Rashba spin-splitting of electrons in asymmetric quantum wells

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We report a comparison of conduction electron spin-splitting in III-V quantum wells caused by asymmetric band edges with that due to applied electric field. Measurements in GaAs/AlGaAs quantum wells and calculations on a range of heterostructures, both symmetric and asymmetric, lead to the conclusion that in a heterostructure with nearly "isomorphous" band edges (i.e., with conduction and valence band-edge potentials related by a constant factor, exemplified by GaAs/AlGaAs) spin splittings will be unmeasurably small even in a highly asymmetric structure. Application of an external electric field or the presence of a Hartree potential gradient in the system will generally break isomorphism and therefore produce a significant spin splitting.

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I. INTRODUCTION

An electron moving in an electric field experiences an effective magnetic field, which removes the spin degeneracy. In semiconductor heterostructures[1,2] such velocity-dependent spin splittings can provide an important degree of freedom in design of spintronic devices where the electron spin is manipulated by external gate voltage.[3] Symmetry arguments indicate that not only electric field but also any breaking of the macroscopic inversion symmetry in a heterostructure, can in principle, generate a spin-splitting known as a Rashba or structural inversion asymmetry (SIA) splitting. This has, for example, led to the belief that asymmetric alloy composition may be used in spintronics to supplement or mimic the effect of an external gate voltage.[4]

The magnitude of Rashba (SIA) splittings has been intensely debated [see Ref. 1 and references therein] and continues to produce surprises, as demonstrated below. For example experiment shows[5] that spin-splitting, in external electric field $\mathbf{F}$, is generally orders of magnitude greater than expected as a result of the effective magnetic field, $(v/c^2)\mathbf{F}$, seen by an electron moving with a typical (thermal or Fermi) velocity, v.[1] This is due to the fact that spin splitting reflects the interplay of both the ion-core and macroscopic structural potentials. It is yet more perplexing that, apparently contradicting the symmetry arguments, Ehrenfest's theorem requires that, for a bound electron state, the expectation value of the potential gradient must vanish, suggesting that spin splitting due to any asymmetry of the confining potential should vanish.[6] This apparent contradiction was discussed theoretically by Lassnig[1,7] who showed that Ehrenfest's theorem applies only to the motion of the electron in the conduction band potential while spin splitting of the electron states is associated with the potential gradient in the valence band.

Here, we investigate the counterintuitive nature of Rashba (SIA) spin-splitting in quantum wells from a new perspective. We compare the magnitude of spin-splitting induced purely by built-in alloy asymmetry with that induced by applied electric field in III-V heterostructures. To make this comparison experimentally we use undoped quantum well structures which contain no free carriers and we determine spin splittings using a combination of optical techniques[5] which involve excitation of low free carrier populations. Consequently the only electric fields involved are externally applied through voltage bias and are not caused by movement of free carriers in the system as for example in modulation-doped structures. In the latter it is difficult to separate the effects of alloy asymmetry from those of electric field.[2] For undoped asymmetric structures one might expect substantial spin-splittings as the built-in potential gradients in the conduction or valence band can readily be made to correspond to an electric field of 100 kV cm^{-1} or more and such a field applied externally is known to give a clearly observable spin-splitting.[5] Surprisingly, our measurements in just such an asymmetric quantum well structure, grown from GaAs/AlGaAs, give an unmeasurably small spin-splitting.

We conclude that this is a consequence of the peculiar nature[1,7] of SIA spin-splitting that may be generalized to the statement that, in a heterostructure where the valence and conduction band edge potential profiles are isomorphous as defined below, Rashba (SIA) splitting will be very small even if the potential is highly asymmetric. By isomorphous we mean that the valence and conduction band edge potential profiles are related by a constant proportionality factor; thus $\partial V_v(z) = \Sigma . \partial V_c(z)$ where $\partial V_v(z)$ and $\partial V_c(z)$ represent the changes of valence and conduction band edge potentials with respect to their values at some arbitrary position along the growth (z) direction and Σ is a constant independent of z. In the absence of any electric field the band edges of heterostructures grown from many material systems, particularly lattice-matched ternary alloys, are quite close to isomorphous in this sense because the ratio of valence to conduction band offset is approximately independent of the composition of the alloy; thus, even if there is a strongly asymmetric spatial variation of the alloy composition the band-edge profiles have an approximately constant ratio. In such material systems it is impossible, although permitted according to the symmetry arguments, to engineer a signifi-

