Magnetobiexcitonic states in a quantum wire

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We report a partly variational calculation of biexcitonic states in a semiconductor quantum wire subjected to a magnetic field. The longitudinal component of the biexciton wave function is computed variationally, while the transverse component is obtained from exact numerical solution of the Schrödinger equation. We find that an external magnetic field squeezes the biexciton wave function, causing the binding energy to increase substantially. The increase is more pronounced in wider wires where the biexcitonic wave function is ''softer'' and hence more squeezable. This increase can enhance the already giant third-order nonlinear susceptibilities predicted for quantum wires, thus making these systems attractive for applications in nonlinear optics. [S0163-1829(96)03531-X]

I. INTRODUCTION

Quasi-one-dimensional excitons and biexcitons (excitonic molecules) in quantum wires have been a subject of considerable theoretical research since they are responsible for the giant third-order nonlinear susceptibility $\chi^{(3)}$ of one-dimensional systems.[1–5] Within the rotating wave approximation,[1] the biexciton binding energy contributes directly to $\chi^{(3)}$. An external magnetic field can increase this binding energy, leading to stronger optical nonlinearities. Additionally, the field can act as an agent to modulate the nonlinear absorption and/or gain in quantum wires, which opens up the possibility of realizing *externally tunable* couplers, limiters, phase shifters, switches, etc.

In the past, the effects of a magnetic field on excitonic complexes have been largely ignored since a magnetic field is technologically less significant than an electric field in device applications. However, a magnetic field has two important advantages. First, it increases the exciton-biexciton binding energy by squeezing the electrons and holes together, while an electric field decreases the energy by spatially separating oppositely charged particles. Therefore, a magnetic field can cause improved nonlinear optical properties. Second, it offers far richer insight into the physics of excitonic complexes, especially for low-dimensional systems.

There have been relatively few theoretical studies of excitons in quantum wires[1–11] and even fewer dealing with biexcitons.[6,10,12–14] To our knowledge, the influence of a magnetic field[15] on these entities has not been explored in sufficient detail. Experimental data in this area are sparse and only recently has the effect of a magnetic field on exciton absorption and magnetophotoluminescence in quantum wires been reported.[16,17] Unambiguous signatures of biexcitons in quantum wires were also observed very recently[18] and it is an important development since the mere existence of these entities in one-dimensional structures has been quite moot.[19]

Unlike the exciton problem, which is straightforward, the biexciton problem in the context of quantum wires is somewhat complex and beset with ambiguities. Recently, Ivanov and Haug[19] claimed that biexcitons do not exist in *strictly* one-dimensional wires because of the strong influence of the polariton effect (although, for very strong binding energies they can exist as a broad resonance). Physically, this result was attributed to the ''topological'' feature of a one-dimensional structure; the exciton inside the biexciton cannot make a complete oscillation. However, this argument breaks down for realistic quantum wires with nonzero lateral dimensions. In our calculations, we consider realistic wires with lateral dimensions larger than the Bohr radius of excitons and the polaritonic effect is not likely to be critical.

Another problem with biexcitons is the lack of a standard model. While there are standard models[20–27] for excitons, the paucity of experimental data for comparison has inhibited the emergence of a single, commonly accepted theoretical model and/or numerical approach for the calculation of biexcitonic states. In most cases, the biexciton problem is treated variationally, even though there is no concrete evidence that this is adequate or justified. The archetypal examples of such calculations are works by Brinkman, Rice, and Bell[28] (bulk), Kleinman[29] (quantum well), and Banyai *et al.*[6] (quantum wire). In a more recent study, Madarasz *et al.*[10,30] calculated exciton and biexciton binding energies variationally for rectangular GaAs quantum wires treating the Coulomb interaction terms exactly in their full three-dimensional form. The trial wave function was chosen to be of the Heitler-London type. However, Ishida, Aoki, and Ogawa[31] presented a study of biexcitons within the framework of a one-dimensional tight-binding model and showed that in their model calculation, the effect of particle correlation over the lattice-constant length scale is important. This makes conventional variational methods or the Heitler-London approximation questionable. In our particular case, the correlation effect is not critical for three reasons. First, unlike Ishida, Aoki, and Ogawa, we are primarily interested in the regime of moderate quantum confinement (in our case wire width L_y=100–700 Å and wire thickness L_z=100–300 Å, which is characteristic of standard samples). Ishida, Aoki, and Ogawa considered biexcitons in strongly confined one-dimensional wires with wire width smaller then the size (effective Bohr radius) of an exciton (about 100 Å for GaAs). For this type of narrow 1D system, the exciton wave function becomes very compact and the behavior of electrons and holes on the atomic length scale assumes critical importance.

Obviously, this is not the case with our structures. Another objection raised by Ishida, Aoki, and Ogawa pertained to the treatment of the Coulomb interaction with its characteristic $1/r$ singularity. Usually, the singularity is dealt with by an arbitrary numerical truncation procedure as in Ref. 6. It was this truncation that was criticized. However, we treat the Coulomb interaction in its full three-dimensional form, which allows us to treat the $1/r$ singularity without the application of any ad hoc truncation procedure. Finally, the discrepancy between the Ishida, Aoki, and Ogawa result and the conventional variational result based on a Heitler London scheme becomes significant (about 50%) when the electron and hole effective masses are similar ($\sigma=m_e/m_h\approx 1$). In our case, we are dealing with effective mass ratios $\sigma\approx 0.1$, which makes the discrepancy much smaller.

