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Conduction-band spin splitting of type-I $Ga_xIn_{1-x}As$/InP quantum wells

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The spin-splitting factor g^* of the electrons at the very bottom of the conduction band in strained $Ga_xIn_{1-x}As$/InP type-I quantum wells is reported. Experimental proof of quantum confinement-dependent anisotropy of g^* is given. Changing the alloy composition at fixed quantization, equivalent to introducing compressive and tensile strain, changes g^*. The values of g^* perpendicular to the quantum-well plane can be explained in a model calculation. Apparently however, no quantitative theory on which to base the calculation of the anisotropic spin splitting is at present available.

Knowledge of the structure of the electronic bands in semiconductors is both of fundamental interest and of importance for the realization of devices based on transport and optical properties. The band structure is well known for most common bulk semiconductors, but for low-dimensional structures (quantum wells, quantum wires, quantum dots) the details are still not understood. The aim is to understand the band structure of low-dimensional systems to such a precision, that theoretical modeling of arbitrary materials and geometries can be performed. In order to realize this, fundamental parameters have to be measured and understood. A band-structure parameter representing a high degree of precision is the effective spin-splitting factor, g^*.

The Landé spin-splitting factor of a free electron, $g = 2.0023$, describes the evolution of spin-up and spin-down electronic levels in a magnetic field. In solids, the free-electron value is changed due to the interaction of the electron with the lattice potential. Depending on the crystal, this interaction can change the spin-splitting factor from very large positive to very large negative numbers. This effective spin splitting g^* can be calculated within most theoretical models. Accurate experimental determination of g^* is, therefore, of fundamental importance in solid-state physics since it provides a sensitive test of band-structure calculations and theoretical concepts in general.[1–3]

Experimental determination of g^* at the band edge has been the subject of numerous investigations over the years. In semiconductor physics, the most precise measurements of the free carrier g^* values have been performed using magnetic resonance techniques. Today, such data are tabulated for most elementary, binary, and, in a few cases, ternary bulk semiconductors.[4] Measurements of g^* values in two-dimensional electron gas (2DEG) systems and quantum-well (QW) structures have also been attempted, using direct as well as indirect techniques.[5–14] Such measurements are, however, complicated by the low signal intensity (small sample volume) in the case of undoped samples, or by dominant, obscuring effects (many-body effects, screening, exchange interaction, etc.) in samples with high electron concentrations. In particular, most attempts to use the optically detected magnetic resonance (ODMR) technique on undoped QW's have failed, the reason being that the radiative recombination times in the type-I QW's (direct in space, the most common type) occurs on a nanosecond time scale. A prerequisite to observe ODMR is the introduction of spin transitions within the lifetime of the decaying system. For the standard cw-ODMR technique, lifetimes must be longer than 0.1 μs. Therefore, it is not surprising that the few, existing experimental results are obtained on type-II QW's or superlattices.[6,10,11,13] Here the electrons and holes are separated in real space. This separation of the carriers leads to recombination times in the microsecond range.

In type-I QW's longer lifetimes have been observed in contacted samples to which an electric field perpendicular to the QW plane was applied.[15] In this paper we will demonstrate, using the $Ga_xIn_{1-x}As$/InP system, that single-sided, p-modulation doping can play an equivalent role in reducing the radiative recombination, thus allowing successful spin resonance measurements of electron g^* values in type-I QW's. The results show clear dependences of g^* on the degree of quantum confinement and on the amount of strain. The data also show a strong, confinement-induced anisotropy of the electron g^* value. A comparison with theoretical calculations calls for further work to be able to establish a refined model of band structures in quantum-confined systems.

The $Ga_xIn_{1-x}As$/InP single QW samples used in this investigation have been grown by low-pressure metalorganic vapor-phase epitaxy at 620 °C on semi-insulating InP:Fe as described elsewhere.[16] The QW's are grown either lattice matched ($x_{Ga} = 0.47$) with varying QW thickness (d), or with varying composition ($0.4 < x_{Ga} < 0.6$) but fixed thickness ($d = 15$ nm). All QW's have a thickness which is below the critical thickness for strain relaxation. The structure is the same for all samples: the QW is grown on top of a 400-nm InP buffer layer followed by a 5-nm spacer and a single-sided p-modulation doping, a 10-nm layer with an acceptor concentration of $1\text{–}2\times10^{18}$ cm^{-3}, and finally a 60-nm capping layer. The quality, and in particular the thickness and the composition, of the QW's were investigated by photoluminescence (PL), magneto-PL, and x-ray diffraction. Hall effect measurements showed hole mobilities of 100–200 cm^2/V s at 77 K.

