Anisotropy of the electron *g* factor in lattice-matched and strained-layer III-V quantum wells

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The influence of quantum confinement and built-in strain on conduction-electron g factors in lattice-matched $GaAs/Al_{0.35}Ga_{0.65}As$ and strained-layer $In_{0.11}Ga_{0.89}As/GaAs$ quantum wells is investigated for well widths between 3 and 20 nm. The magnitude, sign, and anisotropy of the g factors were obtained from quantum beats due to Larmor precession of electron spins in time-resolved, polarization-sensitive, pump-probe reflection at 10 K in magnetic fields applied along and at 45° to the growth axis. Slowly varying shifts of precession frequency, due to buildup of nuclear polarization in the samples over ~1 h and equivalent to up to 0.5 T, occurred for fixed circular pump polarization and oblique applied fields. These Overhauser shifts confirmed the sign of the g factors and were eliminated by modulation of pump polarization to give precise g factors. For both material systems, variation of the g factor with well width follows *qualitatively* the dependence on energy, determined by quantum confinement, calculated from three-band $\mathbf{k}\cdot\mathbf{p}$ theory in the bulk well material. For the lattice-matched system there is excellent *quantitative* agreement with a full three-band $\mathbf{k}\cdot\mathbf{p}$ calculation including anisotropy effects of the quantum-well potential. For the strained-layer system, detailed quantum-well calculations do not exist but $\mathbf{k}\cdot\mathbf{p}$ theory for epitaxial layers predicts 10 times greater anisotropy for wide wells than we observe. This discrepancy is also apparent in previous, less complete, investigations of strained-layer systems and highlights the need for further theoretical effort.

INTRODUCTION

The measurement of carrier g factors in bulk and low-dimensional semiconductor systems is an excellent tool to test the predictions of band-theory calculations giving insight similar to that offered by determination of effective mass. The conduction-electron g factor, g_e, is determined by a balance between the bare-electron contribution (+2.0) and a lattice orbital contribution, which may vary considerably in both magnitude and sign for different structures[1–6] and is expected to show anisotropy due both to quantum confinement potentials[7,8] and to built-in strain.[9] It is also relevant to a variety of areas such as optically detected nuclear resonance, spin electronics, and quantum beats[10] and has particular significance in interpretation of the fractional and integer quantum Hall effects, where the predominance of skyrmion effects in a system is strongly dependent on the ratio of Zeeman and Coulomb energies.[11] In this context the g factor is frequently taken to be isotropic.

In type-I quantum wells, systematic studies of g_e for fields applied perpendicular to the growth axis have been made in the lattice-matched systems $GaAs/Al_xGa_{1-x}As$ (Refs. 1–3) and $In_{0.53}Ga_{0.47}As/InP$ (Ref. 4) and in strained-layer $In_xGa_{1-x}As/GaAs$ (Refs. 5 and 6), showing significant variation with well width L_z and, in the former system, zero crossing for $l_z=5.5\,\mathrm{nm}$.[1] In each case the gross features of the variation can be assigned to the variation of g_e with kinetic energy in the bulk well material (GaAs or $In_xGa_{1-x}As$) due to conduction-band nonparabolicity that is ''sampled'' at different values determined by the quantum-well confinement energy.[1,4,5] Anisotropy of g_e (i.e., $g_\perp - g_\parallel \neq 0$, where the subscripts denote g factors for fields perpendicular and parallel to the growth axis, respectively), induced by built-in biaxial strain and/or the quantum-well potential is expected to be small compared to this gross contribution; in a lattice-matched system the anisotropy should vanish in the limit of wide wells, whereas it should remain finite in a strained-layer system. Detailed $\mathbf{k}\cdot\mathbf{p}$ calculations including anisotropy have been carried out for lattice-matched $GaAs/Al_xGa_{1-x}As$ by Ivchenko and Kiselev[7] and for the effects of built-in strain as a function of x in $In_xGa_{1-x}As$ epilayers on GaAs and InP substrates by Hendorfer and Schneider.[9] Calculations for quantum wells with built-in strain have not been published but the calculations for epilayers suggest that anisotropy induced by strain should be about an order of magnitude greater than that from quantum confinement alone.

