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Two-dimensional excitons in magnetic fields

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We have obtained the energies and the wave functions for two-dimensional (2D) excitons in magnetic fields for the ground and several excited states, using an exact numerical integration of the Schrödinger equation for 2D excitons in a magnetic field. The results of the exact calculation for the ground-state energy and wave functions are in excellent agreement with those of variational and perturbation calculations except at intermediate fields where $\gamma=(\hbar\omega_c/2R)$ is of order 10. Here, R is the exciton Rydberg unit and ω_c is the cyclotron frequency of the exciton. Results are presented for the position of the exciton peak and the magnetoabsorption as a function of the field for 2D excitons.

I. INTRODUCTION

With the development of sophisticated techniques of crystal growth such as molecular-beam epitaxy (MBE), interest has arisen in excitonic effects in semiconducting quantum-well structures in which electrons and holes are confined in their motion perpendicular to the plane of the quantum well.[1,2] When the width of the quantum well becomes less than the exciton Bohr radius, the binding energy of the excitons is considerably enhanced over its value in bulk semiconductors[3–5] and excitonic effects persist at room temperature[6] which makes these structures useful for many possible device applications.[7,8] Magneto-optical studies of excitons in quantum-well structures have been carried out by many authors to help determine the binding energy of excitons in these structures and the exciton reduced mass.[9–14] In addition to the experimental investigations, theoretical calculations have been performed on the exciton binding energies and wave functions as a function of magnetic field in quasi-two-dimensional structures. Because the problem of the hydrogen atom in a square-well potential is not separable, various approximation methods have been used to determine exciton energies and wave functions both in the absence and presence of magnetic fields. The simplest model for quantum-well excitons is that of two-dimensional (2D) excitons. The energy levels and oscillator strengths for 2D excitons in zero magnetic field were obtained by Shinada and Sugano.[15] The problem of 2D excitons in magnetic fields was first treated by Akimoto and Hasegawa[16] using the Wentzel-Kramers-Brillouin (WKB) approximation to obtain the energy levels of the 2D exciton. These results agree, within a numerical factor, with those of a perturbation treatment in both the high- and low-field limits. However, it is not clear from their treatment how the excitonic energy levels shift with magnetic field in the intermediate range of fields, which is in fact the range of fields of experimental interest in quantum-well systems even though these results have been used to correct the energies of interband transitions between Landau levels in recent experiments by Fasolino *et al.*[14] The authors were prevented from obtaining an exact solution of the eigenvalue problem for 2D excitons in magnetic fields because this problem with suitable boundary conditions is beyond the analytic problem of confluent hypergeometric equations.

An alternate approach is to use a variational method to obtain an upper bound on the energy levels of the 2D exciton as a function of magnetic field. In fact, such an approach has been used to calculate the binding energy of excitons as a function of well width for actual quantum-well systems in zero magnetic field.[17–19] Such variational methods have been extended to the problem of quantum-well excitons in magnetic fields using variational wave functions of the hydrogenic[20] and Gaussian[21] type. However, it is well known from calculations of the 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.[22]

Because the variational calculations are very complicated for actual quantum-well structures, calculations were performed for 2D excitons using variational wave functions which were hydrogenic at low magnetic fields and Gaussian at high magnetic fields.[23] These variational calculations yield an upper bound on the exact energy eigenvalues of the Schrödinger equation for 2D excitons in a magnetic field. Moreover, the shift in the exciton peak predicted by such a variational calculation seems to be in agreement with some experimental work.[24] Such calculations are more difficult to perform for the excited excitonic levels. Moreover, the variational method yields better results for the energies of a system than it does for the wave functions, and for excitons in magnetic fields the latter is needed to calculate the behavior of the oscillator strength for 2D excitons as a function of the magnetic field. The magnetic-field dependence of the exciton oscillator strength will enable us to see how the intensity of

the exciton peak changes with increasing magnetic field.

