm 1)$ denotes the parity of the mth-component wave function of the hole state with parity p:

$$p_m = p(-1)^{3/2-m}. \tag{1}$$

At zero wave vector, the valence state is a pure spin state. At a finite in-plane wave vector, the valence state is a linear combination of zero-wave-vector states by the mini $k\cdot p$ method.[7] The conduction- and valence-subband energies and wave functions of an *n*- or *p*-doped quantum well are calculated from a self-consistent potential within the axial approximation.[7] The optical transitions obey the momentum selection rule $s-m=\sigma$, where $\sigma=\pm 1$ for the $\sigma^\pm$ polarizations.

The central thesis of our work is that the incomplete spin relaxation of the photoexcited hole is key to the determination of the polarization of the emitted photon on recombination. In the polarization determination, we neglect the exciton effect[3] since the exciton state reflects the nature of band-edge states with which it is associated. We assume that the conduction electron promoted from the valence band by the excitation laser relaxes to the lowest available conduction-subband state in a time much shorter than the electron-spin relaxation time τ_s or the electron-hole recombination time τ_r.[5] In the n-doped case, the Fermi sea provides equal population of both electron spins for recombination so that the spin state of the photoexcited electron is immaterial. Since the luminescence excitation spectrum is made in Ref. 1 as a measurement of the lowest-energy recombination as a function of the excitation energy, the relative populations of the spin states ($\pm\frac{3}{2}$) of the highest valence-subband states to which the photoexcited hole relaxes are germane. The common assumption of equal hole-spin populations prior to recombination coupled with equal spin populations of electrons in the n-doped well would lead to zero polarization of luminescence as stated above. Since the hole-subband energies are smaller than the optical phonon energy ($\sim$36 meV), relaxation of the hole by emission of the optical phonon is a rare event. Relaxation of the hole through emission of acoustic phonons thus assumes prime importance.

The mirror symmetry of the quantum well is used to classify the hole-phonon scatterings into those which preserve the parity of the hole state and those which change it. An estimate, based on our theory to be detailed elsewhere, of the hole–acoustic-phonon scatterings yields a parity-conserving hole momentum relaxation time τ_+ much shorter than the parity-changing time τ_-. The hole–acoustic-phonon scattering through the deformation potential is given by the strain Hamiltonian[8] of the same symmetry form as the Luttinger Hamiltonian. When the acoustic phonons are classified into even and odd parity, for symmetry reasons the longitudinal-mode and the in-plane transverse-mode phonons favor parity-conserving scatterings and the remaining transverse mode favors parity-flipping scatterings. For the hole–acoustic-phonon scattering through the piezoelectric coupling, all three modes favor the parity-conserving processes. The spin relaxation then comes about because the initial and final hole states are made up of different mixtures of the spin components which are determined by the calculated subband structure. At the top of the valence subbands, the hole-spin relaxation time is taken to be roughly the same as the electron one, τ_s, much longer than $\tau_\pm$.

For the n-doped well, the hole can also relax by shaking up the electron Fermi sea. While the shakeup process is important for the emission with z polarization in the waveguide configuration,[2] it is less than 10% of the simple recombination with $\sigma^\pm$ polarizations. Thus, the Auger process does not supplant the acoustic-phonon scattering.

The numbers of electrons and holes of each spin species produced by the excitation light at a given energy are calculated by the simple optical transition. For the incident light with σ^+ polarization, the generation rates of electrons with spin $s=\pm\frac{1}{2}$ are given by

$$G^e_{-1/2}=3M\sum_\nu g^1_\nu\,|\langle\phi_{1,-1/2}|\zeta_{\nu,+1,-3/2}\rangle|^2 +3M\sum_\nu g^2_\nu\,|\langle\phi_{2,-1/2}|\zeta_{\nu,-1,-3/2}\rangle|^2, \tag{2}$$

$$G^e_{1/2}=M\sum_\nu g^1_\nu\,|\langle\phi_{1,1/2}|\zeta_{\nu,+1,-1/2}\rangle|^2 +M\sum_\nu g^2_\nu\,|\langle\phi_{2,1/2}|\zeta_{\nu,-1,-1/2}\rangle|^2, \tag{3}$$

where g^n_ν is the joint density of states between the nth electron subband and the νth hole subband at the energy of the incident light. Here, we limit the calculation to include only the two lowest conduction subbands. Equation (2) corresponds to the $m=-\frac{3}{2}$ to $s=-\frac{1}{2}$ transitions and Eq. (3) to the $m=-\frac{1}{2}$ to $s=\frac{1}{2}$ transitions. The square of the transition-matrix element for the σ^+ polarization between the bulk Bloch states for the former is given by $3M$ and that for the latter is given by M. Note the parity conservation of the z part of the envelope functions in the optical transitions.

