## "Spin"-flip scattering of holes in semiconductor quantum wells

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We report results of calculations on the "spin"-flip relaxation time of holes in semiconductor quantum wells due to hole interaction with static scatterers such as ionized impurities, alloy fluctuations, and *s*-*d* exchange (in the case of quantum wells based on diluted magnetic semiconductors). We show that size quantization along the growth axis leads to a drastic quenching of the "spin"-flip scattering. This results in hole "spin"-flip relaxation times, which can be much longer than the recombination time when the hole in-plane kinetic energy is small compared with the  $HH_1$ - $LH_1$  separation distance.

Relaxation processes of conduction electrons in semiconductor heterostructures have been the subject of numerous investigations (see, for instance, Refs. 1-7), while hole-scattering mechanisms are in contrast rather rarely documented.<sup>8,9</sup> A particular kind of relaxation is the "spin"-flip scattering process in which energy may or may not be conserved. In what follows we shall denote by hole "spin"-flip scattering the events that couple two different Kramers-degenerate hole states. Due to the large spin-orbit coupling in most of the III-V compounds, the hole spin is not a good quantum number. In bulk materials this leads to a very short "spin" relaxation time. In quantum wells, size quantization along the growth axis gives rise to eigenstates, which are also eigenstates of  $J_z$ , the projection along the growth axis of the (pseudo) total angular momentum J  $(J = \frac{3}{2})$  if the in-plane wave vector (k) vanishes (subbands' edges). The heavy (light) -hole subband edges  $HH_n$  (LH<sub>m</sub>) correspond to  $J_z = \pm \frac{3}{2}$  ( $\pm \frac{1}{2}$ ). Away from the subband edges, the heavy- and light-hole nature of the energy eigenstates becomes mixed—the so-called band-mixing effect.<sup>10-12</sup> One may, however, infer from previous remarks that there should exist in a quantum well an in-plane wave-vector range where the band mixing is not very large and where the energy levels are approximate eigenstates of  $J_z$ . Then, what will matter for the "spin"-flip scattering are the magnitudes of the matrix elements of the perturbing potentials between the two eigenstates of opposite  $J_z$  at each subband edge. It turns out that these matrix elements are zero for several energy-conserving perturbations, such as those due to ionized impurities, alloy fluctuations, or even s-d exchange (when there exist localized magnetic moments in the heterostructure). This should lead to a suppression of "spin"-flip scattering when the hole energy approaches the subband edges. Information on the carrier "spin"-flip scattering can then be extracted from cw or time-resolved photoluminescence experiments.

Very recently, Uenoyama and Sham<sup>13</sup> have shown that it is possible to semiquantitatively interpret the experiments performed on undoped, n-type doped, and p-type doped quantum wells by means of rate equations which incorporate a partial conservation of the holes' "spin" during their relaxation towards  $HH_1$ . The ratio of the (phonon-induced) "spin"-conserving ( $\tau_{sc}$ ) to "spin"-flip ( $\tau_{sf}$ ) hole relaxation times, taken to be energy independent, was fitted to the experiments and found to be equal to 0.46. Our purpose in this communication is to analyze quantitatively the hole "spin"-flip scattering time in quantum wells and to demonstrate that it can be very large near the HH<sub>1</sub> edge, a situation reminiscent of the case of uniaxially stressed bulk materials.<sup>14</sup> We shall limit our consideration to the aforementioned energy-conserving processes, whose Hamiltonians are, respectively,

$$V_{\rm imp}(\mathbf{r}) = \mathbb{1} \frac{2\pi e^2}{\kappa S} \sum_{\mathbf{R}_i} \sum_{\mathbf{Q}} (1/Q) \exp[i\mathbf{Q} \cdot (\boldsymbol{\rho} - \boldsymbol{\rho}_i) - Q|z - z_i|],$$
(1)

