Carrier relaxation and luminescence polarization in quantum wells

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A theory of polarization of luminescence in doped and undoped quantum wells is constructed taking into account the relaxation processes of photoexcited electrons and holes. The hole momentum relaxation by acoustic-phonon interaction yields spin relaxation through the spin mixing of the hole band states, resulting in incomplete spin depolarization in quantum wells. Our theory of hole-phonon interaction in symmetric quantum wells leads to the concept of relaxation through two channels conserving or changing the parity of the hole state. Calculation of the polarization spectra for undoped, *n*-type-doped, and *p*-type-doped GaAs/Al_{1-x}Ga_xAs quantum wells produces an explanation of all the observed features in the spectra.

I. INTRODUCTION

The photoluminescence process may be regarded as composed of three steps. (1) Excitation: A photon is absorbed producing an electron-hole pair or an exciton. (2) Relaxation: The carriers relax, given sufficient time, to quasiequilibrium states. (3) Recombination: The electron-hole pair or the exciton recombines emitting luminescent light. In a quantum well, limiting the incident light normal to the interface plane to a pure circular polarization, say σ^+ , and measuring the emitted light close to the normal separately in the two circular polarizations yields much useful information on the state of the electron-hole pair.¹ The selection rules governing the transition between the conduction subbands and the valence subbands may be regarded as conservation of the z component of the angular momentum, where the normal is chosen to be along the z axis, if the cylindrical $approximation^2$ is used. Since the conduction subbands are simple spin- $\frac{1}{2}$ parabolic bands, the nature of the valence subband state, in addition to its energy, may be deduced from the polarization spectra with the help of the selection rule.

The polarization \mathcal{P} is 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^+ . How the excited electron and hole relax their spins is clearly an important determining factor in polarization. Since a valence state is a mixture of the z-components of the $\frac{3}{2}$ spin, momentum relaxation of the valence hole will also bring about spin relaxation. The natural and common assumption of the complete relaxation of the spin of the photoexcited holes³⁻⁵ leads to a qualitative explanation³ of the polarization spectrum in the undoped quantum wells in terms of the valence-band structure. However, the same assumption in an *n*-type-doped quantum well, in which a Fermi sea of electrons of both spins is present, leads to a featureless polarization spectrum, in contradiction with experiment. In addition, the sign of polarization at the second heavy-hole subband to second conduction subband transitions is reversed in going from an undoped to *n*-type-doped quantum well.⁴ 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 doped wells.⁴ This theory has not been used to produce a quantitative calculation of the polarization spectra and it requires a many-body enhancement of the strength of the conduction-valence exchange.

We have undertaken a theoretical study of the hole relaxation processes, including scatterings by acoustic phonons and shake-up of Fermi seas,⁶ which determine the concomitant spin relaxation. From this study, we are able to set up the rate equations to obtain the spin polarizations of electrons and holes before recombination and, hence, to calculate the polarization spectra for undoped and n- and p-type-doped quantum wells.

Since the spin components of a valence state are mixed, what replaces the spin component label as the quantum number to characterize the state? In zero magnetic field, there are two degenerate hole states.² In a symmetric quantum well, they can be characterized by parity. The parity of the hole state is defined in Sec. II, where the band mixing between light and heavy holes is treated using the Luttinger Hamiltonian.⁷ The subbands in the doped wells are calculated in the self-consistent Hartree approximation.

In Sec. III, we examine the hole relaxation by emission of acoustic phonons in both undoped and doped wells and by shakeup⁶ of the Fermi sea in the doped wells. The change in parity by the relaxation processes is studied by characterizing the parities of the acoustic phonons and the parity changes in the interband transitions during a shakeup. While these processes provide *per se* only momentum and energy relaxation, the different mixings of spin components in different hole states through the $\mathbf{k} \cdot \mathbf{p}$ terms lead to hole spin relaxation. Because of the restrictions of the parity relaxation and the discreteness of the valence subband states in quantum wells, the hole

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spin relaxation is incomplete prior to radiative recombination. This is in agreement with the observation in bulk GaAs that uniaxial stress reduces hole spin relaxation.⁸ In Sec. IV, based on the incomplete hole spin relaxation and the valence subband structure, we construct a unified theory of the polarization spectra for all types of quantum wells, *n*-type-doped, *p*-type-doped, and undoped. In Sec. V, we compare calculated polarization spectra for GaAs/Al_{1-x}Ga_xAs quantum wells with the observations in Ref. 1. The major features in the spectra for all three types of quantum wells are in good agreement with experiment and their locations in energy can be explained in terms of transitions between valence and conduction subbands. We conclude by summarizing what has been accomplished in this work and what needs further work.