The generation rates of holes of parity $p=\pm1$ are given by

$$G^h_{-1}=3M\sum_\nu g^1_\nu\,|\langle\phi_{1,-1/2}|\zeta_{\nu,+1,-3/2}\rangle|^2 +M\sum_\nu g^2_\nu\,|\langle\phi_{2,1/2}|\zeta_{\nu,-1,-1/2}\rangle|^2, \tag{4}$$

$$G^h_1=M\sum_\nu g^1_\nu\,|\langle\phi_{1,1/2}|\zeta_{\nu,+1,-1/2}\rangle|^2 +3M\sum_\nu g^2_\nu\,|\langle\phi_{2,-1/2}|\zeta_{\nu,-1,-3/2}\rangle|^2. \tag{5}$$

In Eq. (4), the first term on the right comes from the same-parity heavy-hole states which, by Eq. (1), also have even-parity $m=-\frac{3}{2}$ components resulting from the $m=-\frac{3}{2}$ to $s=-\frac{1}{2}$ transitions, and the second term comes from the odd-parity light-hole states which, by Eq. (1), have odd-parity $m=-\frac{1}{2}$ components resulting from the $m=-\frac{1}{2}$ to $s=\frac{1}{2}$ transitions. The parity roles are reversed in Eq. (5).

With the carrier generation rates and relaxation times defined above, we set up the rate equations[9] and solve for the steady-state spin populations:

$$n^e_s=\frac{\tau_r}{2}\left[G^e_s+G^e_{-s}+\frac{\tau_s}{\tau_r+\tau_s}(G^e_s-G^e_{-s})\right], \tag{6}$$

$$n^h_p=\frac{\tau_r}{2}\left[G^h_p+G^h_{-p}+\frac{\tau_s}{\tau_r+\tau_s}\,\frac{\tau_--\tau_+}{\tau_-+\tau_+}(G^h_p-G^h_{-p})\right]. \tag{7}$$

In a p-doped quantum well, the presence of equal spin populations of holes means that the luminescence polarization is determined by the electron-spin polarization:[9]

$$\mathcal{P}_p = \frac{G^e_{-1/2} - G^e_{1/2}}{G^e_{-1/2} + G^e_{1/2}} \frac{\tau_s}{\tau_s + \tau_r} . \tag{8}$$

Similarly, in the n-doped well, the luminescence polarization is given by

$$\mathcal{P}_n = \frac{G^h_{-1} - G^h_{1}}{G^h_{-1} + G^h_{1}} \frac{\tau_- - \tau_+}{\tau_- + \tau_+} \frac{\tau_s}{\tau_s + \tau_r} . \tag{9}$$

In the undoped well, the emission intensity depends on the lesser of the numbers of electrons and of holes available for recombination satisfying the momentum selection rule. Thus, the polarization is given by

$$\mathcal{P}_u = \frac{\min(n^e_{-1/2}, n^h_{-1}) - \min(n^e_{1/2}, n^h_{1})}{\min(n^e_{-1/2}, n^h_{-1}) + \min(n^e_{1/2}, n^h_{1})} . \tag{10}$$

Figure 1 shows in order the calculated polarization spectra for a p-doped, an n-doped, and an undoped quantum well in comparison with experiment. The two ratios τ_r/τ_s and τ_+/τ_- are taken to be 0.12 and 0.46 to fit the magnitude of observed polarization[1] in the p-doped and n-doped quantum wells, respectively. The same values are then used for the undoped-well spectrum. The hole subbands are labeled by[1] $1h, 1l, 2h, 2l, \dots$, where the heavy-hole state h refers to $m = \pm\frac{3}{2}$ and the light-hole state l refers to $m = \pm\frac{1}{2}$. The index nC refers to the conduction subband. The marked features are related to the valence-to-conduction-subband transitions. All these features arising out of incomplete hole-spin relaxation are in qualitative agreement with observation[1] and the energies at which they occur are in quantitative agreement, for undoped and doped quantum wells. The ratio of τ_+/τ_- obtained here is larger than expected from theory. However, the marked features are insensitive to the ratio as long as it is less then 0.5.

