CHAPTER 4

TWO-PHOTON EXCITONIC ABSORPTION
IN A GaAs/GaAlAs MULTIPLE QUANTUM WELL
IN A QUANTIZING MAGNETIC FIELD

Part of the work presented in this chapter has appeared in
4.1 Introduction.

The free electron and hole carrier states formed in the interband absorption process interact through the screened coulomb potential to form ‘excitons’. The resulting effective Rydberg measures the binding energy of excitons. The theory of excitons in 3D semiconductors is well documented [1,2]. After first observation by Dingle and co-workers [3,4] excitons in QWs are being studied with great deal of interest in zero and non-zero magnetic fields [5]. The exciton states become Q2D as the well thickness, \( L \), becomes comparable to the exciton Bohr radius, \( a_B \). We discuss mainly excitons in QWs of GaAs/GaAlAs type.

Due to reduction in symmetry along the growth axis of QWs and the presence of energy band discontinuities at the interfaces the valence band degeneracy is removed leading to the formation of heavy hole (HH) and light hole (LH) states and hence HH and LH excitons. A comparison of the sum of the subband energies in the conduction and the valence bands with exciton transition energies gives information about the exciton binding energies.

In QWs the binding energy, \( E_B \), of excitons and their oscillator strengths are enhanced, considerably, over their bulk values [3] due to potential confinement effect. As a result optical absorption spectra of QWs and photoluminescence are dominated by excitonic effects even upto room temperature. This has led to the proposed use of excitonic effects in
optoelectronic devices [6].

With a view to get physical insight into optical properties of these structures, theoretical calculations of optical absorption and binding energy (BE) of excitons have been carried out [7]. The theory of quantum confined excitons is based on the effective mass approximation in the envelope function scheme [8]. Most of the binding energy calculations use variational approach. Early calculations [9 - 11] predicted a monotonic increase of $E_B$ as well width decreases, because the spatial separation of electrons and holes is reduced, with a peak value in the range $3R_y < E_B < 2R$ for $1/4 < L/a_B < 1$, where $R_y$ is the effective 2D Rydberg. For example, maximum $E_B = 9$ meV occurs for $L = 50$ Å and $x = 0.3$. With further decrease of well thickness $E_B$ decreases because the spread of exciton wave function in GaAlAs layers becomes more important and makes $E_B$ approach the GaAlAs bulk value which is not yet observed experimentally.

Recent accurate $E_B$ calculations of Andreani and Pasquarello [12] including important effects such as valence band mixing, coulomb coupling between excitons belonging to different valence bands, non-parabolicity of conduction band and dielectric mismatch agree with the photoluminescence excitation data [13] within few tenths of a meV. Their calculations predict $E_B \approx 17$ meV, which is higher than the 2D limit of four times the bulk Rydberg. $E_B$ is found to increase monotonically with decreasing $L$.
In a quantizing magnetic field, the interband optical excitation leads to the formation of exciton states which are expected to be bound to each of the Landau level. Magneto-exciton effects are studied in bulk semiconductors by Elliot and co-workers [14]. Magneto excitonic studies are being made extensively in GaAs/GaAlAs QWs [15–26] which provide bulk of information about band structure. Theoretical calculations [20,25,26] predict that $E_B$ increases with increasing magnetic field which is due to the further constraint on the motion of the carriers in a magnetic field. Calculations of magneto excitonic properties by Yang and Sham [22] and Bauer and Ando [23] taking account of HH and LH band mixing are in excellent agreement with the high resolution magnetoluminescence spectra by Ossau and co-workers [16] and Potemski et al [24]. $E_B$ calculations for realistic QWs are also in agreement with experimentally observed values [17].

Most of the results on the linear and non-linear optical properties of confined excitons are summarized in several reviews and books. The latest reviews on this topic are given by Schmitt–Rink et al and Cingolani et al [7]. A great deal of interest has been devoted to experimental and theoretical study on the non-linear optical properties of semiconductor QWs. We are interested in one of the non-linear optical properties viz.,
two - photon excitonic absorption in QWs.

4.2 **Two - Photon Excitonic Effect.**

In the study of excitons TPA processes possess certain advantages over OPA processes. One of them is the accurate determination of band edge in TPA for the polarization of light in the plane of the layer. This enables the direct determination of exciton BE unlike the situation in OPA processes. The striking difference is that one - photon transition involve S - state excitons whereas in TPA dipole allowed transitions lead to P - state excitons. TPA transition rates strongly depend on the light polarization through the strong anisotropies in the exciton envelope functions. Hence one can probe the anisotropies in the exciton envelope functions by measuring the TPA spectra.

The zero field TPA spectra are strongly influenced by excitonic effects and are highly polarization dependent [27,28]. The theory of excitonic TPA in 2D systems is first given by Shimizu [29] who estimated two - photon transition rates of exciton states in a simple model. Two - photon transition rates of exciton states in GaAs QWs are calculated by Pasquarello and Quattropani [30], going beyond simple model of Shimizu and are in good agreement with the photoluminescence excitation experiments of Tai et al [27]. A unified theory for the TPA spectra of quasi 0, 1, 2 - D structures is given by Shimizu et al [31].
Fröhlich et al [32] have measured the two-photon magnetoabsorption coefficient in GaAs MQW using circularly polarized light orthogonal to the direction of confinement. In their experimental data, the effect of coulomb interaction is seen on the lowest Landau level. Their fan diagram (resonant energy versus magnetic field) shows a small bending in the low magnetic field region. There is a clear evidence of the 2P excitonic effects with the smallest energy Landau levels \((N' = 0, N = 1; N' = 1, N = 0)\) and results show an increase of exciton BE with increasing magnetic field. The transitions with higher Landau levels are less affected by excitons due to weak coulomb binding between the electrons and the holes. The TPMA study in GaAs MQWs is extended by Nithisooontorn et al [33] using photoconductivity techniques. They have investigated TPMA in GaAs MQWs up to magnetic fields of 7T where a splitting of a 2P state of an exciton is observed for fields higher than 3T. The observation of peaks due to 1S exciton in their experiment is attributed to the second harmonic wave generation. However, no theoretical calculation of two-photon magnetoexcitonic absorption (TPMEA) are made.

In section 4.3 we give the theory of TPMEA in GWs. Also, we calculate the binding energy of 2P magnetoexcitonic state in section 4.4. Calculated TPMEA coefficient and binding energy are compared with the experimental results of Fröhlich et al [32] in section 4.5.
4.3 Theory of Two - Photon Magnetoexcitonic Absorption (TPMEA)

We consider a simple model neglecting the anisotropy of the VB, the coupling of the split-off band, CB non-parabolicity and LH and HH mixing. Also we confine ourselves to a QW with finite height.

With the application of the magnetic field \( \mathbf{B} \), described by the vector potential \( \mathbf{A}(r) \), along the \( z \)-direction the exciton continuum splits into discrete excited excitonic states which are weakly bound to Landau levels and can therefore be described by free electron and hole states. The radius of these magnetoexcitons is assumed to be comparable to the cyclotron radius. The simultaneous observation of the magnetic field dependence of the bound and the continuum states allows a direct determination of the band edge and the exciton ground state energies and hence the exciton BE. A magnetic field applied along the growth axis \( (\mathbf{B} \parallel z) \) does not break the cylindrical symmetry of the zero field states of the excitons in the well. In particular, \( m \), the projection of the orbital angular momentum along the growth axis remains the good quantum number.

Exciton States in a magnetic field.

Because the coulomb potential and the barrier potential are two extremely different potentials, the former is a weak long
range potential, and the latter is a strong short range potential. All exciton states can be expressed in a basis of two particle Bloch states corresponding to an electron in the CB and a hole in the VB [14]

\[ \psi(r_e, r_h) = \sum_{k_e, k_h} a(k_e, k_h) \psi_c(k_e, r_e) \psi_v(k_h, r_h) \ e^{i(k_e \cdot r_e + k_h \cdot r_h)} \]

(4.3.1)

where, \( a(k_e, k_h) \) is the Fourier transform of the exciton envelope function. Since the band gap is direct, in the formation of excitons only the origin of the \( k \) - space with its immediate vicinity becomes important. The \( k \) - dependence of the Bloch states \( \psi_c(k_e, r_e) \) and \( \psi_v(k_h, r_h) \) is weak in this region. Hence they may be replaced by \( \psi_c(0, r_e) \) and \( \psi_v(0, r_h) \), respectively. Thus, equation (4.3.1) can be written as

\[ \psi(r_e, r_h) = \psi_c(0, r_e) \psi_v(0, r_h) \psi(r_e, r_h) \]

where,

\[ \psi(r_e, r_h) = \sum_{k_e, k_h} a(k_e, k_h) e^{i(k_e \cdot r_e + k_h \cdot r_h)} \]

represents the so called envelope function of the exciton satisfying
\[
\begin{align*}
&\left[ \frac{1}{2m} (\hbar \nabla_e + eA(r_e))^2 + \frac{1}{2m} (\hbar \nabla_h - eA(r_h))^2 \\
&+ V_{ew}(z_e) + V_{hw}(z_h) - (E + \frac{e^2}{4\pi\varepsilon_s |r_e - r_h|}) \right] \psi(r_e, r_h) = 0
\end{align*}
\]

where \( A \) is the vector potential and \( V_{ew}(z_e) \) and \( V_{hw}(z_h) \) are the heights of the potential well for the conduction electron and the hole, respectively. The last term in equation (4.3.2) describes the coulomb interaction between the electron and the hole, screened by the static dielectric constant of the medium. The exciton BE is given by the difference of the eigen values of the Hamiltonian with and without this term. In the plane parallel to the interfaces \( k_{exc} = k_e + k_h \), which describes the motion of the exciton, is zero.