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$$V_{\text{alloy}}(\mathbf{r}) = \mathbb{I}\Omega_0 \,\delta V \left[ \sum_{\mathbf{R}_B} x \,\delta(\mathbf{r} - \mathbf{R}_B) - \sum_{\mathbf{R}_C} (1 - x) \delta(\mathbf{r} - \mathbf{R}_C) \right], \quad (2)$$

$$V_{sd}(\mathbf{r}) = J_h \Omega_0 \sum_{\mathbf{R}_i} \delta(\mathbf{r} - \mathbf{R}_i) \mathbf{S}_i \cdot \boldsymbol{\sigma} , \qquad (3)$$

where **Q** is a two-dimensional wave vector;  $\mathbf{R}_i = (\rho_i, z_i)$  is the impurity position;  $\kappa$  is the static dielectric constant; 1 is the 4×4 identity matrix on the  $|\frac{3}{2}, J_z\rangle$  basis;  $\sigma$  is the valence-electron spin (also a 4×4 matrix on the same basis);  $\delta V$  is the strength of the alloy scattering potential averaged over the unit cell (volume  $\Omega_0$ ); x is the C mole fraction in the ternary alloy  $C_x B_{1-x} A$ ;  $J_h$  is the exchange constant for holes in diluted magnetic semiconductors; and  $\mathbf{S}_i$  is the localized spin at site  $\mathbf{R}_i$ .

In the following we use an analytical description of the hole envelope functions. It is obtained by diagonalizing the off-diagonal terms of the Luttinger Hamiltonian in

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the truncated basis spanned by the three lower bound states  $HH_1$ ,  $LH_1$ , and  $HH_2$  of the k=0 problem,<sup>15,16</sup> where k is a two-dimensional in-plane wave vector,

$$\mathbf{k} = (k_x, k_y) = (k \cos\theta, k \sin\theta)$$
.

In doing so, we should be able to describe accurately the topmost subband dispersions of sufficiently narrow quantum wells (typically,  $L \leq 100$  Å in GaAs-Ga<sub>0.7</sub>Al<sub>0.3</sub>As). In the axial approximation,<sup>10</sup> the eigenenergies, each two-fold degenerate, are the solutions of

$$(\varepsilon_{\mathrm{HH}_{1}} - \varepsilon)[(\varepsilon_{\mathrm{LH}_{1}} - \varepsilon)(\varepsilon_{\mathrm{HH}_{2}} - \varepsilon) - |\langle \phi_{1}|b|\chi_{2}\rangle|^{2}] - |\langle \phi_{1}|c|\chi_{1}\rangle|^{2}(\varepsilon_{\mathrm{HH}_{2}} - \varepsilon) = 0, \quad (4)$$

where

$$\varepsilon_{\rm HH_i} = E_{\rm HH_i} - \hbar^2 k^2 (\gamma_1 + \gamma_2) / 2m_0 , \qquad (5)$$

$$\varepsilon_{\mathrm{LH}_i} = E_{\mathrm{LH}_i} - \hbar^2 k^2 (\gamma_1 - \gamma_2) / 2m_0 ,$$
 (6)

$$c = \hbar^2 \sqrt{3} \gamma_2 k^2 e^{-2i\theta} / 2m_0, \tag{7}$$

$$b = -i\sqrt{3}\hbar^2 k e^{-i\theta} \frac{1}{2m_0} \left[ \gamma_3 \frac{d}{dz} + \frac{d}{dz} \gamma_3 \right], \qquad (8)$$

 $\phi_1$ ,  $\chi_1$ , and  $\chi_2$  are the quantum-well envelope functions of the LH<sub>1</sub>, HH<sub>1</sub>, and HH<sub>2</sub> states at k = 0, and  $\gamma_1, \gamma_2, \gamma_3$  are the hosts' Luttinger parameters. With each of the twofold-degenerate eigenenergies, one can associate the two orthogonal wave functions

$$\Psi_{\mathbf{k}\uparrow} = \frac{1}{\sqrt{S}} (1 + a^2 + \eta^2)^{-1/2} \\ \times (ae^{-2i\theta}\chi_1(z), \phi_1(z), 0, i\eta e^{i\theta}\chi_2(x))e^{i\mathbf{k}\cdot\rho} , \qquad (9)$$