cant spin-splitting by simply growing a structure with asymmetric alloy composition. On the other hand, when an electric field is applied to a heterostructure, which contains no free charges, an equal tilt is imparted to the conduction and valence bands and this automatically breaks their isomorphism and induces a significant spin splitting. Similarly, in a structure which contains free charges, for example a modulation-doped structure, there may be a Hartree potential gradient which will again impart a tilt to both bands and so break the isomorphism of the band edges and give a significant spin splitting. We present detailed calculations of spin splitting in a range of systems which verify these assertions.

II. MEASUREMENTS

The spin-splitting for an electron with momentum $\hbar\mathbf{k}$ can be specified by a vector $\mathbf{\Omega}(\mathbf{k})$ representing the Larmor precession in the effective magnetic field seen by the moving electron. Our measurement technique exploits the fact that relaxation of a spin-polarized population of electrons usually occurs via the Dyakonov-Perel (DP) mechanism[8,9] and the spin-relaxation rate for spins oriented along an axis, i, is given by

$$\tau_{s,i}^{-1} = \tau_p^* \langle \Omega_\perp^2 \rangle, \tag{1}$$

where τ_p^* is the momentum scattering time of an electron and $\langle \mathbf{\Omega}_\perp^2 \rangle$ is the mean square component of $\mathbf{\Omega}(\mathbf{k})$ perpendicular to the axis i averaged over the electron momentum distribution. We measure, separately but under the same conditions, the spin-relaxation rate, $\tau_{s,z}^{-1}$, along the growth axis, z, by time-resolved optical Kerr rotation[10] and the scattering time, τ_p^*, by a time-resolved optical spin-grating technique.[11,12] The ratio of these quantities gives, the mean squared component of $\mathbf{\Omega}(\mathbf{k})$ in the quantum well plane. Rashba spin-orbit coupling for asymmetry along the growth axis corresponds to an in-plane component of $\mathbf{\Omega}(\mathbf{k})$ (e.g., $\mathbf{\Omega}_{\mathrm{SIA}}(\mathbf{k}) \sim \mathbf{F} \times \mathbf{k}$ for electric field $\mathbf{F}$) and thereby increases the spin-relaxation rate.

In zincblende structure semiconductors, in addition to any Rashba spin-splitting, $\mathbf{\Omega}_{\mathrm{SIA}}(\mathbf{k})$, which will be oriented in the quantum well plane [Figs. 1(a) and 1(b)], there is an intrinsic component of spin-splitting, $\mathbf{\Omega}_{\mathrm{BIA}}(\mathbf{k})$, due to the underlying crystal structure (bulk inversion asymmetry, BIA);[1,9] the total vector is $\mathbf{\Omega}(\mathbf{k}) = \mathbf{\Omega}_{\mathrm{SIA}}(\mathbf{k}) + \mathbf{\Omega}_{\mathrm{BIA}}(\mathbf{k})$. In standard (100)-oriented quantum wells [Fig. 1(a)], $\mathbf{\Omega}_{\mathrm{BIA}}(\mathbf{k})$ also lies in the quantum well plane and as a result the spin-relaxation along the z axis is relatively short (<100 ps at room temperature) and insensitive to the Rashba component. By contrast, for (110)-oriented quantum wells [Fig. 1(b)], symmetry dictates that $\mathbf{\Omega}_{\mathrm{BIA}}(\mathbf{k})$ lies along the growth axis (z), [13]and therefore, plays no part in spin relaxation along the z axis; in principle, the only contribution to the DP relaxation of spin along the z axis comes from the Rashba component. For symmetrically grown quantum wells with macroscopic mirror symmetry, $\mathbf{\Omega}_{\mathrm{SIA}}(\mathbf{k})$ should be zero and the DP mechanism should be totally suppressed. This is indeed found to be the case; spin-relaxation times as long as 20 ns are observed at room temperature and result from a mechanism other than DP.[14–16] When an electric field F_z is applied, the linear dependence of $\mathbf{\Omega}_{\mathrm{SIA}}(\mathbf{k})$ on F_z should result in an increase of the spin-relaxation rate as F_z^2 according to Eq. (1). Any additional spin-splitting induced, for example, by built-in asymmetry could also be detected as a field-independent contribution to the spin-relaxation rate. When the DP mechanism is suppressed, allowing an alternative mechanism to take over, insertion of the measured values of $\tau_{s,z}$ and τ_p^* into Eq. (1) will yield an apparent r.m.s. spin splitting, which is an *upper* limit for the true spin-splitting.