In this paper we report precision determination of the electron g factor and its anisotropy and sign in both lattice-matched $GaAs/Al_{0.35}Ga_{0.65}As$ and strained-layer $In_{0.11}Ga_{0.89}As/GaAs$ quantum wells for $3 \leqslant L_z \leqslant 20$ nm by means of Larmor beat measurements in a pump-probe reflection geometry. We take precautions to quantify and eliminate nuclear spin polarization (Overhauser) effects, well known in cw optical work,[12] but until recently neglected in time-resolved experiments.[13] This gives improved precision and allows determination of the sign of the g factors. Our measurements indicate that, in contrast to theoretical expectations, the anisotropy induced by built-in strain is small, not significantly greater than that due to quantum confinement in our samples. Measurements by Kowalski *et al.*[4] on 15-nm-wide $In_xGa_{1-x}As/InP$ quantum wells for $0.4 \leqslant x \leqslant 0.6$, in which built-in strain was incorporated in a controlled manner by variation of x about the lattice-matched value 0.53, also indicate a much smaller effect than predicted.

Kalevich, Zakharchenya, and Fedorova[14] made the first measurements of anisotropy in lattice-matched $GaAs/Al_xGa_{1-x}As$ wells with $L_z=8$ and 4.5 nm and, more recently, LeJeune *et al.*,[3] measured both the magnitude and anisotropy of the electron g factor, using time-resolved luminescence to detect electron Larmor precession. The $\mathbf{k}\cdot\mathbf{p}$ calculations[7] fitted their data quite satisfactorily but failed to describe the observed lack of anisotropy for the widest well studied ($L_z = 12$ nm); although the anisotropy must vanish in the limit of infinite well width, the theory appeared to overestimate it greatly for wide wells. We confirm this result and discuss possible reasons for inaccuracy of the calculation. Oestreich and co-workers[8] have investigated anisotropy in a ~5-nm GaAs/AlAs quantum-wire structure and found qualitative agreement with predictions based on Ref. 7. The only previous measurements of anisotropy in the strained-layer system $In_xGa_{1-x}As$/GaAs have been made by Kowalski *et al.*[6] for two samples with well width $L_z = 14.3$ and 14.7 nm and $x = 0.21$; these also indicated a much reduced anisotropy compared to that expected theoretically on the basis of strained-epilayer calculations.[9]

EXPERIMENTAL DETAILS

Each of the samples used in our measurements was grown by molecular beam epitaxy (MBE) on a (100)-oriented substrate without deliberate doping of the wells or barriers. Two $GaAs/Al_{0.35}Ga_{0.65}As$ single-quantum-well samples were examined. The first contained a set of single wells with $3 \leqslant L_z \leqslant 30$ nm separated by 15-nm barriers on a semi-insulating substrate. The second contained a single quantum well of width 9.6 nm on an n^+ substrate that could be biased by means of a transparent top contact to inject a population of $5\times10^{10}\,\mathrm{cm}^{-2}$ free heavy holes into the well.[1,13] For each well L_z was determined from the low-temperature photoluminescence (PL) spectrum by comparison with similar samples of known well width. Three $In_xGa_{1-x}As$/GaAs samples with $x = 0.11\pm0.02$ were studied. One contained three single quantum wells with $L_z = 3$, 6, and 10 nm, while the other two contained 20-period multiple wells with $L_z = 3.8$ and 20 nm, respectively. L_z was determined by transmission electron microscopy and the indium concentration from comparison of PL and photoluminescence excitation (PLE) spectra with Kane model calculations.[15] Results for $In_xGa_{1-x}As$/GaAs in fields applied *perpendicular* to the growth axis ($g_\perp$) have been published elsewhere[5] and are here combined with oblique field data to obtain $g_\parallel$ and the anisotropy of g_e.