II. SUBBAND STRUCTURE

The subbands of the conduction and valence electrons in a quantum well are calculated by the single-band and multiband⁷ effective-mass approximation, respectively. The effect on the carriers due to doping is included in the self-consistent Hartree potential. For the III-V semiconductors, the wave functions are expanded about the Bloch states at Γ , with $\alpha = c$ for the conduction band and $\alpha = v$ for the valence band:

$$\Psi^{\alpha}(r) = \sum_{i=1}^{l_{\alpha}} F_i^{\alpha}(r) u_i^{\alpha}(r) , \qquad (1)$$

where l_{α} is the number of the components of the basis set. The envelope function, $F_i^{\alpha}(r)$, satisfies the effective-mass equation

$$\sum_{j=1}^{l_{\alpha}} [H_{i,j}^{\alpha}(-i\nabla) + V^{\alpha}(r)\delta_{i,j}]F_j^{\alpha}(r) = E^{\alpha}F_i^{\alpha}(r) , \qquad (2)$$

where $H(\mathbf{k})$ is the effective-mass Hamiltonian. The growth axis is taken as the z direction and the in-plane plane-wave motion may be factored out of the envelope function:

$$F_i^{\alpha}(r) = e^{ik_x x + ik_y y} \zeta_i^{\alpha}(z) .$$
(3)

For the conduction band, we consider only the Γ_6 valley, so that the energy is simply parabolic and the envelope function F_s^c has two spin components $(s = \pm \frac{1}{2})$. For the valence band, to include the effects of heavyand light-hole mixing, the wave function is expressed as the basis functions of the Γ_8 valence maximum with the four-component envelope function, $\zeta_m^v(z)$, where the index $m = \frac{3}{2}, \frac{1}{2}, -\frac{1}{2}, -\frac{3}{2}$, taking on the z-components of the spin- $\frac{3}{2}$ angular momentum. The effective Hamiltonian is taken to be the Luttinger Hamiltonian.⁷ With no magnetic field, the state at each subband ν and inplane wave vector **k** is twofold degenerate.² For a symmetric quantum well, it is convenient to classify the two states by a parity number $p = \pm 1$ under the mirror reflection⁹ $z \rightarrow -z$. The Luttinger Hamiltonian, which is invariant under the mirror reflection, has its component $H_{m,m'}$ changing under reflection by a factor $(-1)^{m-m'}$. It follows that the hole state with parity p has the four-component envelope function given by

$$\Phi_{p}(\nu \mathbf{k}, z) = \begin{pmatrix} \zeta_{3/2, p}(\nu \mathbf{k}, z) \\ \zeta_{1/2, -p}(\nu \mathbf{k}, z) \\ \zeta_{-1/2, p}(\nu \mathbf{k}, z) \\ \zeta_{-3/2, -p}(\nu \mathbf{k}, z) \end{pmatrix} .$$
(4)

In other words, the *m*th component of the state with parity p has parity p_m given by

$$p_m = p(-1)^{3/2 - m}.$$
(5)

We follow the mini- $\mathbf{k} \cdot \mathbf{p}$ method² to determine the envelope functions in terms of the states at $\mathbf{k} = \mathbf{0}$ which are single-component, pure *m* states. A component with parity p_m is made up of the $\mathbf{k} = \mathbf{0}$ states with the same parity. The subband index ν for the $\mathbf{k} = \mathbf{0}$ states is composed of (n, α) , where $\alpha = h, \ell$ for heavy and light valence subband states and $n = 1, 2, 3, \ldots$ in order of descending valence subband energies. The parity of the *n*th subband is $(-1)^{n-1}$. Thus, the even-parity component envelope function is the sum of odd-numbered $\mathbf{k} = \mathbf{0}$ wave functions and vice versa. Figure 1 shows an example of the valence subbands of an *n*-type-doped quantum well as a function of the in-plane wave vector calculated in the cylindrical approximation.² The warping is unimportant in the computation of the optical spectra which



FIG. 1. Energy dispersion of the conduction and valence subbands for an n-type-doped quantum well.

TABLE I. Band parameters for a $GaAs/Al_{0.3}Ga_{0.7}As$ quantum well.

GaAs/Al _{0.3} Ga _{0.7} As		
me	0.067	
γ_1	6.85	
γ_2	2.1	
7 3	2.9	
ΔE_{c} (eV)	0.202	
ΔE_{v} (eV)	0.152	
τ_r/τ_s	0.12	
τ_+/τ	0.46	
undoped QW		
L_z (Å)	120	
n-type-doped QW		
$n \ (10^{11} \ \mathrm{cm}^{-2})$	2	
L_z (Å)	120	
p-type-doped QW		
$n \ (10^{11} \ \mathrm{cm}^{-2})$	5.3	
L_z (Å)	90	

sums over the k states in all directions in the plane. The parameters in this calculation are listed in Table 1. For ease of computation, the parameters for the $Al_{1-x}Ga_xAs$ barrier are taken to be the same as GaAs except for the band offsets.

An important example of the role of parity in a valence state is at the top of the valence subband $(1h, \mathbf{k} = 0)$, to which a hole relaxes. There are two degenerate states there. One has the only nonzero component at $m = \frac{3}{2}$, an even function in z. By Eq. (5), the hole state has even parity. The other has the only nonzero component at $m = -\frac{3}{2}$ with the same even function in z. This second state, by Eq. (5), has odd parity. We shall present a theory tracing hole relaxation through its parity. This example then relates the hole parity populations to the hole spin components in the relaxed states.

