Highly nonlinear Zeeman splitting of excitons in semiconductor quantum wells

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We have made a systematic investigation of the Zeeman splitting of $n=1$ heavy-hole excitons in a range of $Al_{0.36}Ga_{0.64}As/GaAs$ and $In_xGa_{1-x}As/GaAs$ ($x=0.08$ and 0.11) quantum wells at 1.8 K and in magnetic fields of up to 6 T applied along the growth axis (001). Calculations of splitting as a function of field were made using an eight-band $\mathbf{K}\cdot\mathbf{P}$ model which reproduce all the main features of the experimental data, including the sign, and give good quantitative agreement. The observed splittings are linear in low field (<1 T), but become nonlinear as field is increased. This behavior is attributed to a spin-dependent field-induced admixture between the light- and heavy-hole valence bands. For the GaAs/AlGaAs system agreement between experiment and theory requires a value for the Luttinger parameter κ in bulk GaAs close to 1.2 which is the generally accepted value, and rules out a lower value (0.7) which was proposed recently. From the theoretical fits to the $In_xGa_{1-x}As/GaAs$ Zeeman data we find that there is significant ''bowing'' of $\kappa(x)$ which can be reproduced accurately using a perturbation theory relating the Luttinger κ and γ parameters, where $\gamma_{1,2,3}$ are obtained from experimentally determined light- and heavy-hole effective masses. [S0163-1829(97)05624-5]

I. INTRODUCTION

Comparison between experimental and theoretical estimates of effective masses in bulk and low-dimensional semiconductors has become a standard technique for the verification of band structure calculations. The spin splittings and corresponding g factors, due to their comparable theoretical status, can be used in a similar manner offering information complementary to cyclotron resonance; however, this approach has received little attention. In this paper we use measurement of exciton spin splittings to obtain band structure information, giving an interesting insight into the effect of light hole-heavy hole mixing on the spin structure of the valence band.

Various magneto-optical investigations have concentrated on the conduction band to obtain precise values of electron g factors using techniques such as conduction electron spin resonance,[1] Hanle depolarization,[2] and, more recently, measurement of quantum beat periods.[3] Information on the valence band spin splittings has been obtained by direct spectroscopic determination, but these studies have been generally limited to high-field regions[4,5,6] (>5 T), due to small splittings in these systems. The only such study to have resolved splittings at fields of less than 1 T by Ossau *et al.*,[7] was limited to relatively wide GaAs wells where the splittings were very small compared to the inhomogeneous linewidth of the spectra. Exciton (as opposed to conduction electron) spin splittings have also been measured accurately by high-resolution laser techniques such as spectral hole burning[8] and detection of quantum beats,[9] but the results are limited to a small number of samples and suffer from an ambiguity of the sign of the splitting. Here we present further results obtained from a method[10,11] which uses polarization selection to separate the Zeeman split spectral lines, and allows splittings of less than 10 μeV to be resolved, with a precision comparable to the high-resolution techniques mentioned above, and with the notable advantage that we are also able to directly measure the sign of the spin splitting. This enables systematic investigation of the behavior of excitonic spin-splittings down to the low-field limit.

We have measured Zeeman splittings up to 6 T for a range of well widths in $Al_{0.36}Ga_{0.64}As/GaAs$ and $In_xGa_{1-x}As/GaAs$ ($x=0.11$ and 0.075) quantum well systems. In previous short papers,[10,11] we have described the asymptotic behavior of the Zeeman splittings in these systems as $B\rightarrow 0$, i.e., the excitonic g factors. Here we are more concerned with the nonlinear behavior of the Zeeman splittings as the field is increased and in particular present calculations using an 8 band $\mathbf{K}\cdot\mathbf{P}$ theory which show good quantitative agreement with the experimental data, reproducing all the main features including the sign of the splitting. By comparing experimental and theory we are able to gain information on the rich spin structure of the valence band, which is not revealed by studies at high fields or by measurement of electron spin splitting alone. We also discuss the validity of using $\mathbf{K}\cdot\mathbf{P}$ theory to describe the fine structure of excitons in these systems.

