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DIAMAGNETIC SHIFTS OF QUANTUM WELL EXCITONS IN STRONG MAGNETIC FIELDS APPLIED IN THE WELL PLANES

N. J. PULSFORD, J. SINGLETON, R. J. NICHOLAS and C. T. B. FOXON*

The Clarendon Laboratory, Parks Road, GB-Oxford OX1 3PU, Great-Britain
Philips Research Laboratories, GB-Redhill RH1 5HA, Surrey, Great-Britain

The behaviour of excitons associated with transitions from the light and heavy hole subbands to the electron subbands has been studied in GaAs-(Ga,Al)As quantum wells, with well widths between 2.2 and 14.5 nm, in strong magnetic fields applied in the plane of the quantum well. The data are successfully modelled using perturbation theory within an envelope function approximation, enabling the higher subband masses to be deduced: the results are in qualitative agreement with the predictions of pseudopotential calculations. Data taken at low fields with the magnetic field parallel and perpendicular to the well planes also reveal the strong non-parabolicity of the first heavy hole subband close to the decoupled (k=0) limit.

In this paper we report measurements of the shift of quantum well energy levels as a result of a strong magnetic field applied in the plane of the quantum well. The MQW samples used were grown on semiinsulating GaAs at Philips Research Laboratories, Redhill. The growth sequence consisted of a nominally undoped GaAs buffer layer, a 130 nm Ga_{0.4}Al_{0.6}As spacer layer, 60 periods of thick (≥15 nm) Ga_{0.4}Al_{0.6}As barriers and GaAs wells, and finally 130 nm of Ga_{0.4}Al_{0.6}As. Five different samples of well widths 2.2, 5.7, 7.5, 11.6 and 14.5 nm were available.

Interband photoconductivity measurements were performed on the samples at 46 Kelvin inside a 16 Tesla superconducting magnet. The photoconductive response is dominated by the GaAs buffer layer, providing a relatively smooth background, on which excitonic transitions appear as absorption minima. Various transition energies were measured as a function of magnetic field applied in the plane of the wells.

In zero magnetic field, the subbands of the quantum well are defined purely by the electric quantisation. In a small magnetic field, the cyclotron radius will be large compared to the well size and so the carriers will not be able to perform cyclotron orbits. As the magnetic field is increased, the cyclotron radius will become a comparable size to the quantum well thickness and hybrid electric-magnetic subbands will evolve [cf 2.3]: it is thus expected that the levels will shift more rapidly with field in wide quantum wells. Finally, in the limit of very high magnetic field, the magnetic quantisation will dominate and the subbands will become bulk GaAs Landau levels: as there is an exact correspondence between the electric and magnetic levels (ie the N=1 electric subband will become the f=0 Landau level etc.), the higher (N=2,3) subbands will shift more quickly with magnetic field than the N=1. Typical experimental results are shown in figures 1 and 2, and it will be seen that the diamagnetic shift both increases with well width and with increasing subband index, as predicted.

The results are modelled within the envelope function approximation [4]. We consider the Hamiltonian for an exciton inside an isolated quantum well. Rather than using an effective mass tensor, the electron and hole are given two masses each - one for in-plane motion (m^I) and the other for motion along the confinement direction (m^L). This both simplifies the problem a great deal and is in keeping with recent results, that a subband mass due to confinement may be different to the mass for in-plane motion [1,5]. In the absence of a magnetic field, the Hamiltonian is,

\[ -\frac{\hbar^2}{2m^I} \left( \frac{\partial^2}{\partial x_e^2} + \frac{\partial^2}{\partial y_e^2} \right) - \frac{\hbar^2}{2m^I} \left( \frac{\partial^2}{\partial x_h^2} + \frac{\partial^2}{\partial y_h^2} \right) - \frac{\hbar^2}{2m^I} \frac{\partial^2}{\partial z_e^2} - \frac{\hbar^2}{2m^I} \frac{\partial^2}{\partial z_h^2} + V_e(z_e) + V_h(z_h) - \frac{q^2}{4\pi\varepsilon_0 L_e - L_h} \]

(1)