III. RELAXATION PROCESSES FOR VALENCE HOLES

Here three scattering processes of holes are considered, by acoustic phonons, by shakeup and by optical phonons. The hole relaxation is followed through not only its energy and in-plane wave vector but also its parity. Then the spin population of the holes before recombination can be determined. In determining the change of parity of the hole state in a scattering event, the phonon or any other scatterer is also classified into even- and odd-parity states under the mirror reflection. The interaction Hamiltonian can then be written as

$$H_{\rm int} = H_{\rm int}^{(+)} + H_{\rm int}^{(-)}.$$
 (6)

The even part, under reflection with $z \rightarrow -z$, has the form of parity

$$H_{\rm int}^{(+)} = \begin{pmatrix} + - + - \\ - + - + \\ + - + - \\ - + - + \end{pmatrix} .$$
(7)

Thus, the scattering matrix element is of the form

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$$\langle \Phi_{p'}(\nu'\mathbf{k}') | H_{\text{int}}^{(+)} | \Phi_p(\nu\mathbf{k}) \rangle \propto \delta_{p,p'}, \qquad (8)$$

showing that on scattering the hole preserves its parity. The odd part of the interaction Hamiltonian has the form

$$H_{\rm int}^{(-)} = \begin{pmatrix} -+ -+ \\ +- +- \\ -+ -+ \\ +- +- \end{pmatrix} , \qquad (9)$$

yielding the scattering matrix element of the form

$$\langle \Phi_{p'}(\nu'\mathbf{k}') | H_{\text{int}}^{(-)} | \Phi_p(\nu\mathbf{k}) \rangle \propto \delta_{p,-p'}, \qquad (10)$$

changing the parity of the hole state.

Since the excited energy of the photocreated hole considered here is generally small or comparable to the optical phonon energy ($\sim 36 \text{ meV}$), scattering by optical phonon is not an important source of hole relaxation. In the doped well, the hole relaxation by shakeup of the Fermi sea is much weaker than the direct recombination.⁶ Thus, the most important process is hole scattering by an acoustic phonon. We consider both the scattering through the deformation potential and through the piezoelectric coupling.

A. Deformation-potential scattering

The hole-acoustic phonon scattering via the deformation potential is given by the strain Hamiltonian¹⁰ of the same form as the Luttinger Hamiltonian. It is given by

$$H(\varepsilon) = \begin{pmatrix} P+Q & -S & R & 0\\ -S^* & P-Q & 0 & R\\ R^* & 0 & P-Q & S\\ 0 & R^* & S^* & P+Q \end{pmatrix}, \quad (11)$$

where

$$P = D_{d} \operatorname{Tr} \varepsilon ,$$

$$Q = -\frac{1}{3} D_{u} (\varepsilon_{xx} + \varepsilon_{yy} - 2\varepsilon_{zz}) ,$$

$$R = \frac{1}{\sqrt{3}} D_{u} (\varepsilon_{xx} - \varepsilon_{yy}) - \frac{2i}{\sqrt{3}} D'_{u} \varepsilon_{xy} ,$$

$$S = -\frac{2}{\sqrt{3}} D'_{u} (\varepsilon_{zx} - i\varepsilon_{zy}) ,$$
(12)

and D_d , D_u , and D'_u are the deformation potentials and ε_{ij} is the strain tensor. For each mode of the acoustic branch of the phonon denoted by the superscript λ , the strain tensor is given in terms of the lattice displacement:

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$$\varepsilon_{ij}^{\lambda} = \frac{1}{2} \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) , \qquad (13)$$

$$\mathbf{u}^{\lambda}(r) = \sum_{q} U(\mathbf{q}, \lambda) \hat{\mathbf{e}}^{\lambda} e^{i\mathbf{q}\cdot\mathbf{r}} (a_{q_{\parallel}q_{\star}\lambda} + a^{\dagger}_{-q_{\parallel}-q_{\star}\lambda}) , \qquad (14)$$

$$U(\mathbf{q},\lambda) = \left(\frac{N\mathbf{q}+1}{2\rho\omega^{\lambda}(\mathbf{q})}\right)^{1/2} , \qquad (15)$$

where \mathbf{q}_{\parallel} and q_z are in-plane and z components of the wave vector \mathbf{q} , a and a^{\dagger} are the annihilation and creation phonon operators, $\omega^{\lambda}(\mathbf{q})$ is the phonon frequency, and ρ is the density of the material. $N_{\mathbf{q}}$ is the phonon distribution, assumed to be zero at low temperatures. $\hat{\mathbf{e}}^{\lambda}$ is the λ mode polarization vector, which, in the isotropic continuum model, may be written for the longitudinal and two transverse modes as, respectively,

$$\hat{\mathbf{e}}^{LA} = i(q_x, q_y, q_z)/|q| = i\mathbf{q}/|q| ,$$
$$\hat{\mathbf{e}}^{TA_1} = i(q_y, -q_x, 0)/|q_{\parallel}| , \qquad (16)$$

$$\hat{\mathbf{e}}^{TA_2} = (-q_x q_z, -q_y q_z, q_x^2 + q_y^2)/|q||q_{\parallel}|,$$

where $|q| = (q_x^2 + q_y^2 + q_z^2)^{1/2}$ and $|q_{\parallel}| = (q_x^2 + q_y^2)^{1/2}$. In a symmetric quantum well, the phonon can be class

sified as an even- or odd-parity phonon in terms of bulk phonons with opposite z components of the wave vectors:

$$b_{q_{\parallel}q_z\lambda+} = a_{q_{\parallel}q_z\lambda} + a_{q_{\parallel}-q_z\lambda} , \qquad (17)$$

$$b_{q_{\parallel}q_{z}\lambda-} = i(a_{q_{\parallel}q_{z}\lambda} - a_{q_{\parallel}-q_{z}\lambda}) , \qquad (18)$$

