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Theory of heavy-hole magnetoexcitons in GaAs-(Al,Ga)As quantum-well heterostructures

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The energy shifts of the ground and "s-like" excited states of the heavy-hole exciton in a modest magnetic field (<4 T) normal to an impenetrable quantum well are calculated by direct numerical integration of the appropriate Schrödinger equation. Good agreement with experiment is obtained for the diamagnetic shift of the heavy-hole exciton for well widths in the range 50–100 Å.

Unlike observations on bulk material, excitonic features are readily seen in the absorption,[1] photoluminescence excitation (PLE), and photoluminescence[2,3] (PL) spectra of GaAs quantum-well heterostructures. In addition to the allowed $\Delta n=0$, heavy- and light-hole transitions, low-temperature spectra often show so-called "forbidden" $\Delta n\neq 0$ excitonic resonances, these transitions gaining optical strength from the valence-band mixing of the light and heavy holes in the plane of the quantum wells.[4–6] Careful spectroscopic studies of the dependence of the observed excitonic transitions as a function of both well width L_z and Al mole fraction x, and most recently, crystallographic orientation has, for example, led to reevaluation of the band offset ratio in this system,[2,3] the empirical determination of GaAs heavy- and light-hole masses in the (001) direction,[2] and the proposal of a new set of GaAs Luttinger parameters to describe the valence-band anisotropy.[7]

The application of a magnetic field perpendicular to the quantum wells is expected to provide further useful band-structure data and several magneto-optical studies have been performed that yield information about the binding energy of the $n=1$ heavy-hole exciton and measure its diamagnetic shift.[8–11] Very recently interband magnetoreflectivity observations[12] have been used to demonstrate the decoupled nature of the heavy- and light-hole bands in narrow GaAs quantum wells.

Previously[13] we have made calculations of the binding energies of the $1s$ and $2s$ states of the heavy-hole exciton in a GaAs quantum well and found excellent agreement with optical determination of the $1s$-$2s$ splittings for a number of well widths in the range 50–100 Å.[13] Thus far theoretical attempts to calculate the behavior of excitons in small magnetic fields[10,14,15] have either not addressed the calculation of the diamagnetic shift[14] or have derived shifts which are much larger than those seen experimentally.[10,15] In this paper we calculate the expected shift of both $1s$ and $2s$ states of the heavy-hole exciton in modest magnetic fields (<4 T) for a number of quantum-well widths by direct numerical integration of the appropriate Schrödinger equation. Using parameters identical to those of Ref. 13, we find gratifying agreement between our calculated diamagnetic shift of the $1s$ state and the experimental observations.

Our method of calculation of the diamagnetic shift is the same in essence as that used to determine the binding energies of the $1s$ and $2s$ states of the heavy-hole exciton[13] with modification of the Hamiltonian to include the effect of the perturbing magnetic field. To be definite, we concern ourselves with impenetrable quantum wells where electron and hole motion is quantized in the [001] direction. Furthermore, we assume that both electron and hole bands are parabolic and isotropic in the plane of the quantum well. The Hamiltonian describing the exciton motion is

$$\left[\frac{p^2}{2m^*}+\frac{e^2A^2}{2m^*}+e\left(\frac{1}{m_h^*}-\frac{1}{m_e^*}\right)\mathbf{A}\cdot\mathbf{p}+V(\mathbf{r})-\frac{2e\hbar}{M}\mathbf{k}\cdot\mathbf{A}\right]\Phi(\mathbf{r})=E\Phi(\mathbf{r}) , \quad (1)$$

where m^* is the reduced mass of the exciton and $\mathbf{A}$ is the vector potential.