In the next section, we present the theory. This is followed by results and finally the conclusion.

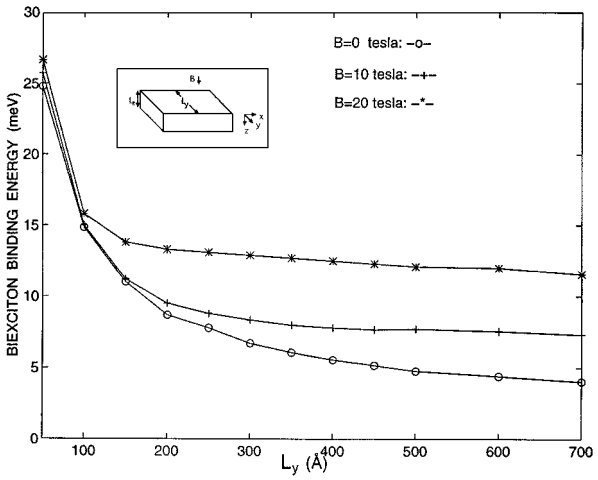

FIG. 1. Biexciton binding energy in a GaAs quantum wire as a function of wire width L_y for three different values of the magnetic flux density B. When there is no field present, the binding energy decreases monotonically with increasing wire width. When the field is present, there the binding energy curves tend to saturate beyond a wire width of about 300 Å. The thickness along the z direction is 200 Å.

II. THEORY

We consider a quantum wire of rectangular cross section as shown in the inset of Fig. 1 with infinite potential barriers located at $y=\pm L_y/2$ and $z=\pm L_z/2$. A magnetic field of flux density B is applied along the z direction.

At the outset, we neglect both correlation and polaritonic effects. Since polaritons arise from the radiative renormalization of a biexciton, we cannot include them perturbationally in the model presented here. Therefore, these effects are reserved for a future study.

For nondegenerate and isotropic bands, the Hamiltonian of a biexciton in a quantum wire subjected to a magnetic field is given in the envelope-function approximation by

$$H^{XX}=\frac{1}{2m_e}(p_{x_1}^2+p_{x_2}^2)+\frac{1}{2m_h}(p_{x_a}^2+p_{x_b}^2)+\frac{1}{2m_e}(p_{y_1}^2+p_{y_2}^2)+\frac{1}{2m_h}(p_{y_a}^2+p_{y_b}^2)+\frac{1}{2m_e}(p_{z_1}^2+p_{z_2}^2)+\frac{1}{2m_h}(p_{z_a}^2+p_{z_b}^2)$$

$$+\frac{1}{2m_e}[e^2B^2(y_1^2+y_2^2)-2eB(y_1p_{x_1}+y_2p_{x_2})]+\frac{1}{2m_h}[e^2B^2(y_a^2+y_b^2)+2eB(y_ap_{x_a}+y_bp_{x_b})]+V_{\text{Coulomb}}+V_{\text{conf}}, \tag{1}$$

where V_{conf} is the confinement potential along the y and z directions, and V_{Coulomb} is the Coulomb interaction between various charged entities. The electron coordinates are subscripted by the numerals 1 and 2 while hole coordinates are subscripted by the letters a and b. In Eq. (1) we neglected the Zeeman splitting since the Landé g factor is small for GaAs.

For weak Coulomb interaction (characteristic of relatively narrow gap materials with large dielectric constant), the wave function of the biexciton in a z-directed magnetic field can be written as a product of particle-in-a-box states along the z direction, magnetoelectric states along the y direction, and a Heitler-London-type symmetric linear combination of Gaussian orbitals (involving variational parameters) along the x direction. Because of this particular choice of the wave function, it is convenient to use the following relative and center-of-mass coordinates to transform the Hamiltonian in Eq. (1):

$$X=[m_e(x_1+x_2)+m_h(x_a+x_b)]/2(m_e+m_h),$$

$$x_{1a}=x_1-x_a,\quad x_{2b}=x_2-x_b,\quad x_{ab}=x_a-x_b.$$

To rewrite the biexciton Hamiltonian in the new coordinate system, we utilize the usual canonical transformations for the momentum operators. We also drop the kinetic energy operators associated with the center-of-mass motion since the Hamiltonian is invariant in X so that P_X is a good quantum number. We are only interested in obtaining biexcitonic states that can be accessed optically, and under optical excitation, the center-of-mass motion can be neglected since the photon momentum is too small to create states of significant center-of-mass kinetic energy. Consequently, the transformed Hamiltonian becomes

