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Effects of interface asymmetry on hole subband degeneracies and spin-relaxation rates in quantum wells

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We consider the subband dispersions and the spin relaxation times for holes in semiconductor heterostructures without a common atom. We show that the interface asymmetry in such structures leads to a breaking of the parity degeneracy of the Luttinger dispersions. We calculate that the resulting spin splitting induces a D'yakonov-Perel' spin relaxation mechanism that can be up to five orders of magnitude faster than the one considered hitherto. [S0163-1829(97)52844-X]

It was recently realised that symmetry reduction due to interfaces can dramatically influence the optical properties of a broad class of semiconductor quantum wells (QW's).[1–4] In these so-called ''no common atom (NCA)'' heterostructures, such as InAs-GaSb, (InGa)As-InP, or (InAl)As-InP, both the well and barrier cations ($C1$,$C2$) and anions ($A1$,$A2$) are different. It was shown experimentally that in such QW's the absorption of light propagating along the [0,0,1] growth axis is strongly anisotropic with respect to the in-plane polarization direction,[1–3] and that this strong dichroism can be tuned by an axial electric field (this is the ''quantum confined Pockels effect'').[3] Although these effects are group-theoretically allowed by the reduced C_{2v} symmetry of NCA heterostructures, their large size is rather surprising, as they are strictly forbidden (i) in the classical envelope function theory (EFT) of QW structures,[5] and (ii) in the D_{2d} symmetry group corresponding to ''common anion'' QW's, which at first sight seem not so different. *Ab initio* tight-binding calculations[2] have established the intrinsic character of these effects, which are a genuine consequence of the different chemical bonding at subsequent interfaces: the left interface involves $C1A1$ bonds all lying in the $(-1,1,0)$ plane and $A1C2$ bonds lying in the perpendicular (1,1,0) plane, while the right interface consists of $C2A2$ bonds in the $(-1,1,0)$ plane and $A2C1$ bonds in the (1,1,0) plane. It is this interface asymmetry that reduces the QW point group from D_{2d} to C_{2v}. In the latter, the projection of the total angular momentum on the quantization axis J_z is not a good quantum number, even at the minizone center: any valence state has a mixed heavy-hole/light-hole character.[2] This result contrasts with the EFT, which is ''oversymmetric'' because it does not take into account the details of the interface bonding.

The interface effects can be included in the envelope function framework either through generalized boundary conditions[6] or in a perturbation scheme called the ''H_{BF} model'' (where H_{BF} stands for the perturbation Hamiltonian associated with the asymmetric interfaces).[3] The latter has the merit of very high flexibility, contains only two adjustable parameters (two interface potentials), and explains in a straightforward manner the observed optical anisotropy in terms of a k-independent coupling of the heavy- and light-hole states induced by the symmetry-breaking interface perturbation. Any term in the Hamiltonian coupling the $J_z = \pm\frac{1}{2}$ (light hole) and $J_z = \pm\frac{3}{2}$ (heavy-hole) states is expected to reduce the hole spin lifetime.[7] It is usually considered that the dominant mechanism for hole spin relaxation in symmetric QW's is an Elliot-Yafet type of mechanism where direct spin-flip collisions are allowed by the nondiagonal terms of the Luttinger Hamiltonian.[8,9] Here, we reexamine this question in the case of NCA QW's, using the H_{BF} model to account for the actual symmetry of the Hamiltonian. We prove that the symmetry breaking lifts the parity degeneracy of the hole subbands, and that the resulting k-dependent spin splitting induces a D'yakonov-Perel' spin relaxation mechanism that is several orders of magnitude faster than the Elliot-Yafet relaxation of parity degenerate states.

In what follows we will consider spin-flip and spin-conserving scattering events for holes in NCA QW's. We consider holes rather than electrons as the former are much more affected by H_{BF}. The numerical calculations will be done for InP-$In_{0.53}Ga_{0.47}As$. We shall limit our study to the energy-conserving scattering processes due to ionized impurities and alloy fluctuations. The latter mechanism is of particular importance for $In_{0.53}Ga_{0.47}As$ wells.