Introducing, relative coordinate \( r = (r_e - r_h) \) and center of mass coordinate \( R_\pm = \frac{m^* r_e + m^* r_h}{m^* + m_\pm} \) equation (4.3.2) can be reduced to a single particle equation

\[
\begin{align*}
\left\{ - \left( \frac{\hbar^2}{2\mu} \right) \nabla^2 - \left( \frac{\hbar}{\mu^*} \right) eA.\nabla + \frac{(e\hbar)^2}{2\mu} - \left[ E + \frac{e^2}{4\pi\varepsilon_s |R_\pm|} + V_{ew}(z_e) + V_{hw}(z_h) \right] \right\} F(r) = 0, \quad (4.3.3)
\end{align*}
\]

The function \( F(r) \) is defined through
\[ \psi(r_e, r_h) = \exp \left( \frac{i}{\hbar} A(R_\pm) \cdot r \right) F(r). \] (4.3.3 a)

In equation (4.3.3)
\[
\frac{1}{\mu_\pm} = \frac{1}{m_e} + \frac{1}{m_\pm} ; \quad \frac{1}{\mu'_\pm} = \frac{1}{m} - \frac{1}{m_\pm}
\] (4.3.3 b)

where
\[
\frac{1}{m_\pm} = \frac{1}{m_0} (\gamma_1 \pm \gamma_2)
\] (4.3.3 c)

In equation (4.3.3) the quantity in \{ \} is the relative coordinate Hamiltonian for the exciton in the magnetic field and \( \gamma_1 \) and \( \gamma_2 \) are well known Kohn - Luttinger parameters.

For the magnetic field coinciding with the \( z \) - axis, in cylindrical coordinates, \( A \) takes the form \( A = (B/2) r \hat{e}_\phi \).

Equation (4.3.3) may now be written as
\[
\left[ \frac{1}{\rho} \frac{\partial}{\partial \rho} \rho \frac{\partial}{\partial \rho} + \frac{1}{\rho^2} \frac{\partial^2}{\partial \phi^2} + \frac{\mu_\pm}{m_e} \frac{\partial^2}{\partial z_e^2} + \frac{\mu_\pm}{m_\pm} \frac{\partial^2}{\partial z_h^2} + E + \frac{2}{\rho} - \gamma L_z - \frac{1}{4} \gamma^2 \rho^2 - V_{e_w}(z_e) - V_{h_w}(z_h) \right] F(\rho, z, \phi) = 0 \] (4.3.4)

The dimensionless parameter \( \gamma \) is a measure of the magnetic field given by
\[ \gamma = \frac{e \hbar B}{2 \mu CR_y^\pm} . \]  
\hfill (4.3.4 a)

\[ R_y^\pm, \text{ is the effective Rydberg, given by} \]

\[ R_y^\pm = \frac{e^4 \mu^\pm}{2 \pi \hbar^2} . \]  
\hfill (4.3.4 b)

The exciton Hamiltonian is in dimensionless form where all energies have been expressed in term of effective Rydbergs \( R_y^\pm \) and all lengths in terms of effective Bohr radius \( a_\pm \), given by

\[ a_\pm = \frac{\pi \hbar^2}{\mu^\pm e^2} , \]  
\hfill (4.3.4 c)

\( L_z \) is the \( z \)-component of the angular momentum in units of \( \hbar \).

The explicit forms of \( V_{ew}(z_e) \) and \( V_{hw}(z_h) \) are

\[ V_{ew}(z_e) = \begin{cases} 0 & |z_e| \leq L/2 \\ V_e & |z_e| > L/2 \end{cases} \]  
\hfill (4.3.4 d)

\[ V_{hw}(z_h) = \begin{cases} 0 & |z_h| \leq L/2 \\ V_h & |z_h| > L/2 \end{cases} \]
The center of mass is assumed to be at rest at any time. In equation (4.3.3) if we set coulomb interaction to zero we recover series of Landau levels (See Appendix C).

Here the origin of the coordinate system is chosen to be at the center of the well, so that the wave functions are characterized by a well defined parity. The exact solution of the above Schrodinger equation is not possible and therefore we resort to solve it variationally.

If we ignore the image potential and band bending near the interface the energy level spectrum is hydrogenic. We expand the wave function $F$, in spherical harmonics taking full advantage of the symmetry, each of which is multiplied by a complete set of radial function taken to be the sum of Gaussians.

The trial wave function $F$ can be written as

$$F(\rho, \phi, z_e, z_n) = f_C(z_e) f_V(z_n) G(\rho, \phi, z) \quad (4.3.5)$$

where $f_C(z_e)$ and $f_V(z_n)$ are quantum well eigen functions of the CB and VB respectively, given by equations

$$f_C(z_e) = \begin{cases} \cos(k_{e} z_e) & |z_e| < L/2 \\ A_{e} \exp\left(-\kappa_{e}|z_e|\right) & |z_e| > L/2 \end{cases} \quad (4.3.6)$$
and a similar expression holds for the hole wave function. The parameters $k_e$ and $k_h$ are obtained from the energies of the first subband and $A_i$ and $x_i (i = e, h)$ are obtained from $k_e$ and $k_h$ requiring the continuity of $f_i (i = n_e, n_h)$ and its derivative at the interface. $G(\rho, \phi, z)$ describes the internal state of the exciton. For wells less than several Bohr radii in width, the factorization (4.3.5) is particularly appropriate because it enables the separation of the relatively large square well energy that would otherwise overwhelm the smaller coulomb energy contribution.

Owing to the definite parity of the wave function $G(\rho, \phi, z)$ can be written in the form (19)

$$G(\rho, \phi, z = z_e - z_h) = z^q \rho^{|m|} e^{im\phi} \sum_{i,j} A_{ij} G_{ij}(\rho, z) \quad (4.3.7)$$

where $m$ is the component of the angular momentum (in units of $h$) along the axis of symmetry and $m + q$ (where $q = 0, 1$) together with $f(z)$, defines the parity of the state. We consider two different cases corresponding to $q = 0$ and $q = 1$.

Following Greene and Bajaj (19) we choose for the basis functions $G_{ij}(\rho, z)$ the form

$$G_{ij}(\rho, z) = e^{-\alpha_i z^2} e^{-(\alpha_j + \delta)\rho^2} \quad (4.3.8)$$
which is the product of the Gaussians in $\rho$ and $z$ variables. Here $\alpha_i$ and $\delta$ are the variational parameters which are adjusted to minimize energy. The set of parameters $\alpha_i$ are taken from the results of Huzinaga [35]. $\delta$ is the non-linear variational parameter primarily determined by the value of the magnetic field. We restrict the basis functions by requiring

$$A_{ij} = 0 \text{ for } |i - j| > 0. \quad (4.3.9)$$

The excitonic transitions are determined by the set of quantum numbers $\beta_{\text{exc}} = (n_e, n_h, m, \nu)$ where $n_e$ and $n_h$ are the subband quantum numbers in the CB and VB, respectively and $\nu$ refers to the principal quantum number of the exciton. Since the magnetic quantum number $m$, is a good quantum number, as a function of the magnetic field the exciton lines must join smoothly to the zero field exciton states with the same angular momentum components.

**Two - Photon Magneto Exciton Absorption Coefficient.**

We consider a circularly polarized radiation as described by equation (3.2.20) and the perturbation due to the two - photon field is

$$H_1 = (e/m_0c) [ A_1 . P + A_2 . P ] \quad (3.3.7 \text{ b})$$

The TPMEA coefficient for photon of energy $\hbar \omega_1$, when $\hbar \omega_1$
and \( \hbar \omega_2 \) are simultaneously present is given by

\[
\alpha = \left( \frac{2n_1}{eN_1} \right) \sum_{\text{in}} W_{\text{in}}(v + \beta_{\text{exc}}),
\]

where \( n_1 \) is the index of refraction, \( N_1 \) is the density of photons of energy \( \hbar \omega_1 \). The sum over 'in' stands for sum over all the initial states of the system.

The transition probability for the two photon excitonic absorption is obtained from the second order perturbation theory and is given by

\[
W_{\text{in}}(v + \beta_{\text{exc}}) = \left( \frac{2n_1}{\hbar} \right) \sum_{\beta_{\text{exc}}} |M|^2 \delta(E_{\beta_{\text{exc}}} - E_v - \hbar \omega_1 - \hbar \omega_2)
\]

where

\[
|M|^2 = |P_0 e \cdot P_2|^2 \left| \sum_{\beta'_{\text{exc}}} \left[ \frac{\langle \beta_{\text{exc}} \rangle \langle e_{1, e} \cdot P | \beta'_{\text{exc}} \rangle \langle \beta'_{\text{exc}} \rangle \langle e_{1, e} \cdot P | v \rangle}{E_{\beta_{\text{exc}}} - E_v - \hbar \omega_2} + \frac{\langle \beta_{\text{exc}} \rangle \langle e_{1, e} \cdot P | \beta'_{\text{exc}} \rangle \langle \beta'_{\text{exc}} \rangle \langle e_{1, e} \cdot P | v \rangle}{E_{\beta'_{\text{exc}}} - E_v - \hbar \omega_1} \right|^2 \right|
\]

The sum over \( \beta'_{\text{exc}} \) stands for a sum over all the intermediate excitonic eigen states. In equation (4.3.11) \( E_v \) is the energy of the electron in the valence band and \( E_{\beta_{\text{exc}}} \) is the energy of the
In the following we evaluate matrix elements appearing in equation (4.3.11 a), in the dipole approximation, which give the selection rules for the allowed transitions.