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II. SAMPLE DETAILS AND EXPERIMENT

Four $In_xGa_{1-x}As/GaAs$ samples were investigated. They were undoped and grown by molecular-beam epitaxy (MBE) on (001)-oriented semi-insulating GaAs substrates with a 0.5-μm GaAs buffer layer. One of the samples contained three quantum wells of nominal thickness 3, 6, and 10 nm separated by 30-nm GaAs barriers, while the other three contained single 4, 8, and 12 nm quantum wells, respectively. Transmission electron microscopy studies showed that the nominal well widths for the three-well sample were correct to within ± 5 Å, and we have assumed that this is the case for the other wells. For all the samples the nominal indium concentration was 0.11. To obtain independent estimates of the concentrations we carried out photoluminescence excitation measurements at 1.8 K of the light-hole (LH) and heavy-hole (HH) exciton energies. We compared these with calculations based on the Kane model using parameters given by Warburton *et al.*[5,12] This comparison indicated that the indium concentration for the three-well sample was 0.11 ± 0.02, and for the other three samples was 0.08 ± 0.01. Five undoped $Al_xGa_{1-x}As/GaAs$ samples were studied. They were all 60-period multiple quantum wells grown by MBE, also on (001)-orientated semi-insulating GaAs substrates. X-ray diffraction and photoluminescence excitation measurements[13] have shown that the aluminum concentration for these wells is $x = 0.36 \pm 0.01$ and that the well widths are 25.7, 56, 73.4, 112.5, and 149 Å with an uncertainty of around 1%.

Measurements were made at 1.8 K and at fields of up to 6 T applied along the sample growth direction. Excitation was by linearly polarized laser light at an energy well above the electron-hole continuum edge, incident along the growth axis, and backward luminescence was collected and detected by either a 0.25 or 0.5 m grating spectrometer connected to a photomultilplier. The four basis states of the exciton are separated into two doublets by the spin-dependent exchange Hamiltonian.[14] Electric-dipole-allowed recombination occurs only from one of these doublets, with emission of oppositely circularly polarized photons along the z axis, the other two states being nondipole allowed. The HH exciton emission line then shows a splitting with g factor

$$g_{\mathrm{ex}} = g_e + g_h . \tag{1}$$

For low fields the splittings are linear in field and the g factor constant. With increasing field, however, we can expect that the spin splittings become nonlinear, i.e., the g factor becomes field dependent. Although it is well known that the conduction band g factor increases with increasing field, the dominant effect comes from the valence band. The applied field causes a strong admixture of HH and LH states, and because the HH and LH splittings are very different this causes a strong nonlinearity in the spin splittings. This is the effect we focus on here. The predominance of this effect is controlled mainly by the light-heavy hole splitting, a factor influenced by the presence of strain, material composition, and the level of quantum confinement.

The fact that the two Zeeman components have different circular polarizations allows us to measure them separately and determine their splitting, despite the fact that the splitting is much less than the inhomogeneous linewidth. Our measurements utilize a 50 kHz photo elastic modulator in conjunction with either gated photon counting[11] or lockin detection[10] to record simultaneously σ^+ and σ^-polarized luminescence from the sample, giving the two Zeeman components in a single spectrometer scan. Both recording methods have the advantage of excellent noise rejection, although the use of lockin detection was found to improve on the precision gained with photon counting. The limiting statistical uncertainty for determination of one Zeeman component is $\Gamma/\sqrt{N}$, where Γ is the inhomogeneous linewidth and N is the total photon count of the component. We used a previously developed computational technique[11] to establish the difference in the first moments, which gives a precision approaching this limit for individual measurements. For the AlGaAs/GaAs data, however, systematic uncertainties from run to run, presumably arising from well width fluctuations across the sample, led to variations of order ± 10 to ± 20 μeV at 2 T. These fluctuations were not present in the InGaAs/GaAs measurements and, indeed, the overall accuracy of these measurements was of the order of ± 2 μeV. The sign of the splittings was determined directly by comparison with a standard σ^+-polarized beam.

III. THEORY

To calculate the Zeeman splittings of the quantum well structures we make the starting assumption that the splittings are primarily influenced by modifications to the bulk band structure from quantum confinement and strain. Excitonic effects are therefore neglected as far as the splittings are concerned, since it is extremely difficult to perform a calculation which includes both valence band mixing and the Coulomb interaction. This approach is reasonable provided that the energy separation between the various confined states is large compared to the excitonic binding energy as in this case the Coulomb potential will not mix these states significantly. The assumption will break down only in the valence band should the LH and HH confined states be energetically close on the scale of the exciton binding energies (5–10 meV). As we discuss below, this will occur for wide GaAs quantum wells and very narrow InGaAs wells. Otherwise, the approximation should be valid.