where b_{\pm} are the even- and odd-parity phonon annihilation operators. The edge modes can be similarly classified. In terms of the even and odd phonons, the lattice displacements for the three acoustic modes are given by

$$\mathbf{u}^{\mathrm{LA}}(\mathbf{r}) = \sum_{q_z \ge 0, q_{\parallel}} \frac{U(\mathbf{q}, \mathrm{LA})}{|q|} e^{i\mathbf{q}_{\parallel} \cdot \mathbf{r}_{\parallel}} i \left[\begin{pmatrix} q_x \cos(q_z z) \\ q_y \cos(q_z z) \\ iq_z \sin(q_z z) \end{pmatrix} (b_{q_{\parallel}q_z \mathrm{LA}+} + b^{\dagger}_{-q_{\parallel}q_z \mathrm{LA}+}) + \begin{pmatrix} q_x \sin(q_z z) \\ q_y \sin(q_z z) \\ -iq_z \cos(q_z z) \end{pmatrix} (b_{q_{\parallel}q_z \mathrm{LA}-} + b^{\dagger}_{-q_{\parallel}q_z \mathrm{LA}-}) \right],$$
(19)

$$\mathbf{u}^{\mathrm{TA}_{1}}(\mathbf{r}) = \sum_{q_{z} \ge 0, q_{\parallel}} \frac{U(\mathbf{q}, \mathrm{TA}_{1})}{|q_{\parallel}|} e^{i\mathbf{q}_{\parallel} \cdot \mathbf{r}_{\parallel}} i \left[\begin{pmatrix} q_{y} \cos(q_{z}z) \\ -q_{x} \cos(q_{z}z) \\ 0 \end{pmatrix} (b_{q_{\parallel}q_{z} \mathrm{TA}_{1}+} + b^{\dagger}_{-q_{\parallel}q_{z} \mathrm{TA}_{1}+}) + \begin{pmatrix} q_{y} \sin(q_{z}z) \\ -q_{x} \sin(q_{z}z) \\ 0 \end{pmatrix} (b_{q_{\parallel}q_{z} \mathrm{TA}_{1}-} + b^{\dagger}_{-q_{\parallel}q_{z} \mathrm{TA}_{1}-}) \right],$$
(20)

$$\mathbf{u}^{\mathrm{TA}_{2}}(r) = \sum_{q_{z} \geq 0, q_{\parallel}} \frac{U(\mathbf{q}, \mathrm{TA}_{2})}{|q||q_{\parallel}|} e^{i\mathbf{q}_{\parallel}\cdot\mathbf{r}_{\parallel}} i \left[\begin{pmatrix} -q_{x}q_{z}\sin(q_{z}z) \\ -q_{y}q_{z}\sin(q_{z}z) \\ -iq_{\parallel}^{2}\cos(q_{z}z) \end{pmatrix} (b_{q_{\parallel}q_{z}\mathrm{TA}_{2}+} + b^{\dagger}_{-q_{\parallel}q_{z}\mathrm{TA}_{2}+}) \\ + \begin{pmatrix} -q_{x}q_{z}\cos(q_{z}z) \\ -q_{y}q_{z}\cos(q_{z}z) \\ -iq_{\parallel}^{2}\sin(q_{z}z) \end{pmatrix} (b_{q_{\parallel}q_{z}\mathrm{TA}_{2}-} + b^{\dagger}_{-q_{\parallel}q_{z}\mathrm{TA}_{2}-}) \right].$$
(21)

Substitution of the z dependence of each mode into the strain Hamiltonian, Eq. (11), shows that, for the even phonons of the LA and TA₁ modes and for the odd phonon of the TA₂ mode, the Hamiltonian has the same behavior as the even interaction Hamiltonian, Eq. (7), under reflection. The hole parity is conserved on scattering by these phonon modes. For the odd phonons of the LA and TA₁ modes and for the even phonon of the TA₂ mode, the opposite pertains: the strain Hamiltonian is of the same form as the odd interaction Hamiltonian, Eq. (9), and scattering by these modes changes the parity of the hole state.

B. Piezoelectric scattering

Through the piezoelectric coupling, the lattice vibration gives rise to an electric potential¹⁰ at wave vector q:

$$\varphi_{\mathbf{q}} = -\frac{8\pi i}{\epsilon_0 q^2} e_{14} (q_x \varepsilon_{yz} + q_y \varepsilon_{zx} + q_z \varepsilon_{xy}), \qquad (22)$$

which, being a long-range potential, occurs in the diagonal terms of the Luttinger Hamiltonian.¹⁰ The factor e_{14} is the appropriate piezoelectric coefficient. Symmetry consideration similar to that in the last subsection shows that the roles of the phonon modes are reversed. For the piezoelectric coupling, the even phonons of the LA and TA₁ modes and the odd phonon of the TA₂ mode change the hole parity, whereas the odd phonons of the LA and TA₁ modes and the even phonon of the TA₂ mode preserve the hole parity.

C. Optical phonon scattering

Although the energy restriction makes this mechanism unimportant for hole relaxation except for high-energy excitations, we record here the results of an analysis similar to that for the acoustic phonon scattering. From the Fröhlich coupling¹¹ of the electron with the longitudinal phonon, it is straightforward to see that the even optical mode gives parity-conserving scattering and the odd mode changes the hole parity.