Larmor precession was detected at 10 K using time-resolved pump-probe nonlinear optical reflection at near-normal incidence. The pump and probe beams, provided by a mode-locked Ti-sapphire laser tuned to the $n = 1$ heavy-hole exciton absorption, consisted of 2-ps pulses with 80 MHz repetition frequency, had average powers of about 1.5 mW and 200 μW and were focused to spot sizes of 50 and 40 μm, respectively.

The samples were mounted in the bore of a superconducting magnet, as shown in the inset to Fig. 1. The sample holder S and stearing mirror M could be independently rotated about a vertical axis, allowing the growth axis of the sample to be oriented at any angle to the field while keeping the angles of incidence of pump and probe beams on the sample fixed. The pump was circularly polarized to produce a photoexcited carrier population with spins initially polarized along the growth direction.

The probe was linearly polarized enabling the evolution of this spin polarization to be monitored via the induced rotation of the plane of polarization on reflection. The rotation, being proportional to the difference of excited spin populations parallel and antiparralel with the growth axis, showed oscillations at the Larmor frequency. In principle the latter is the Larmor frequency of excitons, but as discussed below, under the conditions of these measurements, the spins of electrons and holes are uncorrelated[10] and the observed oscillations occur at the electron Larmor frequency, the precession of the holes being absent (for perpendicular fields) and/or damped out in the first few picoseconds.