In equation (4.3.11 a) the matrix element \( \langle \beta'_{\text{exc}} | e_j, P | \nu \rangle \) is due to the transition between valence state and an intermediate exciton state. They are given by

\[
\langle \beta'_{\text{exc}} | e_j, P | \nu \rangle = |F(0)|^2 |e \cdot P_{\text{cv}}|^2 \delta_{m,0} \tag{4.3.12}
\]

where

\[
P_{\text{cv}} = \int u_c(o, r_e) \ P \ u_v(o, r_h) \, dr. \tag{4.3.12 a}
\]

Oscillator strength (OS) is equal to the probability density at the origin i.e. \(|F(0)|^2\) which can be obtained from equation (4.3.5).

The matrix element for a transition from the intermediate state \( \beta'_{\text{exc}} = (n'_e, n'_h, m', \nu') \) to the state \( \beta_{\text{exc}} = (n_e, n_h, m, \nu) \), due to circularly polarized radiation is given by
\[ \langle \beta_{\text{exc}} | P^\pm | \beta^\prime_{\text{exc}} \rangle = \frac{m_o \hbar}{\sqrt{2}} \sum_i I(\alpha_i) (\alpha_i + \delta) \delta_{m-m'}, 1 \]

(4.3.13)

where

\[ P^\pm = (P_x \pm iP_y)/\sqrt{2} \]

and

\[ I(\alpha_i) = \int_0^\infty z^2 e^{-\alpha_i z^2} |f_{n_e}^c(z_e)|^2 |f_{n_h}^v(z_h)|^2 dz_e dz_h \]  

(4.3.13 a)

Here plus and minus signs refer to left and right circular polarizations, respectively.

Combining equation (4.3.12) and (4.3.13) we find that only P - excitons are involved in TPMEA. Thus in the two - photon excitonic absorption, the first photon induces a virtual interband transition to the \( \nu = 1 \) exciton or a higher lying state with \( S - \) like envelope function. The second photon completes the transition by coupling to the \( \nu = 2 \) exciton or higher lying state with \( P - \) like envelope function of the electron - hole pair. Since the interband transition changes the parity of the envelope function, the lowest energy final state is the 2P exciton. Because of the small OS \( \Xi (\nu - 1/2)^{-1/2} \), the experimentally observed lines correspond to the lowest value of \( \nu \). For the
higher values of \( \nu \) in the exciton bound state spectrum, the lines are broad and they overlap which causes a smooth spectrum.

\[ E_{\nu} \] in the argument of \( \delta \)-function in equation (4.3.11) is given by \( E_{\nu} = E_C - E_B(\nu P) \) where \( E_C \) is the energy in the conduction band with which a series of excitonic states is associated and it is given by

\[ E_C = E_{0C} + E_{n_C} + (N' + \frac{1}{2}) \alpha_C \hbar \omega_L \]

and \( E_B(\nu P) \) is the BE of the exciton involved in the transition. The argument of the \( \delta \)-function is now found to be

\[ E_{\nu} - E_C - \hbar \omega_1 + \hbar \omega_2 = E_g + E_{n_C} + E_{n_P} + (N' + \frac{1}{2}) \alpha_C \hbar \omega_L + (N + \frac{1}{2}) \alpha_P \hbar \omega_L - E_B(\nu P) + \hbar \omega_1 - \hbar \omega_2. \]

As seen in chapter III, in the absence of excitonic effects, the free Landau level transitions obey the selection rules \( N' = N \pm 1 \) where + and - signs correspond to left and right circular polarizations, respectively. Hence the argument of the delta function becomes

\[ E_g + E_{n_C} + E_{n_P} + (N + \frac{1}{2})(\alpha_C + \alpha_P) \hbar \omega_L + \alpha_C \hbar \omega_L - E_B(\nu P) - \hbar \omega_1 - \hbar \omega_2. \]

As discussed in section 4.2 the excitonic effects are only with the lowest Landau level, \( N = 0 \) (1), of the lowest subband in the case of left (right) circular polarization for
which the argument of the delta function is

$$E_g(\nu P) \pm \alpha C \hbar \omega_L - \hbar \omega_1 - \hbar \omega_2$$

(4.3.14)

where

$$E_g(\nu P) = E_g + E_{oc} + E_{av} + (\frac{\ell}{2}) (\alpha_C + \alpha_v) \hbar \omega_L - E_B(\nu P)$$

(4.3.15)

with $\ell = 1 (3)$ for LCP (RCP) and plus (minus) sign corresponds to LCP (RCP).

Now, substituting equations (4.3.12), (4.3.13) in equation (4.3.11) and replacing the Dirac - delta function by Lorentzian distribution function, we get for TPMEA coefficient

$$\alpha^\pm = \alpha' \left| \sum_{\nu, \nu} (\alpha_{\nu} + \delta_{\nu, 0}) I(\alpha_{\nu}) \left[ (e_{\nu} . P_{CV}) \left\{ E_B(\nu P) + \hbar \omega_1 \right\}^{-1} \right. \right.$$

$$+ \left. (e_{\nu} . P_{CV}) \left\{ E_B(\nu P) + \hbar \omega_2 \right\}^{-1} \right]^{2} \right. \left( \frac{\Gamma}{\pi} \right)$$

$$\times \frac{E_g(\nu P) \pm \alpha_C \hbar \omega_L - \hbar \omega_1 - \hbar \omega_2}{(E_g(\nu P) \pm \alpha_C \hbar \omega_L - \hbar \omega_1 - \hbar \omega_2)^2 + \Gamma^2},$$

(4.3.16)

where all the symbols have the same meaning as defined in section (3.3).

For $(\hbar \omega_1 + \hbar \omega_2) > E_g + E_{ne} + E_{nh} + \frac{\ell}{2} (\alpha_C + \alpha_v) \hbar \omega_L$, we get free electron - hole transitions which are treated in chapter III.

From equation (4.3.14) it is evident that the threshold
energy for the excitonic absorption is given by

\[
(h\omega_1 + h\omega_2)_{\text{th}} = E_g + E_{oc} + E_{ov} + \frac{\epsilon}{2}(\alpha_c + \alpha_v)h\omega_L \pm \alpha_c h\omega_L - E_B(\nu P)
\]

(4.3.17)

Hence the peaks are expected to show up in the TPMEA spectrum whenever the frequency condition given by equation (4.3.14) is satisfied. The exciton energy is characterized by the set of quantum numbers \((n_e, n_h, m, \nu)\). Thus what in an ordinary magnetoabsorption spectrum is supposed to show up as one Landau peak is now resolved into an infinite set of lines which reflects the discrete character of the new quantum number. Thus although it appears that, as the threshold condition is satisfied for \(P\) excitons corresponding to different \(\nu\) values, the lines should appear in TPMEA spectrum at each of these threshold values, experimentally observed lines correspond to the lowest value of \(\nu\) with largest OS. As such only the lowest line is expected to show up in the entire spectrum.

From equation (4.3.14) it follows that the threshold for the excitonic transition is lowered by the BE of the excitons allowed in the transition. Hence the TPMEA studies enable the determination of BE of the exciton from the difference between the threshold values corresponding to the excitonic and the free
electron - hole transition.

In the above treatment excitons are assumed to be of quasi two - dimensional character. The amount of 2D nature of the exciton in a layer is determined by the ratio \( L/a_o \), where \( a_o \) is the bulk exciton diameter. For very thick layers \( (L/a_o \gg 1) \) the excitons are nearly 3D whereas for thin layers \( L/a_o < 1 \) and they approach their 2D limit. For extremely thin layers, however, the leakage of the wave function into the barrier regions eventually causes the exciton to look more 3D with an attendant fall in its BE. It is found that the enhancement of BE is substantial only in the range \((1/4) < L/a_o < 1\) [36]. For GaAs and for the well width of the specimen of Fröhlich et al \((L/a_o) \sim 0.4\). Hence the pronounced excitonic effects are expected in the specimen of Fröhlich et al.

Since the BE of the 2P exciton (unresolved) appears as a parameter in the expression for the absorption coefficient we now proceed to calculate it.

4.4 Binding Energy of 2P Exciton in a Quantizing Magnetic Field:

BE of the ground state of both the LH and HH excitons are calculated by Greene and Bajaj [19] in the presence of an arbitrary magnetic field, treating the valence band structure in terms of two decoupled LH and HH bands. This decoupled nature in
narrow GaAs QWs is demonstrated by interband magnetoreflectivity observations [37]. Yang and Sham [22] and Bauer and Ando [23] take account of the effect of magnetic field together with the valence band mixing. These authors have shown that mixing of the valence band structure is important in interpreting the experimental magneto-optical results. Bauer and Ando use, as their set of basis functions, the hydrogenic functions to calculate the excitonic energies which limits the validity of their calculations to low fields while Yang and Sham have performed their calculations using a set of Landau level wave functions as basis functions.