First we look at how H_{BF} changes the band structure as obtained within the EFT. We start from the same Hamiltonian as used in Ref. 8, i.e., the Luttinger Hamiltonian (H_{Lutt}) projected on the truncated basis spanned by the three lower bound states $H1$, $L1$, and $H2$ of the $\mathbf{k}=\mathbf{0}$ problem, where $\mathbf{k}$ is the two-dimensional in-plane wave vector. To this (6×6) Hamiltonian we add then H_{BF}; the rules of its projection are given in Ref. 3. Within the proposed basis $\{\chi_1\cdot H^+,\phi_1\cdot L^-,\chi_2\cdot H^-,\chi_1\cdot H^-,\phi_1\cdot L^+,\chi_2\cdot H^+\}$, where χ_1, ϕ_1, and χ_2 are the envelope functions of the first heavy hole, the first light hole, and the second heavy hole, and $H^{\pm}$ and $L^{\pm}$ the zone-center Bloch functions of the Γ_8 basis associated to the heavy and light holes, H_{BF} becomes

$$H_{\mathrm{BF}}=\begin{bmatrix} \Delta H_1 & A_{11} & 0 & 0 & 0 & B_{12} \\ A_{11}^* & \Delta L_1 & 0 & 0 & 0 & A_{12} \\ 0 & 0 & \Delta H_2 & B_{12} & A_{12} & 0 \\ 0 & 0 & B_{12} & \Delta H_1 & A_{11}^* & 0 \\ 0 & 0 & A_{12}^* & A_{11} & \Delta L_1 & 0 \\ B_{12} & A_{12}^* & 0 & 0 & 0 & \Delta H_2 \end{bmatrix}$$

with

$$\Delta H_1=(a_0/4)\{[\chi_1(z_1)]^2(V_2'-V_2)+[\chi_1(z_2)]^2(V_1'-V_1)\},$$

$$\Delta L_1=(a_0/4)\{[\phi_1(z_1)]^2(V_2'-V_2)+[\phi_1(z_2)]^2(V_1'-V_1)\},$$

$$\Delta H_2=(a_0/4)\{[\chi_2(z_1)]^2(V_2'-V_2)+[\chi_2(z_2)]^2(V_1'-V_1)\},$$

$$A_{11}=-ia_{11}=i(a_0/4\sqrt{3})[\chi_1(z_1)\phi_1(z_1)(V_2'-V_1)+\chi_1(z_2)\phi_1(z_2)(V_1'-V_2)]$$

$$A_{12}=ia_{12}=-i(a_0/4\sqrt{3})[\chi_2(z_1)\phi_1(z_1)(V_2'-V_1)+\chi_2(z_2)\phi_1(z_2)(V_1'-V_2)]$$

$$B_{12}=(a_0/4)[\chi_1(z_1)\chi_2(z_1)(V_2'-V_2)+\chi_2(z_2)\chi_1(z_2)(V_1'-V_1)].$$

These matrix elements [in which a_0 is the unit-cell parameter and $z_{1(2)}$ the coordinate of the left (right) interface] depend on the interface potentials $dV_1=V_1'-V_1$ and $dV_2=V_2'-V_2$ [see insert of Fig. 2(b)]. A second important characteristic is that they are **k** independent, so existing even at $\mathbf{k}=\mathbf{0}$. In the following we use $dV_1=-650$ meV and $dV_2=10$ meV, as these values lead to the same confinement energies (to within 2 meV) at $\mathbf{k}=\mathbf{0}$ as obtained by the tight-binding calculation of Ref. 2 for a 45 Å well. Optical anisotropy measurements in a variety of $In_xGa_{1-x}As$-InP QW's indicate values for $|dV_1+dV_2|$ in the 0.5–1 eV range.[10] Once a value is assigned to the interface potentials the total Hamiltonian can be numerically diagonalized. The resulting in-plane dispersions of an 80-Å-thick $In_{0.53}Ga_{0.47}As$ QW are shown in Figs. 1(a) and 1(b) with [Fig. 1(b)] and without [Fig. 1(a)] taking H_{BF} into account.