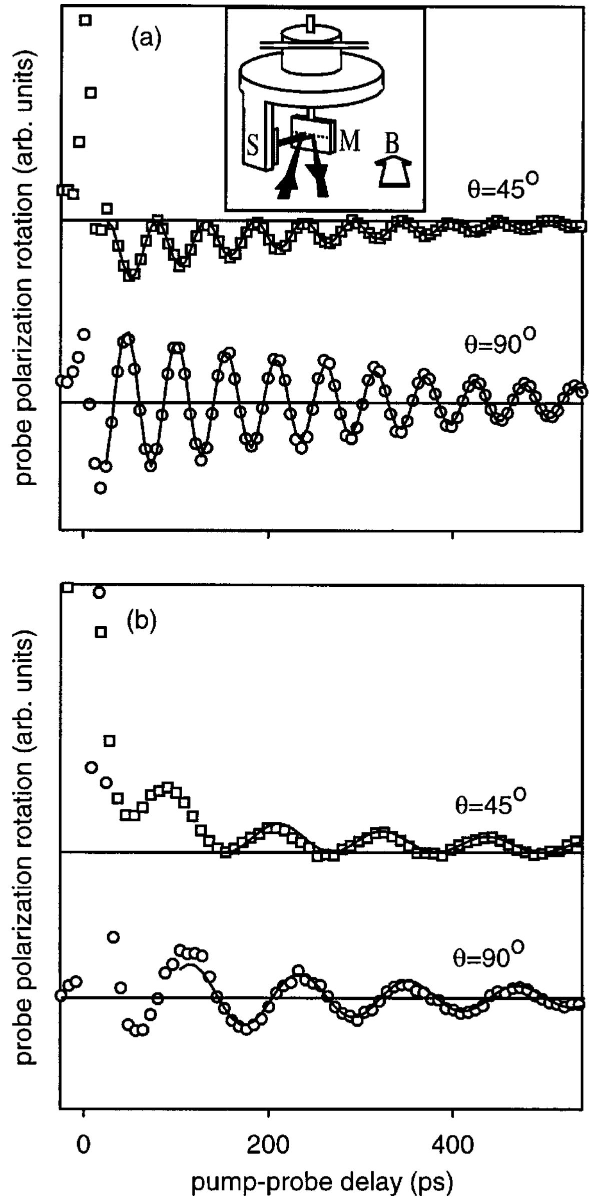

FIG. 1. Pump-induced probe-polarization rotation signals at 10 K for magnetic fields applied at an angle $\theta = 90°$ (circles) and 45° (squares) to the sample growth axis in (a) a 16.4-nm $GaAs/Al_{0.35}Ga_{0.65}As$ single quantum well at 4 T and (b) a 20-nm $In_{0.11}Ga_{0.89}As$/GaAs multiple quantum well at 1 T. Solid curves are best fits to the data of the form $Ae^{-t/\tau}\cos(\Omega t+\phi)$ for circles and $Be^{-t/\tau}[1+\cos(\Omega t+\phi)]$ for squares, respectively. The inset shows the experimental arrangement, allowing nearly normal incidence over a wide range of field angles.

Rotation of the probe polarization after reflection was monitored using a Faraday modulator, driven at frequency f_{probe}, followed by an analyzer crossed with the incident polarization and detection with a *p-i-n* photodiode. By modulation of the pump at frequency f_{pump} either in intensity by mechanical chopping with fixed circular polarization or between left and right circular polarizations by means of a linear polarizer and rotating $\lambda/4$ plate, the pump-induced probe-polarization rotation was extracted via lock-in detection at a frequency $f_{\mathrm{probe}}+f_{\mathrm{pump}}$, usually about 2 kHz.

RESULTS AND DISCUSSION

Figure 1 shows typical data for the two material systems with fields applied at angles $\theta=90°$ (circles) and 45° (squares) to the growth axis. In each trace there is an initial transient lasting up to 20 ps followed by oscillations decaying over several hundred picoseconds. For $\theta=90°$ the oscillations are symmetrical about zero whereas for $\theta=45°$ they are ''one-sided,'' between a finite value and zero. To interpret this behavior we note that initially the pump-induced spin polarization is along the growth direction. Thus a magnetic field applied at 90° to the growth axis induces the electron spins to precess on a disk between parallel and antiparallel orientations with respect to the growth axis, giving probe-polarization rotations oscillating between positive and negative values at the Larmor frequency. For fields applied at 45° to the growth axis the electron spins precess on a cone of half-angle 45° and therefore become perpendicular (rather than antiparallel) to the growth axis at one extreme of the precession. The parallel-spin state gives a finite rotation of probe polarization, whereas the perpendicular orientation corresponds to a coherent equal superposition of parallel- and antiparallel-spin states, giving zero rotation. The observed oscillations (squares) are thus at the electron Larmor frequency appropriate to the 45° field orientation, but now one-sided, between a finite value and zero.[3]