With the development of methods for numerically integrating differential equations, it has become possible to numerically integrate the Schrödinger equation with appropriate boundary conditions to obtain exact energy eigenvalues and eigenfunctions for 2D excitons in a magnetic field. Moreover, these numerical methods can be applied to yield the eigenfunctions and eigenvalues for both the ground and excited states of the exciton as a function of magnetic field. In this paper, we wish to present the results of such an exact numerical integration of the Schrödinger equation for 2D excitons in a magnetic field using a program called SLEIGN which was written by Bailey *et al.*[25] for solving Sturm-Liouville eigenvalue equations. We obtain the exact energy eigenvalues and eigenfunctions of the 2D exciton as a function of the magnetic field and use this information to calculate the absorption of light as a function of photon frequency and magnetic field for such 2D excitons. In our calculations, we will neglect complications connected with the energy levels of real quantum-well excitons such as those due to the finite width of the quantum well, the nonparabolicity of the valence bands, and the mixing of the heavy- and light-hole bands even though they are important in real materials such as GaAs. Andreani and Pasquarello[26] have taken account of valence-band mixing in calculating the binding energies, wave functions, and oscillator strengths of quantum-well excitons in the absence of magnetic fields, while Yang and Sham[27] and Bauer and Ando[28] take account of the effect of magnetic fields together with the valence-band mixing. These authors have shown that the mixing of the valence-band structure is important in interpreting the experimental magneto-optical results. Bauer and Ando[28] use as their set of basis functions to calculate the exciton energies, hydrogenic wave functions which limits the validity of their calculations to low magnetic fields, while Yang and Sham[26] have performed their calculations using as basis functions a set of Landau-level wave functions. As in the case of earlier variational calculations neglecting band mixing, it is not clear whether the use of either hydrogenic or Gaussian basis functions will yield good results at intermediate fields where the regime of transition between the two kinds of basis functions occurs. Therefore, exact calculations of the exciton energy levels and oscillator strengths, using the simplified model of two-dimensional excitons with the neglect of band nonparabolicity and valence band mixing, will increase our understanding of the transition in magnetic fields between hydrogenic and free Landau-level behavior of the electron-hole states.

II. THEORY

For a 2D exciton in a magnetic field we have axial symmetry, and the angular and center-of-mass coordinate dependence of the wave function can be separated off, so that the Schrödinger equation reduces to a second-order ordinary differential equation of the form

$$\frac{d^2f}{d\rho^2}+\frac{1}{\rho}\frac{df}{d\rho}+\left[\frac{2}{\rho}-\frac{m^2}{\rho^2}-m\gamma-\frac{1}{4}\gamma^2\rho^2+E\right]f=0\ , \tag{1}$$

where E is in Rydberg units, $R=(e^2/2\kappa a)$ is the exciton Rydberg unit, ρ is in units of exciton Bohr radii, $a=(\kappa\hbar^2/e^2\mu)$ is the exciton Bohr radius, $\gamma=(\hbar\omega_c/2R)$ is a dimensionless magnetic field, $\omega_c=(eB/\mu c)$ is the cyclotron frequency, κ is the relative dielectric constant of the material, m is the quantum number specifying the orbital angular momentum along the magnetic-field direction, and $\mu=(m_e m_h/(m_e+m_h))$ is the reduced mass of the electron-hole pair, which are bound together into the exciton.

In the limit of zero magnetic field, the energy eigenvalues and eigenfunctions of Eq. (1) can be obtained exactly and are[15]

$$E_n=-(n-\tfrac{1}{2})^{-2},\quad n=1,2,3,\ldots \tag{2}$$

and

$$f_{n,m}(\rho)=\frac{2}{(n-\frac{1}{2})}\left[\frac{(n-|m|-1)!}{(n+|m|-1)!(2n-1)}\right]^{1/2}\times\left[\frac{2\rho}{n-\frac{1}{2}}\right]^{|M|}\times\exp\left[\frac{-\rho}{n-\frac{1}{2}}\right]L_{n-|m|-1}^{2|m|}\left[\frac{2\rho}{n-\frac{1}{2}}\right], \tag{3}$$

respectively, where $L_p^q(\rho)$ is the associated Laguerre polynomial. In the limit of zero Coulomb interaction, the exact energy eigenvalues and eigenfunctions of Eq. (1) can also be obtained exactly and are

$$E_n=(2n+1)\gamma,\quad n=0,1,2,\ldots \tag{4}$$

and

$$f_{n,m}(\rho)=\left[\frac{(n-|m|)!\gamma}{n!}\right]^{1/2}\left[\frac{\gamma\rho^2}{2}\right]^{1/2}\times\exp\left[\frac{-\gamma\rho^2}{4}\right]L_{n-|m|}^{|m|}\left[\frac{\gamma\rho^2}{2}\right], \tag{5}$$

respectively.