Various approximation methods have been used to determine exciton energies and wave functions both in the absence and presence of magnetic fields. In the following we calculate the BE of 2P exciton employing two different techniques viz.,

1. Variational method following the approach of Greene et al [19]

2. Perturbation method applicable to the excited states [34]

4.4 a) Variational Method

This procedure results in our variational BE's being rigorous upper bounds to the true BE's.

We use the complete basis function
\[ \psi_1(\rho, z, \phi) = f_{n_e}^C f_{n_h}^V \rho^{\left| m \right|} e^{im\phi} G_1(\rho, z) \]  

(4.4.1)

as the trial function, with

\[ G_1(\rho, z) = z^n e^{-\alpha_1 z^2} e^{-(\alpha_i + \delta)^2} \rho^2 \]  

(4.4.2)

The coulombic term essentially binds the electron - hole reduced motion in the plane of the layer plane, as described by the exponential term, whereas the exciton motion along the growth axis is mostly dominated by the confining potentials at \( z = -L/2 \) and \( z = L/2 \) corresponding to the one particle wave function \( f_{n_e}(z) \) and \( f_{n_h}(z) \).

Using the trial functions given by equation (4.4.1) and the Hamiltonian given by equation (4.3.4), the energy eigen values are obtained by solving the following determinantal equation

\[ \det \left( H - E \Delta \right) = 0 \]  

(4.4.3)

The expression thus obtained is minimized variationally and the lowest of the eigen values thus obtained gives an upper bound to the true eigen function corresponding to the Hamiltonian (4.3.4).
In equation (4.4.3), $H$ and $\Delta$ are the Hamiltonian and overlap matrices with the corresponding matrix elements given, respectively, by

$$H_{ij} = \int \psi_i^* H \psi_j \, d\tau$$ \hspace{1cm} (4.4.4a)

$$\Delta_{ij} = \int \psi_i^* \psi_j \, d\tau$$ \hspace{1cm} (4.4.4b)

The condition given in equation (4.3.9) renders Hamiltonian and overlap matrices to be two subscript quantities which in the absence of that condition are 4-subscript quantities of the form

$$H_{ij}^{i', j'} = \int \psi_i^* H \psi_{i', j'} \, d\tau$$

The quantities $i$ and $j$ in the equation (4.4.4) are chosen large enough to ensure convergence.

The BE of the $P$-exciton associated with the lowest Landau level of the lowest subband is then given by

$$E_B = E_{OC} + E_{OV} + \frac{\epsilon}{2} \left( \alpha_c^2 + \alpha_v^2 \right) \hbar \omega_L \pm \alpha_c \hbar \omega_L - E$$ \hspace{1cm} (4.4.5)

In equation (4.4.5), $E$ is the eigen value of the Hamiltonian
corresponding to the level under consideration. In the absence of
the coulomb interaction, the total energy $E$ is just the sum of
$E_{oc}$ and $E_{ov}$ and the Landau level energy. The coulomb interaction
between the electron and the hole lowers $E$ and leads to the
formation of an exciton.

The values of $E_{oc}$ and $E_{ov}$ are obtained by numerically solving the following transcendental equations.

\[
\left[ \frac{E_{oc}}{V_e} \right]^{1/2} = \cos \left\{ \left[ \frac{m^*}{2h} \frac{E_{oc}}{L} \right] \right\}
\]

(4.4.6)

\[
\left[ \frac{E_{ov}}{V_h} \right]^{1/2} = \cos \left\{ \left[ \frac{m^\pm}{2h} \frac{E_{ov}}{L} \right] \right\}
\]

The Hamiltonian and overlap matrix elements are obtained using $\psi_i$'s given by equation (4.4.1). The Hamiltonian matrix element is
found to be

\[
H_{ij} = 2\pi \left\{ |\lambda_e|^2 |\lambda_h|^2 \left[ \frac{1}{(\alpha_{ij\delta})^2} \left( \frac{A}{\alpha_{ij\delta}} + \frac{B}{2} \right) \sigma_0 - \left( \frac{1}{m_e} + \frac{1}{m_h} \right) \right] \times \right. \\
\times \frac{\mu_s \alpha_i^2}{(\alpha_{ij\delta})^2} \left[ 3\sigma_1 - 2\sigma_2 \right] + \left[ \frac{1}{(\alpha_{ij\delta})^2} \left( \frac{A}{\alpha_{ij\delta}} + \frac{B'}{2} \right) \sigma_0 + \sigma_0 \right]
\]

\[ 97 \]
\[ + \left( \frac{1}{m_e} + \frac{1}{m_t} \right) \frac{2\mu_\pi \alpha_j^2}{\alpha_{ij}^2} \kappa_1 + \frac{\mu_\pi \alpha_j}{\alpha_{ij}^2} \left\{ \frac{k_e}{m_e} \chi + \frac{k_h}{m_t} \chi' \right\} - 2\sigma_3 \]

(4.4.7)

where

\[ \sigma_m = \int dz_e \int dz_h z^m + 2q \exp(2k_e z_e) \exp(2k_h z_h) \exp\left\{ - (\alpha_{ij})z^2 \right\}, \]

for \( m = 0, 1 \)

\[ \sigma_n = \int dz_e \int dz_h z^n + 2q \cos^2(k_e z_e) \cos^2(k_h z_h) \exp\left\{ - (\alpha_{ij})z^2 \right\}, \]

\[ \kappa_2 = \int dz_e \int dz_h z^{2q} \exp(2k_e z_e) \exp(2k_h z_h) \exp\left\{ - (\alpha_{ij})z^2 \right\} \times \]

\[ \times \left( \frac{1}{(\alpha_{ij})^{3/2}} \left\{ 2z^2 \text{erfc}(z) - \Gamma(3/2, \alpha_{ij} z^2) \right\} \right) \]

\[ \sigma_3 = \int dz_e \int dz_h z^{2q} \exp(2k_e z_e) \exp(2k_h z_h) \times \]

\[ \times \exp\left\{ - (\alpha_{ij})z^2 \right\} \left( \frac{1}{(\alpha_{ij})^{-3/2}} \left\{ 2z^2 \text{erfc}(z) - \Gamma(3/2, \alpha_{ij} z^2) \right\} \right), \]

\[ \chi = \int dz_e \int dz_h z^{2q + 1} \sin(2k_e z_e) \cos^2(k_h z_h) \exp\left\{ - (\alpha_{ij})z^2 \right\} \]

\( \chi' \) is obtained from \( \chi \) by interchanging \( k_e \) and \( k_h \).
In the above equations,

\[
\alpha_{ij} = \alpha_i + \alpha_j; \quad \alpha_{ij\delta} = \alpha_{ij} + 2\delta; \quad z = z_e - z_h
\]

\[
A = \frac{1}{4} \gamma^2 - 2 \alpha_{ij\delta}
\]

\[
B = 4 \alpha_{ij\delta} + \frac{\mu_+}{m} (\alpha_{ij} + k_e^2) + \frac{\mu_-}{m} (\alpha_{ij} + k_h^2) + \gamma
\]

\[
B' = 4 \alpha_{ij\delta} - \frac{\mu_+}{m} (-\alpha_{ij} + z_e^2) - \frac{\mu_-}{m} (-\alpha_{ij} + z_h^2) + V_e + V_h + \gamma
\]

erfc is the complementary error function and \( \Gamma \) is the incomplete Gamma function.

The overlap matrix element \( \Delta_{ij} \) is given by

\[
\Delta_{ij} = \pi \frac{1.3.5...(2|m|-1)}{2|m| \alpha_{ij\delta}} \sqrt{\frac{\pi}{\alpha_{ij\delta}}}
\]

\[
\times \int dz_e \int dz_h \, z_e^2 \, |f_{n_e}^{c}(z_e)|^2 \, |f_{n_h}^{v}(z_h)|^2 \, \exp\left\{ - \alpha_{ij} z_e^2 \right\}
\]

(4.4.8)
4.4 b) **Perturbation Method**:

It treats the coulomb interaction between the electron-hole pair as a perturbation on the free Landau levels in the magnetic field.

For the perturbative method we rewrite the total Hamiltonian as follows.

\[
H = H_0 + H' = H_e + H_h + H_B^0 + H'
\]

(4.4.9)

where,

\[ H_e(z_e) = -\frac{\mu_{\pm}}{m} \frac{\partial^2}{\partial z_e^2} + V_e(z_e) \]  

(4.4.9 a)

\[ H_h(z_h) = -\frac{\mu_{\pm}}{m_{\pm}} \frac{\partial^2}{\partial z_h^2} + V_h(z_h) \]  

(4.4.9 b)

\[ H_B^0 \text{ is the magnetic field dependent part of the Hamiltonian given by} \]

\[
H_B^0 = \left[ \frac{1}{\rho} \frac{\partial}{\partial \rho} \rho \frac{\partial}{\partial \rho} + \frac{1}{\rho^2} \frac{\partial^2}{\partial \phi^2} - \gamma L_z - \frac{1}{4} \rho^2 \right], \]  

(4.4.9 c)

\[ H' \text{ is a perturbation for the system given by} \]
\[
H' = \frac{-2}{\\left(\rho^2 + (z_e - z_h)^2\right)^{1/2}}
\]  

(4.4.9 d)

In the above equations all energies are in units of Rydbergs, lengths in units of Bohr radius and angular momenta are in units of \(\hbar\).