The $\mathbf{K}\cdot\mathbf{P}$ Hamiltonian adopted is given in Ref. 15 with all terms relating to inversion asymmetry omitted. Solutions for the quantum wells are sought subject to the boundary conditions that the envelope function $\underline{F}$ and the multiband analog of $1/m^* \, d\underline{F}/dz$ are continuous across the interfaces. The material parameters for the InGaAs/GaAs system are listed in Ref. 12 where the same computational procedure was used to model cyclotron resonance measurements on InGaAs/GaAs quantum wells. For the GaAs/AlGaAs wells we use a conduction-valence band offset ratio of 70:30 and a standard set of material parameters.

IV. EXPERIMENTAL RESULTS AND ANALYSIS

The results obtained for the $Al_{0.30}Ga_{0.64}As/GaAs$ samples are shown in Figs. 1(a) and 1(b). For the 56 and 73.4 Å samples, the data and theory are in good agreement. The data for the 25.7 Å sample are complicated by level crossings between one of the optically active states with one of the optically inactive states at ~1.1 and 4.0 T.[11,14] These cross-

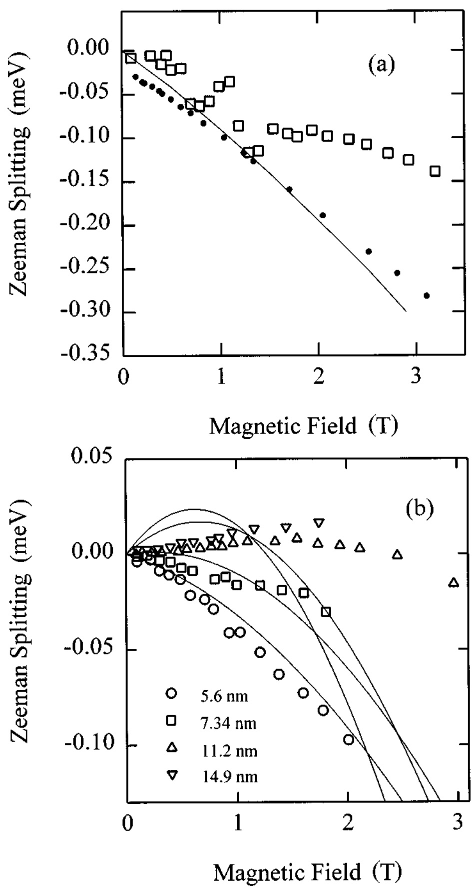

FIG. 1. Zeeman splitting of $n=1$ heavy hole excitons in $GaAs/Al_{0.36}Ga_{0.64}As$ multiple quantum wells (MQW's): Experimental data (points) and **K**·**P** calculations (curves) for (a) the 2.57 nm MQW and (b) the other 4 MQW's. In (a) the squares are measurements of Zeeman splitting by the polarization sensitive technique described in the text and the dots are results of quantum beat measurements (see text for discussion).

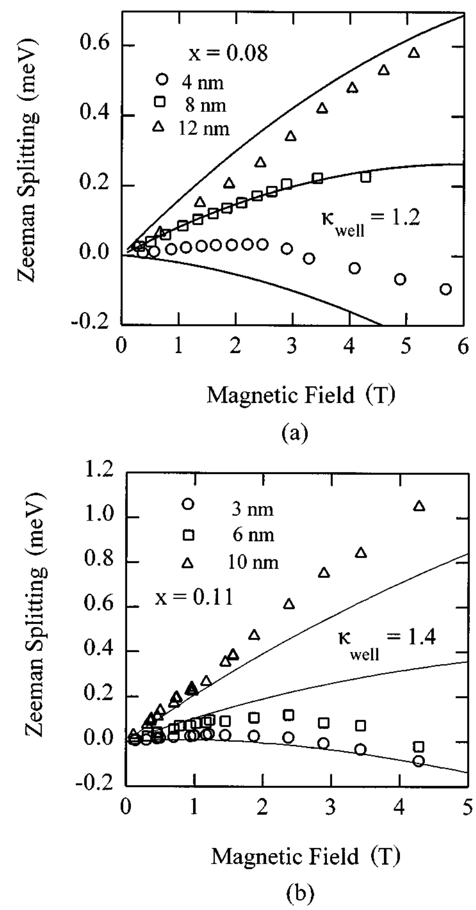

FIG. 2. Zeeman splitting of $n=1$ heavy-hole excitons in (a) $In_{0.08}Ga_{0.92}As/GaAs$ and (b) $In_{0.11}Ga_{0.89}As/GaAs$ quantum wells: Experimental (points) and **K**·**P** theory (curves).