To obtain the eigenvalues and eigenfunctions for the 2D exciton in a magnetic field for the general case, Eq. (1) is numerically integrated using SLEIGN. The eigenvalue problem given by Eq. (1) is singular in two senses. First, the interval $(0,\infty)$ in which it is defined is semi-infinite. Second, several of its coefficient functions have a singularity at the origin. These facts governed our choice of SLEIGN, since it is the only Sturm-Liouville code we know of which can handle such problems. The SLEIGN approach is to assume that the nth eigenvalue of a singular problem can be approximated arbitrarily closely by the nth eigenvalue of a nonsingular problem obtained by truncating the original interval. In order to solve the nonsingular problem, the classic Prufer substitution is used to transform from a second-order linear equation to a first-order nonlinear equation in the phase. This substitution also allows the boundary conditions at each end to be converted to initial conditions. Initial-value problems are then solved inwardly from each end point and matched at the center of the interval to obtain the correct

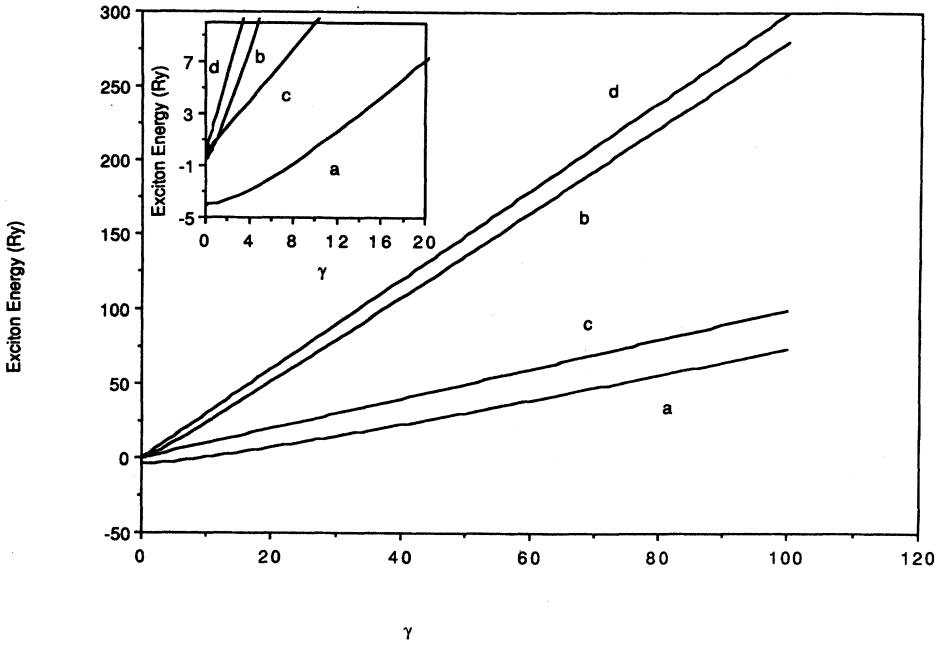

FIG. 1. The exciton energy in Rydberg units is shown as a function of the normalized magnetic field γ for *a*, the ground, and *b*, the first-excited states. The Landau energy levels for the ground and first-excited states of a free-electron-hole pair are shown by curves *c* and *d* for comparison.

eigenvalue. For more details see Ref. 25.

The energy eigenvalues of Eq. (1) obtained using SLEIGN are shown as a function of the normalized magnetic field γ in Fig. 1 for the ground and first-excited s states ($m=0$) for the 2D exciton. At high magnetic fields, the energy becomes a linear function of magnetic field, but the energy of the first-excited $m=0$ state increases much more rapidly with field that the ground $m=0$ state. In fact, while the energy for the $1s$ state approaches asymptotically the energy of the first Landau level ($n=0$) for a noninteracting electron-hole pair, the energy for the $2s$ state approaches that of the second Landau level ($n=1$) for a noninteracting electron-hole pair. This is in agreement with the results of Akimoto and Hasegawa[16] using the WKB approximation to find the energy eigenvalues, although the WKB approximation is in error as far as determining the exact energy eigenvalues as a function of magnetic field, as can be seen by comparing the WKB results of Akimoto and Hasegawa[16] with the perturbation theory results in the limit of both very weak and very strong magnetic fields. The energy eigenvalue for the first p state ($m=1$) has also been calculated. The first-excited $m=1$ state, which is degenerate with the first-excited $m=0$ state in the absence of the field, also has its energy increase linearly with field at high fields, but although it is no longer degenerate with the first-excited s state, its energy lies close to that of the $2s$ state at all values of the magnetic field. Therefore on the scale on which the exciton energies for the ground and first-excited $m=0$ states are plotted in Fig. 1, the results for the $m=1$ state would almost be indistinguishable from the first excited $m=0$ state. In addition, the energy for the $2p$ state also approaches the energy of the first-excited Landau level asymptotically with increasing magnetic field and lies in value between the energy of the $2s$ state and the $n=1$ Landau level.