The solutions for unperturbed part \(H_0\) are exactly known. The eigenvectors for the unperturbed Hamiltonian \(H_0\) are

\[
\psi_{\nu,m} = f_{n_e}^C(z_e) f_{n_h}^V(z_h) \psi_{\nu,m}^B(\rho, \phi)
\]

(4.4.10)

where \(f_{n_e}^C(z_e)\) and \(f_{n_h}^V(z_h)\) are given by equations of the type (4.3.6), and

\[
\psi_{\nu,m}^B(\rho, \phi) = \frac{1}{\sqrt{2\pi}} \exp(i m \phi) \left( \frac{1}{\alpha_c} \right) \left( \frac{(\nu - 1)!!}{2 \left( \nu + |m| - 1 \right)!} \right)^{1/2} \times
\]

\[
\times \exp\left(-\frac{\rho^2}{4\alpha_c^2}\right) \rho^{|m|} L_{\nu + |m| - 1}^{\frac{1}{2}}(\rho^2/2\alpha_c^2)
\]

(4.4.10 a)

Here \(L_{\nu + |m| - 1}^{\frac{1}{2}}(\rho^2/2\alpha_c^2)\) are the associated Laguerre polynomials

while \(|m| < \nu\) and \(\alpha_c = \left\lceil \frac{\hbar}{\mu \omega_c} \right\rceil^{1/2}\)

101
The corresponding eigen energies are given by

\[ E = E_{oc} + E_{ov} + E_{\nu,m} \]

\[ = E_{oc} + E_{ov} + \hbar \omega_c \left( \nu + \frac{|m|}{2} + \frac{m}{2} - \frac{1}{2} \right) \]  
(4.4.11)

where \( \nu = 1, 2 \ldots \)

The solutions for the unperturbed part \( H_0 \) are exactly known. From equation (4.4.11) we see that the \( \nu = 2 \) state of the exciton is two fold degenerate corresponding to \( m = 0 \) and \( m = -1 \). From equation (4.4.10), the eigen functions corresponding to the two degenerate energy eigen values are

\[ \chi_1 = f_{n_e}^C(z_e) f_{n_h}^V(z_h) \frac{1}{\sqrt{2\pi}} \frac{1}{\alpha_c} \exp \left( -\frac{\rho^2}{4\alpha_c^2} \right) L^0_1(\frac{\rho^2}{2\alpha_c^2}) \]  
(4.4.12 a)

and

\[ \chi_2 = f_{n_e}^C(z_e) f_{n_h}^V(z_h) \frac{1}{\sqrt{2\pi}} \frac{1}{4\alpha_c^2} \times \rho \exp(i\phi) \exp \left( -\frac{\rho^2}{4\alpha_c^2} \right) L^1_2(\frac{\rho^2}{2\alpha_c^2}) \]  
(4.4.12 b)

Let the required linear combination be

\[ \psi^{(0)} = c_1 \chi_1 + c_2 \chi_2 \]  
(4.4.13)
The first order correction to eigen value is obtained by solving

\[
\begin{vmatrix}
H_{11} - E_1 & H_{12} \\ H_{21} & H_{22} - E_1
\end{vmatrix}
= 0, \quad (4.6.14)
\]

where

\[
H_{mn} = \langle \chi_m | H' | \chi_n \rangle \quad (4.4.15)
\]

For the levels under consideration (lowest subband electron and subband hole, \( n_e = 1, n_h = 1 \), \( \nu = 2 \), \( m = 0 \)), the explicit expression for the first order matrix elements is given in appendix D.

The second order correction is obtained by solving

\[
\sum_n \left| \frac{\langle \psi_{2,0} | H' | \psi_{n,0} \rangle}{E_{2,0} - E_{n,0}} \right|^2 - \left( \sum_n \left| \frac{\langle \psi_{2,0} | H' | \psi_{n,0} \rangle \langle \psi_{n,0} | H' | \psi_{2,-1} \rangle}{E_{2,0} - E_{n,0}} \right| \right)
\]

\[
\sum_n \left| \frac{\langle \psi_{2,-1} | H' | \psi_{n,-1} \rangle \langle \psi_{n,-1} | H' | \psi_{2,0} \rangle}{E_{2,0} - E_{n,0}} \right|^2 - \left( \sum_n \left| \frac{\langle \psi_{2,-1} | H' | \psi_{n,-1} \rangle}{E_{2,0} - E_{n,0}} \right| \right)^2 - E_2 = 0 \quad (4.4.16)
\]

The prime on \( \sum \) indicates omission of the summation over \( n = 2 \).
In the second order correction we have calculated analytically the corrections up to the first three leading terms. The expression for the matrix elements appearing in equation (4.4.16) are as under:

\[
\sum_n \frac{\langle \Psi_{2,0} | H' | \Psi_{n,0} \rangle}{E_{2,0} - E_{n,0}} = \frac{\langle \Psi_{2,0} | H' | \Psi_{1,0} \rangle}{E_{2,0} - E_{1,0}} + \frac{\langle \Psi_{2,0} | H' | \Psi_{3,0} \rangle}{E_{2,0} - E_{3,0}} + \frac{\langle \Psi_{2,0} | H' | \Psi_{4,0} \rangle}{E_{2,0} - E_{4,0}}
\]

\[
\sum_n \frac{\langle \Psi_{2,-1} | H' | \Psi_{n,-1} \rangle}{E_{2,-1} - E_{n,-1}} = \frac{\langle \Psi_{2,-1} | H' | \Psi_{1,-1} \rangle}{E_{2,-1} - E_{1,-1}} + \frac{\langle \Psi_{2,-1} | H' | \Psi_{3,-1} \rangle}{E_{2,-1} - E_{3,-1}} + \frac{\langle \Psi_{2,-1} | H' | \Psi_{4,-1} \rangle}{E_{2,-1} - E_{4,-1}}
\]

The explicit expressions for the different matrix elements appearing in the above equation are given in appendix E. Contribution from higher order terms is found to be insignificant. The BE of the exciton is given by

\[
E_B = -E_1 - E_2.
\]
4.5 Results and discussion

We present the numerical calculations for the specimen of Frohlich et al [32]. For the energy range of radiation used in their experiment only $n_e = n_h = 1$ subbands are involved in the transition. Since only the transitions allowed with lowest Landau levels are influenced by excitonic effects, in their experiment, we consider only $1 \rightarrow 0 (0 \rightarrow 1)$ inter Landau level transition in our calculations for left (right) circular polarization (In these transitions first and second numbers represent Landau levels in CB and VB, respectively). The values of different quantities used in our calculations are $E_g = 1.518 \, \text{eV}$, $m_c = 0.07 \, m_0$, $m_v = 0.22 \, m_0$, $s = 12.5$, $L = 84.5 \, \tilde{A}$, $\gamma_1 = 7.36$ and $\gamma_2 = 2.57$. The values of $m_c$ and $m_v$ are determined from peak positions for left and right circular polarization and subband energy $(E_{oc} + E_{ov}) = 65 \, \text{meV}$ is treated as parameter to fit data of Frohlich et al involving higher Landau level transition. (See section 3.4.).

In calculating $V_e$ and $V_h$, in equation (4.4.6) we have assumed the CB and VB discontinuities at the interface between the two semiconductors at the $\Gamma$ point to be 85% and 15%, respectively, of the total band gap difference $\Delta E_g$, the expression for which in terms of aluminium mole fraction $x$ of $\text{Al}_x\text{Ga}_{1-x}\text{As}$ is given by $\Delta E_g = 1.155x + 0.37x^2$, in units of eV.

From equation (4.3.16) we find that absorption peaks arise
for the photon energy given by equation \( E_g(2\text{P}) \pm \alpha \hbar \omega \). Treating \( E_B(2\text{P}) \) in equation (4.3.15) as a fitting parameter we have numerically evaluated two-photon threshold energy \( (\hbar \omega_1 + \hbar \omega_2)_\text{th} \), as a function of magnetic field. In figure 4.1 we have shown \( (\hbar \omega_1 + \hbar \omega_2)_\text{th} \) as a function of magnetic field. The broken curve is due to our calculations with excitonic effects and the continuous curve is without excitonic effects. Points are experimental values of Fröhlich et al. Figure 4.1a) and 4.1b) are, respectively, due to left and right circular polarizations. We obtain a very good fit for the range of the magnetic field 3.3 - 6.6 T. The BEs of 2P excitons thus obtained by fitting to the data, for different magnetic fields are given in Table I. We find that the BE of 2P excitons increases with increasing magnetic field, as expected from physical grounds, due to the increased confinement provided by larger magnetic fields. The separation between the broken and continuous curve goes on increasing with increasing magnetic field indicative of the increase in BE with increasing magnetic field.