ings perturb the luminescence line shape of the optically active level involved compromising the accuracy of our splitting determination procedure. Indeed, there is a pronounced minimum in the magnitude of the (apparent) splitting at the 1.1 T level crossing and so we believe that the actual splitting of the excitonic line lies closer to the lower limit of the data in this region. This assumption has been proven to be correct by quantum beat spectroscopy of this sample where the magnitude of the Zeeman splitting has been determined unambiguously.[16] The splittings obtained by measuring the quantum beat period are shown as dots in Fig. 1(a). Thus there is excellent agreement between experiment and theory for well widths up to 73.4 Å.

For wider wells the theory no longer reproduces the experimental splittings so well and this may be understood by consideration of the magnitude of the light-hole heavy-hole splitting. In this system, the LH-HH splitting peaks at a well width of around 40 Å, and for wider wells, decreases to such an extent that it becomes comparable to the exciton binding energy.[7] In fact, using the results of a calculation of the HH exciton binding energy at zero field[18] we estimate that the LH-HH hole splitting equals the excitonic binding energy at a well width of 150 Å. For wells of this width it thus seems reasonable to suppose that the Coulomb potential will mix the light- and heavy-hole states leading to a change in the Zeeman splitting from that calculated by our theory which, being a purely band calculation, does not include treatment of the Coulomb interaction. It is interesting to note that Bauer and Ando[19] find good agreement with the spin splitting data of Ossau *et al.*[7] for $Al_{0.3}Ga_{0.7}As/GaAs$ quantum wells wider than 120 Å by using an effective mass approximation which includes a consideration of the exciton binding energies, although this agreement may be fortuitous since the calculations used a 15–85 band offset ratio and an outdated material parameter set.

The nonlinear behavior of the Zeeman splitting at high fields, which can be seen clearly in the experimental data of both Figs. 1 and 2 and which is very well reproduced by the theory, can be ascribed principally to spin-dependent coupling between the heavy- and light-hole valence bands (calculated Zeeman splitting of the conduction band is essentially linear over this field range). As the magnetic field is increased the light- and heavy-hole states are linearly shifted and split by matrix elements of the field which are diagonal terms, while off-diagonal terms in the **K**·**P** Hamiltonian lead to level repulsion and the nonlinearity. However, because of symmetry restrictions, although the first $+\frac{3}{2}$ state is strongly influenced by this interaction, the first $-\frac{3}{2}$ state is not. This results in the crossover of these states and sign reversal of the Zeeman splitting in some cases. Symmetry considerations[20] show that the states must transform as either the Γ_3 (spin-up) or Γ_4 (spin-down) representation of the group of the star of k, C_s, and such off-diagonal mixing is only possible between states of the same representation. Thus the heavy hole $+\frac{3}{2}$ state is strongly repulsed by approach to the light hole $+\frac{1}{2}$ state as the field is increased, whereas the $-\frac{3}{2}$ state remains well separated from the $-\frac{1}{2}$ state and no such repulsion occurs. The field dependence of the $-\frac{3}{2}$ state is thus strikingly linear as can be seen from the calculations of Ref. 5.

A recent investigation[6] has reported a Luttinger parameter value for bulk GaAs of $\kappa=0.7$, rather than the widely accepted $\kappa=1.2$.[21] Our results do not support this conclusion; all our calculations were carried out using $\kappa=1.2$, and substitution of 0.7 results in extremely large discrepancies between experiment and theory for all the samples studied.

In the $In_xGa_{1-x}As$ system, because there is evidence that there is considerable bowing in the indium concentration dependence of κ (Refs. 22 and 23) we have performed **K·P** calculations of Zeeman splitting using κ as an adjustable parameter. The agreement between data and theory [Figs. 2(a) and 2(b)] is satisfactory for all the wells, within the constraints of well width and concentration uncertainties. Unlike the $Al_{0.30}Ga_{0.64}$/GaAs quantum wells in $In_xGa_{1-x}As$/GaAs heterostructures there is a strain-induced splitting between the light and heavy holes, with the consequence that the light holes are weakly type II, for the indium concentrations we have here. This means that the energy level of the light holes is independent of the level of quantum confinement. Consequently the LH-HH splitting for a given indium concentration is controlled solely by the amount that the heavy-hole energy is raised by quantum confinement. This means, therefore, that the nonlinearity in the Zeeman splitting as a function of field, due to admixture of the LH and HH states, will be strongest in narrow wells. This can be clearly seen from the data shown in Fig. 2. Mixing of the LH and HH states via the Coulomb potential, as observed in the GaAs wells, may be present in very narrow InGaAs wells where the LH-HH splitting is decreased by quantum confinement. Pragmatically, the generally good agreement between our data and theory suggests that the model is good for well widths greater than 30 Å. From a more fundamental point of view, the splitting between the confined HH level and the LH band edge in the barrier is comparable to the exciton binding energy only for very narrow well widths, ~10 Å. This means that a strong HH-LH mixing from the Coulomb potential is not expected.