We have also calculated the exciton energy for the $3s$ and $4s$ exciton states using SLEIGN. At zero field, the results for the energies of these exciton states are those given by Eq. (2) while in strong fields, they asymptotically approach the energies of the $n=2$ and $n=3$ Landau levels, respectively. Since several authors have used the energies of the higher Landau levels to try to extrapolate the value of the zero-field exciton binding energy, it is of interest to see how the Coulomb binding effects the energies of Landau levels with increasing magnetic field. The Coulomb binding energy of a particular Landau level can be obtained by subtracting the exciton energy calculated for the exciton associated with a particular Landau level from the energy of that particular Landau level given by Eq. (4), i.e., $E_b=(2n+1)\gamma-E$. The results are shown in Fig. 2 for the first four $m=0$ exciton states. It can be seen that the exciton binding energy increases with magnetic field for each of these states and that for a fixed field, the binding energy decreases as we go to higher-lying exciton states. However, the binding energy is not negligible even for the highest state ($4s$) for which we have done calculations.

In Fig. 3 the exciton energy obtained for the ground state using SLEIGN is shown together with the results of a simple variational calculation using hydrogenic and Gaussian-type wave functions. It can be seen that for values of γ less than unity, the ground-state energy using a hydrogenic variational wave function yields good agreement with the exact ground-state energy. On the other hand, for values of γ greater than 20, the Gaussian varia-

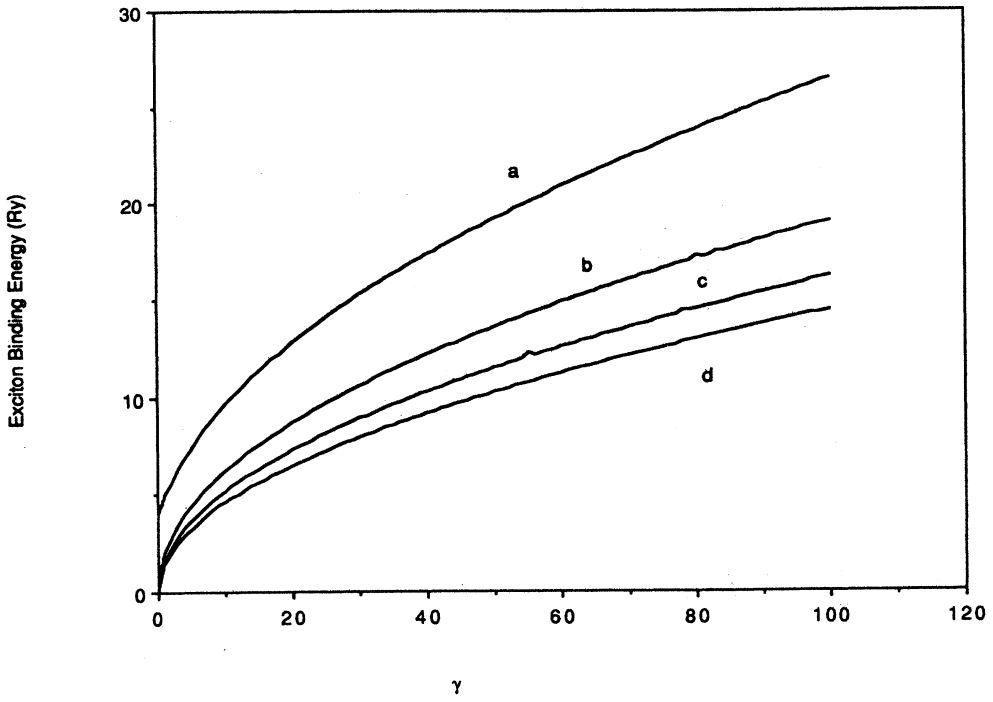

FIG. 2. The exciton binding energy in Rydbergs units is shown as a function of γ for the $1s$, $2s$, $3s$, and $4s$ exciton states.