A PL spectrum from a $Ga_xIn_{1-x}As$/InP QW (x_{Ga}=0.47, $d = 15$ nm) shows a single line, peaking at 0.83 eV and with full peak width a half maximum of 8 meV. The emission is dominated by the recombination between the first electron and heavy-hole subbands (e_1hh_1). The peak position is slightly shifted from that observed in symmetrical QW's due to the electric field ($\approx 10^5$ V/cm) (Ref. 17) induced by the single-sided modulation doping. Qualitatively the same PL results are observed for all the other compositions and QW thicknesses.

The magnetopolarization properties of the emission were investigated by illuminating the samples with unpolarized laser light (514.5 nm, < 0.3 W/cm^2), and analyzing the right- ($I_{\sigma-}$) and left- ($I_{\sigma+}$) circularly polarized components of the emission (Faraday configuration). The magnetic field and temperature dependences of this magnetic circularly polarized emission (MCPE) ($= I_{\sigma+} - I_{\sigma-}$) signal showed an unequal occupation of the Zeeman-split states. The degree of polarization, which did not have a perfect Boltzmann distribution, was typically a few percent. The ODMR was observed as a decrease in the MCPE signal at the magnetic field at which the microwaves partially equalize the difference in spin occupation according to $\Delta E_{\mu w} = \mu_B g^* B$ ($S = \frac{1}{2}$, μ_B is the Bohr magneton). The ODMR experiments were performed at $T = 1.6$ K in a 4-T magneto-optical system using 24- and 36-GHz microwaves. As an example, the experimental result obtained with the magnetic field parallel to the normal of the $Ga_{0.47}In_{0.53}As$/InP ($d = 15$ nm) QW (100) plane is shown in Fig. 1. The resonance signal has a Lorentzian line shape, characteristic for homogeneous broadening and $g_{\parallel}^* = -3.27 \pm 0.04$. The sign of the g value was determined by analyzing the polarization as a function of temperature and magnetic-field strength and direction. Rotating the sample through an angle θ from $B||\langle 001\rangle$ towards $B||\langle 110\rangle$ results in a significant change of the resonance position as shown in Fig. 1 (inset). The data are analyzed using the standard expression for a g tensor in axial symmetry, $g^*(\theta) = (g_{\parallel}^{*2}\cos^2\theta + g_{\perp}^{*2}\sin^2\theta)^{1/2}$, and a best fit (solid line) is obtained using $g_{\perp}^* = -1.88 \pm 0.04$. A rotation from $B||\langle 001\rangle$ towards $B||\langle 100\rangle$, gave the same result, indicating that the g^* value is isotropic with B in the QW plane.

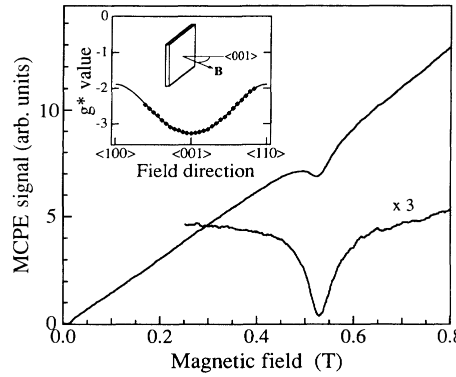

FIG. 1. ODMR (24 GHz) detected on the magnetic circularly polarized emission signal of a 15-nm $Ga_{0.47}In_{0.53}As$/InP single QW at $T = 1.6$ K. In the blowup the background magnetization has been subtracted. The inset shows the anisotropy of the effective g factor. Solid circles are experimental data and the solid line is a fit using the expression for axial symmetry (see text).

The strained samples with different compositions in the QW ($d = 15$ nm) showed substantial changes of the $g_{\parallel}^*$ and the $g_{\perp}^*$ values (see Fig. 2). Both $g_{\parallel}^*$ and $g_{\perp}^*$ increase approximately linearly with Ga content in the investigated composition range. However, the anisotropy ratio $g_{\parallel}^*/g_{\perp}^*$ remains almost constant.