We have also calculated absorption coefficient as a function of two-photon energy for different magnetic fields and is shown in figure 4.2 for left circular polarization. We find that absorption coefficient increases with increasing magnetic field. Since absorption coefficient is proportional to oscillator strength \( |F(0)|^2 |e \cdot p_{cv}|^2 \), which increases with magnetic field resulting in increase in the intensity of the excitonic peaks, a
Fig. 4.1 Variation of threshold energy as a function of magnetic field for the specimen of Fröhlich et al [32]. Closed circles represent experimental data. Continuous (broken) curve represents our calculations without excitons (with excitons). In 1-0 and 0-1 transitions first and second numbers represent the Landau levels in the conduction band and valence band, respectively.

a) Due to left circular polarization.
b) Due to right circular polarization.
Fig. 4.2 Two-photon magnetoexcitonic absorption spectrum for the specimen of Frohlich et al. [27] as a function of photon energy for left circular polarization at different magnetic fields.

a) $B = 3\ T$  
b) $B = 4\ T$  
c) $B = 5\ T$
behaviour similar to that observed in the case of free-electron hole transitions.

Figure 4.3 shows the two-photon excitonic absorption spectrum at $B = 5.5 \, \text{T}$ for the specimen of Fröhlich et al as a function of two-photon energy. Curve a is due to our calculation. Closed circles represent experimental data. A good fit is obtained with Lorentzian of width $\Gamma = 2 \, \text{meV}$. The curve b is without excitonic effect. Figures 4.3 a) and 4.3 b) are due to left and right circular polarizations, respectively. The figures show the decrease in the threshold energy for absorption due to excitonic effects. The difference between the peak positions of the two curves in each figure 4.3 a) and 4.3 b) gives the binding energy of the 2P exciton at magnetic field of 5.5 T.

We have calculated the BE of the unresolved 2P - excitons for different magnetic fields using both variational method and the perturbation technique for the values of the specimen of Fröhlich et al for $B = 5.5 \, \text{T}$. We have employed the basis functions given by equation (4.4.1) in the variational calculations and have gone up to the fifth order determinantal equations in Hamiltonian and overlap matrices. Hamiltonian and overlap matrices of still higher order do not change the value of BE appreciably. We have used the values of $\alpha_j$ corresponding to 2P excitons given in ref [35]. The calculated values of BE are given in Table I along with the BEs extracted by fitting $(\hbar \omega_1 + \hbar \omega_2)_\text{th}$.
Fig. 4.3 a) Two-photon magnetoabsorption spectrum as a function of two-photon energy for the specimen of Fröhlich et al [27] at $B = 5.5$ T for left circular polarization. The circles represent the experimental data of Fröhlich et al. Curve a (Curve b) is with excitons (without excitons).
Fig. 4.3 b) Two-photon magnetoabsorption spectrum as a function of two-photon energy for the specimen of Fröhlich et al [27] at $B = 5.5$ T for right circular polarization. The circles represent the experimental data of Fröhlich et al. Curve a (Curve b) is with excitons (without excitons).
to the experimental data for comparison.

Table I

BE of 2p excitons in a magnetic field

<table>
<thead>
<tr>
<th>B</th>
<th>BE of 2p excitons (in meV)</th>
<th>BE calculated using variational method (q = 0) (in meV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>4 T</td>
<td>2.63</td>
<td>3.201</td>
</tr>
<tr>
<td>5 T</td>
<td>3.00</td>
<td>3.810</td>
</tr>
<tr>
<td>6 T</td>
<td>3.73</td>
<td>4.782</td>
</tr>
</tbody>
</table>

As can be seen from Table I both the calculated BE and BE extracted from the experimental data increase with the increasing magnetic field. This is expected because finite magnetic fields compress the excitonic wave functions by reducing the cyclotron radius compared to the exciton Bohr radius. With the increase in magnetic field the exciton wave function is more compressed in the QW leading to increased binding. The BE calculated using perturbation technique is in close agreement with that deduced from the experimental data. It is found that the difference in
the values of the BE calculated variationally and those deduced from the experimental data vary from 57 % to 25 % when the order of the matrix is increased from 3 to 5. The exciton energies converge then onwards. The discrepancy between the experimental values and our theoretical values is attributed to the oversimplified model used by us, wherein screening of the coulomb interaction, complex nature of the VB etc are neglected (which is beyond the scope of this work).

The accuracy of the results with the variational method depends to a large extent on the form of the trial wave function chosen. However a set of Gaussians in \( \rho \) and \( z \) variables is assumed to provide a better description of the exciton system throughout the range of the magnetic field considered here [38]. The variation of the expectation value of the ground state energy of a purely two dimensional exciton in a magnetic field is given by Marasas and Spector [38] where the hydrogenic wave function is found to be more favourable for magnetic fields corresponding to the values of \( \gamma < 10 \). In their approach the \( z \) dependence of the coulomb interaction is relaxed as the excitons are assumed to be purely two dimensional. However, it is well known from calculations of energy levels of hydrogenic atoms in the 3D case that the best variational wave functions change their nature from being a set of hydrogenic functions at low fields to being a set of Gaussian functions at high fields [39].
In figure 4.4 we have shown the energy of the purely two-dimensional P-exciton as a function of the normalized magnetic field $\gamma$ using a hydrogenic variational wave function (dashed curve) and a Gaussian variational wave function (continuous curve) with the corresponding BE being shown in figure 4.5 choosing the best lower bound at a particular field. We see from the graph that the Gaussian is a more desirable wave function throughout the entire range of the magnetic field considered.

Table II

<table>
<thead>
<tr>
<th>BE of 2p excitons in a magnetic field</th>
</tr>
</thead>
<tbody>
<tr>
<td>BE</td>
</tr>
<tr>
<td>-----</td>
</tr>
<tr>
<td>$4 \text{T}$</td>
</tr>
<tr>
<td>$5 \text{T}$</td>
</tr>
<tr>
<td>$6 \text{T}$</td>
</tr>
</tbody>
</table>

In addition to the BE, expectation values of $|z_e - z_h|$ and $\rho^2$ are relevant quantities which are helpful to ascertain the
Energy of a purely two-dimensional $P$-exciton as a function of the normalized magnetic field $\gamma$.

Dashed curve: Hydrogenic variational wave function.
Continuous curve: Gaussian variational wave function.
Points are shown to guide the eye.
Fig. 4.5  Binding energy of a purely two-dimensional $P$-exciton as a function of the normalized magnetic field $\gamma$ choosing the best lower bound at a particular field. Points are shown to guide the eye.
accuracy of the trial wave function as they provide some insight into its spatial extension. Even a one parameter wave function yields results which are accurate as long as the wells are deep. Normally the choice is between separable and the non-separable wave function depending on the well thickness and then between Gaussian and hydrogenic depending on the strength of the magnetic field. Accordingly there are four different wave functions to tackle. We investigate the thickness and the magnetic field dependence of the two quantities, the lateral extension of the exciton \( \left( \langle \rho^2 \rangle \right)^{1/2} \) and the extension in the growth direction \( \left( \langle z^2 \rangle - \langle z_h^2 \rangle \right)^{1/2} \), in units of Bohr radius, for the following four different wave functions.

\[
\Psi_1(\rho, z, \phi) = f_{n_e}(z_e) f_{n_h}(z_h) \rho^m e^{i m \phi} G_1(\rho, z)
\]

\[G_1(\rho, z) = e^{-\alpha_1 z^2} e^{-(\alpha_1 + \delta) \rho^2}\] Gaussian separable (q = 0) \hspace{1cm} (4.5.1 a)

\[G_1(\rho, z) = z e^{-\alpha_1 z^2} e^{-(\alpha_1 + \delta) \rho^2}\] Gaussian separable (q = 1) \hspace{1cm} (4.5.1 b)

\[G_1(\rho, z) = e^{-\alpha_1 (z^2 + \rho^2)^{1/2}}\] Hydrogenic non-separable \hspace{1cm} (4.5.1 c)
\[ G_i(\rho, z) = e^{-\alpha_i \rho} e^{-(\alpha_i + \delta) \rho} \]

Hydrogenic separable

(4.5.1 d)

The expressions for \( (\langle \rho^2 \rangle)^{1/2} \), for four different wave functions, are given by

\[ \left( \langle \rho^2 \rangle \right)^{1/2} = \left[ \frac{1}{2} \left( |m| + 1 \right) \frac{1}{(\alpha_j + \delta)} \right]^{1/2} \] (4.5.2 a)

\[ \left( \langle \rho^2 \rangle \right)^{1/2} = \left[ \frac{(|m| + 1)(2|m| + 3)}{2(\alpha_j + \delta)^2} \right]^{1/2} \] (4.5.2 b)

\[ \left( \langle \rho^2 \rangle \right)^{1/2} = |A|^{-2} \left[ \frac{\Gamma_{m+2}}{\sqrt{\pi \alpha_i |m| + 3/2}} \right] \int_{z_e} dz_e \int_{z_h} dz_h \times 
\]
\[ \times \left| f_{n_e}^{\psi}(z_e) \right|^2 \left| f_{n_h}^{\psi}(z_h) \right|^2 \mathcal{K} |m| + 5/2 \left( 2\alpha_i (z_e - z_h) \right) \] (4.5.2 c)

\[ \left( \langle \rho^2 \rangle \right)^{1/2} \] is same for wave function given by equations (4.5.2 a) and (4.5.2 b).