$$H^{XX}=-\frac{\hbar^2}{2\mu}\left(\frac{\partial^2}{\partial x_{1a}^2}+\frac{\partial^2}{\partial x_{2b}^2}\right)-\frac{\hbar^2}{m_h}\left(\frac{\partial^2}{\partial x_{ab}^2}-\frac{\partial^2}{\partial x_{1a}\partial x_{ab}}+\frac{\partial^2}{\partial x_{2b}\partial x_{ab}}\right)-\frac{\hbar^2}{2m_e}\left(\frac{\partial^2}{\partial {y_1}^2}+\frac{\partial^2}{\partial {y_2}^2}+\frac{\partial^2}{\partial {z_1}^2}+\frac{\partial^2}{\partial {z_2}^2}\right)$$
$$-\frac{\hbar^2}{2m_h}\left(\frac{\partial^2}{\partial {y_a}^2}+\frac{\partial^2}{\partial {y_b}^2}+\frac{\partial^2}{\partial {z_a}^2}+\frac{\partial^2}{\partial {z_b}^2}\right)+\frac{e^2B^2}{2}\left(\frac{y_1^2+y_2^2}{m_e}+\frac{y_a^2+y_b^2}{m_h}\right)+\frac{eBi\hbar}{m_e}\left(y_1\frac{\partial^2}{\partial x_{1a}}+y_2\frac{\partial^2}{\partial x_{2b}}\right)$$
$$+\frac{eBi\hbar}{m_h}\left(y_a\frac{\partial^2}{\partial x_{1a}}+(y_b-y_a)\frac{\partial^2}{\partial x_{ab}}+y_b\frac{\partial^2}{\partial x_{2b}}\right)+V_{\text{Coulomb}}+V_{\text{conf}}. \tag{2}$$

where μ is the reduced mass. The Coulomb interaction term in Eq. (2) is given by

$$V_{\text{Coulomb}}=\frac{e^2}{4\pi\epsilon}\left\{\frac{(-1)}{\sqrt{x_{1a}^2+r_{1a}^2}}+\frac{(-1)}{\sqrt{x_{1b}^2+r_{1b}^2}}+\frac{(-1)}{\sqrt{x_{2b}^2+r_{2b}^2}}\right.$$
$$\left.+\frac{(-1)}{\sqrt{x_{2a}^2+r_{2a}^2}}+\frac{1}{\sqrt{x_{ab}^2+r_{ab}^2}}+\frac{1}{\sqrt{x_{12}^2+r_{12}^2}}\right\}, \tag{3}$$

where

$$r_{\alpha\gamma}^2=(y_\alpha-y_\gamma)^2+(z_\alpha-z_\gamma)^2,$$

$$x_{\alpha,\gamma}=x_\alpha-x_\gamma,$$

with $\alpha,\gamma=1,2,a,b$, and $\alpha\neq\gamma$.

In choosing the variational wave function, we are motivated by many considerations. In the regime of weak quantum confinement, where the lateral dimensions $L_{y,z}$ of the wire are much larger than the effective Bohr radius a_B^* of a free exciton, the Coulomb interaction energy is dominant over the confinement energy. In that case, one may apply the ''free boson'' model to determine the different electron-hole-pair states. The ''free boson'' model is based on the Heitler-London approximation in which the biexciton wave function is written as a linear combination of atomic orbitals in analogy with the hydrogen molecule. The linear combination is chosen symmetric in space so that it corresponds to the singlet state, which is known to be lower in energy than the triplet state.[19]

In the regime of strong quantum confinement, $L_{y,z}$ are smaller than the effective Bohr radius a_B^*. In this case, the confinement energy is the dominant contribution. Electrons and holes are confined separately, and the Coulomb potential may be neglected in comparison with the large kinetic energy caused by the strong confinement. In our case, we are primarily interested in the regime of moderate quantum confinement. Therefore, we have to adopt a physically realistic trial wave function that would allow us to use the Heitler-London approach while taking into account the difference in the motion along confined and ''free'' directions. We achieve this by considering the electrons and holes to be independently quantized along y and z directions, while applying the Heitler-London approximation along the x direction. Following the approach in Refs. 10 and 12 we choose a trial wave function as a singlet state with the electron-hole-pair contributions given by the Gaussian-type ''orbital'' functions:

$$\Psi=\frac{1}{S}\{\psi_{1a}\psi_{2b}+\psi_{2a}\psi_{1b}\}G_{ab}(x_{ab}), \tag{4}$$

where

$$\psi_{\alpha\gamma}=g_{\alpha\gamma}\phi_\alpha(y_\alpha)\chi_\alpha(z_\alpha)\phi_\gamma(y_\gamma)\chi_\gamma(z_\gamma), \tag{5}$$

with

$$G_{ab}(x_{ab})=e^{-x_{ab}^2/\tau^2}, \tag{6}$$

and

$$g_{1a}=e^{-x_{1a}^2/\eta^2}, \tag{7}$$

$$g_{2b}=e^{-x_{2b}^2/\eta^2}, \tag{8}$$

$$g_{2a}=e^{-(x_{2b}-x_{ab})^2/\eta^2}, \tag{9}$$

$$g_{1b}=e^{-(x_{1a}+x_{ab})^2/\eta^2}. \tag{10}$$

The wave functions $g_{\alpha\gamma}$ are Gaussian orbitals whose ''spread'' η and τ are variational parameters. It is obvious that these parameters physically correspond to the electron-hole and hole-hole separations along the length of the quantum wire. The quantity S is a normalization constant, which can be evaluated as described in Ref. 12. The quantities $\phi_{e,h}(y_{e,h})$, are y components of the wave functions of independently confined electrons and holes in the quantum wire under a magnetic field. They are magnetoelectric states and are to be calculated independently by solving the Schrödinger equation. This is achieved by following the prescription given in Ref. 32. The quantities $\chi_{e,h}(z_{e,h})$ are the z components of the wave function of independently confined electrons and holes. These quantities are not affected by the magnetic field (the magnetic field is oriented along the z direction) and are therefore represented by particle-in-a-box states.