The Luttinger Hamiltonian alone gives rise to twofold spin-degenerate bands, which is a general characteristic for symmetric QW's. In this approximation one can associate for each **k** and for each subband n ($n=H_i$ or L_j) two wave functions, $\Psi_{u,n}$ and $\Psi_{d,n}$, with the degenerate energy eigenvalues $\varepsilon_{u,n}=\varepsilon_{d,n}$: $H_{\mathrm{Lutt}}\Psi_{u(d),n}=\varepsilon_{u(d),n}\Psi_{u(d),n}$. Analytical expressions for the Ψ_u and Ψ_d are given in Ref. 8. The mean values of the projection of the total angular momentum along the growth axis for these two states have opposite signs: $\langle\Psi_u|J_z|\Psi_u\rangle=-\langle\Psi_d|J_z|\Psi_d\rangle$ [at $\mathbf{k}=\mathbf{0}$, $\langle\Psi_u(\Psi_d)|J_z|\Psi_u(\Psi_d)\rangle=\frac{3}{2}\,(-\frac{3}{2})$ for the ground heavy states and $\frac{1}{2}\,(-\frac{1}{2})$ for the light states]. Therefore it is appropriate to associate these two degenerate levels to the ''up'' and ''down'' ''spin'' sublevels.

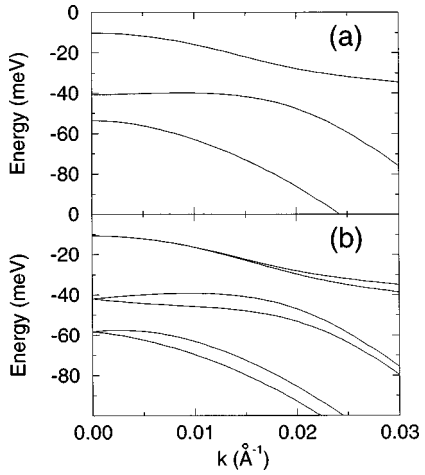

FIG. 1. (a) In-plane dispersions of the topmost valence subbands of an 80-Å-thick InP-$In_{0.53}Ga_{0.47}As$ quantum well, calculated within the Luttinger description. (b) Same as (a), but including H_{BF} in the Hamiltonian.

The most important effect of the asymmetry-related H_{BF} is the breaking of the parity degeneracy [Fig. 1(b)]. For small interface potentials an analytical expression for the energies can be obtained by considering H_{BF} as a perturbation on H_{Lutt} and by diagonalizing H_{BF} within the degenerate eigenstates Ψ_u and Ψ_d of H_{Lutt}. One obtains in a straightforward way that within this basis, to the first order of perturbation:

$$H_{\mathrm{tot}}=H_{\mathrm{Lutt}}+H_{BF}=\begin{bmatrix}\varepsilon_1 & \delta\\ \delta^* & \varepsilon_1\end{bmatrix} \tag{1}$$

with $\varepsilon_1=\varepsilon_u+\langle\Psi_u|H_{BF}|\Psi_u\rangle=\varepsilon_u+N^2(a^2\Delta H_1+\Delta L_1+\eta^2\Delta H_2+2aa_{11}\sin 2\theta)$. Here θ is the in-plane azimuth of the wave vector, and expressions for the **k**-dependent N, a, and η can be found in Ref. 8. The spin splitting is then given by $2|\delta|=4\eta N^2\sqrt{a_{12}^2+B_{12}^2+2a_{12}B_{12}\sin 2\theta}$. This formula gives the correct θ dependence (''warping''), also for higher orders of perturbation.

It should be noticed that the spin splittings we calculate here can be quite large (4 meV at $k=0.03$ Å^{-1} for the $H1$ band and up to 7 meV for the $L1$ and $H2$ band). Recently there has been considerable interest in the experimental and theoretical study of the spin splitting in GaAs-$Al_xGa_{1-x}As$ heterostructures.[11–14] The values we obtain here for NCA QW's are several times larger, and should therefore be detectable by spin resonance,[12] Raman scattering[11] or transport[13] measurements. We emphasize that interface asymmetry as the origin of spin splitting has not yet been discussed in the literature: this asymmetry lies somewhere in between the so-called bulk inversion asymmetry and structure inversion asymmetry.[14]