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For an in-plane magnetic field, \( \mathbf{B} = (B,0,0) \), a suitable magnetic potential is \( \mathbf{A} = (0,-Bz,0) \), which adds four terms to the Hamiltonian:

\[
- \frac{qBz}{m_e^*} \frac{\partial}{\partial y_e} + \frac{qBz}{m_h^*} \frac{\partial}{\partial y_h} + \frac{e^2B^2z^2}{2m_e^*} + \frac{e^2B^2z^2}{2m_h^*}
\]

(2)

For a 1s exciton state, the two terms linear in \( B \) do not produce any shift. The diamagnetic shifts are therefore calculated by treating just the quadratic terms as a perturbation on zero field wavefunctions. Normally, such a perturbation approach is only valid if the energy shifts due to the perturbation are much less than the separation of the unperturbed levels. In this case, although the diamagnetic shifts are much less than the electric subband separation, they can be comparable to the binding energy, so a perturbation approach would appear unsuitable. However, due to the thickness of the wells compared with the spatial extent of a bulk exciton (Bohr radius of Wannier GaAs exciton \( \approx 13\text{nm} \)), the Coulomb term should not increase the z-confinement of the electrons and holes very much, but act mainly in the x-y directions. Thus the free carrier shift should be very similar to the excitonic shift, or equivalently, the exciton binding energy should not be significantly altered by an in-plane magnetic field. This is supported both by experimental data and a numerical calculation. For two of the samples, it was possible to measure the energy of the heavy-hole \( N=1 \) subband (H1) to electron \( N=1 \) subband (C1) free carrier transition [1,6] together with the excitonic transition. It was found that, to within experimental error, the two diamagnetic shifts were very similar, indicating a virtually constant binding energy. In addition, some diamagnetic shifts were calculated numerically both for free carriers and for excitons using the variational wavefunctions of [7]; the parameters for the exciton wavefunctions were taken from [7]. The difference between the free-carrier transition and exciton shifts was typically about 1%, indicating that the perturbation approach is indeed valid.

Assuming therefore a constant binding energy, the unperturbed wavefunctions can be taken to be simply the product of electron and hole finite square-well eigenfunctions. The appropriate well and barrier wavevectors (from terms in \( m_e^* \) and \( m_h^* \) in the Hamiltonian) for the confined states are obtained by fitting all the observed zero field transitions using Bastard's envelope function model [4] for an isolated quantum well and treating the well width, hole masses and electron non-parabolicity as variable parameters: several forbidden (\( \Delta N=1 \)) transitions are included so that a reliable cross-check of the confinement energies can be made. The final parameters which are needed are the in-plane subband edge effective masses: \( m_e^* \) and \( m_h^* \).

Values for the H1 and C1 masses have previously been determined from perpendicular field interband optical data [1]. The good agreement between theory and experiment provides a useful cross-reference between the two different lines of approach. Using the same C1 subband edge effective mass, the first light-hole subband (L1) mass is now varied to fit the L1−E1 diamagnetic shifts. For all the samples, this required an electron-like L1 effective mass as the E1 shift alone was consistently greater than the measured shift. This ties in with recent calculations of the in-plane valence band dispersion relationships which indicate that due to repulsion between L1 and H1, the L1 subband is electron-like at the zone-centre and only becomes hole-like further out into the zone [8,9]: evidence for this is also seen in previous magneto-optical data [1,6]. Such anti-crossing effects also produce a heavy H1 mass, but the H2 mass should be much lighter.

Higher transitions are harder to fit both because of the lack of knowledge of higher subband edge effective masses and because the broadness of the transition increases the experimental error. For the widest wells (14.4 nm), both the C1−H3 and C3−H3 shifts were measured, allowing values for the H3 and C3 masses to be deduced separately. However, all the other transitions that were considered obey the \( \Delta n=0 \) selection rule, which makes it impossible to determine the two relevant subband masses independently. Therefore, to fit the results, the electron effective mass (\( m_{e,\text{obs}}^* \)) was treated using a development [5] of the 3-band k.p expression originally derived by Palik et al.[10]:

\[
(1/m_{e,\text{obs}}^*) = (1/m_0^*)(1+2K_{xz}(T_z/E_g))
\]

(3)

where \( m_0^* \) is the band-edge effective mass, \( K_{xz} \) is the non-parabolicity seen in motion in the well-plane due to confinement and \( T_z \) is related to the confinement energy (\( E_{\text{conf}} \)) by:

\[
E_{\text{conf}} = T_z(1+K_{xz}(T_z/E_g))
\]