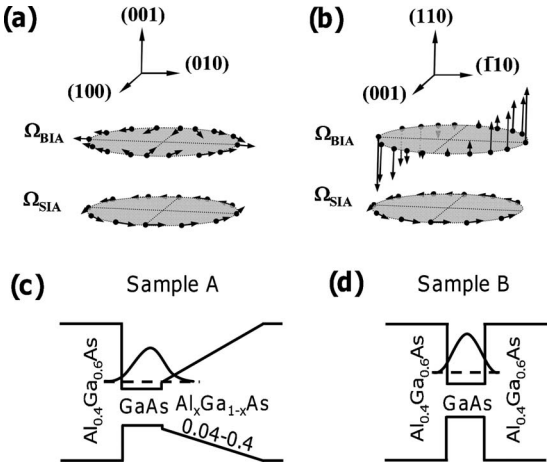

FIG. 1. (a) and (b), illustrating the BIA and Rashba (SIA) components of spin-orbit splitting vector for electrons in (001)- and (110)-oriented quantum wells respectively, as a function of wave vector, $\mathbf{k}$, distributed on the perimeter of a disk in the quantum well plane; the growth axis (z) is vertical. To first approximation, the magnitude of the vector increases linearly with $|\mathbf{k}|$. (c) and (d), band potential profiles and schematic electron probability density for samples A and B respectively.

We present measurements for three different multi-quantum-well samples, grown by molecular beam epitaxy on semi-insulating (110)-oriented GaAs substrates. Samples A and B are completely undoped with asymmetric and symmetric alloy composition respectively. Sample A [Fig. 1(c)] is made up of 5 repeats of 8 nm GaAs quantum wells each with a 30nm graded upper interface where the aluminum fraction, x, is varied from 0.04 to 0.4, followed by a 12 nm barrier. Sample B [Fig. 1(d)] comprises 20 repeats of 7.5 nm GaAs quantum wells with abrupt 12 nm $Al_{0.4}Ga_{0.6}As$ barriers. Sample C is a *p-i-n* device structure with undoped symmetrical quantum wells nominally identical to those of sample B grown in the insulating (i) region, so that an external electric field F_z can be applied to them; maximum field is 80 kV cm^{-1}, limited by increasing (avalanche) photocurrent in reverse bias.[5,16]

Measurements were made of the spin-relaxation rate $\tau_{s,z}^{-1}$ from time-resolved Kerr rotation and the electron scattering time τ_p^* from the spin-grating decay at temperatures between 80 and 300K in all the samples. For sample C $\tau_{s,z}^{-1}$ was also measured for values of applied electric field up to 80 kV cm^{-1}. The optically excited carrier density in the measurements was kept sufficiently low that the effect of any Hartree potential gradients due to the photoexcited carriers can be neglected. Figure 2(a) illustrates the spin decay along the growth axis from the Kerr rotation technique at 200K.[10] The spin-grating measurements[12] showed that all three

