Theoretical studies of the effects of a magnetic field on excitonic nonlinear optical properties of quantum wires

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(Received 12 February 1996)

The magnetic-field dependence of the third-order excitonic nonlinear susceptibility $\chi^{(3)}$ in a quantum wire is explored within the rotating wave approximation. Both the real and imaginary parts of $\chi^{(3)}$, arising from population saturation of the excitonic state under optical pumping, are calculated for a GaAs wire as a function of magnetic field and pump-probe detuning frequencies. The imaginary part of $\chi^{(3)}$ exhibits a negative peak associated with the bleaching of the excitonic resonance and a positive, broad, off-resonance absorption peak associated with biexciton formation. The amplitude, line shape, and spectral frequency of both these peaks can be modulated by a magnetic field which indicates the possibility of using such a field to probe the mechanism underlying optical nonlinearity in a quantum wire. Furthermore, the field can also be used to tune the optical nonlinearity over a range of frequencies which has device applications. [S0163-1829(96)04228-2]

I. INTRODUCTION

It is well known that quasi-one-dimensional systems (quantum wires) exhibit giant third-order nonlinear susceptibility $\chi^{(3)}$ under optical pumping because of quantum confinement of excitons and polyexcitonic complexes.[1,2] A magnetic field further increases the confinement by localizing the electron and hole wave functions, leading to even larger $\chi^{(3)}$. This allows one to modulate the nonlinear refractive index and absorption (or gain) in quantum wires with a magnetic field, thereby opening up the possibility of realizing *externally tunable* couplers, limiters, phase shifters, switches, etc. Furthermore, the field can also be used as an experimental tool to extract the specific mechanism responsible for the optical nonlinearity in the system.

In this paper, we will investigate the effects of a magnetic field on optical nonlinearity (and the associated $\chi^{(3)}$) in a quantum wire caused by exciton-exciton interaction and formation of excitonic molecules (specifically biexcitons). This interaction is likely to be the dominant mechanism for optical nonlinearity, and the leading contribution to $\chi^{(3)}$ in quantum wires of most technologically important semiconductors. Recently some researchers[2,3] reported experimental observations of giant optical nonlinearity in quantum wires which they attributed to this mechanism. The enhanced nonlinearity is undoubtedly caused by quantum confinement which increases the binding energy of all excitonic complexes. Additionally, the oscillator strength for the lowest-energy exciton-to-biexciton transition increases and gives rise to huge third-order susceptibilities. This oscillator strength is already significant because the biexciton radius is very large,[4] and a second photon (two-photon absorption) in the volume of an excitonic molecule can be easily found and absorbed to create the molecule.

In Sec. II, we outline the theoretical framework that was used for calculating $\chi^{(3)}$ associated with population saturation of the excitonic state and biexciton formation. Section III presents the results of our numerical computation followed by a discussion. We also compare the results that we obtain (in the absence of any magnetic field) with the theoretical calculations of Ref. 4, and with available experimental data. Conclusions are given in Sec. IV.

II. THEORETICAL MODEL

We consider a rectangular GaAs quantum wire of the geometry shown in the inset of Fig. 1. An external magnetic field of flux density B is applied perpendicular to the wire axis, as indicated in the inset. In order to calculate $\chi^{(3)}$ for this system, we make the following assumptions: (i) $\chi^{(3)}$ is measured in a nondegenerate pump-and-probe spectroscopy experiment with nearly resonant pumping of the excitonic state; (ii) the exciton gas is sufficiently dilute that higher-order complexes (beyond the biexcitonic state) can be neglected; (iii) the rotating wave approximation[5] is valid; and (iv) in the frequency range of interest, the lowest-lying states are the major contributors to $\chi^{(3)}$ and therefore we can treat the system approximately as a two-level system.