The chosen trial wave function implies that the ''true'' wave function is more sensitive to the separation between the holes than between the electrons. Since holes have higher effective masses, this assumption is physically realistic.

The biexciton binding energy E_B^{XX} is given by

$$E_B^{XX}=\min\langle\Psi|H^{XX}|\Psi\rangle-2\min\langle\psi|H^X|\psi\rangle, \tag{11}$$

where $\min\langle\Psi|H^{XX}|\Psi\rangle$ is found by minimizing the expectation value of the Hamiltonian H^{XX} with respect to the hole-hole variational parameter τ, and $\min\langle\psi|H^X|\psi\rangle$ is found by minimizing the expectation value of the exciton Hamiltonian with respect to the electron-hole variational parameter η as

described in Ref. 11. As pointed out by Klepfer, Madarasz, and Szmulowicz,[12] introduction of a third variational parameter, which allows for relaxation of the electron-hole pair within the biexciton, would not seriously change the biexciton binding energy when there is no magnetic field present. We tacitly assume that the same is true when a magnetic field is present. Mathematical details of computing the expectation values of the exciton and biexciton Hamiltonian are provided in the Appendix. One should note that every imaginary term in the expectation value of the Hamiltonian vanishes exactly, which makes the expectation value strictly real and shows that the trial wave function space is admissible.

Before concluding this section, we should make a few remarks about the computational details. The evaluation of the integrals in the Coulomb term of the expectation value (see Appendix) is not straightforward owing to the $1/r$ singularity. References 5 and 10 avoided dealing with this by applying a Fourier transform of the Coulomb term. This is not possible in our case since we have a magnetic field present, which transforms particle-in-a-box states along the width of the quantum wire (y direction) into magnetoelectric states whose wave functions $\phi_{e,h}(y_{e,h})$, are calculated numerically. Fortunately, unlike in the case of the strictly one-dimensional Coulomb potential, the Coulomb term treated in its full three-dimensional form is not pathological, and can be integrated in real space using the extended (multidimensional) definition of the improper integral that exists for this situation. This procedure involves evaluation of five- and seven-dimensional integrals in real space. We checked for the convergence of the integrals by setting all but the free coordinates to zero and letting the relative coordinates along the x direction gradually vanish over a hypersphere in order to ensure that the volume element approaches zero faster than the singularity.

III. RESULTS AND DISCUSSION

All the results that we present are for GaAs quantum wires. The electron and hole effective masses are assumed to be $0.067m_0$ and $0.5m_0$, respectively. The relative permittivity of GaAs is taken as 12.9.

In Fig. 1, we present the biexciton binding energy as a function of wire width L_y for three different values of the magnetic field. The binding energy always decreases with increasing wire width as expected because of decreasing electrostatic confinement. For narrow widths $\leqslant$100 Å, the magnetic field has little effect. Only at large widths does it cause the rate of decrease to decelerate. These features can be explained by invoking the complementary roles of electrostatic and magnetostatic confinement. At narrow widths, the electrostatic confinement is predominant and the magnetic field has little or no effect. Once the wire width significantly exceeds the magnetic length $\sqrt{\hbar/eB}$, the electrostatic confinement becomes weaker and yields dominant sway to the magnetostatic confinement. Since the latter depends only on the magnetic field strength and is independent of the wire width, the dependence of the binding energy on the wire width becomes much weaker. In the limit of very wide wires (approaching the two-dimensional limit), the electrostatic confinement will gradually vanish. The magnetoelectric states will condense into pure Landau orbits and the confinement will be entirely of magnetostatic origin. In that case, we expect the binding energy to become independent of the wire width so that the top two curves in Fig. 1 will saturate at a constant value. This is exactly what happens asymptotically as $L_y \to \infty$.

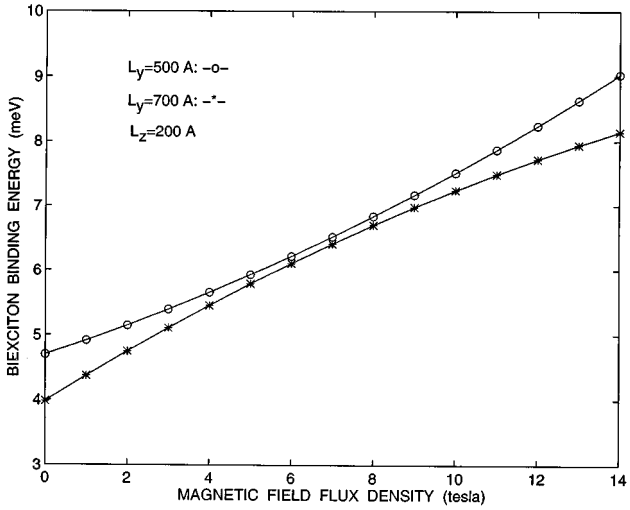

FIG. 2. Biexciton binding energy in a GaAs quantum wire as a function of magnetic flux density for two values of the wire width: L_y=500 Å and L_y=700 Å. The thickness along the z direction is 200 Å.