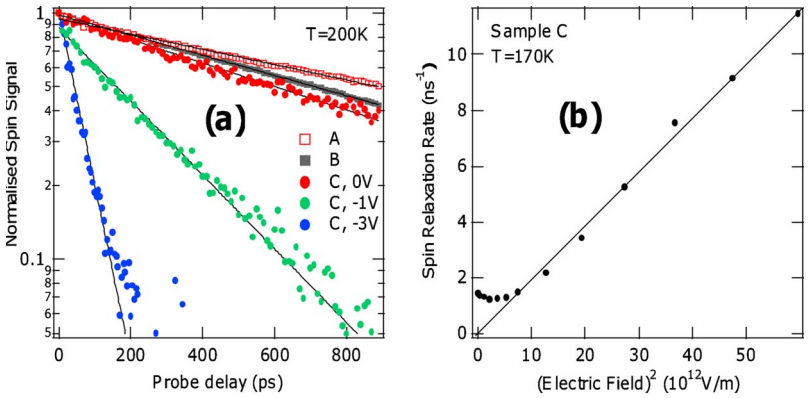

FIG. 2. (Color online) (a) Decay of spin polarization from time resolved Kerr rotation measurements at 200 K in samples A (decay time 1397 ± 12 ps) and B (1058 ± 10 ps) and for sample C with three different bias voltages (0 V, 914 ± 13 ps; −1V, 290 ± 8 ps; −3V, 62 ± 5 ps). (b) Variation of spin-relaxation rate with square of electric field in sample C at 170K.

samples have very similar values of momentum relaxation time, τ_p^*, and temperature dependence between 70K and 300K close to $1/T$.[17] Thus, the spin decay rates at a given temperature give an immediate indication of the relative magnitudes of the mean square spin-splittings for different samples. Figure 2(a) shows that in both the symmetric (B) and the asymmetric (A) quantum wells the spin-relaxation rates are <1 ns^{-1}, indicating that the asymmetry in A does not produce a large increase of the spin splitting. By contrast, applied electric field has a profound effect; sample C with zero bias voltage (corresponding to $F_z=25$ kV cm^{-1}) has spin decay rate almost as low as in samples A and B but at −3V bias ($F_z=80$ kV cm^{-1}) it increases more than 10-fold to ~ 16 ns^{-1}.

Figure 2(b) shows the spin-relaxation rate in sample C as a function of the square of applied electric field (F_z) at 170 K; at high fields the variation follows a straight line extrapolating to the origin while below 30 kV cm^{-1} it flattens out and becomes almost constant. The same form of variation was observed at all temperatures from 80 K to 230 K.[5] In agreement with Eq. (1), the quadratic dependence on the electric field unambiguously indicates dominance of the DP spin-relaxation mechanism, the spin-splitting being linear in field; under these circumstances the true value of the spin-splitting is given by the ratio of spin-relaxation rate to the scattering time [Eq. (1)]. The fact that the variation with F_z^2 extrapolates to the origin demonstrates that there is no field-independent contribution to the spin-splitting, as one expects in this sample which has symmetrical quantum wells.

Figure 3 shows the apparent r.m.s. Rashba spin-splittings, obtained for the different samples between 80 and 300K by combining spin relaxation and scattering time data. In sample C at 80 kV cm^{-1} the splitting increases approximately linearly with temperature. The dotted curve shows the calculated variation for a Boltzmann distribution of electron energies and temperature-dependent electric-field-splitting coefficient, as discussed in Ref. 5. In sample C with zero field applied and also for samples A and B it appears that the DP mechanism is so weak that an alternative mechanism, most likely Bir-Aronov-Pikus[14,15] has taken over; the weak dependence on temperature as shown in Fig. 3 and the fact that the spin relaxation rates and, hence, the apparent spin-splittings increase significantly with excitation density (not shown here, see Ref. 10) are both incompatible with the DP mechanism. The apparent spin-splittings for these cases therefore set an *upper* limit on the true spin-splittings. The striking fact is that sample A which has asymmetric quantum wells and potential gradients in conduction and valence band of order 50–100 kV/cm has r.m.s. spin-splitting less than 40 μeV.