Next we consider the hole spin relaxation, both with and without taking H_{BF} into account. For valence states spin-orbit interactions are important, and nonmagnetic scattering centers can induce hole diffusions with both a change in spin state and orbital state ($\Delta\mathbf{k}\neq 0$): this is the Elliot-Yafet (EY) mechanism for spin flips.[8,9] Spin-conserving scatterings are also allowed; we denote, respectively, by τ_{sc} and τ_{sf} the **k**-dependent spin-conserving and spin-flip scattering times in

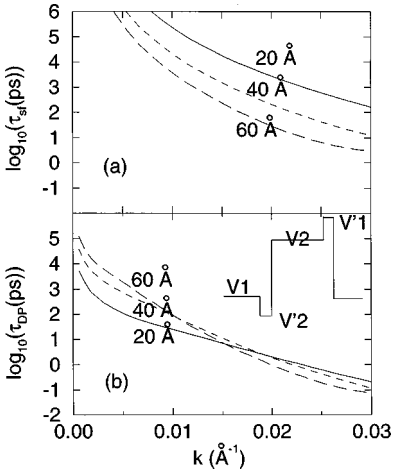

FIG. 2. (a) k dependence of the alloy-fluctuation-assisted spin-flip scattering time calculated within the Luttinger approximation for three quantum wells. (b) Same as (a), but with inclusion of H_{BF} in the theory. The insert represents the valence-band maximum in well and barrier, and defines the interface potentials as introduced in Ref. 3.

the Luttinger description. For scattering within the nth subband $\tau_{\mathrm{sf}}(\mathbf{k})$ is calculated under the Born approximation with Fermi's golden rule:

$$h/\tau_{\mathrm{sf}}(\mathbf{k})=2\pi\sum_{k'}\ |\langle\psi_{un\mathbf{k}}|V|\psi_{dn\mathbf{k}'}\rangle|^2\,\delta(\varepsilon_{un\mathbf{k}}-\varepsilon_{dn\mathbf{k}'}). \quad (2)$$

An analogous expression (involving two up eigenstates) yields $\tau_{\mathrm{sc}}(\mathbf{k})$. τ_{sf} as calculated by formula (2) is the EY scattering time. The matrix elements contain the scattering potentials (V) for diffusion induced by ionized impurities or alloy fluctuations.[5] Since τ_{sf} is directly linked to a measurable quantity if n coincides with the lower hole subband ($H1$), we restrict the calculations to this case.

Figure 2(a) shows the $\mathbf{k}$ dependence of the hole spin-flip relaxation times calculated within the Luttinger approximation [formula (2)]. The scattering which is considered for these results is the short-range alloy scattering; the calculations are done for wells with thicknesses = 20, 40, and 60 Å. As is a general property for symmetric QW's, there is a strong dependence of τ_{sf} upon k, i.e., on the band mixing: when $k=0$ the spin lifetime diverges (Ψ_u and Ψ_d are eigenstates of J_z at the zone center), while for finite k it strongly decreases due to band mixing, which makes EY spin-flip scattering allowed.

If one takes the whole Hamiltonian into account, this picture changes drastically. First, due to the energy splitting, it is not possible anymore to define up and down energy eigenstates. One readily proves that for any eigenstate Ψ of the total Hamiltonian the matrix element $\langle\Psi|J_z|\Psi\rangle$ equals zero for any $\mathbf{k}\neq\mathbf{0}$, which means that the eigenstates are completely depolarized. In this case one expects that a D'yakonov-Perel' mechanism may be operative for the hole relaxation. That this is indeed so follows from the fact that the total Hamiltonian (1) can be written under the form

$$H_{\mathrm{tot}}=\varepsilon_1\mathbf{1}+\frac{h}{2}\,\sigma\cdot\Omega, \quad (3)$$

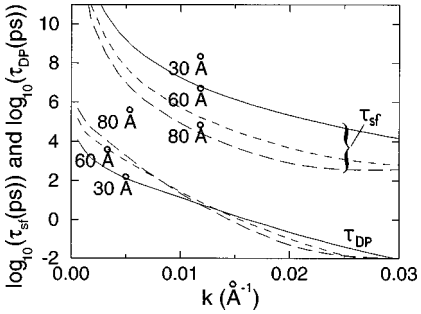

FIG. 3. Same relaxation times as in Figs. 2(a) and (b), but for impurity-assisted scattering.