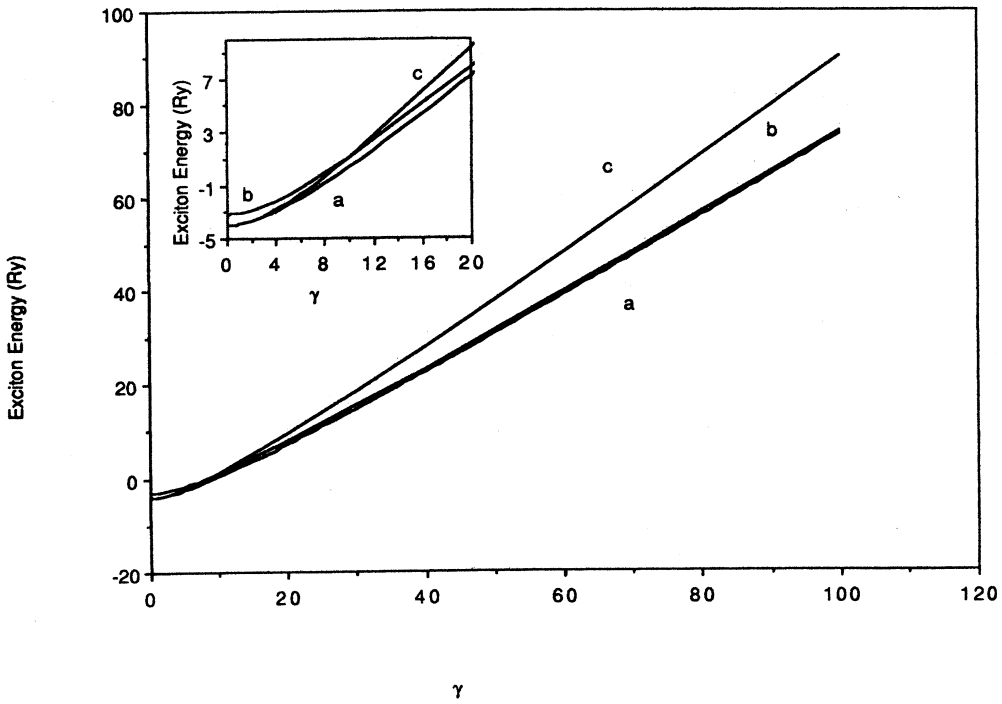

FIG. 3. The ground-state exciton energy as a function of normalized magnetic field calculated using SLEIGN (*a*) is shown in comparison with the results of variational calculations using Gaussian (*b*) and hydrogenic (*c*) variational wave functions.

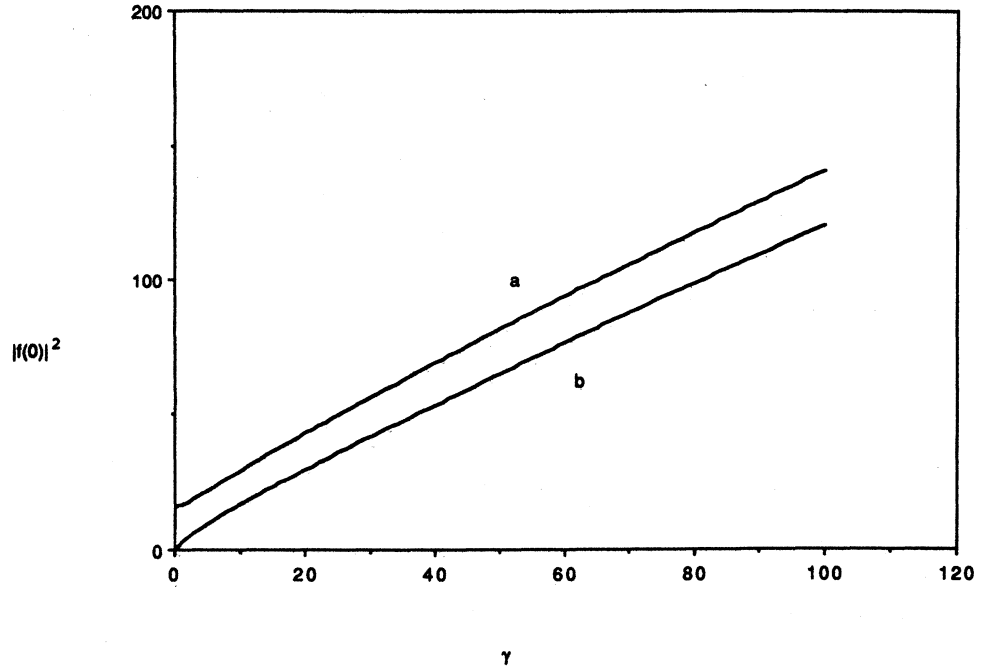

FIG. 4. The normalization coefficient $|f(0)|^2$ is shown as a function of γ for the $1s$ and $2s$ exciton states.

tional wave function yields ground-state energies which agree with the results of the exact calculations to less than a few percent. Therefore, except for intermediate values of the magnetic field, simple hydrogenic or Gaussian variational wave functions will give very good results for the ground-state exciton energy.