At 10 K, with resonant excitation, we expect the photogenerated population of carriers to be dominated by excitons. However, it does not follow that the observed precession frequency will be that of excitons, that is, of correlated electron and hole spins. Amand and co-workers[10] have demonstrated that the individual particle spins are uncorrelated if $\tau_h \ll \hbar/\delta$, where τ_h is the single-particle hole-spin relaxation time, and δ is the exciton exchange energy; in this regime the spins precess independently and the Larmor beats of the electrons persist at times longer than τ_h,[10] decaying due to population and spin relaxation of the electrons. For bulk GaAs, δ is approximately 12 μeV ($\hbar/\delta=44$ ps) and the value is enhanced by quantum confinement in the GaAs/Al_xGa_{1-x}As system to about 130 μeV ($\hbar/\delta=4$ ps) in the narrowest wells studied here (3 nm).[16] Hole-spin relaxation is very dependent on the kinetic energy of the holes and hence τ_h falls dramatically with increasing temperature and excitation density.[17] Under the conditions of our measurements—excitation density greater than $10^{10}\,\mathrm{cm}^{-2}$ and $T=10$ K—we expect $\tau_h \leqslant 4$ ps in GaAs/Al_xGa_{1-x}As (Ref. 17) so that the beats observed at longer times can be unambiguously assigned to electron, and not exciton or hole, Larmor precession. In the case of In_xGa_{1-x}As/GaAs, the enhancement of δ due to confinement is expected to be somewhat smaller,[16] favoring decoupling of the spins; we estimate that for the narrowest wells $\hbar/\delta \sim 9$ ps. On the other hand, spin relaxation of holes is inhibited compared to GaAs/Al_xGa_{1-x}As due to strain-induced decoupling of light- and heavy-hole bands; Dareys *et al.*[18] found $\tau_h \sim 17$ ps for $L_z=7$ nm for excitation density $\sim 10^{10}\,\mathrm{cm}^{-2}$ at 1.7 K. This would indicate correlation of electron and hole spins; however, we expect that τ_h will be reduced in our experiment due to the higher temperature and greater excitation density[17] so that the conditions for uncorrelated spins are still likely to be fulfilled. The observed initial transients in the $\theta=45°$ traces in Fig. 1 may then be assigned to effects of hole-spin precession and rapid relaxation. Other possible transient effects might arise due to nonideality of the pump polarization,[19,20] but these have decay times much faster than the observed electronic spin relaxation. By beginning the fitting of our oscillations from some sufficiently positive pump-probe delay, we ensure that we model only the longer-lived electron Larmor oscillations. This picture of uncorrelated electron and hole spins is also supported by the fact that our values of g factor are clearly different from those obtained from transient linear birefringence[19] and Zeeman[15,21,22] measurements with much lower excitation density and temperature, where excitonic effects dominate, and are close to previously obtained values for the electron g factor in GaAs/Al_xGa_{1-x}As and In_xGa_{1-x}As/GaAs QW.[1–5] It should also be noted that for field applied perpendicular to the growth axis ($\theta=90°$) the heavy holes ($J_z=\pm\frac{3}{2}$) will show no linear Zeeman splitting so that at fields where the precession frequency exceeds $\delta/\hbar$ the exciton Larmor frequency would, in any case, be very close to that of a free electron.[3,5]

In some experiments, where a fixed circular polarization of pump and an oblique magnetic field were used, it was found that the observed precession frequency changed over time periods of order 1 h after the experiment was set up. We attribute this to buildup of nuclear polarization via contact hyperfine interaction. In an oblique field, where the electron spins precess on a cone, a net electron spin polarization, parallel to the applied field, persists over the lifetime of the spin oscillations. The interaction of the resulting net electron spin with the nuclear spins of the lattice via the hyperfine interaction $A\mathbf{I}\cdot\mathbf{S}$ builds up a nuclear spin alignment $\langle\mathbf{I}\rangle$.[12] This, in turn, reacts back on the electron spins as an effective (Overhauser) magnetic field $\mathbf{B}_N$, given by

$$\mathbf{B}_N=\frac{A\langle\mathbf{I}\rangle}{g_e\beta}, \tag{1}$$

which augments or reduces the total field experienced by the electron spins. The time scale and magnitude of the changes in precession frequency were consistent with buildup of nuclear polarization via spin diffusion that we have previously investigated in detail in the 9.6-nm GaAs/Al_xGa_{1-x}As quantum-well sample.[13] For all isotopes of the III-V elements the hyperfine coupling constant A is positive so that the sign of B_N gives the sign of g_e. The signs of g factors obtained from B_N in this work confirmed previous determinations via oblique field Hanle measurements in similar quantum wells.[1,5] The observed Overhauser shifts in precession frequency were substantial, corresponding to B_N of the order of 0.5 T, the neglect of which would seriously affect