Following Ishihara[2] and Madarasz *et al.*,[4] we can write the third-order susceptibility as follows:

$$\chi^{(3)}=\frac{-2}{\pi\sqrt{2\pi}}\frac{\tau}{\eta^2}\frac{N_0e^4}{m_0^2\omega_{g0}^4}E_p^2\left[\frac{1}{(\omega_1-\omega_{g0}+i\Gamma_{g0})}-\frac{1}{(\omega_1-\omega_{g0}+\omega_b+i\Gamma_{bg})}\right]\times\sum_{r=1}^{2}\left\{\frac{1}{\hbar^3(\omega_r-\omega_2+i\gamma)}\left[\frac{1}{(\omega_{g0}-\omega_2+i\Gamma_{g0})}+\frac{1}{(\omega_r-\omega_{g0}+i\Gamma_{g0})}\right]\right\}, \quad (1)$$

where ω_2 and ω_1 are the pump and probe frequencies, $\hbar\omega_{g0}$ is the exciton ground-state energy, $\hbar\omega_b$ is the biexciton binding energy, m_0 is the rest mass of a free electron, and N_0 is the average areal density of unit cells. The quantities Γ_{ij} and γ are the transverse and longitudinal broadening parameters (or damping constants), and E_p is the Kane matrix

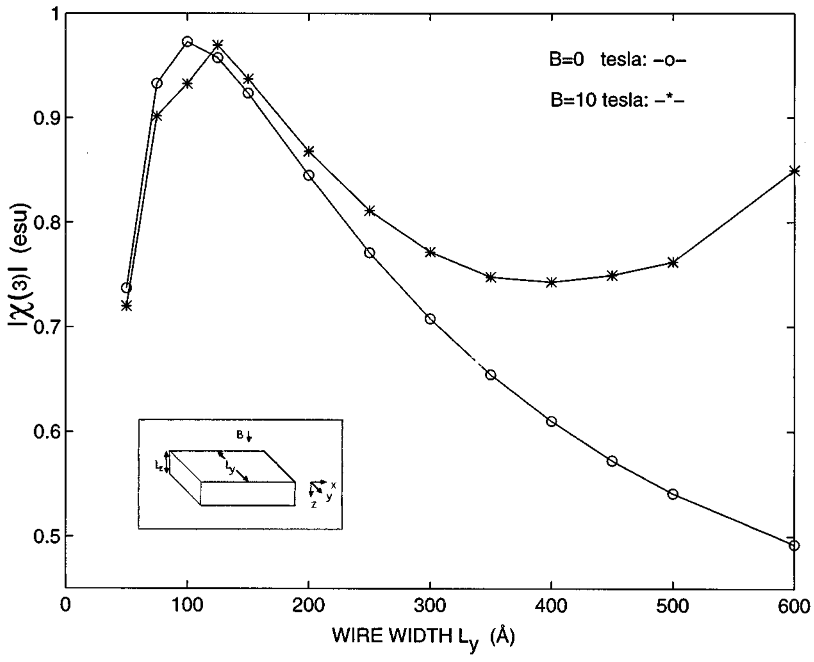

FIG. 1. The absolute value of the third-order susceptibility $\chi^{(3)}$ (for resonant excitation) as a function of wire width for two different values of magnetic field. The inset shows the wire geometry and the orientation of the magnetic field. The wire thickness along the z direction is 200 Å .

element. The indices i or j indicates system ground state (0), exciton ground state (g), and biexciton ground state (b). The values of the parameters used are listed in Table I, and correspond to GaAs. Parameters η and τ physically correspond to the exciton and biexciton correlation lengths (electron hole and hole hole mean separations in the two cases), and have to be determined variationally for each magnetic-field strength and for each set of wire dimensions following the prescription given in Refs. 6 and 7.

The exciton ground-state energy $\hbar\omega_{g0}$ is defined as follows:

$$E_g^X=\hbar\omega_{g0}=E_G+E_{e1}+E_{\mathrm{hh1}}-E_B^X, \tag{2}$$

where E_G is a bulk band gap of the material, E_{e1} and $E_{\mathrm{hh}\,1}$ are the lowest electron and the highest heavy-hole magnetoelectric subband bottom energies in a quantum wire (measured from the bottom of the bulk conduction band and the top of the bulk valence band) respectively, and E_B^X is the ground-state exciton binding energy which is also determined variationally.[6,7]

One should note from Eq. (1) that $\chi^{(3)}$ is a strong function of the transverse and longitudinal broadening parameters Γ_{ij} and γ. Physically, γ is related to the population decay rate of the excitonic states. The smaller the value of γ, the larger the lifetime of excitons and the higher the probability of forming a biexciton in a two-step photon absorption. The transverse broadening parameters Γ_{ij} represent, for $i\neq j$, the phenomenological coherence decay rate of the ij transition, while, for $i=j$, they describe the population decay of the state i. The population decay rate, in its turn, is determined by the dominant scattering mechanism in the sample. In most cases, the values of Γ_{ij} and γ are difficult to obtain experimentally, and fairly difficult to estimate theoretically. Moreover, these parameters could be strong functions of the confinement, population density of excitons, magnetic field, and temperature. In view of little experimental data available, and in order to simplify the calculations, we assume that $\Gamma_{ij}=\Gamma$ for all i and j.