Alternating the QW width at constant alloy composition ($x_{Ga} = 0.47$) changes both the g values as well as the $g_{\parallel}^*/g_{\perp}^*$ ratio (see Fig. 3). In the quasi-three-dimensional case ($d = 100$ nm) a $g^* = -4.01 \pm 0.04$ resonance is obtained, which is isotropic within the experimental uncertainty. With increasing quantization (decreasing QW width) the anisotropy increases to $g_{\parallel}^*/g_{\perp}^* = 4$ for $d = 6$ nm, the thinnest QW for which ODMR could be observed.

The ODMR signal is caused by spin resonance of the conduction electrons at the very bottom of the conduction band. The laser-excitation produces in our experiments $< 10^9$ cm^{-2} electron-hole pairs in the QW. The photogenerated electrons are located at the bottom of the conduction band or, if a magnetic field is applied, in the lowest spin-split Landau level. The photogenerated holes, however, only marginally affect the $\approx 10^{12}$ cm^{-2} hole concentration already present in the QW due to the single-sided modulation doping. The quasi-Fermi-energy

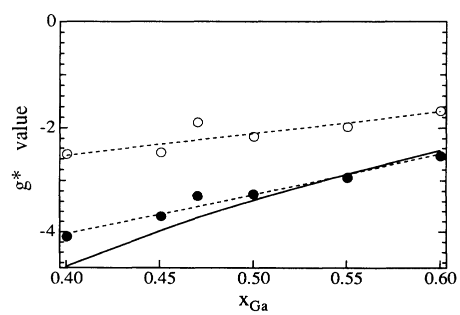

FIG. 2. $g_{\parallel}^*$ (solid circles) and $g_{\perp}^*$ (open circles) in $Ga_xIn_{1-x}As$/InP 15-nm QW's. The solid line is the result of a calculation (see text), and the dashed lines are a guide for the eye.

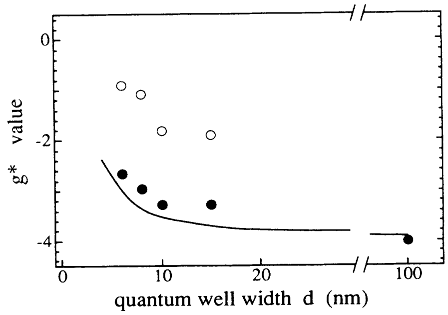

FIG. 3. $g^*_{\parallel}$ (solid circles) and $g^*_{\perp}$ (open circles) for different nominal well widths in $Ga_{0.47}In_{0.53}As/InP$ QW's. The solid line is the result of a calculation (see text).

level of these holes is located several meV below the valence-band edge, and a hole resonance (at $k = k_F$) is highly unlikely since the ODMR is detected on the band-edge PL ($k = 0$). The observed anisotropy in the spin resonance signal could, in accordance with previous studies of type-II systems, be taken as evidence for a hole spin resonance.[10,11] For holes an analysis in the spin-$\frac{3}{2}$ formalism, valid for the valence band for $x_{Ga} \leq 0.5$, where the electron–heavy-hole transition dominates the PL, gives $g^*_{\parallel} = 4$ and $g^*_{\perp} = 0$.[10,13] The discrepancy between the present results and these values is therefore a further indication that holes are not involved. Another indication is the absence of change in the anisotropy at the x value at which a transition from heavy-hole-dominated PL to ligh-hole-dominated PL takes place ($x_{Ga} \approx 0.5$). The conclusion that the spin resonance signal originates in the free electrons located at the bottom of the conduction band is, on the other hand, supported by the values of the spin splitting at spin resonance (≈ 0.1 meV), the thermal energy (≈ 0.14 meV), and the electron quasi-Fermi energy (< 0.1 meV).

The observation of ODMR signals in type-I QW structures is apparently facilitated by the spatial separation of the electron and hole wave functions caused by the single-sided modulation doping. Self-consistent calculations of overlap integrals give an increase of approximately two orders of magnitude, corresponding to an increase of the optical lifetime from the intrinsic ≈ 1 ns to ≈ 0.1 μs, the region where spin-flip transitions can be made to occur. Another possible reason may be the admixture of p-type states in the pure spin-up and spin-down s-type wave functions of the conduction band due to the electric field, $\mathbf{k} \cdot \mathbf{p}$ interaction and inversion asymmetry.[18] This could, in principle, lead to a relaxation of the pure magnetic dipole selection rules and an increased spin-flip probability through electric dipole spin transitions. Time-resolved measurements to further elucidate this question are in progress.