The third term in the Hamiltonian is the Zeeman term which makes no contribution so long as we concern ourselves with excitons of s symmetry. The term involving the scalar product of $\mathbf{k}$ with $\mathbf{A}$ describes the coupling of the center-of-mass motion of the exciton to the magnetic field, for sufficiently small $\mathbf{k}$ this term can be neglected.[16]

If we choose the cylindrical gauge such that

$$\mathbf{A}=\tfrac{1}{2}\mathbf{B}\cdot\mathbf{r} , \quad (2)$$

then the form of the Hamiltonian which we need to solve is (in cylindrical polar coordinates)

$$\left\{\frac{-\hbar^2}{2m^*}\left[\frac{1}{\rho}\frac{\partial}{\partial\rho}\left(\rho\frac{\partial}{\partial\rho}\right)+\frac{1}{\rho^2}\frac{\partial^2}{\partial\phi^2}\right]+\frac{e^2}{8m^*}B^2\rho^2+\frac{e}{2}\left(\frac{1}{m_h^*}-\frac{1}{m_e^*}\right)Bm\hbar+V(\rho)\right\}\Phi(\rho,\phi)=E\Phi(\rho,\phi) , \quad (3)$$

where $V(\rho)$ is given by

$$V(\rho)=\frac{-e^2}{4\pi\varepsilon_0\varepsilon_r}\int\int\frac{dz_e dz_h\,|Z_e(z_e)|^2|Z_h(z_h)|^2}{[\rho^2+(z_e-z_h)^2]^{1/2}} . \quad (4)$$

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$Z_e(z_e)$ and $Z_h(z_h)$ are just the sinusoidal solutions to the particle in an impenetrable box problem. Rather than seek a variational solution to Eq. (3) we have integrated it numerically. This is particularly advantageous when one wishes to calculate the excited states of the exciton, as we do below, since it circumvents the necessity of inventing a trial wave function for each state. The materials parameters we used were $\varepsilon=12.5$, $m_e=0.0665m_0$, and an in-plane heavy-hole mass of $0.18m_0$. This value of in-plane heavy-hole mass is the near $\mathbf{k}=0$ value for a 50-Å decoupled well with barriers having an Al mole fraction of 0.35, calculated using a sophisticated $\mathbf{k}\cdot\mathbf{p}$ band structure[17] that includes the light- and heavy-hole mixing. The effect of nonparabolicity of the electron effective mass on the exciton reduced mass was included as in Ref. 13. The necessary confined electron energy levels were calculated assuming an Al mole fraction of 0.35 and a conduction- to valence-band offset ratio of 67:33.[3,18]

The shifts of the $1s$ and $2s$ heavy-hole excitons in applied magnetic fields of up to 4 T for quantum-well widths of 50, 75, and 100 Å are shown in Fig. 1. Clearly the $2s$ state moves to higher energies much faster in the applied field than does the $1s$. The increasing energy difference between these states with applied field means an enlarged binding energy of the heavy-hole exciton in the applied field due to the increased confinement of the electron and hole in the plane of the quantum wells. Figure 2 shows

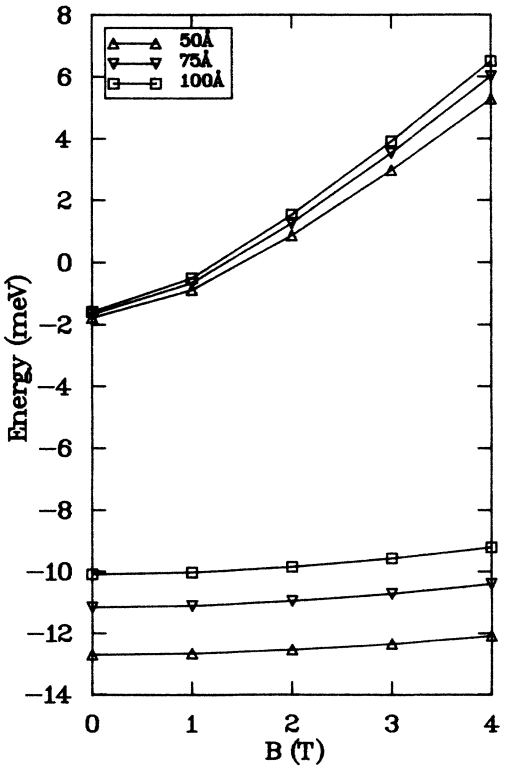

FIG. 1. Calculated shift of the $1s$ and $2s$ levels of the heavy-hole exciton in a magnetic field applied perpendicular to the plane of the quantum wells. (Calculated points are joined by straight lines.)