The expressions for \( \left( \langle (z_e - z_h)^2 \rangle \right)^{1/2} \) for the first three wave functions are straightforward. For the hydrogenic non-separable wave function it is given by
\[
\frac{1}{2} = |A|^{-2} \left( \frac{2}{\alpha} \right) \left( \frac{\alpha}{\alpha_i} \right)^{1/2} \int dz_n(z_e - z_h) |m| + 7/2 \times
\]
\[
\times |f_n(z_e)|^2 |f_{n}\', (z_h)|^2 \kappa |m| + 3/2 \left( 2\alpha_i(z_e - z_h) \right)
\]

where

\[
|A|^{-2} = \left( \frac{\alpha}{2} \right) \left( \frac{\alpha}{\alpha} \right)^{1/2} \int dz_e \int dz_n(z_e) \left( f_n(z_e) \right)^2 \left( f_{n}\', (z_h) \right)^2 \times
\]
\[
\times (z_e - z_h)^{|m| + 3/2} \kappa |m| + 3/2 \left( 2\alpha_i(z_e - z_h) \right)
\]

with \( \kappa |m| + 3/2 \) is the modified Bessel function of order \(|m| + 3/2\).

We examine the behaviour of \( \left( \frac{1}{2} \right)^{1/2} \) and \( \left( \frac{1}{2} \right)^{1/2} \) in the two limiting cases as \( L \to 0 \) and \( L \to \infty \), respectively and compare them respectively with the purely 2D and 3D cases. In figures 4.6 and 4.7, we have shown the extension in the \( xy \)-plane \( \left( \frac{1}{2} \right)^{1/2} \) and the extension in the \( z \)-direction, \( \left( \frac{1}{2} \right)^{1/2} \), respectively, for the Hydrogenic separable and non-separable wave functions. Both the wave functions extrapolate to the exact 2D value of \( \sqrt{3/8} a_B \) at vanishing \( L \) and \( B \). Also, at \( B = 5.5 \) T, both the wave functions exhibit a similar behaviour for the values of \( L \) below 1.5, in units of Bohr radius, extrapolating to the value of 0.742 very close to the exact value of 0.75, at \( B = 5.5 \) T in a purely 2D case. On the other hand at large \( L \), the Hydrogenic separable wave function diverges whereas the non-
Fig. 4.6 Thickness dependence of the quantities $\left(\langle \rho^2 \rangle \right)^{1/2}$ in units of Bohr radius.

Curve a: Hydrogenic separable wave function.

Curve b: Hydrogenic non-separable wave function.
Fig. 4.7 Thickness dependence of the quantities $\left(\langle z_e - z_h \rangle^2 \right)^{1/2}$ in units of Bohr radius.

Curve a: Hydrogenic separable wave function.
Curve b: Hydrogenic non-separable wave function.
 separable wave function tends to converge to a 3D limit. This behaviour is attributed to the fact that the non-separable exponential factor in the spatial coordinates of equation (4.4.1) ensures some amount of coulomb binding even when the QW is very wide as compared to the variable separated factor. Thus the separable wave function is correct only for thin well thicknesses whereas the non-separable wave function provides a suitable interpolation between the limits of thin and thick wells. Thus we conclude that for the well thicknesses and the magnetic fields considered in our calculations the separable wave function can equally well describe the exciton formation as non-separable wave function. Similar behaviour is observed for the extension in the z-direction $\langle(z_e - z_h)^2\rangle^{1/2}$ (figure 4.7) where for large $L$, hydrogenic separable tends to diverge in contrary to the non-separable wave function converging to a bulk limit. The extension in the z-direction, $\langle(z_e - z_h)^2\rangle^{1/2}$, increases linearly with $L$ for the hydrogenic separable wave function but for the non-separable wave function it departs from the straight line for $L \geq 0.7 a_B$ and smoothly reaches the exact 3D value $3/4 a_B$ at large $L$.

In figure 4.8 we have shown the magnetic field dependence of the dimensionless quantity $\langle\rho^2\rangle^{1/2}$ for $L = 84.5 \AA$. From figure 4.8 we see that both the wave functions extrapolate to the exact 2D value at vanishing $B$. At large magnetic fields Gaussian is more closer to the exact value of 0.68. From figure 4.8 it can be
Fig. 4.8 Magnetic field dependence of the quantities \( \langle \phi^2 \rangle^{1/2} \) in units of Bohr radius for \( L = 84.5 \) Å.

Curve a: Hydrogenic separable wave function.
Curve b: Gaussian separable wave function with \( q = 0 \).
concluded that the hydrogenic wave function is correct only for thin wells and small magnetic fields whereas the Gaussian provides a suitable interpolation between the limits of thin and thick wells and small and large magnetic fields. Thus, the Gaussian trial wave function (4.5.1a) and (4.5.1b) used in our BE calculations are more appropriate than the hydrogenic wave function. Moreover, as expected from physical grounds the difference between the wave functions is reflected more in the spatial extent of the wave function than in energies. Thus, in order to overcome the cumbersome calculations involved, we employ Gaussian separable wave function in our BE calculations.

The lateral extension of the exciton increases monotonically with L as a consequence of the weakening of the effective in-plane attraction. In the z-direction the extensions approach asymptotically (as L increases) an effective 3D limit where the electron and the hole are confined to move in different planes at a distance L. The extension in the growth direction is an indicative of the degree of confinement of the functions $|f_{n_e}^{C}(z_e)|^2$ and $|f_{n_h}^{y}(z_h)|^2$; accordingly with that the larger overlap of two functions in equation (4.5.2) gives a larger value of the effective in-plane coulomb interaction and a shortening of the electron-hole average distance in the parallel direction. The ratio $\frac{\langle z \rangle^2}{\langle \rho \rangle^2}$ represents the degree of compression of the wave function along the z-axis by the potential wells and is equal
to 0.5 for the spherical ground state of bulk excitons. In figure 4.9 we have shown the variation of $\frac{\langle \rho \rangle^2}{\langle z \rangle^2}$ as a function of well width.

Since in the exciton formation the probability of finding the electron and the hole at the same point $r = 0$ is more, the wave function with the less spatial extension seems to describe the situation very well. In figures 4.10a) and 4.10b), the spatial extension of the wave function is shown for Gaussian and hydrogenic wave functions. From the figures it is clear that the Gaussian with the less spatial extension is the desirable function for the exciton formation. We have also shown the extension of the wave functions in the $\rho - z$ plane for the four wave functions given by equations (4.5.1a) through (4.5.1d) in figures 4.11a) - 4.11d), respectively. These figures also confirm that Gaussian with less spatial extension is desirable.

In figure 4.12 the variation of the coulomb integral $I$ as given by

$$I = \sum_{\ell} \left[ \frac{2}{\sqrt{\rho^2 + (z_e - z_h)^2}} \right] \left| f_{n_e}^{\ell}(z_e) \right|^2 \left| f_{n_h}^{\ell}(z_h) \right|^2 \left| G_{\ell}(\rho, z) \right|^2 d\tau$$

is plotted as a function of QW width at different magnetic fields. As expected on physical grounds the coulomb integral decreases exponentially as the well width is increased. This is
Fig. 4.9 Variation of dimensionless quantity $\frac{\langle \rho \rangle^2}{\langle z \rangle^2}$ as a function of well width for Gaussian wave function with $q = 0$. 

\[ \text{WELL-WIDTH (BOHR RADIUS)} \]
Fig. 4.10 a) Spatial extension of the Gaussian wave function with $q = 0$ in $x-y$ plane.
Fig. 4.10 b) Spatial extent of the Hydrogenic separable wave function in x-y plane.
Fig. 4

a) Spatial extension in the $\rho - z$ plane of the Gaussian wave function with $q = 0$. 

Gaussian ($q=0$)
Gaussian \((q=1)\)

Fig. 4.11 b) Spatial extension in the \(\rho - z\) plane of the Gaussian wave function with \(q = 1\).
Fig. 4.11 c) Spatial extension in the $\rho - z$ plane of the Hydrogenic separable wave function.
Fig. 4.11 d) Spatial extension in the $\rho - z$ plane of the Hydrogenic non-separable wave function.
**Fig. 4.12** Variation of coulomb integral as a function of well width at different magnetic fields.

Curve a : \( B = 5 \text{ T} \).

Curve b : \( B = 10 \text{ T} \).

Curve c : \( B = 20 \text{ T} \).
because the electron-hole overlap decreases as the well width is increased and hence the electron becomes more localized in to the well thereby the coulomb interaction between the electron and the hole is reduced considerably. As can be seen from curves a, b, and c, the value of I increases with increasing magnetic field as expected, due to the increase in confinement provided by the magnetic field. The value of I remains relatively constant beyond the well width \( L = 5 \) in units of Bohr radius.

In conclusion we have given a simple theory to account for the part of the TPMA spectrum affected by excitons for the specimen of Fröhlich et al. The calculated BE of 2P magnetoexciton at different fields are in reasonably good agreement with those extracted from TPMA spectra of Fröhlich et al. BE calculated by perturbation method are closer to the experimental values.
Appendix C

Deduction of Eigen Values of Energy In the Absence of Coulomb Interaction In a Magnetic Field

In equation (4.3.3) if we neglect the coulomb term for a moment, we recover the Schrödinger equation for a charged particle moving in an electromagnetic field if \( \mu = \mu' \). The eigen values associated with this problem are well known and are given by

\[
E = \left( \frac{e\hbar}{\mu} \right) B \left\{ \nu + \frac{1}{2} + \frac{|m|}{2} \left[ 1 \pm \frac{\mu^* - \mu}{\mu^* + \mu} \right] \right\} + E_{sn}
\]

\[(c1)\]

where \( E_{sn} \) corresponds to the energy of the nth subband in the sth band.