The experimental technique used in this investigation is based on an occupation difference in the spin population. It is, however, well known that the relaxation of the electronic spin polarization is accompanied by a build up of a nuclear spin polarization via flip-flop processes (Overhauser effect).[19] The result is that the polarized nuclei produce a magnetic field that introduces a shift of the spin resonance position. An estimate[20] gives in our case a 10-mT shift, corresponding to $< 1\%$ uncertainty in g^*.

Another possible source of misinterpretation of g^* has to do with the electric field. First, the electric field shifts the energy levels compared to the zero-field case, introducing corrections of the g^* values. Second, the electric field induces a spin splitting.[21] This electric-field spin splitting strongly affects the measured g^* values at low magnetic fields. However, since our measurements at 24 and 36 GHz give identical results we conclude that we are safely above this magnetic-field region. These effects, for instance, cause the isotropic g^* value in the 100-nm QW to deviate from the true $Ga_{0.47}In_{0.53}As$ bulk value. An extrapolation to the true bulk value (using the model calculation described below), taking this electric-field effect into account, gives $g^* = -4.1$. The electric-field splitting, in combination with low microwave frequency measurements could, in fact, be the reason for the weak $|g^*| = 5.6$ signal observed in a lattice-matched 15-nm QW.[5] This is supported by an independent investigation on exactly the same sample in which an electron-hole separation in the QW was reported.[22] Another reason for their result may be that they measured on a bound electron state. The details of the electric-field-induced spin splitting are not known, and form an interesting subject for further research.

The $Ga_{0.47}In_{0.53}As$ bulk value in the present work, $g^* = -4.1$, is in agreement with the value suggested in Ref. 8 using electrically detected spin resonance in a 2DEG, but is quite different from the $|g^*| = 5.2$ value reported in Ref. 23.

From the observed dependence of the $g^*_{\parallel}$ values on alloy composition for $d = 15$ nm QW's assuming the dependence to be linear, the approximated value for an InAs QW is $g^*_{\parallel} = -7$. This is reasonably consistent with the only available data,[24] an indirect measurement of a 10-nm InAs QW in GaSb, which gave $7.8 \leq |g^*_{\parallel}| \leq 8.7$, compared to $g^* = -15$ for bulk InAs.[25] The extrapolation to GaAs results in a $g^*_{\parallel}$ value close to zero, in agreement with literature.[12,19]

The observed confinement-induced anisotropy of the g^* value of the conduction electrons is a direct experimental verification of a recent theoretical prediction by Ivchenko and Kiselev.[3] In this model the effect of quantization is introduced as a change of the band gap, which in the Kane model[26] introduces a correction of the $g^*_{\parallel}$ value. Even though the theory can be criticized for oversimplifying the problem by choosing bulk concepts in the treatment of a two-dimensional problem, it seems to explain the essence of the experimental results.

A calculation of the $g^*_{\parallel}$ values as a function of QW width for the unstrained $Ga_{0.47}In_{0.53}As/InP$ system, using a more realistic model leads to quantitatively good agreement with the experimental data (see Fig. 3). In this self-consistent, subband calculation an 8×8 Kane Hamiltonian is used to describe the interaction between the Γ_{6c}, the Γ_{8v}, and the spin-orbit-split Γ_{7v} band, as well as a correction for interactions with higher conduction bands. In the calculation of the spin splitting, the Zeeman interaction, the effects of the electric field, and the interface effects, are taken into account. The latter effects are the reasons for the electric-field-induced spin splitting. The resulting two coupled equations are solved numerically using the self-consistently calculated potential and the boundary conditions described in Ref. 27. The material parameters are taken from Ref. 4, and the bulk Zeeman splitting factor $g^* = -4.1$ was chosen. The agreement with experiment is quite satisfactory, bearing in mind that the inversion-asymmetry spin splitting, proportional to k^3, has not been taken into account. It is, however, a difficult task to calculate the $g^*_\perp$ value, since an accurate choice of basis functions is nontrivial.

The calculation of $g^*_\parallel$ values in the case of $d = 15$ nm QW's with varying alloy composition is in principle possible, but at present the information regarding interband coupling terms in the case of compressive and tensile strain is not available. The calculation of $g^*_\parallel$, including electric-field, quantum-confinement, and strain effects, approximating the interband coupling terms from a linear interpolation between the lattice-matched $Ga_{0.47}In_{0.53}As$ and the binaries, gives the solid line in Fig. 2. The calculated result is in reasonable agreement for higher x_{Ga} values, but deviates for smaller x_{Ga} values. The calculation of the $g^*_\perp$ value is, for the above-mentioned reasons, even more difficult in this case.

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