TABLE I. Physical parameters for GaAs.

$E_G=1.519$ eV
$\hbar\Gamma=3$ meV
$E_p=23$ eV
$N_0=7.89\times10^{14}/\mathrm{cm}^2$

Since in this work we are interested in the modulation of the nonlinear response of quantum wires with a magnetic field, the influence of the field on the above parameters is especially important. The value of Γ in quantum wires is primarily determined by carrier-phonon interaction.[8] As shown in Ref. 8, the scattering rates associated with these interactions can be affected by a magnetic field at any given kinetic energy of an electron or hole. However, when the rates are averaged over the energy, the magnetic-field dependence turns out to be quite weak. As a first approximation, we can therefore consider the rates to be independent of the magnetic field. We also neglect thermal broadening of the damping parameters, since it is less important in quantum confined systems than in bulk.[9] An important property of Eq. (1) is the following. If all the transverse relaxation parameters are assumed to be equal (like in our case) and the biexciton binding energy $(\hbar\omega_b)$ approaches zero, then $\chi^{(3)}$ vanishes. This is a reflection of the well-known fact that noninteracting ideal independent bosons do not show any nonlinearity.[10] Consequently, exciton-exciton interaction, leading to biexciton formation, is necessary for the existence of nonlinearity.

A calculation of the excitonic contribution to $\chi^{(3)}$ requires that the exciton and biexciton binding energies be obtained first. Additionally, the parameters η and τ need to be found. For details of computing these energies and these parameters in the case of a quantum wire subjected to a magnetic field, we refer the reader to our past work.[6,7,11] Once these quantities are evaluated, we can calculate $\chi^{(3)}$ from Eq. (1) as a function of magnetic field, wire width, and pump-probe detuning frequencies.

III. RESULTS AND DISCUSSION

All results in this paper are pertinent to GaAs quantum wires. In Fig. 1, we plot the absolute value of $\chi^{(3)}$ as a function of the wire width for a fixed wire thickness of 200 Å , with and without a magnetic field. The susceptibility peaks at about 0.97 esu, corresponding to a wire width of about 100–120 Å . The sharp drop at smaller wire widths is caused by a fast rise in electron and hole confinement energies E_{e1} and E_{hh1} with shrinking wire width. This rise is faster than the rise in the exciton binding energy E_B^X, which eventually leads to a decrease in $|\chi^{(3)}|$. For dimensions larger than 120–150 Å , the ground-state energy varies little, and the behavior of $\chi^{(3)}$ is primarily determined by the effective exciton and biexciton correlation lengths τ and η. When no magnetic field is present, both τ and η increase with increasing wire width, but η increases at a faster rate. Consequently, the term (τ/η^2) decreases monotonically with increasing wire width, making $\chi^{(3)}$ decrease. This decrease is somewhat offset by the variation of the ground-state exciton energy, which causes the roll-off rate to be more gentle than at small wire widths. For a nonzero magnetic field (of 10 T), the exciton radius η in a quantum wire has a *nonmonotonic* dependence on wire width which results in a well-resolved maximum in η. This rather surprising behavior was reported by us earlier,[11] and explained in terms of the complementary roles of electrostatic and magnetostatic confinement. The biexciton radius τ also has a maximum, but it is much broader and shallower than the one associated with η. Consequently, there exists a minimum in the ratio τ/η^2 which causes $\chi^{(3)}$ to exhibit a nonmonotonic dependence on the wire width (past the maximum) when a magnetic field is present. This accounts for the broad valley in the curve when a magnetic field of 10 T is applied.