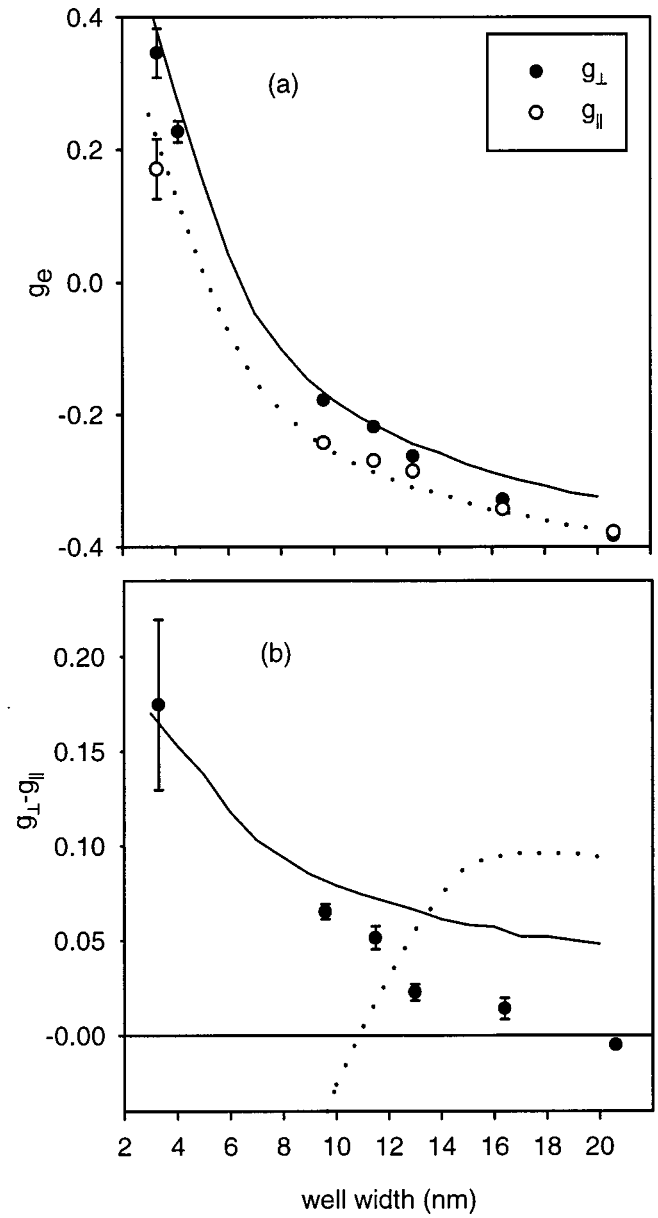

FIG. 2. (a) Measured values of $g_\perp$ (filled circles) and $g_\parallel$ (open circles) against well width for $GaAs/Al_{0.35}Ga_{0.65}As$ quantum wells compared with $\mathbf{k}\cdot\mathbf{p}$ calculations from Ref. 7 (curves) using the heavy-hole effective-mass ratio 0.4. (b) Measured anisotropy $(g_\perp - g_\parallel)$ (points) compared with calculations from Ref. 7 as in (a) with hole mass ratio 0.4 (solid curve) and with infinite hole mass ratio (dotted curve).