$$\Psi_{kl} = \frac{1}{\sqrt{S}} (1 + a^2 + \eta^2)^{-1/2} \\ \times (-i\eta e^{-i\theta} \chi_2(z), 0, \phi_1(z), a e^{2i\theta} \chi_1(z)) e^{i\mathbf{k}\cdot\boldsymbol{\rho}}$$
(10)

relative to the basis  $(\frac{3}{2}, -\frac{1}{2}, \frac{1}{2}, -\frac{3}{2})$  and where

$$a = \hbar^2 \sqrt{3} k^2 \gamma_2 \langle \phi_1 | \chi_1 \rangle / [2m_0(\varepsilon - \varepsilon_{\mathrm{HH}_1})], \qquad (11)$$

$$\eta = \hbar^2 \sqrt{3}k \left\langle \chi_2 \left| \left[ \gamma_3 \frac{d}{dx} + \frac{d}{dz} \gamma_3 \right] \right| \phi_1 \right\rangle \frac{1}{2m_0(\varepsilon - \varepsilon_{\mathrm{HH}_2})}$$
(12)

One readily checks that  $\langle \Psi_{k\uparrow} | J_z | \Psi_{k\uparrow} \rangle$  ( $\langle \Psi_{k\downarrow} | J_z | \Psi_{k\downarrow} \rangle$ ) extrapolates to  $+\frac{3}{2}$  ( $-\frac{3}{2}$ ) for the lowest-lying hole states (the HH<sub>1</sub> branch). Moreover,  $J_z$  has no nonvanishing matrix element between any  $\Psi_{k\uparrow}$  and  $\Psi_{k\downarrow}$  corresponding to the same energy. This has led us to label the hole eigenstates according to the "spin" ( $\downarrow,\uparrow$ ), even though neither  $\Psi_{k\uparrow}$  nor  $\Psi_{k\downarrow}$  are eigenstates of  $\sigma_z$  or  $J_z$  at finite k. Figures 1(a) and 1(b) show the calculated dispersion relations and averages of  $J_z$  over the  $\uparrow$  and  $\downarrow$  eigenstates for a 80-Å GaAs-(Ga,Al)As quantum well (valence-band offset  $|V_P|=177 \text{ meV}, \gamma_1=6.85, \gamma_2=2.1, \gamma_3=2.9$ ). As expected on intuitive grounds and already discussed else-



FIG. 1. (a) Calculated in-plane dispersion relations of the topmost valence subbands in a 80-Å-thick GaAs-Ga<sub>0.7</sub>Al<sub>0.3</sub>As single quantum well. Solid line, three-level model; dashed lines, inclusion of all the levels bound in the well at k = 0. The arrow locates the maximum of the LH<sub>1</sub> subband. (b) Calculated dependence upon the in-plane wave vector of the  $J_z$  averages over the energy eigenstates in the three-level model. The notation  $\uparrow, \downarrow$  refers here to the dominant character at k = 0. They coincide with (are opposite to) those used in the text for the HH<sub>1</sub> (LH<sub>1</sub>, HH<sub>2</sub>) subbands.

where,<sup>11</sup> the  $J_z$  averages deviate from  $\pm \frac{3}{2}$   $(\pm \frac{1}{2})$  for the heavy (light) -hole branches when they anticross. We also show for comparison in Fig. 1(a) the dispersion curves obtained by retaining more subbands in the k = 0 basis. It is seen that over a significant k range the three-level model gives a fair account of the HH<sub>1</sub> dispersion.