The optical-absorption coefficient can be related to the imaginary part of the dielectric function. For a two-dimensional gas of free carriers and excitons, Lederman and Dow[29] have shown that the imaginary part of the dielectric function is given by

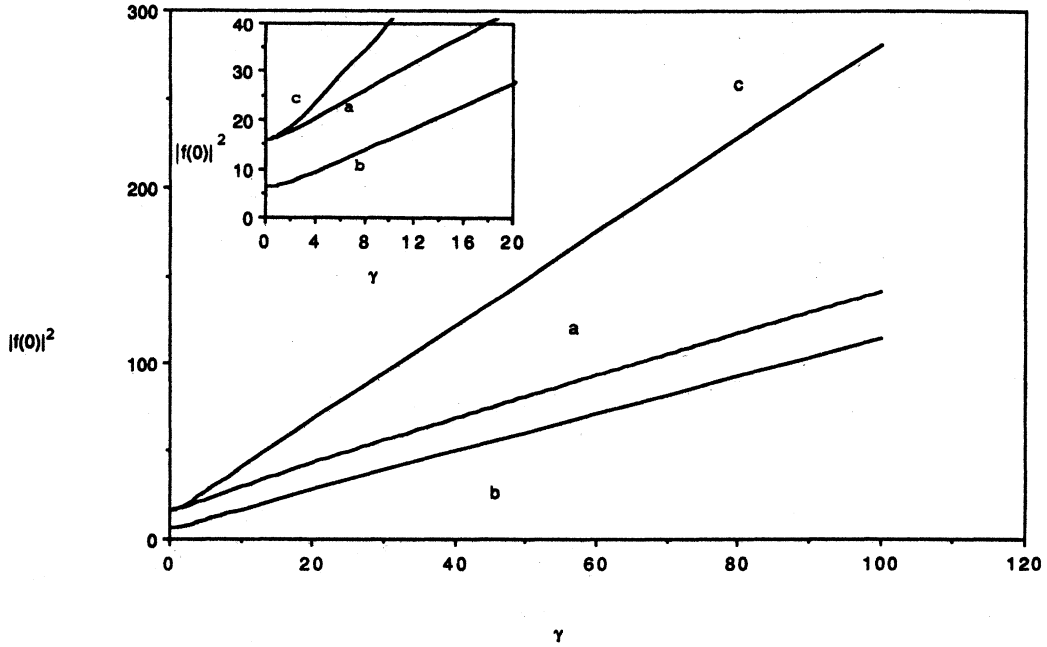

FIG. 5. The dependence of $|f(0)|^2$ on γ calculated using SLEIGN (a) is shown in comparison with the results of variational calculations using Gaussian (b) and hydrogenic (c) variational wave functions.

$$\epsilon_2(\omega)=C_{02}|f(0)|^2S(E)\ , \tag{6}$$

where $S(E)$ is the density of states for the relevant particles, $|f(0)|^2$ is the square of the envelope function for vanishing separation of the electron-hole pair, $E=\hbar\omega-E_g$, ω is the photon frequency, E_g is the band gap of the semiconductor, and