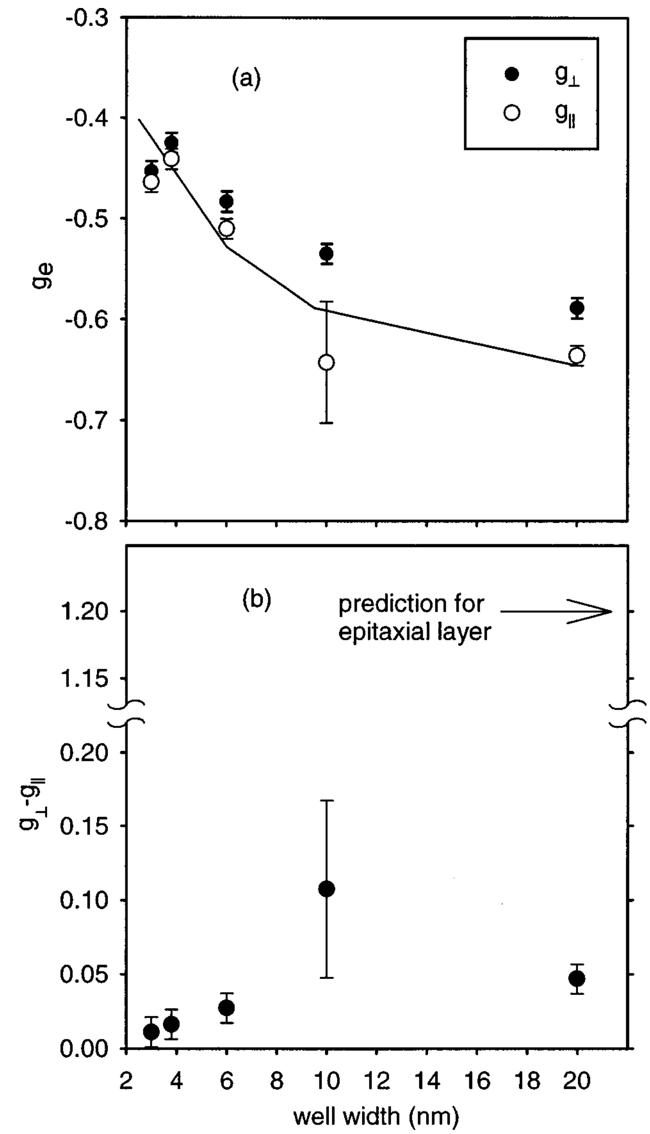

FIG. 3. (a) Measured values of $g_\perp$ (filled circles) and $g_\parallel$ (open circles) for $In_{0.11}Ga_{0.89}As/GaAs$ quantum wells. The solid curve is a three-band $\mathbf{k}\cdot\mathbf{p}$ approximation to g_e (see text). (b) Measured anisotropy $(g_\perp - g_\parallel)$ compared with predicted anisotropy for infinite well width, from Ref. 9, indicated by the arrow (note the broken vertical scale).

the accuracy of g-factor measurements. Modulation of the polarization of the pump at 85 Hz by the rotating λ/4 plate, a time scale that is fast compared to the observed accumulation of nuclear polarization, completely eliminated the drifts of precession frequency with time, and data taken in this mode were used to measure the magnitude of the g factors.

In Fig. 1 the fitted curves have the form $Ae^{-t/\tau}\cos(\Omega t+\phi)$ and $Be^{-t/\tau}[1+\cos(\Omega t+\phi)]$ for $\theta=90°$ and 45° respectively, allowing accurate determination of the frequencies Ω. Measurements of Ω were made at several values of field for $\theta=90°$ and $\theta=45°$ in each of the quantum wells and it was found to be linear in applied fields up to 6 T,[5] consistent with free-electron precession. The magnitude of the g factor is given by

$$|g_e| = \frac{\hbar\Omega}{\beta B}, \tag{2}$$

where β is the Bohr magneton and B the applied field. Figures 2(a) and 3(a) show the measured values of $g_\perp$ and also values of $g_\parallel$ given by $g_\parallel^2 = 2g_{45}^2 - g_\perp^2$ (Ref. 3) in the two material systems, while Figs. 2(b) and 3(b) show the anisotropy $(g_\perp - g_\parallel)$ as a function of well width. In the narrower $GaAs/Al_xGa_{1-x}As$ wells (Fig. 2) the precision is limited by the electron-spin relaxation rate, which reduces the number of observable oscillations for a given applied field, and where g was close to zero it was not possible to observe enough oscillations with the maximum available field (6 T) to determine g. In the wide wells, with long electron-spin lifetimes, the estimated errors are of order ± 0.001 for $g_\perp$ and ± 0.004 for $g_\parallel$. For $In_xGa_{1-x}As/GaAs$ (Fig. 3) the error bars are ± 0.01 for $g_\perp$ and $g_\parallel$ for most of the wells, again limited by the electron-spin lifetime. The difference in g factors between the 3.0- and 3.8-nm $In_xGa_{1-x}As/GaAs$ wells is consistent with the uncertainty in x.