The "spin"-flip scattering time  $\tau_{nk}$  within the *n*th subband is calculated under the Born approximation by using the Fermi golden rule:

$$\hbar/2\pi\tau_{nk} = \sum_{\mathbf{k}'} |\langle \Psi_{nk\uparrow} | V_{def} | \Psi_{nk\downarrow\downarrow} \rangle|^2 \delta(\varepsilon_{nk\uparrow} - \varepsilon_{nk\downarrow\downarrow}) , \quad (13)$$

where  $V_{def}$  is the scattering potential and where an average over the random location of the scatterers has to be



FIG. 2. Calculated dependence upon the in-plane wave vector of the impurity-assisted "spin"-flip scattering times  $(\tau_{sf})$  within the HH<sub>1</sub> and LH<sub>1</sub> subbands for three GaAs-AlAs quantum wells.

performed. Note that  $\tau_{nk}$  is directly linked to a measurable quantity when *n* coincides with the lower hole subband (HH<sub>1</sub>). In fact, it is easy to check by using rate equations that any initial hole polarization  $(n_{k\uparrow} - n_{k\downarrow})$ , as e.g., is created by optical spin orientation, decays with a time constant  $\tau_{nk}/2$  due to the defect-induced spin-flip scattering. It is therefore of importance to get quantitative estimates of  $\tau_{nk}$  in order to decide whether photoluminescence polarization results only from electron disorientation as in bulk materials (because the hole polarization decays almost instantaneously or because the holes which participate in the recombination process exist prior to the illumination and are unpolarized) or whether size quantization complicates this simple picture.

Figure 2 shows the k dependence of the "spin"-flip relaxation time within the HH<sub>1</sub> and LH<sub>1</sub> subbands for three GaAs-AlAs quantum wells with thicknesses L = 30, 50,and 70 Å ( $|V_P| = 0.5 \text{ eV}$ ). The scatterers are ionized impurities ( $N_{\text{imp}} = 10^{10} \text{ cm}^{-2}$ ), assumed to be localized on one interface of the quantum well. The dominant trend seen in Fig. 2 is a very strong dependence of the "spin"flip relaxation time upon k, i.e., upon the band mixing. Note, in particular, that  $\tau_{nk}$  diverges as k tends to zero, where the energy eigenstates are also eigenstates of  $J_z$ and thus characterized by forbidden "spin" flip. As k increases, intra-HH1 or -LH1 "spin"-flip scattering becomes allowed and  $\tau_{nk}$  decreases sharply. As expected from the physical origin of the "spin"-flip scattering, the  $\tau_{nk}$  drop with **K** is steeper for the thicker wells, since the energy separation between the k = 0 levels decreases with increasing L. Over a significant **k** range the  $\tau_k$  for HH<sub>1</sub> is longer than 100 ps, i.e., becomes comparable to the electron-hole recombination time. We are thus led to conclude that any hole polarization created in this k range will not decay quickly enough, which invalidates the interpretation of the photoluminescence polarization experiments in terms of an instantaneous hole depolarization. On the other hand, when the band mixing is large (say, k > 0.02 Å  $^{-1}$  for  $L \ge 50$  Å) the "spin"-flip time becomes short ( $\leq 10$  ps), a bulklike situation. Two singularities (very short  $\tau$ 's) are noticeable in the LH<sub>1</sub>-related curves. They are linked to the one-hump shape of the LH<sub>1</sub> dispersion relations which induces a very large (in principle, infinite) density of states in the vicinity of the local maximum and thus a very short "spin"-flip time. The singularity is absent in the 30-Å-thick well because the energy separation between  $LH_1$  and  $HH_2$  is too large to allow a relative maximum in the  $LH_1$  dispersion (at least in our three-level analysis). We cannot compare the "spin"-flip time with the equivalent "spin"-conserving one due to the long-range nature of the Coulombic potential which leads to an infinite cross section for the "spin"-conserving processes.

Such is not the case for the short-range alloy scatteran important scattering mechanism ing, in Ga<sub>0.47</sub>In<sub>0.53</sub>As-InP heterolayers. Figure 3 shows a plot of the intra-HH1 "spin"-flip and "spin"-conserving relaxation times versus k due to alloy and impurity scatterings for two quantum-well thicknesses (30 and 50 Å,  $|V_p| = 0.365 \text{ eV}$ ). The "spin"-flip curves display versus k the same sort of large variations as discussed in Fig. 2. We again find that over a significant  $\mathbf{k}$  range the hole "spin"-flip relaxation time is comparable to, or larger than, the recombination lifetime. The "spin"-conserving transitions are instead fast and almost k independent. The latter feature is reminiscent of that already discussed for electrons, where the alloy-scattering-limited lifetime is inversely proportional to the density of states,<sup>11</sup> a constant for quasi-two-dimensional motion. Notice that at large k the "spin"-flip and "spin"-conserving times are of the same order of magnitude, a quasibulk situation. In fact, we also show in Fig. 3 the k dependence of the