In Figs. 2–4 we have calculated Im$\chi^{(3)}$ for a two-beam experiment in which the frequency of one beam, the pump, is fixed, and that of the other, the probe, is allowed to vary over a frequency range of $\hbar\Delta\omega = 40$ meV centered around the pump frequency. In Figs. 2 and 4, the pump frequency is chosen to be resonant with the exciton ground-state transition, and in Fig. 3 the pump is detuned from the exciton resonance by a frequency $-\Gamma/(2^{1/2}\hbar)$. It is important to remember that since the ground-state exciton binding energy is a function of magnetic field, the pump should be retuned every time the magnetic field changes. In all figures, the imaginary part of the third-order susceptibility is plotted for four values of magnetic field. The significance of Im$\chi^{(3)}$ is in that it is proportional to the differential change in the optical transmission or in the absorption coefficient $\Delta\alpha$. This relation is given by the formula[12]

$$\mathrm{Im}\chi^{(3)} = \frac{c^2 n_0^2 \Delta\alpha(\omega)}{8\pi^2 \omega I(\omega)}, \tag{3}$$

where n_0 is a linear refractive index, c is the speed of light, and $I(\omega)$ is the intensity of a resonant monochromatic light beam. Positive peaks in Im$\chi^{(3)}$ will correspond to strong absorption and negative peaks to the spectral regions of strong transmission (bleaching bands).

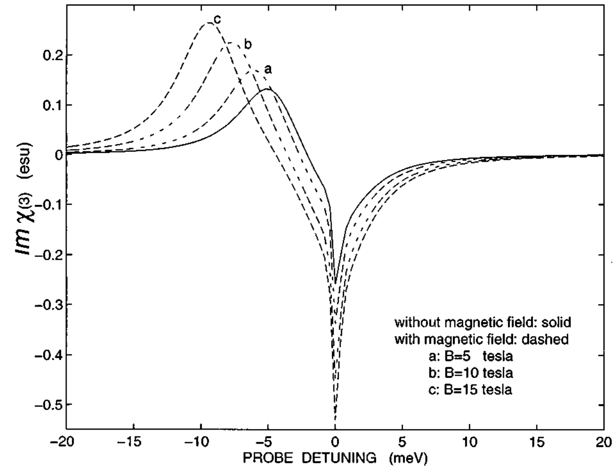

FIG. 2. The imaginary part of the third-order susceptibility as a function of the probe detuning energy for a dual-beam pump-probe experiment. The pump is set at exciton resonance for each value of magnetic field, and the longitudinal broadening parameter is assumed to be one-tenth the value of the transverse broadening parameters.

Figure 4 illustrates the dependence of third-order susceptibility on the longitudinal broadening parameters γ (attention should be paid to the change of scale along the vertical axis compared to Fig. 2). The difference between Figs. 2 and 4 is that, in the former case, the longitudinal broadening parameter is one-tenth that of the transverse broadening parameter, whereas in the latter figure they are equal. Im$\chi^{(3)}$ is extremely sensitive to the magnitude of γ: varying this damping parameter from $\gamma = 0.1\Gamma$ to $\gamma = \Gamma$ changes the value of Im$\chi^{(3)}$ by more than an order of magnitude.

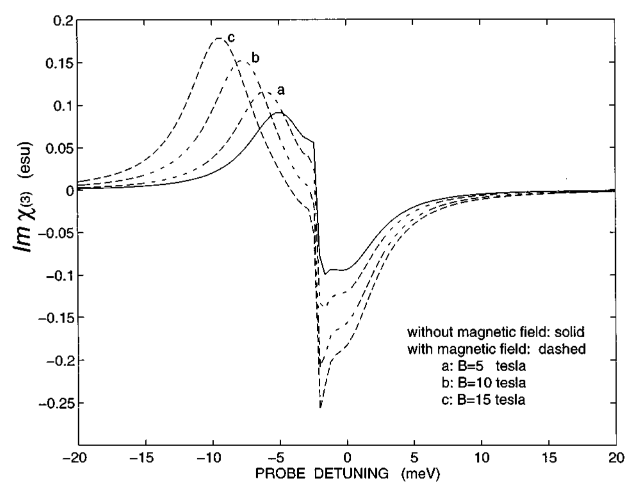

FIG. 3. The imaginary part of the third-order optical susceptibility as a function of the probe detuning energy for a dual-beam pump-probe experiment. The pump is detuned slightly below the exciton resonance for each value of magnetic field. Again, the longitudinal broadening parameter is one-tenth the value of the transverse broadening parameters as in Fig. 2.