D. Shakeup process

The hole can relax by exciting the electron Fermi sea 6 in an n-type-doped quantum well or by exciting the hole Fermi sea¹² in a p-type-doped well. The latter is an Auger process. Figure 2 provides two examples of the shakeup process in an n-type-doped well. By the parity conservation in the electron-hole Coulomb interaction matrix element, part (a) shows that when the electron parity is unchanged the hole parity is also unchanged. Part (b) shows that the intersubband transition of the electron from one parity to another leads to the hole changing its parity. Since the valence subband energy is much smaller than the conduction subband energy, the intrasubband Auger process may occur more frequently than the intersubband one. Thus, hole relaxation via shakeup preserves its parity more often than not. While the shakeup process is important for the emission with zpolarization in the wave-guide configuration,⁶ it is estimated to be less than 10% of the simple recombination emitting light with σ^{\pm} polarizations. While it strength-

(b)

2c

(a)



ens somewhat the parity conserving hole relaxation, the shakeup process is much less important than the acoustic phonon scattering. In the application described below, this process is neglected.

IV. CARRIER RELAXATION AND LUMINESCENCE POLARIZATION

Based on the above investigation of the hole relaxation mechanisms, we establish in this section a new theory of the relaxation times of valence holes in quantum wells. This is crucial to the determination of the spin populations of holes just before recombination and, hence, the polarization of the emitted light. For the conduction electrons, we follow the traditional method.¹³ In this paper, we confine our attention to the free carriers and neglect the exciton effect in polarization, noting that the exciton state reflects the nature of the band-edge states with which it is associated.³

A. Carrier generation rates

We examine first the electron and hole generation rates by excitation light of a given polarization and a given energy, which are one of the determining factors of luminescence polarization. The optical transitions obey the angular momentum selection rule:

$$s - m = \sigma, \tag{23}$$

where m is the z component of the valence hole state and $\sigma = \pm 1$ corresponds to the σ^{\pm} polarizations. The transitions can be classified into sums exciting from a valence state with component and parity (m, p_m) to a conduction state with spin and parity (s, p_m) obeying parity conservation, given by

$$G(m, p_m \to s, p_m)$$

$$= |2m| M \sum_{\mu,\nu,\mathbf{k}}^{\delta} |\langle \zeta_{s,p_m}^e(\mu, \mathbf{k}) | \zeta_{m,p_m}(\nu \mathbf{k}) \rangle|^2,$$
(24)

where the superscript δ of the summation sign denotes the limitation to energy conservation processes at a given excitation light energy, $\zeta_{s,p}^{e}(\mu, \mathbf{k})$ denotes the z dependent part of the wave function of the conduction subband with spin s, parity p, band μ , and in-plane wave vector \mathbf{k} , and the hole wave function component is given by Eq. (4). The factor |2m|M represents the square of the transition matrix element between the bulk Bloch states, 3M for the heavy holes, and M for the light holes. If the polarization of the excitation light is σ^+ , there are four such sums for the transitions $(m, p_m \rightarrow s, p_m)$:

$$(-\frac{3}{2}, p \to -\frac{1}{2}, p)$$
,
 $(-\frac{1}{2}, p \to +\frac{1}{2}, p)$, (25)

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with $p = \pm 1$. The conduction subband index μ in the sum is odd for even-parity transitions and even for odd-parity transitions.

The generation rate of spin s electrons, G_s^e , at a given excitation energy, is given by

$$G_{-1/2}^{e} = \sum_{p=\pm 1} G(-\frac{3}{2}, p \to -\frac{1}{2}, p) ,$$

$$G_{1/2}^{e} = \sum_{p=\pm 1} G(-\frac{1}{2}, p \to +\frac{1}{2}, p) .$$
(26)

The generation rate of holes with parity p at a given excitation energy, G_p^h , is given by

$$G_p^h = G(-\frac{3}{2}, p \to -\frac{1}{2}, p) + G(-\frac{1}{2}, p \to +\frac{1}{2}, p).$$
(27)

B. Electron relaxation

The electron in a conduction subband promoted from a valence subband by the excitation laser relaxes by optical and acoustic phonon emissions¹⁴ to the lowest available state (the bottom of the conduction band in the undoped and *p*-type-doped quantum wells and the Fermi level in the *n*-type-doped well) in a time τ_e (in the picosecond range⁵) much shorter than the spin relaxation time τ_s and recombination time τ_r (in the nanosecond range). Electron spin relaxation can occur both during the energy relaxation and in the thermalized state at the conduction-band minimum or Fermi level before the recombination.

In the *n*-type-doped case, the Fermi sea provides equal populations of both electron spins for recombination so that the spin state of the photo-excited electron is immaterial. For the undoped or *p*-type-doped quantum well, the number of spin *s* electrons is governed by the rate equation¹⁵

$$\frac{dn_s^e}{dt} = G_s^e - \frac{n_s^e}{\tau_r} - \frac{n_s^e - n_{-s}^e}{2\tau_s} \quad (s = \pm \frac{1}{2}).$$
(28)

The second and third terms on the right-hand side of the equation account for the loss of the spin s electrons through recombination and for the change in spin polarization through spin flip to and from -s, respectively. The generation rate on the right-hand side of Eq. (28), $G_{s,1}^{e}$ is given by Eq. (26).