Figure 2 shows the biexciton binding energy as a function of the magnetic field for different values of wire width L_y. For relatively wide wires (L_y=700 Å), the binding energy increases sublinearly with the magnetic field, whereas for narrower wires (L_y=500 Å), the increase is superlinear. For wider wires, the magnetostatic confinement takes over the dominant role at relatively low magnetic field strengths and hence increasing the magnetic field to very high values will not have a dramatic effect. This causes the sublinearity. For narrower wires, the increase is superlinear since at low values of the field, the electrostatic confinement is dominant and the field has no effect. Only at relatively high fields, the binding energy increases as the magnetostatic confinement becomes increasingly dominant.

Figures 3(a) and 3(b) show the probability density distribution of the biexciton as a function of relative coordinates x_{1a} and x_{2b} (the separation along the wire between oppositely charged particles) with and without magnetic field. These two relative coordinates are the separations between one electron and one hole, and between the other electron and the other hole. The presence of two peaks can be easily understood as follows. Energetically, the most favored configuration in a quasi-one-dimensional system is ''e-h-e-h,'' which means that no two like charges are physically contiguous. Consequently, there are two favored values of x_{1a} and x_{2b}—the distance between nearest-neighbor particles and the distance between the two extreme particles in the above configuration. The effect of the magnetic field is to squeeze the biexciton wave function along the free (x axis) direction for both relative coordinates. This is a consequence of magnetostatic confinement.

The dependence of the ratio of biexciton to exciton bind-

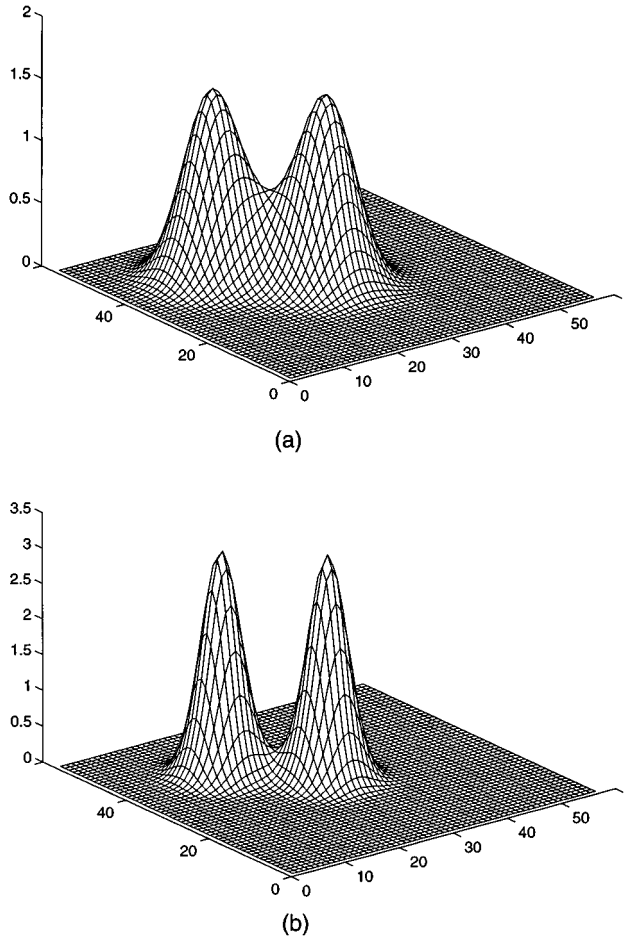

FIG. 3. Plot of the biexciton probability density as a function of relative coordinates x_{1a},x_{2b} for a quantum wire with and without magnetic field. The wire dimensions are L_z=200 Å, L_y=700 Å.

ing energy E_B^{XX}/E_B^X on the mass ratio $\sigma=m_e/m_h$ has traditionally been a subject of significant interest and has been investigated by various authors for quasi-one-dimensional systems,[6,31] for two-dimensional systems,[29] and for bulk.[28,33–37] The primary findings until now have been that (i) the ratio decreases monotonically with increasing σ from $\sigma=0$ (hydrogen molecule limit) to $\sigma=1$ (positronium limit), (ii) the ratio versus σ plot is symmetric about $\sigma=1$, (iii) the ratio at any value of σ decreases with increasing dimensionality (quantum wire→quantum well→bulk), and (iv) the plot has zero slope at $\sigma=1$. All of these features are unaffected by a magnetic field as shown in Fig. 4. The slight nonzero slope at $\sigma=1$ is a consequence of numerics and also the Heitler-London-type approximation for the variational wave function.