In Fig. 2(a) the curves are the $\mathbf{k}\cdot\mathbf{p}$ calculations of Ivchenko and Kisilev[7] for $GaAs/Al_{0.35}Ga_{0.65}As$. The calculation assumes a heavy-hole effective mass ratio of 0.4, which affects the curve for $g_{\parallel}$ but not $g_{\perp}$. The agreement with the measurements is remarkably good except that the measured anisotropy tends to zero more rapidly with increasing well width than calculated. This is indicated directly in Fig. 2(b), where the curve is the difference between the two calculated curves in Fig. 2(a) LeJeune *et al.*[3] found no evidence of anisotropy of g_e in a 12-nm $GaAs/Al_xGa_{1-x}As$ QW; with better precision, we find that anisotropy within their uncertainty does exist for our 11.5-, 13-, and 16.5-nm QW's but becomes unmeasurable for our widest 20.6-nm well. For narrow wells there is some discrepancy between our measurements of g_e and those of LeJeune *et al.*, which may be associated with uncertainties in the well widths. A possible explanation of the discrepancy between the calculations and experiments for wide wells is suggested by the dotted curve in Fig. 2(b), which shows the anisotropy calculated for infinite heavy-hole mass.[7] This clearly worsens the discrepancy for wide wells, suggesting that a reduction of hole mass ratio from 0.4 might improve the accuracy of the calculation in this region. Such a change would be justified by magnetooptical measurements[23] that have shown hole mass ratios of order 0.14. Unfortunately Ref. 7 contains insufficient detail to allow this modification to be made to the calculation but, in any case, we conclude that the $\mathbf{k}\cdot\mathbf{p}$ theory gives an extremely good account of the electron g factor and its anisotropy in this lattice-matched system.

The curve in Fig. 3(a) is the result of a three-band $\mathbf{k}\cdot\mathbf{p}$ model[1,5] of the energy dependence of the g factor in bulk $In_xGa_{1-x}As$ but neglects effects of strain, anisotropy due to the quantum-well potential, and penetration of the electron wave function into the barriers. It can be seen that this represents an adequate first approximation for the g factor in these wells. Figure 3(b) shows the anisotropy; as expected for a strained-layer system, it tends to a finite value in wide wells but the limiting anisotropy for wide wells is about 10 times less than that predicted for an epitaxial $In_{0.11}Ga_{0.89}As$ layer on a GaAs substrate[9] as indicated by the arrow in Fig. 3(b). In contrast to $GaAs/Al_xGa_{1-x}As$, the anisotropy is much reduced in narrow wells, which suggests a partial cancellation of effects of the quantum-well potential and built-in strain in this system.

CONCLUSIONS

In conclusion, we have presented precision measurements of the conduction-electron g factor, sign, and anisotropy in lattice-matched $GaAs/Al_xGa_{1-x}As$ and in strained-layer $In_{0.11}Ga_{0.89}As/GaAs$ quantum wells. The *qualitative* dependence on well width of both magnitude and anisotropy are as expected. The lattice-matched system is *quantitatively* described by $\mathbf{k}\cdot\mathbf{p}$ theory, including anisotropy due to the quantum-well potential,[7] except for well widths greater than 12 nm where the anisotropy is overestimated. This discrepancy may be removed by use of a more realistic value of heavy-hole effective mass in the calculation. Although we have no detailed theory for comparison with our data for the strained-layer system, it appears that the effects of built-in strain are far less than expected theoretically; $\mathbf{k}\cdot\mathbf{p}$ calculations of the g-factor anisotropy in strained epilayers[8] exceed our limiting value for wide wells by an order of magnitude. Measurements by Kowalski *et al.* on $In_{0.21}Ga_{0.79}As/GaAs$ wells[6] and on slightly strained $In_xGa_{1-x}As/InP$ wells[4] also support the conclusion that existing theory greatly overestimates the effect of strain. Renewed theoretical investigation of this problem is therefore desirable. Our experiments have also highlighted the importance of taking account of nuclear polarization effects in time-resolved polarization-sensitive measurements.

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