FIG. 3. Calculated dependence upon the in-plane wave vector of the impurity- and alloy-fluctuation-assisted "spin"-flip (sf) and "spin"-conserving (sc) scattering times within the HH<sub>1</sub> subband of  $Ga_{0.47}In_{0.53}As$ -InP quantum wells and bulk  $Ga_{0.47}In_{0.53}As$ .

"spin"-flip and "spin"-conserving relaxation times for bulk  $Ga_{0.47}In_{0.53}As$  when  $k_z$  is set equal to that corresponding to the HH<sub>1</sub> bound state in the 50-Å-thick well,

$$-\hbar^2 k_z^2 (\gamma_1 - 2\gamma_2)/2m_0 = E_{\rm HH_1}$$

These times have been calculated with neglect of the band warping ( $\gamma_2 = \gamma_3$  in the Luttinger Hamiltonian) and found to be equal to

$$\hbar/2\pi\tau_{\rm sf} = [m_0\Omega_0/32\pi h^2(\gamma_1 - 2\gamma_2)] \\ \times x (1-x)(\delta V)^2 (k_z^2 + k^2)^{1/2} , \qquad (14)$$

$$K/2\pi\tau_{\rm sc} = [m_0\Omega_0/8\pi h^2(\gamma_1 - 2\gamma_2)] \\ \times x (1-x)(\delta V)^2 (k_z^2 + k^2)^{1/2} \\ \times \left[1 - \frac{3}{8}\frac{k_z^2}{k_z^2 + k^2}\right].$$
(15)

Over the k range shown in Fig. 3,  $k \ll k_z$  and both  $\tau_{\rm sc}$  and  $\tau_{\rm sc}$  in the bulk are nearly constant and close to  $\tau_{\rm sc}$  in the well. It is remarkable that under very comparable k and  $k_z$  conditions a quantum well and the corresponding bulk material can be characterized by "spin"-conserving recombination times which are of the same order of magnitude, while the "spin"-flip relaxation times are several decades longer in the well than in the bulk.

Let us now discuss the case of quantum wells based on diluted magnetic semiconductors<sup>17</sup> such as

$$Cd_{1-x}Mn_xTe-Cd_{1-y}Mn_yTe$$
.

These heterostructures contain efficient spin-flip scatterers: the localized magnetic moments  $(Mn^{2+}, S = \frac{5}{2})$ whose effect on the spin-flip scattering of conduction electrons has recently been shown<sup>18</sup> to lead to intrasubband spin-flip relaxation times falling in the range 10-100 ps, irrespective of the in-plane electron kinetic energy. Time-resolved photoluminescence polarization experiments have yielded evidence for a very fast decay of the photoluminescence polarization,<sup>19</sup> independently of the location of the scatterers (well or barrier). For "spin"-flip scattering within the HH<sub>1</sub> subband induced by a Heisenberg coupling between the valence and d electrons, one should examine the matrix elements of  $\sigma$  between  $\Psi_{\mathbf{k}\uparrow}$  and  $\Psi_{\mathbf{k}\downarrow}$ . At  $k = 0 \sigma$  has no nonvanishing elements between the  $|\frac{3}{2}, \pm \frac{3}{2}\rangle$  eigenstates, in marked contrast with the conduction-band case where the band-edge Bloch functions are pure spin states.  $\sigma_{+}$  and  $\sigma_{-}$  couple  $|\frac{3}{2},\pm\frac{3}{2}\rangle$  with  $|\frac{3}{2},\pm\frac{1}{2}\rangle$  while  $\sigma_z$  is diagonal on the  $|\frac{3}{2},J_z\rangle$ basis. Thus, at finite **k**,  $\mathbf{S}_i \cdot \boldsymbol{\sigma}$  will have nonzero matrix elements between  $\Psi_{k\uparrow}$  and  $\Psi_{k\downarrow}$  only to the extent that these wave functions display an admixture of HH<sub>1</sub> and LH<sub>1</sub> band-edge states. Therefore, the narrower the well, the smaller will be the band-mixing effects, because the energy separation between HH<sub>1</sub> and LH<sub>1</sub> will increase, and thus the more inhibited will be the "spin"-flip scattering in the HH<sub>1</sub> branch. This process will end when, by further decreasing the quantum-well thickness, the distance between  $HH_1$  and  $LH_1$  will be made to decrease until, at zero thickness, HH1 and LH1 will coin-