The magnitude of the energy ratio is of course material dependent since it depends on the Coulomb interaction[31] and hence on the dielectric constant. It should be noted that the ratio is always quite large, which makes the so-called ''adiabatic approximation,'' where the biexciton is treated as a molecule of two structureless excitons,[19] questionable since this approximation is founded on the assumption that the exciton is much more tightly bound than the biexciton. The dependence of the ratio on σ is strongest (i.e., the slope of the plot is largest) for the highest magnetic field and the intercept is also largest for the highest field. Because of these

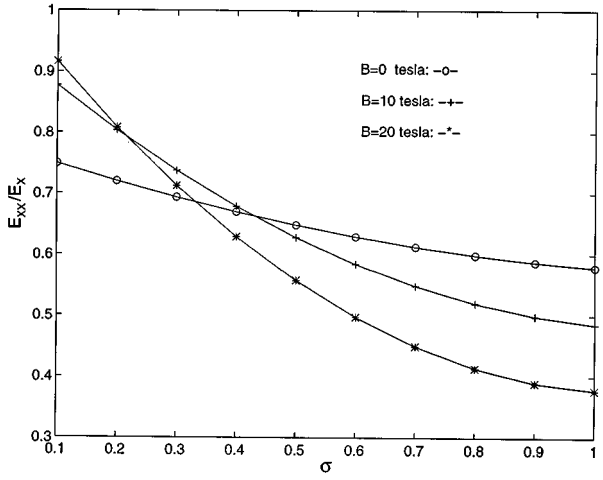

FIG. 4. The ratio of the biexciton to exciton binding energy as a function of electron-hole mass ratio σ for different values of magnetic flux density. The wire dimensions are L_z=200 Å, L_y=700 Å.

two features, there are crossings between the three characteristics in Fig. 4. The ratio of the binding energies increases with magnetic field for small values of σ, but decreases with the magnetic field as σ approaches unity (i.e., the electron and hole effective masses become equal).

In order to verify the verity of our model, we have compared our results for the binding energy (without any magnetic field) with those given in Refs. 5, 10, and 12. The ground-state binding energies are in excellent agreement over the whole range of wire width L_y=150–700 Å. The agreement deteriorates slightly at small values of L_y and this happens because of the different values of effective masses used in the different calculations. We have also compared our results with those experimentally measured and reported in Refs. 16 and 17. Since Ref. 17 used T-shaped edge quantum wires rather than wires of rectangular cross sections, a direct quantitative comparison is not possible. Nontheless, there is excellent qualitative agreement.

IV. CONCLUSION

In this paper, we presented numerical calculations of the biexciton ground-state binding energies in a quasi-one-dimensional structure subjected to a magnetic field. The field increases the binding energies and this can be utilized to modulate the nonlinear optical properties, specifically the third-order nonlinear susceptibility $\chi^{(3)}$, in a quantum wire. We found that even a modest change in the binding energy, caused by a relatively modest magnetic field of 1 T, can result in a significant change in the differential nonlinear refractive index and differential absorption in a quantum wire. Therefore, a magnetic field is a useful entity and has a practical application in this respect (calculations of refractive index and absorption will be reported in a forthcoming publication). The effect of the magnetic field is most pronounced in the widest wires because the wave function of both electrons and holes are ''softest'' in them and hence most ''squeezable'' by a magnetic field.[38] Consequently, wider wires are more suitable for magneto-optic modulators in

which a magnetic field is used to tune $\chi^{(3)}$ and therefore the nonlinear refractive index.

ACKNOWLEDGMENTS

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APPENDIX

In this appendix, we derive the expectation value of the Hamiltonian for both an exciton and a biexciton in a quantum wire subjected to a magnetic field.

For nondegenerate and isotropic bands, the Hamiltonian of a free Wannier exciton is given within the envelope-function approximation by

$$H^X=\frac{P_X^2}{2M}+\frac{p_x^2}{2\mu}+\frac{p_{y_e}^2+p_{z_e}^2}{2m_e}+\frac{p_{y_h}^2+p_{z_h}^2}{2m_h}+\frac{eB(y_e-y_h)}{M}P_X+eB(y_e/m_e+y_h/m_h)p_x+\frac{e^2B^2}{2}(y_e^2/m_e+y_h^2/m_h)$$

$$+V_{\rm conf}(y_e,y_h,z_e,z_h)-\frac{e^2}{4\pi\epsilon[x^2+(y_e-y_h)^2+(z_e-z_h)^2]^{1/2}}+sg^*\mu_B B. \tag{A1}$$

The last term (Zeeman splitting) will be neglected since the Landé g factor is very small in GaAs ($g^*=-0.445$). Following Ref. 11, we adopt the standard variational approach and write the exciton trial wave function as

$$\psi\equiv\psi(x,y_e,y_h,z_e,z_h)=g_t(x,\eta)\phi_e(y_e)\phi_h(y_h)\chi_e(z_e)\chi_h(z_h), \tag{A2}$$

where $g_t(x,\eta)$ is chosen to be the Gaussian-type ''orbital'' function:

$$g_t(x,\eta)=\frac{1}{\eta^{1/2}}\left(\frac{2}{\pi}\right)^{1/4}e^{-(x/\eta)^2} \tag{A3}$$

in which η is a variational parameter that determines the electron-hole separation. The variables $\chi_{e,h}(z_{e,h})$ are the z components of the wave functions, which are not affected by the magnetic field. They are given by particle-in-a-box states

$$\chi_{e,h}(z_{e,h})=\sqrt{\frac{2}{L_z}}\cos\left(\pi\frac{z_{e,h}}{L_z}\right). \tag{A4}$$

The electron and hole wave functions along the y direction, $\phi_{e,h}(y_{e,h})$, are to be calculated numerically when a magnetic field is present. This is done by solving the Schrödinger equation directly following the prescription given in Ref. 32.

Calculation of the expectation values of the exciton and biexciton Hamiltonians

First, we calculate the expectation value of the exciton Hamiltonian in Eq. (A1) with a trial wave function given by (A2)–(A4). For clarity, each term is treated separately.