The steady-state electron spin populations, n_s^e , are, from Eq. (28),

$$n_{s}^{e} = \frac{\tau_{r}}{2} \left(G_{s}^{e} + G_{-s}^{e} + \frac{\tau_{s}}{\tau_{r} + \tau_{s}} (G_{s}^{e} - G_{-s}^{e}) \right) .$$
(29)

Since the minority carriers for the *p*-type-doped quantum well are the electrons in the conduction band, the polarization of the luminescence \mathcal{P}_p is given in terms of the electron spin polarization as,

$$\mathcal{P}_{p} = \frac{n_{-1/2}^{e} - n_{1/2}^{e}}{n_{-1/2}^{e} + n_{1/2}^{e}} = \frac{G_{-1/2}^{e} - G_{1/2}^{e}}{G_{-1/2}^{e} + G_{1/2}^{e}} \frac{\tau_{s}}{\tau_{s} + \tau_{r}}$$
$$= \mathcal{P}_{p}^{0} \frac{\tau_{s}}{\tau_{s} + \tau_{r}} \quad . \tag{30}$$

The expression for \mathcal{P}_p^0 is the same as the polarization in Ref. 3 whose authors' assumption of equal hole polarization holds trivially for the *p*-type-doped quantum well. Since the transition probability between the heavy-hole state Γ_8 and the electron state Γ_6 is three times that between the light-hole state Γ_8 and the electron state Γ_6 [from the |2m|M factor in Eq. (24)], the polarization of photoexcited electrons is 50% in the bulk case in the limit of infinite τ_s . This simple limit is used in extracting relaxation times from the Hanle effect measurements.⁵ However, we see from Eq. (30) that the electron polarization is very much dependent on the subband structure through the generation rate.

C. Hole relaxation

From Sec. III, we see that hole scattering which changes its momentum and energy also changes its spin components because of the heavy- and light-hole mixing. Here we attempt to construct a simple theory of the hole spin polarization as a result of energy and momentum relaxation. There are two hole scattering channels, one conserving the hole parity and one flipping the parity. We average over the scattering rates for each channel and define two relaxation times for holes: τ_+ the hole momentum relaxation rate conserving parity and τ_- changing parity. These times should be in the picosecond range.⁵

The density of relaxed holes, n_p^h , of parity p at the top of the valence subband (heavy hole) is established in two stages. First, the density of holes of parity p generated, N_p^h , is given by

$$\frac{dN_p^h}{dt} = G_p^h - \left(\frac{1}{\tau_+} + \frac{1}{\tau_-}\right) N_p^h , \qquad (31)$$

where G_p^h is the hole generation rate given by Eq. (27) and the two remaining terms are losses due to relaxation conserving or changing the hole parity. Second, from the hole density generated the relaxed hole density is deduced:

$$\frac{dn_p^h}{dt} = \frac{N_p^h}{\tau_+} + \frac{N_{-p}^h}{\tau_-} - \frac{n_p^h}{\tau_r} - \frac{n_p^h - n_{-p}^h}{2\tau_s^h} .$$
(32)

The first two terms come from hole momentum relaxation from the two parity channels. The third term is the loss due to recombination. The last term is the spin relaxation at the top of the heavy-hole valence subband, as opposed to the spin change due to momentum scattering, and the intrinsic hole spin relaxation τ_s^h is of the same order of magnitude as the electron spin relaxation time τ_s because the spin-flip mechanisms for these two times are similar. In the following, for simplicity, we assume these two times to be equal. Solution of the rate equations yields the steady-state hole density:

$$n_{p}^{h} = \frac{\tau_{r}}{2} \left(G_{p}^{h} + G_{-p}^{h} + \frac{\tau_{s}}{\tau_{r} + \tau_{s}} \frac{\tau_{-} - \tau_{+}}{\tau_{-} + \tau_{+}} (G_{p}^{h} - G_{-p}^{h}) \right) .$$
(33)

From the example worked out at the end of Sec. II, n_p^h

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has spin- $\pm \frac{3}{2}$ for $p = \pm 1$.

In the *n*-type-doped quantum well, where there are equal populations of electrons of both spins, the luminescence polarization is determined by the hole polarization, i.e.,

$$\mathcal{P}_{n} = \frac{n_{-1}^{h} - n_{1}^{h}}{n_{-1}^{h} + n_{1}^{h}} = \frac{G_{-1}^{h} - G_{1}^{h}}{G_{-1}^{h} + G_{1}^{h}} \frac{\tau_{-} - \tau_{+}}{\tau_{-} + \tau_{+}} \frac{\tau_{s}}{\tau_{s} + \tau_{r}}$$
$$= \mathcal{P}_{n}^{0} \frac{\tau_{-} - \tau_{+}}{\tau_{-} + \tau_{+}} \frac{\tau_{s}}{\tau_{s} + \tau_{r}} . \tag{34}$$

For the undoped well, the recombination events depend on the smaller of the numbers of available electrons and of holes:

$$\mathcal{P}_{u} = \frac{\min(n_{-1/2}^{e}, n_{-1}^{h}) - \min(n_{1/2}^{e}, n_{1}^{h})}{\min(n_{-1/2}^{e}, n_{-1}^{h}) + \min(n_{1/2}^{e}, n_{1}^{h})} , \qquad (35)$$

where the function $\min(x, y)$ takes on the smaller of x and y.