From the comparison between data and theory for the samples studied here we obtain estimates of $\kappa=1.2\pm0.1$ for 8% indium and $\kappa=1.4\pm0.1$ for 11% indium and these values are plotted in Fig. 3. In order to understand the bowing of $\kappa(x)$ we relate the Luttinger parameters $\gamma_{1,2,3}$ and κ using the perturbation theory result[24]

$$\kappa=-\frac{1}{3}\gamma_1+\frac{2}{3}\gamma_2+\gamma_3-\frac{2}{3} \qquad (2)$$

which assumes that the Dresselhaus parameter H2 and the Luttinger parameter q are both zero. In order to interpolate the γ parameter between GaAs and InAs we make the following assumptions. The HH effective mass along the (001) direction which is given by $(\gamma_1-2\gamma_2)^{-1}$, is found experimentally to be the same for both GaAs and InAs (0.34), so we assume that it remains constant for all indium concentrations x. The LH mass in this direction $(\gamma_1+2\gamma_2)^{-1}$, is found experimentally to depend on x. The variation is given by $0.0942-0.062x$ for small x (Ref. 12) and for larger x a quadratic term must be included to obtain the correct InAs LH masa of 0.0275 at $x=1$. These two pieces of experimental data determine $\gamma_1(x)$ and $\gamma_2(x)$. The HH mass along (111) is given by $(\gamma_1-2\gamma_3)^{-1}$. To estimate $\gamma_3(x)$ we linearly interpolate between (111) HH masses of 0.73 for GaAs and 0.92 for InAs.[12] The value of $\kappa(x)$ obtained using these concentration-dependent γ's in Eq. (2) is plotted in Fig. 3, together with our experimentally determined values of κ and values of $\kappa=1.8$ for $x=0.18$ obtained by Warburton *et al.*[22] and the values for GaAs (Ref. 21) and InAs.[25] The experimental bowing is most satisfactorily reproduced by the perturbation theory result.

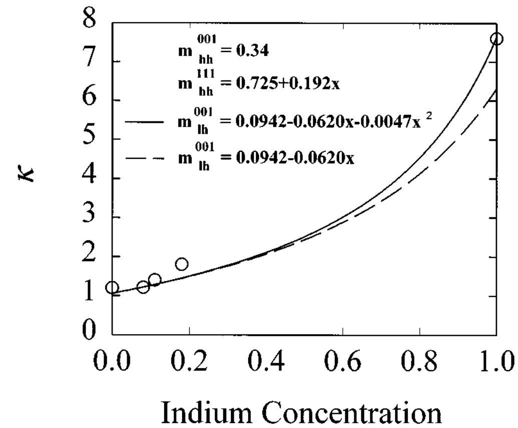

FIG. 3. Experimentally determined Luttinger κ in $In_xGa_{1-x}As$ compared with the perturbation theory result $\kappa=-\frac{1}{3}\gamma_1+\frac{2}{3}\gamma_2+\gamma_3-\frac{2}{3}$.

V. CONCLUSIONS

Measurements of Zeeman splittings of the $n=1$ heavy-hole excitons were made for a range of $Al_{0.36}Ga_{0.64}As$/GaAs and $In_xGa_{1-x}As$/GaAs quantum wells. The splittings show a linear field dependence at low fields (<1 T), but become highly nonlinear at high fields. Calculations were made using an eight band **K·P** model which show good quantitative agreement and reproduce all the main features of the experimental data, except in the case where the LH-HH splitting is comparable in magnitude to the exciton binding energy. The nonlinearity can be interpreted as a spin dependence of the field-induced admixture of the heavy- and light-hole states. Comparison between data and theory lends strong support to the value of 1.2 for the Luttinger parameter κ in bulk GaAs and we have also experimentally verified a perturbation theory connection between the Luttinger κ and γ parameters.

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