The first term, $\langle\psi|\hat{P}_X^2/2M|\psi\rangle$, is zero since the wave function does not depend on X. The kinetic term of relative motion along the x direction can be integrated analytically using the fact that $\phi_e,\phi_h,\chi_e,\chi_h$ are normalized:

$$\left\langle\psi\left|\frac{p_x^2}{2\mu}\right|\psi\right\rangle=-\frac{\hbar^2}{2\mu}\int_{-\infty}^{+\infty}g_t g_t''\,dx$$

$$=-\frac{\hbar^2}{2\mu}\frac{2}{\eta^3}\left(\frac{2}{\pi}\right)^{1/2}\times\int_{-\infty}^{+\infty}e^{-2(x/\eta)^2}[2(x/\eta)^2-1]dx=\frac{\hbar^2}{2\mu\eta^2}.$$

In the above, a prime denotes spatial derivative (single prime is the first and double prime the second derivative). The kinetic term along the y direction is affected by the magnetic field and is given by

$$\left\langle\psi\left|\frac{p_{y_{e,h}}{}^2}{2m_{e,h}}\right|\psi\right\rangle=-\frac{\hbar^2}{2m_{e,h}}\int_{-\infty}^{+\infty}g_t^2\,dx\int_{-L/2}^{L/2}\phi_{h,e}^2\,dy_{h,e}\times\int_{-L/2}^{L/2}\phi_{e,h}\phi_{e,h}''\,dy_{e,h}$$

$$=-\frac{\hbar^2}{2m_{e,h}}\int_{-L/2}^{L/2}\phi_{e,h}\phi_{e,h}''\,dy_{e,h}.$$

Integrating by parts and taking into account that $\phi_{e,h}$ are zero at the boundaries, we can eliminate the second derivatives

$$\left\langle\psi\left|\frac{p_{y_{e,h}}{}^2}{2m_{e,h}}\right|\psi\right\rangle=\frac{\hbar^2}{2m_{e,h}}\int_{-L/2}^{L/2}(\phi_{e,h}')^2\,dy_{e,h}.$$

Along the z direction we have the regular confinement energy

$$\left\langle \psi \left| \frac{p_{z_{e,h}}{}^2}{2m_{e,h}} \right| \psi \right\rangle = \frac{\hbar^2 \pi^2}{2m_{e,h} W^2}.$$

The next two terms in the expectation value of the Hamiltonian vanish. The term $\langle \psi | [eB(y_e - y_h)/M] P_X | \psi \rangle$ is zero because, again, the wave function does not depend on X and the other term is zero due to the fact that the function g_t and its derivative are orthogonal to each other:

$$\langle \psi | eB(y_e, m_e + y_h/m_h) p_x | \psi \rangle = i\hbar eB(y_e/m_e + y_h/m_h) \int_{-\infty}^{+\infty} g_t g_t' \, dx = 0.$$

Note that the vanishing of these two paramagnetic terms makes the expectation value of the total Hamiltonian strictly real and shows that the trial wave function space is admissible. The expectation value of the remaining diamagnetic term is given by the sum of two integrals

$$\left\langle \psi \left| \frac{e^2 B^2}{2} (y_e^2/m_e + y_h^2/m_h) \right| \psi \right\rangle = \frac{e^2 B^2}{2} \int_{-\infty}^{+\infty} g_t^2 \, dx \int_{-L/2}^{L/2} \phi_e^2 \phi_h^2 (y_e^2/m_e + y_h^2/m_h) dy_e \, dy_h$$

$$= \frac{e^2 B^2}{2m_e} \int_{-L/2}^{L/2} \phi_e^2 y_e^2 \, dy_e + \frac{e^2 B^2}{2m_h} \int_{-L/2}^{L/2} \phi_h^2 y_h^2 \, dy_h.$$

Combining all the above terms with the Coulomb term, we obtain the expectation value of the exciton Hamiltonian as

$$\langle \psi | H^X | \psi \rangle = \frac{\hbar^2}{2\mu\eta^2} + \frac{\hbar^2 \pi^2}{2\mu W^2} + \frac{\hbar^2}{2m_e} \int_{-L/2}^{L/2} (\phi_e')^2 \, dy_e + \frac{\hbar^2}{2m_h} \int_{-L/2}^{L/2} (\phi_h')^2 \, dy_h + \frac{e^2 B^2}{2m_e} \int_{-L/2}^{L/2} (\phi_e y_e)^2 \, dy_e$$

$$+ \frac{e^2 B^2}{2m_h} \int_{-L/2}^{L/2} (\phi_h y_h)^2 \, dy_h - \frac{e^2}{4\pi\epsilon} \int_{\Omega} \frac{g_t^2(x,\eta) \phi_e^2 \phi_h^2 \chi_e^2 \chi_h^2}{[x^2 + (y_e - y_h)^2 + (z_e - z_h)^2]^{1/2}} \, d\zeta, \tag{A5}$$

where $d\zeta = dx \, dy_e \, dy_h \, dz_e \, dz_h$. The integration of the last (Coulomb) term is carried out over a hyper-rectangle Ω, which has an infinite interval along the x direction and limited by $\pm L_y/2$ and $\pm L_z/2$ along the y and z directions, respectively.