V. LUMINESCENCE OF GaAs/Al_{1-x}Ga_xAs QUANTUM WELLS

Our theory of hole relaxation and luminescence polarization is applied to the $GaAs/Al_{1-x}Ga_xAs$ quantum wells. From Eq. (34), we see that the competition between the parity-conserving and the parity-changing momentum relaxations determines the sign of the polarization in the *n*-type-doped well. We estimate the ratio τ_+/τ_- of the relaxation times through the two channels. We neglect shakeup and the optical phonon scatterings and use only hole-acoustic phonon scatterings through both the deformation potential and the piezoelectric coupling, which apply to both doped and undoped wells. In general, because the diagonal part of the interaction Hamiltonian conserving parity has a cosine dependence on z and the parity-changing counterpart has a sine dependence, the parity-conserving scattering is stronger, leading to the ratio τ_+/τ_- less than unity. A detailed calculation of the deformation-potential scatterings¹⁶ using the wave functions in a well of 120 Å width and impenetrable walls yields (i) within the intrasubband transitions 1h to the top of the band (see Fig. 1) the average ratio of 0.08; (ii) for the intersubband transitions from the $\mathbf{k} = \mathbf{0}$ state of 1ℓ to the states in the 1h subband an average value of 0.73; (iii) for the transitions from the $\mathbf{k} = \mathbf{0}$ state of 2h to the states in 1ℓ subband an average value of 0.52. For the piezoelectric coupling,¹⁷ the corresponding values are 0.005, 0.14, 0.45. The calculations confirm the conclusion that $\tau_+/\tau_- < 1$.

We note that in Eq. (30) for the luminescence polarization for the *p*-type-doped well, the relaxation times form a factor modulating the polarization spectrum, whose dependence on the excitation energy is given by \mathcal{P}_p^0 , the part of the polarization determined by the generation rates which are calculated from the subband energies and wave functions within the self-consistent potential effective-mass approximation. The ratio τ_r/τ_s is used to adjust the vertical scale of the calculated spectrum to fit the measured one,¹ resulting in the value of 0.12. Comparison of theory with experiment is shown in Fig. 3(a).

We assume the same value for the relaxation time ratio τ_r/τ_s in the *n*-type-doped quantum well. From Eq. (34), again the generation rate factor, \mathcal{P}_n^0 , is calculated from the *n*-type-doped well subband wave functions and energies, leaving only the vertical scale to be determined by τ_r/τ_s and by the ratio of the hole momentum relaxation times τ_+/τ_- . Fit to experiment shown in Fig. 3(b) is shown for $\tau_+/\tau_- = 0.46$. We assume the same two relaxation times ratios for the undoped well and calculate its polarization spectrum with no more adjustable parameters, shown in Fig. 3(c). Excitonic effects are neglected in our calculation.

We note the general agreement for all the spectra, especially for the excitation energies of the band-to-band transition thresholds, which are marked. The steplike features could be rounded off by including particle life-time or inhomogeneous broadening due to varying thickness of the well. In the following we provide a detailed analysis of these thresholds to gain a physical understanding of the polarization spectra.

(1) At the onset of the $(1\ell \rightarrow 1c)$ transitions, the polarization in all three wells decreases rapidly. The incident light creates pairs of $s = \frac{1}{2}$ electrons and $m = -\frac{1}{2}$ holes. The holes relax, predominantly preserving even parity, to the $m = \frac{3}{2}$ states in the 1*h* band. Emission of σ^- polarization results from recombination between $s = \frac{1}{2}$ electrons and $m = \frac{3}{2}$ holes. For the undoped and p-type-doped wells, the high joint density of states near the threshold,¹⁸ which comes from the 1c conduction subband and the 1ℓ valence subband having curvature of the same sign, results in the negative polarization.³ Thus, we interpret the dip in the spectra at the threshold as experimental evidence for the high joint density of states. A feature in photoreflectance was assigned to the same critical singularity.¹⁹ In the *n*-type-doped well, the Fermi sea blocks the $(1\ell \rightarrow 1c)$ transitions near the threshold and thus prevents the polarization reversal at that energy.

(2) Consider the disparate behavior for the three wells at the $(1\ell, 2h \rightarrow 2c)$ threshold which was first noted by Ruckenstein et al.⁴ In the p-type and undoped wells, the polarization increases at the threshold. In the n-typedoped well, the polarization decreases so much that it changes sign. The proximity of the 1ℓ and 2h hole subbands causes strong $\mathbf{k} \cdot \mathbf{p}$ mixing and gives rise to a strong $m = \pm \frac{3}{2}$ component in both bands. The odd-parity component $(n = 2, m = -\frac{3}{2})$ permits the optical excitation from $m = -\frac{3}{2}$ to $s = -\frac{1}{2}$ of the odd-parity conduction band 2c. By Eq. (5), the odd-parity $m = -\frac{3}{2}$ component at the (n = 2) subband means that the corresponding hole states in both the 1ℓ and 2h bands must have even parity. Via the parity-conserving scattering channel, a larger portion of these holes relax to the top heavy-hole (1*h*) even-parity state with $m = \frac{3}{2}$. In the *n*-type-doped well, such hole states recombine with $s = -\frac{1}{2}$ electrons to give a strong σ^- luminescence and, hence, the polariza-



FIG. 3. Polarization spectra under σ_+ excitation for (a) a *p*-type-doped well, (b) an *n*-type-doped well, and (c) an undoped well. The excitation energy is measured from the band gap in the well.

tion reversal. In the *p*-type-doped well, the same optical excitation generates $s = -\frac{1}{2}$ electrons which increase the σ^+ luminescence and the paucity of the $s = \frac{1}{2}$ electrons limits the σ^- luminescence. In the *n*-type well, a minor portion of the optically excited even-parity holes relax via the parity-changing scatterings to the odd-parity, $m = -\frac{3}{2}$ holes which recombine with the dominant spindown electrons to give the σ^+ luminescence. This step would disappear if there were no parity-changing hole relaxation.