$$C_{02}=(4\pi e^2/m^2\omega^2Ra^2)|\langle c|\hat{\epsilon}p|v\rangle|^2$$

for a two-dimensional system. Here $\hat{\epsilon}$ is a unit vector in the direction of polarization of the light and ρ is the momentum operator. Therefore the oscillator strength for the optical transition and the intensity of the exciton peak are proportional to the factor $|f(0)|^2$. The intensity of the exciton absorption peak, therefore, depends upon the value of the wave function $f(\rho)$ at the origin, i.e., on the probability of the electron and hole being found at the same point in space. For excitons in zero magnetic fields, $|f(0)|^2=2/(n-\frac{1}{2})^3$ while for a free-electron-hole pair in the presence of the magnetic field but in the absence of the Coulomb interaction between them, $|f(0)|^2=\gamma$. SLEIGN also enables us to obtain the value of the wave function at the origin for arbitrary magnetic fields since we are able to obtain numerically the eigenfunctions of Eq. (1) as well as its eigenvalues. In Fig. 4 $|f(0)|^2$ is shown as a function of the normalized magnetic field γ for the $1s$ and $2s$ states. The value of $|f(0)|^2$ is larger for the ground state than it is for the excited states, and for both the ground and excited states, $|f(0)|^2$ increases with magnetic field. This means that the intensity at the exciton peaks should also increase with magnetic field. In Fig. 5 we compare the value of $|f(0)|^2$ obtained here to those obtained as a result of the simple variational calculations using hydrogenic and Gaussian wave functions. The value of $|f(0)|^2$ as a function of magnetic field obtained using SLEIGN starts out at the value obtained using a hydrogenic variational wave function at low magnetic fields. At high magnetic fields, its value tends to approach the value obtained using the Gaussian variational wave function although it never quite reaches that value for finite magnetic field. However, it can be seen from Figs. 3 and 5, that the variational method yields better agreement with the exact numerical results obtained using SLEIGN for the exciton energies than for $|f(0)|^2$ and that this agreement exists over a wider range of fields. For the states for which m is not equal to zero, $|f(0)|^2$ vanishes and therefore such states would not contribute to the intensity at the exciton peaks for allowed transitions in direct-band-gap semiconductors.

The magnetoabsorption coefficient which is related to $\epsilon_2(\omega)$ depends upon the density of states. For a system having discrete energies, the density of states is given by a sum of δ functions whose arguments are related to these discrete energies. To obtain values for the magnetoabsorption coefficient as a function of photon energy at fixed magnetic field, we replace the δ functions by Lorentzians which give an absorption coefficient K of the form

$$K(\omega)=A|f(0)|^2[(E+E_{\rm ex})^2+\Gamma^2]^{-1}\ , \tag{7}$$

where A is a coefficient which contains the parameters of the material and Γ is the exciton linewidth. In Fig. 6 the normalized absorption coefficient is shown as a function of E for various values of the magnetic field. The value used for Γ is 0.5 in exciton Rydberg units and is assumed in our calculations to be independent of the magnetic field. From Fig. 6 we see that the exciton peak shifts to higher energies E with increasing magnetic field and that the intensity of the exciton peak, assuming a Lorentzian for the exciton linewidth, also increases with increasing magnetic field. In our calculations, the contribution of both the $1s$ and $2s$ exciton states to the exciton peak are taken into account. From Fig. 6 we see that the absorption peak due to the $2s$ state is barely observable in zero magnetic field, but that the height of this peak relative to that of the $1s$ absorption peak increases dramatically with increasing magnetic fields. Even for the case where $\gamma=1$, when there is a negligible shift in energy of the $1s$ absorption peak, the $2s$ absorption peak clearly stands out. Although we have not shown the peaks arising from the higher-excited exciton s states, which would be almost unobservable in the absence of the magnetic field, they should behave similarly. In fact, in the strong-field re-

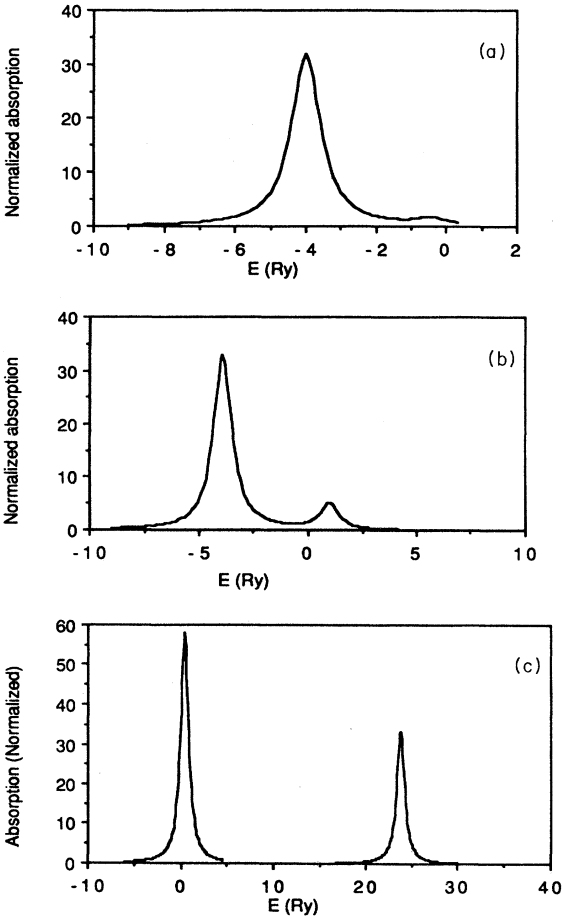

FIG. 6. The normalized optical-absorption coefficient is shown as a function of E for $\gamma=0$ (a), $\gamma=1$ (b), and $\gamma=10$ (c), respectively. Only the $1s$ and $2s$ states have been taken into account in this calculation.