In order to calculate the expectation value of the biexciton Hamiltonian in Eq. (2), we rewrite the trial wave function in (4)–(10) in a form given below:

$$\Psi = \frac{1}{S} \{ e^{-(x_{1a}^2 + x_{2b}^2)/\eta^2} + e^{-[(x_{2b} - x_{ab})^2 + (x_{1a} + x_{ab})^2]/\eta^2} \} e^{-x_{ab}^2/\tau^2} \phi_\alpha(y_\alpha) \chi_\alpha(z_\alpha).$$

Denoting

$$F_1(\mathbf{x}) = e^{-(x_{1a}^2 + x_{2b}^2)/\eta^2},$$

$$F_2(\mathbf{x}) = e^{-[(x_{2b} - x_{ab})^2 + (x_{1a} + x_{ab})^2]/\eta^2},$$

and omitting normalized components $\phi_\alpha(y_\alpha)\chi_\alpha(z_\alpha)$, we take the derivatives analytically:

$$\frac{\partial^2}{\partial x_{1a}^2} \Psi = \frac{-2}{S\eta^2} \left\{ F_1(\mathbf{x}) \left(1 - 2\frac{x_{1a}^2}{\eta^2} \right) + F_2(\mathbf{x}) \left(1 - 2\frac{(x_{1a} + x_{ab})^2}{\eta^2} \right) \right\} G_{ab}(x_{ab}), \tag{A6}$$

$$\frac{\partial^2}{\partial x_{2b}^2} \Psi = \frac{-2}{S\eta^2} \left\{ F_1(\mathbf{x}) \left(1 - 2\frac{x_{2b}^2}{\eta^2} \right) + F_2(\mathbf{x}) \left(1 - 2\frac{(x_{2b} - x_{ab})^2}{\eta^2} \right) \right\} G_{ab}(x_{ab}), \tag{A7}$$

$$\frac{\partial^2}{\partial x_{ab}^2} \Psi = \frac{-2}{S} \left\{ 2F_2(\mathbf{x}) \left(\frac{1}{\eta^2} - \frac{(x_{2b} - 2x_{ab} - x_{1a})^2}{\eta^4} + \frac{2(x_{2b} - 2x_{ab} - x_{1a}) x_{ab}}{\eta^2 \tau^2} \right) + [F_1(\mathbf{x}) + F_2(\mathbf{x})] \left(\frac{1}{\tau^2} - \frac{2x_{ab}^2}{\tau^4} \right) \right\} G_{ab}(x_{ab}), \tag{A8}$$

$$\frac{\partial^2}{\partial x_{1a} \partial x_{ab}} \Psi = \frac{-2}{S\eta^2} F_2(\mathbf{x}) \left\{ 1 + \frac{2(x_{1a} + x_{ab})(x_{2b} - 2x_{ab} - x_{1a})}{\eta^2} \right\} G_{ab}(x_{ab}) + \frac{4x_{ab}}{S\tau^2\eta^2} \{ F_1(\mathbf{x}) x_{1a} + F_2(\mathbf{x})(x_{1a} + x_{ab}) \} G_{ab}(x_{ab}), \tag{A9}$$

$$\frac{\partial^2}{\partial x_{2b} \partial x_{ab}} \Psi = \frac{2}{S\eta^2} F_2(\mathbf{x}) \left\{ 1 + \frac{2(x_{2b} - x_{ab})(-x_{2b} + 2x_{ab} + x_{1a})}{\eta^2} \right\} G_{ab}(x_{ab}) + \frac{4x_{ab}}{S\tau^2\eta^2} \{ F_1(\mathbf{x}) x_{2b} + F_2(\mathbf{x})(x_{2b} - x_{ab}) \} G_{ab}(x_{ab}). \tag{A10}$$

Using the above results for the derivatives, we can calculate the expectation value of the biexciton Hamiltonian $\langle\Psi|H^{XX}|\Psi\rangle$.

The only remaining exercise is to show that the expectation values of the imaginary terms vanish. To show this, we prove the general result that the trial wave function is orthogonal to all its first derivatives along the x axis. Consider the integral

$$\int_{\Omega} \Psi \frac{\partial}{\partial x_{1a}} \Psi \, d\mathbf{x}. \tag{A11}$$

Substituting for the first derivative

$$\frac{\partial}{\partial x_{1a}} \Psi = \frac{-2}{S\eta^2} \{F_1(\mathbf{x}) x_{1a} + F_2(\mathbf{x})(x_{1a}+x_{ab})\} G_{ab}(x_{ab}), \tag{A12}$$

and integrating over y_α, z_α, and x_{1a} we obtain

$$\frac{1}{2S^2} \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} e^{-2x_{ab}^2/\tau^2} [F_1(\mathbf{x}) + F_2(\mathbf{x})]^2 \Big|_{x_{1a}=-\infty}^{x_{1a}=+\infty} dx_{2b} \, dx_{ab}. \tag{A13}$$

Based on the definition of $F_{1,2}(\mathbf{x})$, it is easy to see that the integrand in the last equation is zero. By analogy, we can prove that the expectation values of all other imaginary terms in the Hamiltonian vanish. Again, this makes the expectation value of the biexciton Hamiltonian strictly real so that the trial wave function space is admissible.

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