(3) The $(2\ell \to 2c)$ transitions promote the electrons from the $m = -\frac{1}{2}$ valence states to the $s = \frac{1}{2}$ conduction states. The majority of the holes states, being of odd parity (p = -1), relax to the odd-parity state (p = -1) in the 1*h* band which, by Eq. (5), has the even-parity $m = -\frac{3}{2}$ component (at n = 1). In the *n*-type-doped well, these holes provide the σ^+ emission and, thus, change the polarization to positive again. In the undoped and *p*-type-doped well, the simultaneous production of the $s = \frac{1}{2}$ electrons provides the σ^- emission and the lack of s = -1/2 electrons limits the σ^+ emission. Thus, at this threshold the polarization is reduced.

(4) No polarization anomalies occur at the $(3h \rightarrow 3c)$ transition threshold since by symmetry it is analogous to the $(1h \rightarrow 1c)$ transition.

All these features arising out of incomplete hole spin relaxation are in good agreement with observation¹ and the energies at which they occur are in quantitative agreement, for all three quantum wells, undoped, *n*-type doped, and *p*-type doped.

VI. CONCLUSION

By investigating the relaxation of holes through the complex valence subband structure in the quantum wells, we replace the usual assumption of complete hole depolarization by establishing a method to calculate the hole spin polarization from two channels of hole relaxation through two sets of degenerate spin-mixed states. For the symmetric quantum wells, the two channels are conveniently classified by the hole parity. A theory of luminescence polarization is based on the rate equations including the momentum and intrinsic spin relaxations of both electrons and holes. Since hole spins are changed by momentum relaxation, we use the word intrinsic to denote spin changes due to the usual nonmomentum processes. The ability of the theory to explain the physical processes in all the salient features of the observed polarization spectra, whether the well is intrinsic, *n*- or *p*-type, demonstrates the indispensability of the incomplete hole depolarization. Since all luminescence processes involve hole relaxation, our theory is of crucial importance in a wide range of optical phenomena. A brief summary of our work has been given elsewhere.²⁰

The theory should be further tested by varying the excitation conditions, such as from circular to plane polarization or from continuous excitation to pulse, by applying external magnetic or electric fields and by changing the temperature. Plane-polarized excitation light leads to the so-called optical alignment¹³ in which the electronhole pair retains the linear polarization. This is evident from Eqs. (30) and (34) for the doped wells because of the factor depending on the equal generation rates of both polarizations independent of the hole orientation due to $\tau_+ < \tau_-$ and it is true to a lesser extent for the undoped wells.

Our theory can also explain why the polarization of luminescence in an *n*-type-doped quantum well is insensitive to the weak transverse magnetic field, the Hanle effect.⁴ There are equal numbers of conduction electrons of both spin components so that the polarization is dominated by the spin populations of holes at the valenceband maximum. Since the hole relaxes its spin through the momentum relaxation, which is much faster than intrinsic spin relaxation with time τ_s or recombination with time τ_r . The characteristic hole momentum relaxation time τ_p , given by

$$\frac{1}{\tau_p} = \frac{1}{\tau_+} + \frac{1}{\tau_-},\tag{36}$$

determines the magnetization time. During such a small time τ_p , the Larmor precession of each photoexcited hole

cannot decrease the polarization enough as compared with the Hanle effect due to the electrons in the conduction band. In deducing the recombination time and the electron spin relaxation time from the Hanle effect in the undoped quantum well,⁵ the polarization in Eq. (35) should be used as the zero field value, rather than the 50% actually used. Equation (35) at high excitation energies is about 23%, which is close to the experimental result 20%.⁵ A future direction of our work is to deduce relaxation times from polarization measurement within the framework of our theory.

In a high magnetic field normal to the interface plane of the quantum well, the subbands are split into Landau levels. Luminescence polarizations have been measured in high magnetic fields for excitation light of both polarizations in an undoped well.²¹ Calculation of the polarization based on our theory for high magnetic fields in the limit of dominant parity-conserving hole relaxation²² vields the correct sign for the luminescence polarization for each of the Landau levels measured. The difference with the theoretical interpretation of Ref. 21 is that the authors assumed complete hole depolarization and we do not. A simple test of our theory would be to measure the luminescence polarization in high magnetic fields in an *n*-type-doped quantum well. Further examination of the relaxation times for both the electrons and holes between Landau levels is under way.

An electric field normal to the interface plane will mix the parities of the hole states and change the relaxation times of the two channels of degenerate states. This effect will be an important test of our concept of hole relaxation. We plan to pursue our theory to include the electric field. Finite-temperature effects will also be studied in future.

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FIG. 2. The shake-up processes in an *n*-type-doped well: (a) the parities of the carriers are preserved; (b) the parities are changed.