with $\sigma=(\sigma_x,\sigma_y)$ (Pauli matrices) and $h\Omega/2=[\mathrm{Re}(\delta),-\mathrm{Im}(\delta)]$. D'yakonov-Perel' (DP) have shown[15] (originally for electrons) that due to the form (3) a very effective spin-flip mechanism exists. An additional condition, of which we have explicitly verified the validity, is that $h|\Omega|=2|\delta|\ll(h/\tau_p)$, where τ_p is a generalized momentum relaxation time that can be approximated by the spin-conserving scattering time τ_{sc}.[16,9] The latter condition expresses that the spin splitting should be much smaller than the level broadening induced by the various scattering mechanisms. The physical meaning of formula (3) is then the following: the up and down eigenstates of H_{Lutt} are no longer eigenstates of the total Hamiltonian and so the spin precesses between these two pure states in presence of an in-plane effective magnetic field $\mathbf{B}_{\mathrm{eff}}$ proportional to the $\mathbf{k}$-dependent Ω. In other words, the spin relaxation is governed by a precession of the spin around $\mathbf{B}_{\mathrm{eff}}$, the direction of which changes randomly and rapidly in a time of the order τ_p. The spin-flip scattering time should now be calculated using the expression[16,9] $1/\tau_{\mathrm{DP}}(\mathbf{k})=\frac{1}{2}\tau_p|\Omega|^2$. Figure 2(b) displays τ_{DP} calculated according to this formula, for the same QW's and scattering potential as in Fig. 2(a).

The point we want to stress here is that the spin relaxation that one calculates when including the interface asymmetry in the Hamiltonian is much faster than the one obtained for the unperturbed Hamiltonian. This is in accordance with the additional mixing of the wave functions induced by H_{BF}. The k range that matters for spin-polarization experiments, say, $k<0.01$ Å^{-1}, is also the k range where the DP condition $2|\delta|\ll h/\tau_p$ is best fulfilled. In this range τ_{DP} is about 40 times smaller than τ_{sf} for a 60 Å well, while for a 20 Å well the difference is more than three decades. For sufficiently large k, τ_{DP} becomes smaller than typical electron hole recombination times of a few hundreds of ps, which means that the depolarization should be detectable by optical polarization experiments.

Even more salient results are obtained when considering scattering on ionized impurities. In Fig. 3 the spin relaxation times for this diffusion mechanism are shown for a 30, 60, and 80 Å well. In the calculations we assumed the scatterers to be localized at one interface. We used an impurity density of 10^{10} cm^{-2} and a screened Coulomb potential with Bohr radius of 100 Å.[5] Both the relaxation times with and without inclusion of H_{BF} are represented. Again one observes a huge difference between the two results: for the 30 Å well it is

more than five decades. This clearly establishes the importance of the interface asymmetry in the spin relaxation of NCA QW's.

Another remarkable result is that if one makes full account of the interface asymmetry one obtains spin-flip relaxation times which decrease with decreasing well thickness [Figs. 2(b) and 3], at least in the k range of importance. One is used to precisely the opposite tendency in usual (common anion) QW's [Ref. 8 and Figs. 2(a) and 3]. On the other hand, it seems logical that the effect of the interface-related H_{BF} is the stronger the thinner the well. We believe that spin polarization experiments (e.g., of the type considered in Ref. 7) in which one compares NCA QW's with different well thicknesses would provide a decisive test of our model. An important depolarization in a thin (say 30 Å thick) $In_{0.53}Ga_{0.47}As$-InP QW would be a clear-cut signature of the interface asymmetry that we have studied here.

In conclusion, we have shown that the hitherto considered models for hole spin relaxation have omitted an asymmetry-related term in the Hamiltonian that very efficiently depolarizes the hole states in NCA structures. This term is also responsible for a new type of spin splitting of the bands, and its effect is the stronger the larger the ratio of the number of interface atoms to the number of bulk atoms.

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