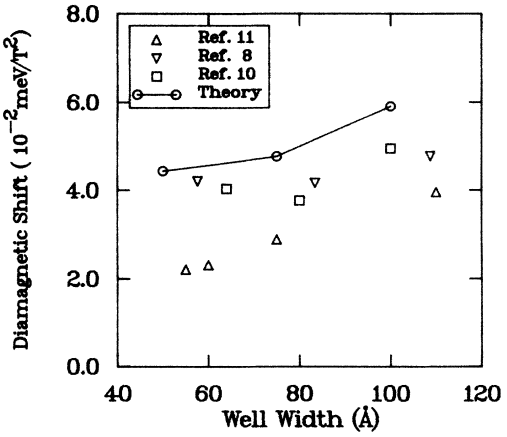

FIG. 2. Variation of the computationally deduced quadratic shift of the $1s$ levels of the heavy-hole exciton as a function of quantum-well width. Experimental data from Refs. 8, 10, and 11 are included for comparison.

the calculated variation in the diamagnetic shift (determined between 0 and 1 T) of the heavy-hole exciton as a function of well width. Also shown for comparison are the results of experimental observations on GaAs quantum wells in the range 55–110 Å.[8,10,11] There is a variation in the Al fraction x of the measured samples ($0.3 \leq x \leq 1.0$) and this must to some degree explain the variation in the shifts seen by the different groups. However, this variation is not likely to account for the almost factor of 2 difference, at the narrowest well widths, between the work of Rogers *et al.*[11] and those of Refs. 9 and 10 where the Al fraction in the barriers is very similar. Examination of the experimental data of Ref. 9 (Fig. 2) for their 65-Å sample shows a rather poor correspondence between the derived diamagnetic shift and the experimental points over the range 0–4 T, indeed the authors suggest that this sample may indeed be slightly inhomogeneous.[9] As for the diamagnetic shift deduced by Miura, Iwasa, Tarucha, and Okamoto,[8] this data is derived from experimental observations at pulsed magnetic fields up to 40 T. No experimental data is available at fields intermediate between 0 and ~8 T, and since the diamagnetic shifts ought to be deduced at low fields, then this experimental data must be regarded with some caution. As for the photocurrent spectroscopy data of Rogers *et al.*,[11] measured on samples with an Al fraction of 0.36, the authors do not explicitly state over what range of field their quadratic shifts were obtained, but they have observed excitonic peaks as low as 1 T. Given the uncertainty in the experimental determination of the diamagnetic shifts it is fair to claim good agreement between the calculations presented here and available experimental data.

Ossau, Jakel, and Bangert[10] have also calculated the quadratic shift of the $1s$ exciton in a magnetic field using a philosophy similar to ours. The principal differences between their approach and this one being (i) use of a variational wave function for the ground state, (ii) their inclusion of the finite barrier height, and (iii) the use of an in-plane heavy-hole mass of $\sim 0.14m_0$ for a 50-Å sample.

The authors cite an underestimate of electron nonparabolicity, a possibly inadequate variational wave function, and the use of the spherical approximation in the Luttinger Hamiltonian as reasons for the discrepancy between calculation and observation. From a comparison with our own calculations it seems clear that it is the larger in-plane hole mass which is essential to reproducing the measured shifts. We believe that combination of the newly proposed Luttinger parameters[7] and the $\mathbf{k}\cdot\mathbf{p}$ calculation of Ref. 17 provides *a posteriori* justification for our use of $0.18m_0$ in calculating exciton binding energies[13] and supports its continued use in the current magnetoexciton problem.