FIG. 4. Calculated dependence upon the in-plane wave vector of the *s*-*d* exchange-assisted "spin"-flip (sf, dashed lines) and "spin"-conserving (sc, solid lines) scattering times within the  $HH_1$  subband of a  $Cd_{0.935}Mn_{0.065}Te-Cd_{0.62}Mn_{0.38}Te$  quantum well.

cide with the top of the well where a fully threedimensional situation will be restored.

With this in mind, we again have to expect a large variation of the "spin"-flip scattering time with k when the quasi-two-dimensional situation prevails. This is illustrated in Fig. 4 where we show the k dependence of the intra-HH<sub>1</sub> "spin"-flip and "spin"-conserving relaxation times in a  $Cd_{1-x}Mn_xTe-Cd_{1-y}Mn_yTe$  [x=0.065, y=0.38,  $|V_p|=98$  meV,  $J_h=0.88$  eV (Ref. 20)] quantum well with L=86 Å. We have neglected the strain effects that are due to the elastic accommodation of the lattice mismatch between the two hosts on the grounds that the exact strain state depends sensitively on the exact sample configuration (nature of the substrate, buffer thickness, etc.). We have also assumed that the scattering rates are proportional to the corresponding mole fractions x or y



FIG. 5. Same as in Fig. 4 but for different well thicknesses and "spin"-flip scattering only.

whereas it may be argued<sup>18</sup> that the only spins available for the "spin"-flip scattering are those which are not antiferromagnetically locked to a nearest neighbor into a nonmagnetic singlet. In any case the numerical results displayed in Figs. 4 and 5 can be scaled to the appropriate Mn concentration. As in the electron case,<sup>18</sup> we find that the spins located in the well are considerably more efficient than those located in the barrier, as a result of the strong localization of the hole eigenstates in the well. The s-d exchange scattering shows many common features with the alloy scattering; in particular, its shortrange nature leads to "spin"-conserving relaxation times which are fast and almost k independent. The "spin"-flip scattering times again display a very large decrease with increasing k as a result of band-mixing effects. The krange where the "spin"-flip times are comparable to, or larger than, the recombination time is significant  $(10^6)$  $cm^{-1}$  for "spin"-flip scattering occurring in the well, more than  $2.5 \times 10^6$  cm<sup>-1</sup> for that occurring in the barrier). In Fig. 5 we show the k dependence of the "spin"flip scattering time for the same material parameters as used in Fig. 4 but for several quantum-well thicknesses. With increasing L, band mixing becomes more important, which results in a faster "spin"-flip scattering for the localized spins in the well. For those located in the barrier, the inverse trend takes place due to the prevalent

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effect of decreasing wave-function penetration into the barrier with increasing L.

In conclusion, we have shown that the current assumption of fast hole depolarization is not correct in semiconductor quantum wells and that there exists instead a significant k range where the hole "spin"-flip relaxation time is comparable to, or longer than, the recombination time. We recall that the "spin"-flip scattering in the lower heavy-hole branch HH<sub>1</sub> arises entirely from the admixture at finite k between the HH<sub>1</sub> and LH<sub>1</sub> band-edge states. It is thus natural that the suppression of "spin"flip scattering will be the more efficient in the narrower wells, provided, of course, that the assumption of a x strong quasi-two-dimensionality remains valid. Finally, we believe the suppression of hole "spin" flip should be taken into account in order to interpret the spinorientation experiments.

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