III. THEORETICAL INTERPRETATION

While the vanishing of spin-splitting for a highly asymmetric heterostructure such as sample A seems counterintuitive when modest electric field induces a large splitting in a symmetrical structure such as C, it can nonetheless be understood from the following argument based on a multiband analysis. Spin-splitting induced by inversion asymmetry reflects the coupled motion of electrons and holes.[1,7] When these motions are decoupled, e.g., by means of a unitary transformation, the Hamiltonian acting in the subspace of the electron states acquires an extra term, which is exactly the Rashba form

$$H_R = \alpha\, \partial V_v (\sigma_x k_y - \sigma_y k_x) \tag{2}$$

where σ_x and σ_y denote Pauli spin matrices, and ∂V_v is the z-component of the gradient of the potential of the valence

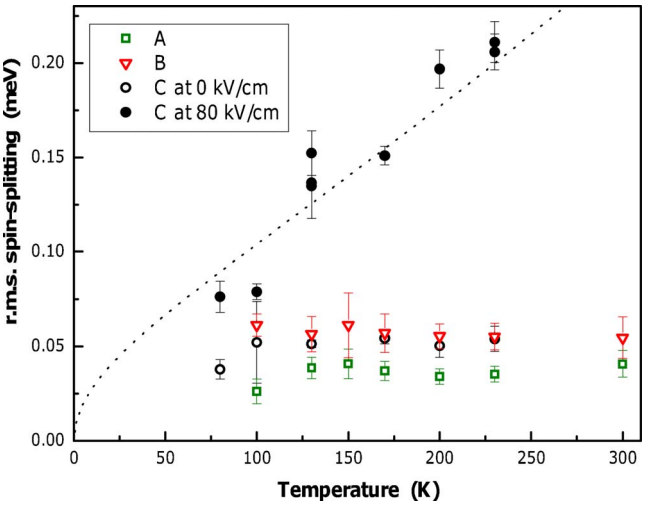

FIG. 3. (Color online) Apparent r.m.s spin splitting for samples A, B and C obtained from time resolved Kerr and spin-grating measurements using Eq. (1). For A, B, and for C in zero electric field the measurements give an upper limit. For sample C at 80 kV cm^{-1} the points are a true measure of the splitting.

band V_v. This decoupling is analogous to the derivation of the Pauli spin-orbit coupling from the Dirac equation and it results in terms that have the same form,[1,18] though for Dirac particles it is usually not meaningful to consider the possibility that particles and antiparticles see different potentials. The prefactor α is material specific, but is independent of the geometry of an individual sample.[1] The important point is that the spin splitting for electrons in the conduction band is given by the expectation value of the gradient of the potential in the *valence* band $\langle \partial V_v \rangle$ between the conduction band states not that of the conduction band gradient, $\langle \partial V_c \rangle$.

In general, V_v and V_c depend only on the z coordinate and are each made up of two contributions,

$$V_v(z) = V_{v,int}(z) + V_H(z) \quad \text{and} \quad V_c(z) = V_{c,int}(z) + V_H(z), \tag{3}$$

where $V_{v,int}(z)$ and $V_{c,int}(z)$ are internal contributions which reflect the position-dependent band edges in the sample, while $V_H(z)$ includes the Hartree potential from the charge distribution in the system and any externally applied voltage. It is important to note, first, that $V_H(z)$ contributes equally to both the conduction and valence band potentials and, second, that the potentials $V_{v,int}(z)$ and $V_{c,int}(z)$ are often isomorphous (or nearly so), meaning that the position-dependent gradients of the conduction and valence band edges have a constant ratio,