gime, the intensity of the peaks due to the higher excitonic levels should approach the same value as the peaks arising from the free Landau levels.

III. SUMMARY

Exact numerical integration of the Schrödinger equation for 2D excitons in a magnetic field yields the energy eigenvalues and eigenfunctions of the excitons which enables us to obtain the optical absorption due to such 2D excitons as a function of magnetic field. The results of this numerical integration yield ground-state exciton energies which are in excellent agreement with those obtained using a variational approach except at intermediate values of magnetic field, such that γ is of order 10. In low fields our results for both the exciton energies and oscillator strengths agree with those for 2D excitons in zero magnetic field, while in high fields they agree with those for the free Landau levels both for the ground and excited states. With increasing magnetic field, the exciton states go over to the free Landau states. The exciton energies are found to shift almost linearly with field for high magnetic fields, while the oscillator strengths are also found to increase linearly with field for high fields. Our calculations predict a shift of the exciton peak with increasing field together with an increase in the intensity at the peak. We also predict that the intensity of exciton peaks due to excited exciton states also will increase with field so that these peaks will become more prominent as the field increases. Our predictions seem to be in good agreement with the experiments of Davis *et al.*[24] in which the shift in the exciton peak was observed in a GaAs/GaAlAs multiple-quantum-well system in magnetic fields up to 27 T. The only deviations between our theoretical calculations and the experiments occurred at the highest magnetic fields where nonparabolicities due to valence-band mixing would be expected to be important.

Our results agree with the general behavior at low and high magnetic field predicted by Akimoto and Hasegawa.[16] However, the exact numerical values of the exciton energy, which they predict using the WKB method, are in disagreement with the exact numerical results at both low and higher magnetic fields. In both the low- and high-field limits, their use of the WKB approximation leads to results which overestimate the exciton binding energy. In the low-field limit, their predicted diamagnetic shift of the exciton energy in the ground state is a factor of 2 smaller than the predictions of a perturbation calculation and our results arising from an exact numerical integration of the wave equation. In the high-field limit, their calculations predict a binding energy which is 20% larger than our exact results and those of a perturbation calculation which treats the Coulomb interaction between the electron-hole pair as a perturbation on the free Landau levels in the magnetic field. Their values of the exciton oscillator strength are also in disagreement with our exact results since we obtain a stronger variation of the oscillator strength as a function of field for the ground state than do they.

Finally, we have presented our results in terms of dimensionless quantities, i.e., the exciton and cyclotron energies are given in terms of exciton Rydberg units. Therefore, our results are applicable to excitons in any semiconducting quantum-well structures independent of the material parameters as long as the approximations of our simple model hold, i.e., neglect of the finite width of the well and of the mixing of the light- and heavy-hole valence bands. The material parameters in this case come in only in determining the value of the exciton Rydberg units and the cyclotron frequency ω_c. In $GaAs/Ga_{1-x}Al_xAs$ quantum-well structures, where the light- and heavy-hole exciton reduced masses are $0.051m_0$ and $0.041m_0$, respectively and the relative dielectric constant is 11.6, the exciton Rydberg unit is 3.8 meV for the heavy-hole exciton and 4.85 meV for the light-hole exciton. Therefore $\gamma=1$ corresponds to a magnetic field of 2.7 T, while $\gamma=10$ corresponds to a magnetic field of 27 T for the heavy-hole exciton. Therefore in $GaAs/Ga_{1-x}Al_xAs$ quantum-well systems, the energy of the heavy-hole exciton increases almost linearly with magnetic fields for magnetic fields above 10 T. However, even at much higher fields, the exciton energy still differs by quite a bit for the ground state from the lowest free Landau level. On the other hand, at fields of only a few Tesla, the energies of the excited exciton states have already approached the energies of their related free Landau levels. This is due to the decrease of the exciton binding energy of the higher states which is shown in Fig. 2.

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