Recently Yang and Sham[19] have published a theory of magnetoexcitons in high magnetic fields which explicitly includes the effect of light- and heavy-hole mixing.[19] These authors point out that all the observable optical transition lines in a finite magnetic field are all bound exciton lines,[19] and furthermore they state that all optically active components of the exciton will be *s* like. Application of their theory to the published work of Maan *et al.*[20] resulted in identification of all the strong observable transitions as originating from either the ground ($1s$) or *s*-like excited states ($2s$, $3d$, $4d$, etc.) of the heavy- or light-hole excitons. However, the authors do note that their calculation is not in very good quantitative agreement with the experimentally observed shifts; the calculations overestimating the movement in the applied field. In a complementary study Bauer and Ando[15] have made similar calculations at fields up to 10 T and compare their theory favorably for well widths >120 Å with the experimental data of Ossau, Jakel, and Bangert.[10] In particular, they reproduce successfully the small spin splitting of the heavy-hole exciton claimed to be seen in the experiments. Neither of the above papers looks at small well widths at low magnetic fields, this we do below.

To illustrate the results obtained by numerically integrating Eq. (3), we present calculations of some of the excited states of the heavy-hole exciton one might expect to see in a 75-Å GaAs quantum well at applied magnetic fields up to 4 T. The results are shown in Fig. 3 and compared with the optical transitions seen in low-temperature ($\sim$1.8 K) magnetoreflectivity by Plaut *et al.*[12] from a 75-Å GaAs quantum well surrounded by $Al_{0.36}Ga_{0.64}As$ barriers. Calculations are presented for the $1s$ to $5s$ states of the heavy-hole exciton. The input parameters for the calculation are as described above. A number of points are noteworthy (i) the zero-field $1s$-$2s$ splitting is excellently reproduced, (ii) the diamagnetic shift of the lowest exciton state is similarly well described, and (iii) the one to one correspondence between the calculated excited states and the observations.

The excellent agreement between the calculation and the observation for the $1s$ state illustrates the difficulty associated with the determination of the diamagnetic shift. The calculated shift is 47.6 μeV/T^2 compared to the value

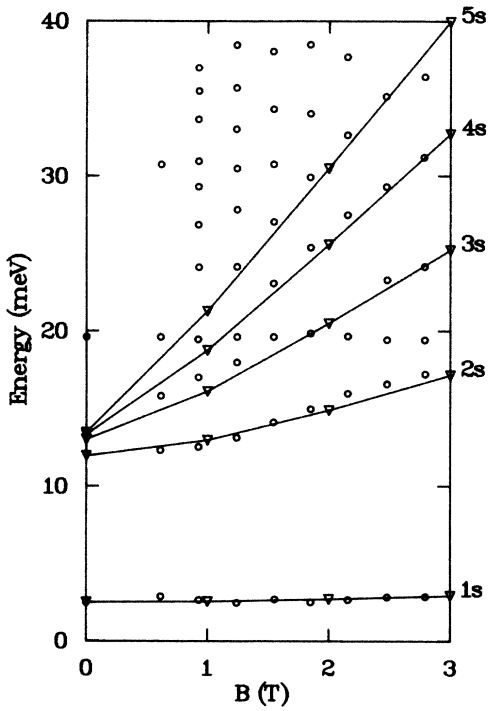

FIG. 3. Calculated shift of the ground and some excited states of the heavy-hole exciton in a magnetic field applied perpendicular to the plane of a 75-Å quantum well. The open circles are magnetoreflectively data reproduced from Ref. 12.

of 26 μeV/T^2 derived by Rogers *et al.*[11] by photocurrent spectroscopy on the same sample. Furthermore, Fig. 3 reinforces Yang and Sham's assertion that all the observable states are *s* like.

To summarize, we have made calculations of the ground and excited states of the heavy-hole exciton in a GaAs quantum well confined by infinite potential barriers. Computation of the energies of the states is not performed variationally but rather by direct numerical integration of the appropriate Schrödinger equation. Comparison of the calculated quadratic shifts of the $1s$ exciton as a function of well width with available experimental data is gratifyingly good and achieved by making *no* changes to the parameters used previously to successfully calculate the heavy-hole exciton binding energy.

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