For \( \mu \) to be equal to \( \mu' \), \( m_\pm \to \infty \) and the above equation reduces to

\[
E = \left( \frac{e\hbar}{\mu^*} \right) B \left\{ \nu + \frac{1}{2} + \frac{|m|}{2} \right\} + E_{sn}.
\]

If we put \( N = \nu + \frac{|m|}{2} (1 \mp 1) \) we get,

\[
E = (N + \frac{1}{2}) \hbar \omega_c + E_{sn}
\]

\[(c2)\]

which takes the familiar form of the Landau energies. Hence each exciton level contributes all \( m \geq 0 \) states, with the same \( \nu \) to the Landau level.
Appendix D

Expressions for the First Order Matrix Elements In the Evaluation of the BE of 2P Magnetoeexciton Using Perturbation Theory

The explicit expressions for the first order matrix elements appearing in equation (4.4.14) are given by

\[ H_{11} = - \left( \frac{1}{8\alpha_c^2} \right)^{1/2} \int dz_e \int dz_h |f_{n_e}(z_e)|^2 |f_{n_h}(z_h)|^2 \exp(\beta^2) \left( \Gamma(5/2,\beta^2) - 2(\beta^2 - 1) \Gamma(3/2,\beta^2) + \sqrt{\pi} (\beta^2 + 1)^2 \text{erfc}(\beta) \right) \]

(d1)

\[ H_{22} = - \left( \frac{1}{32\gamma_2\alpha_c^3} \right) \int dz_e \int dz_h |f_{n_e}(z_e)|^2 |f_{n_h}(z_h)|^2 \exp(\beta^2) \left( \Gamma(11/2,\beta^2) - (5\beta^2 + 12) \Gamma(9/2,\beta^2) + 2(5\beta^4 + 24\beta^2 + 24) \Gamma(7/2,\beta^2) - 2(5\beta^6 + 36\beta^4 + 72\beta^2 + 36) \Gamma(5/2,\beta^2) + 2(5\beta^8 + 48\beta^6 + 144\beta^4 + 144\beta^2 + 36) \Gamma(3/2,\beta^2) - \sqrt{\pi} \beta^2 (\beta^8 + 12\beta^6 + 48\beta^4 + 72\beta^2 + 36) \text{erfc}(\beta) \right) \]

(d2)

In the above equations erfc is the complementary error function and \( \Gamma \) is the incomplete gamma function.
Appendix E

Expressions for the Second Order Matrix Elements In the Evaluation of the BE of 2P Magnetoexciton Using Perturbation Theory

The explicit expressions for the matrix elements appearing in equation (4.4.17) are given by

\[
\langle \Psi_{2,0} | H' | \Psi_{1,0} \rangle = - \left( \frac{1}{8 \alpha_c^2} \right)^{1/2} \int dz_e \int dz_h \left| f_{n_e}(z_e) \right|^2 \left| f_{n_h}(z_h) \right|^2 \\
\exp(\beta^2) \left\{ \gamma \Gamma(3/2, \beta^2) - \gamma \Gamma(1+\beta^2) \text{erfc}(\beta) \right\}
\]

\( (e1) \)

\[
\langle \Psi_{2,0} | H' | \Psi_{3,0} \rangle = - \left( \frac{1}{32 \alpha_c^2} \right)^{1/2} \int dz_e \int dz_h \left| f_{n_e}(z_e) \right|^2 \left| f_{n_h}(z_h) \right|^2 \\
\exp(\beta^2) \left\{ -\gamma \Gamma(7/2, \beta^2) - (5 + 3\beta^2) \Gamma(5/2, \beta^2) - 2(6 + 10\beta^2 + 3\beta^4) \Gamma(3/2, \beta^2) + \gamma \Gamma(2 + 6\beta^2 + 5\beta^4 + \beta^6) \text{erfc}(\beta) \right\}
\]

\( (e2) \)

\[
\langle \Psi_{2,0} | H' | \Psi_{4,0} \rangle = - \left( \frac{1}{288 \alpha_c^2} \right)^{1/2} \int dz_e \int dz_h \left| f_{n_e}(z_e) \right|^2 \left| f_{n_h}(z_h) \right|^2 \\
\exp(\beta^2) \left\{ \gamma \Gamma(9/2, \beta^2) - 2(5 + 2\beta^2) \Gamma(7/2, \beta^2) + 3(9 + 10\beta^2 + 2\beta^4) \Gamma(7/2, \beta^2) - 2(12 + 27\beta^2 + 15\beta^4 + 2\beta^6) \Gamma(3/2, \beta^2) + 2(6 + 24\beta^2 + 27\beta^4 + 10\beta^6 + \beta^8) \gamma \text{erfc}(\beta) \right\}
\]

\( (e3) \)
\[ \langle \Psi_{2,-1} | H' | \Psi_{1,-1} \rangle = - \left( \frac{1}{32a^3_c} \right) \int dz_e \int dz_h \left| f_{n_e}^+(z_e) \right|^2 \left| f_{n_h}^-(z_h) \right|^2 \]

\[ \exp(\beta^2) \left\{ - \Gamma(9/2, \beta^2) - 4(2 + \beta^2) \Gamma(7/2, \beta^2) - 6(3 + 4\beta^2 + \beta^4) \right. \]

\[ \Gamma(5/2, \beta^2) - 4(3 + 9\beta^2 + 6\beta^4 + \beta^6) \Gamma(3/2, \beta^2) - \forall \beta^2 \left( 12 + \\
\left. + 18\beta^2 + 8\beta^4 + \beta^6 \right) \text{erfc}(\beta) \right\} \]

(e4)

\[ \langle \Psi_{2,-1} | H' | \Psi_{3,-1} \rangle = - \left( \frac{1}{48\gamma^6 a^3_c} \right) \int dz_e \int dz_h \left| f_{n_e}^+(z_e) \right|^2 \left| f_{n_h}^+(z_h) \right|^2 \]

\[ \exp(\beta^2) \left\{ - \Gamma(13/2, \beta^2) + 6(3 + \beta^2) \Gamma(11/2, \beta^2) - (114 + 90\beta^2 + \\
15\beta^4) \Gamma(9/2, \beta^2) - 4(78 + 114\beta^2 + 45\beta^4 + 5\beta^6) \Gamma(7/2, \beta^2) - (360 + \\
936\beta^2 + 684\beta^4 + 180\beta^6 + 15\beta^8) \Gamma(5/2, \beta^2) + 6(24 + 120\beta^2 + 156\beta^4 + \\
764\beta^6 + 15\beta^8 + \beta^{10}) \Gamma(3/2, \beta^2) - \forall \beta^2 \left( 144 + 360\beta^2 + 312\beta^4 + \\
+ 114\beta^6 + \beta^8 \right) \text{erfc}(\beta) \right\} \]

(e5)

\[ \langle \Psi_{2,-1} | H' | \Psi_{4,-1} \rangle = - \left( \frac{1}{384\gamma^2 a^3_c} \right) \int dz_e \int dz_h \left| f_{n_e}^+(z_e) \right|^2 \left| f_{n_h}^+(z_h) \right|^2 \]

\[ \exp(\beta^2) \left\{ \Gamma(15/2, \beta^2) + (26 + 7\beta^2) \Gamma(13/2, \beta^2) + (246 + 676\beta^2 + \\
21\beta^4) \Gamma(11/2, \beta^2) - (-1080 + 1230\beta^2 + 390\beta^4 + 35\beta^6) \Gamma(9/2, \beta^2) - \right. \]

\[ - \left. (540 + 2310\beta^2 + 1890\beta^4 + 315\beta^6) \Gamma(7/2, \beta^2) - (240 + 1470\beta^2 + 1260\beta^4 + 210\beta^6) \Gamma(5/2, \beta^2) + \\
+ 6(120 + 702\beta^2 + 630\beta^4 + 90\beta^6) \Gamma(3/2, \beta^2) - \forall \beta^2 \left( 216 + 1260\beta^2 + 1158\beta^4 + 210\beta^6 \right) \text{erfc}(\beta) \right\} \]

(e6)
\[
(2280 + 4320\beta^2 + 2460\beta^4 + 520\beta^6 + 13\beta^8) \Gamma(7/2, \beta^2) - (2160 +
6840\beta^2 - 6480\beta^4 + 1230\beta^6 + 390\beta^8 + 21\beta^{10}) \Gamma(5/2, \beta^2) - (720 + 4320\beta^2
+ 6840\beta^4 - 4320\beta^6 + 1230\beta^8 + 156\beta^{10} + 7\beta^{12}) \Gamma(3/2, \beta^2)
\]
\[
\sqrt{\pi} \beta^2 (720 + 2160\beta^2 + 2280\beta^4 - 1080\beta^6 + 246\beta^8 + 26\beta^{10} + \beta^{12}) \times
\]
\[
\times \text{erfc}(\beta)
\]

(e6)
References.