$$\partial V_{v,int}(z) = \Sigma \,.\, \partial V_{c,int}(z), \tag{4}$$

where Σ is approximately constant. This is true in many alloy systems where the ratio of the conduction to the valence band offset is nearly independent of alloy composition. In direct-gap GaAs/AlGaAs, for example, $\Sigma \sim -0.54$.[19] (Note also that the condition $\Sigma = 1$ corresponds to a position-independent band gap and therefore absence of heterojunctions in the structure). According to Ehrenfest's theorem the expectation value of the gradient of the conduction band potential must vanish, reflecting the fact that no net force can act on an electron in a bound state, so that $\langle \partial V_c \rangle = \langle \partial (V_{c,int} + V_H) \rangle = 0$ and

$$\langle \partial V_{c,int} \rangle = -\langle \partial V_H \rangle. \tag{5}$$

This must be true for symmetric or asymmetric structures; the electron wave function will simply adjust, becoming more or less asymmetric, to ensure the equality Eq. (5). This point can be proved by integrating the Schrödinger equation. On the other hand spin splitting of the electron states is determined, to first order, by the expectation value of the gradient of the valence band potential, which may be written using Eq. (4) as

$$\langle \partial V_v \rangle = \langle \partial (V_H + V_{v,int}) \rangle = \langle \partial V_H \rangle + \Sigma \langle \partial V_{c,int} \rangle \tag{6}$$

Then combining Eqs. (5) and (6), to first order, the spin-splitting is proportional to

$$\langle \partial V_v \rangle = (1 - \Sigma) \langle \partial V_H \rangle. \tag{7}$$

In a case where Σ is a constant, this shows that the splitting will be proportional to $\langle \partial V_H \rangle$ and so will be zero unless there is an applied voltage and/or gradient of the Hartree potential.

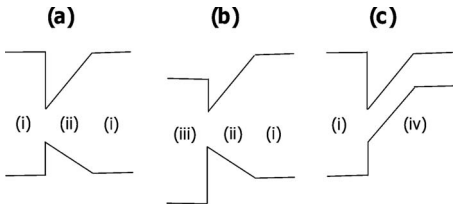

FIG. 4. Schematic band potential profiles of three structures used for full calculations of conduction band spin splitting in the $InP/In_{0.47}(Ga_xAl_{1-x})_{0.53}As$ material system; (i) indicates $x=0$, (ii) x graded linearly from 1 to 0 over 35 nm and (iii) InP. (iv) has $x=1$ together with a theoretical charge distribution to give the potential profile shown.

It also suggests that in structures which have no free charges or external bias to generate a Hartree potential gradient, there should be no Rashba (SIA) spin splitting even if the confining potential defined by the alloy composition is highly asymmetric. In real alloy systems the band offset ratio and therefore Σ is usually composition dependent to some degree due to different bowing of the conduction and valence band edges but our detailed calculations described below indicate that the resulting spin splittings are often relatively small.

To support this simplified argument, we present results of self-consistent numerical calculations for six quantum well potentials in real material systems based on diagonalization of the 8×8 Kane Hamiltonian; the theoretical framework and technical details are given in Ref. 1. These accurate and realistic calculations take into account SIA and BIA spin splitting to all orders in $\mathbf{k}$. They include variations (with bowing) of band parameter values (energies, effective masses, etc.) with alloy composition.[19] From the numerically calculated sub-band dispersion curves we obtain the spin sub-band densities $N_{\pm}$ by means of analytic quadratic Brillouin zone integration.[20] Finally, we obtain explicit expressions for the Rashba coefficients using the relation

$$\alpha \langle \partial V_v \rangle = \sqrt{\pi} \langle \hbar^2 / \mu_c \rangle \{ \sqrt{N_+} - \sqrt{N_-} \}, \tag{8}$$

where μ_c is the effective mass.

We first consider three structures in alloy systems where the conduction and valence bands are approximately isomorphous:

(1) For the GaAs/AlGaAs structure of sample A [Fig. 1(c)] we obtain $\alpha \langle \partial V_v \rangle = 0.91$ μeV.nm corresponding to r.m.s spin-splitting at 300K of 0.2 μeV. The value is non-zero which is in agreement with symmetry arguments, but it is extremely small compared to the sensitivity of our measurements (cf. Fig. 3).