The physical origins of these features will be described in detail in a long paper. The most striking feature[4] is the different polarization behavior for different doping at the transition range of $(1l, 2h \rightarrow 2C)$. The proximity of the $1l$ and $2h$ hole subbands causes strong $k \cdot p$ mixing and strong $\zeta_{v,-1,-3/2}(z)$ components in both bands. The odd-parity component permits the optical excitation from $m = -\frac{3}{2}$ to $s = -\frac{1}{2}$ of the odd-parity conduction band $2C$. By Eq. (1), the odd-parity $m = -\frac{3}{2}$ component means that the hole states in both the $1l$ and $2h$ band must have even parity. By the parity-conserving processes, they relax to the top heavy-hole $(1h)$ state with $m = \frac{3}{2}$. In the n-doped well, such hole states lead to a strong σ^- luminescence and, hence, polarization reversal. In the undoped well, the same process generates $s = -\frac{1}{2}$ electrons and the paucity of the $s = \frac{1}{2}$ electrons limits the σ^- luminescence. The similarity of the p-doped behavior of this feature to the undoped one is also evidence for the role of holes in the n-doped well.

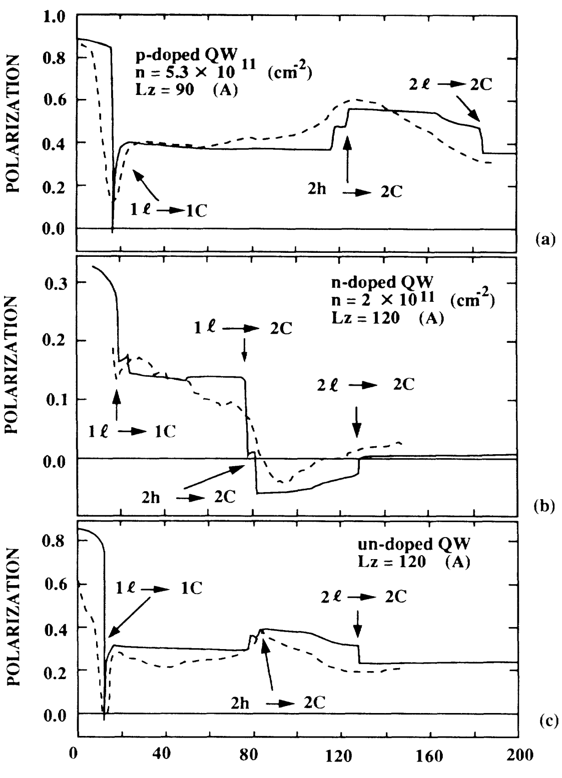

FIG. 1. Comparison of calculated polarization spectra (solid lines) with experiment (dashed lines) for (a) a p-doped quantum well, (b) an n-doped well, and (c) an undoped well. The excitation energy is measured relative to the $1h \rightarrow 1C$ transition threshold.

The polarization reversal should be insensitive to a weak in-plane magnetic field[1] (Hanle effect) since it is caused by hole momentum relaxation and not by electron polarization. A high magnetic-field was recently observed to lengthen the spin relaxation time in an undoped well[10] and the measured luminescence polarization of the Landau levels agree well with our calculation though the interpretation of the relaxation times differ. Details will be given in a long paper.

The ability of our theory to explain key features in the polarization spectra of all types of quantum wells gives support to our conclusion of incomplete electron-spin and hole-spin relaxation. Because our theory contains some fast hole-spin relaxation, such relaxation-time behavior is not inconsistent with the measured relaxation times and the observed lack of optical alignment effects.[5] The next step is to study the quantitative effects of finite relaxation times on the polarization, to calculate these times, and to compare the theory with future experiments in time-resolved luminescence polarization spectra.

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