(4)

with \( K_{xz} \) the non-parabolicity observed in motion perpendicular to the well due to confinement. \( K_{xz} \) and \( K_{zz} \) are assumed to be equal to -0.7 [5]. This defines the electron subband edge effective mass (\( m_{e,\text{obs}}^* \)) for each confined state; the hole masses are treated as variable parameters: the fitted values are shown in table 1. The choice of -0.7 for the electron non-parabolicity might appear somewhat
arbitrary: however, the exact value is not important as any reasonable value still gives the same trends in the hole subband masses. The H2 and H3 masses obtained are consistently half, or less than half, the H1 mass; whereas the L2 mass appears to be very heavy, but neither definitely electron or hole like. As mentioned above, pseudopotential calculations [9] do predict a light H2 mass, but they also predict a heavy, electron-like H3 mass and a small, hole-like L2 mass, in contradiction to the experimental results. The reason for the discrepancy might be the fact that the measured H3 and L2 subband separation is only 2-3 meV for the 7.5, 11.6 and 14.6 nm wells, whereas in the pseudopotential calculation the H3 and L2 subbands are about 25 meV apart for a 10nm well. In view of the actual closeness of the H3 and L2 levels, it is likely that some form of anti-crossing behaviour is taking place, yielding the large L2 mass: as the pseudopotential calculation may give energies for the higher subbands which are a few percent inaccurate [9], these effects will not be predicted.

The final point of note is that the H1-C1 transition shows an anomalously high diamagnetic shift at very low magnetic field. The effect, which is also observed in the diamagnetic shift of the H1 to C1 excitons in magnetic fields applied perpendicular to the well plane (figure 3), is another manifestation of the complex H1 dispersion relationship. Close to B=0 (the decoupled limit in the Luttinger Hamiltonian), the H1 level will have very small in-plane effective mass [8,9], giving a rapid diamagnetic shift. As B (and therefore the wavevector in the well plane) increases, anticrossing effects will give rise to the heavy L1 and H1 effective masses seen in the high-field data. This spectacular low-field heavy-hole non-parabolicity has also recently been observed in inter-band Landau level transitions seen in reflectivity [11].

In summary, by measuring the shifts of the quantum-well excitons in a magnetic field applied in the plane of the quantum well layers, and by modelling the shifts using a envelope function model, we have obtained values for the effective masses of the higher hole subbands. The relative sizes of the masses are in qualitative agreement with pseudopotential calculations, apart from those of the L2 and H3 subbands, which are found to be much closer in energy than predicted, leading to a very large L2 effective mass. In addition, the low-field diamagnetic shifts of the H1 to C1 exciton demonstrate the large H1 non-parabolicity close to k=0, as recently observed in reflectivity measurements [11].

<table>
<thead>
<tr>
<th>Subband</th>
<th>2.2 nm</th>
<th>5.7 nm</th>
<th>7.7 nm</th>
<th>11.7 nm</th>
<th>14.2 nm</th>
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<tr>
<td>H1</td>
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<td>0.55</td>
<td>0.85</td>
<td>0.85</td>
<td>1</td>
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<tr>
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<td>0.45</td>
<td>0.4</td>
<td>0.4</td>
<td>0.5</td>
</tr>
<tr>
<td>H3</td>
<td>0.4</td>
<td>-0.5</td>
<td>-0.4</td>
<td></td>
<td>0.4</td>
</tr>
<tr>
<td>L1</td>
<td>-0.3</td>
<td>-1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>L2</td>
<td>-1</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 1: Fitted effective masses for the various hole subbands in the wells. The electron effective masses were calculated using m=0.066m_e and Kxz=-0.7 (see equations (3) and (4)).

References
11] Plaut A.S. et al, to be published
Fig 1) The diamagnetic shifts of different heavy hole (HN) to electron (CN) subband transitions for a 14.2 nm quantum well in an in-plane magnetic field. The solid lines are a theoretical fit to the experimental data.

Fig 2) The diamagnetic shifts of the C1-H1 subband transition for different quantum well widths in an in-plane magnetic field. The solid lines are a theoretical fit to the experimental data.

Fig 3) The low field diamagnetic shift of the C1-H1 subband transition in a magnetic field perpendicular to the well plane showing the anomalously high shift close to B=0.