(2) A similar, unmeasurably small value was calculated for a graded GaAs/AlGaAs structure like sample A but without the 8 nm wide region where $x=0$.

(3) For a graded well consisting of a 35 nm layer of the lattice-matched alloy $In_{0.53}(Ga_xAl_{1-x})_{0.47}As$, see Fig. 4(a), having barriers with $x=0$ and with x varying linearly from 1 to 0 across the well we find $\alpha \langle \partial V_v \rangle = 65.6$ μeV.nm. In this system Σ is approximately independent of x and is ~ -0.37,[19] giving nearly isomorphous band edges; the calculated spin splitting corresponds to an r.m.s. value of 14.8 μeV at 300K

and is again very small compared to our measurement sensitivity.

For each of the above three structures the potential gradients in the conduction and valence bands correspond to of order 100 kV cm^{-1} yet the spin splitting is far less than we have measured with a similar externally applied electric field.

Consider next calculated spin splittings in three structures where the band edges are not isomorphous either because the band offset ratio Σ depends on position z or because a Hartree potential gradient is present:

(4) For a graded well as depicted in Fig. 4(b), i.e., similar to Fig. 4(a) but with an InP (rather than $In_{0.53}Al_{0.47}As$) barrier on the left side of the well, we calculate $\alpha\langle\partial V_v\rangle = 7130$ μeV.nm. This is a lattice-matched structure but, since for the InP/$(In_{0.53}Ga_{0.47})$As interface $\Sigma \sim -1.31$,[19] the band edges are not isomorphous and indeed, compared to Fig. 4(a), the calculated r.m.s. spin splitting at 300K is increased dramatically to 1.61 meV and would be easily measurable.

(5) For a hypothetical GaAs/AlGaAs structure with the same conduction band profile as sample A but with the valence band running parallel to the conduction band in the graded region we calculate $\alpha\langle\partial V_v\rangle = 600$ μeV.nm. This structure corresponds to the right hand barrier in Fig. 1(c) having a constant alloy composition, $x=0.04$, plus a distribution of charges to give a Hartree potential gradient equivalent to the conduction band gradient in sample A. The calculated r.m.s. splitting of 135 μeV at 300K would again be readily measurable.

(6) For a second hypothetical structure, shown in Fig. 4(c), based on a single $InGa_{0.53}As/InAl_{0.47}As$ heterojunction engineered by incorporation of a Hartree potential to give a 35 nm triangular well in the conduction band $\alpha\langle\partial V_v\rangle = 8160$ μeV.nm giving an easily detectable r.m.s. spin splitting of 1.84 meV at 300 K.

IV. CONCLUSIONS

We conclude that lack of mirror symmetry along the growth axis of a heterostructure does not necessarily cause significant Rashba spin-splitting for conduction electrons although such a splitting is allowed on symmetry arguments; the spin-splitting will be very small in situations where the band edges are isomorphous (i.e., their potentials are related by a constant factor). Thus in many material systems (including ternary alloys exemplified by GaAs/AlGaAs) strongly asymmetric alloy composition does not, by itself, generate a significant Rashba spin-splitting. The isomorphous relationship of the band edges can be broken to give a significant spin splitting by the presence of an electric field due to external applied voltage or from an asymmetric distribution of charges (Hartree potential gradient). To engineer a spin splitting in a structure without an electric field the isomorphous relationship of band edges can most readily be broken by using an asymmetric quaternary alloy system such as example (4) above. Our theoretical arguments also show that the presence of heterointerfaces is required for a significant Rashba-type splitting, i.e., Σ must differ from unity in Eq. (3). Thus, for example, we expect that *n-i-p-i* doping structures,[21] in which the potential wells are defined by inbuilt Hartree fields alone, should show no significant Rashba splitting either for applied bias voltage or even if the doping profile lacks inversion symmetry. These results clarify longstanding fundamental questions concerning the spin-orbit coupling in heterostructures as well as helping to define the ground rules for the design of spintronic devices.

ACKNOWLEDGMENTS

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