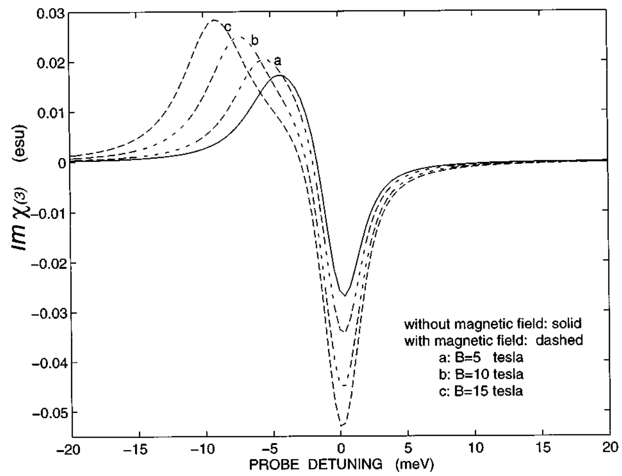

FIG. 4. The imaginary part of the third-order optical susceptibility as a function of the probe detuning energy for a dual-beam pump-probe experiment. The pump is set at exciton resonance at each value of a magnetic field as in Fig. 2, but the longitudinal broadening parameter is equal to the transverse broadening parameters. Attention should be paid to the change of scale along the vertical axis compared to Fig. 2.

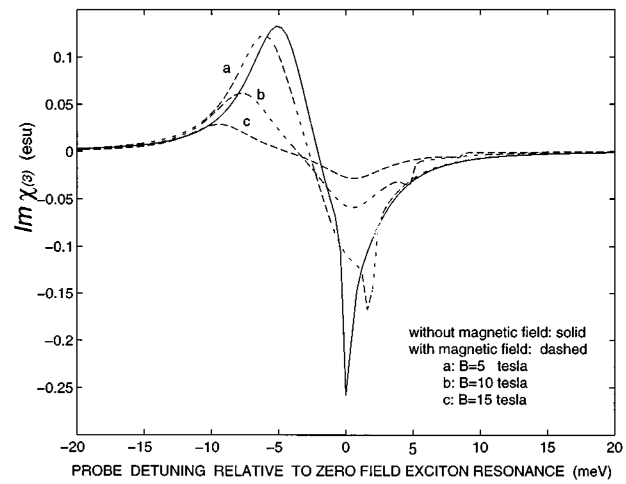

FIG. 5. The imaginary part of the third-order susceptibility as a function of the probe detuning energy for a dual-beam pump-probe experiment. The pump is now set at the exciton resonance at zero magnetic field and not retuned every time the magnetic field changes. Again, the longitudinal broadening parameter is one-tenth the value of the transverse broadening parameters as in Fig. 2. Due to the pump detuning at nonzero magnetic field, both exciton and biexciton resonances are quenched.

A pronounced negative peak is present in all of the spectra. It represents a strong transmission which is due to a saturation (or bleaching) of the excitonic state. Physically, the initial exciton population created by the pump beam tends to amplify the probe beam when its energy is tuned at or near the exciton ground state (this corresponds to the linear gain peak). A magnetic field makes the peak deeper, without significant broadening, thus enhancing transmission further. At first glance, it may surprise the reader that the peak is not shifted in frequency by the magnetic field even though the exciton binding energy depends on the magnetic field. The reason for this is that the probe beam is retuned to the exciton resonance for each value of the magnetic field, so that no frequency shift should arise.

Another feature of interest in all of these plots is in the region of positive $\mathrm{Im}\chi^{(3)}$ that corresponds to optical absorption. This absorption may be attributed to the formation of the excitonic molecule (biexciton).[4,13,14] The initial exciton population enables the probe to be more strongly absorbed when its energy matches the exciton-biexciton transition energy $\hbar(\omega_{g0}-\omega_b)$. Consequently, at zero magnetic field, the positive peak is separated from the exciton resonance by approximately -5 meV, which corresponds to the biexciton binding energy for this case. At a magnetic flux density of 5 T the peak separation is about -7.5 meV, which again corresponds to the biexciton binding energy, this time for a flux density of 5 T. Such a dependence of the energy difference between transmission (negative peak) and absorption (positive peak) on a magnetic field can be used to modulate the optical properties of a quantum wire with an external field. It can also be used as a means to determine the particular mechanism causing nonlinearity in a quantum wire. Note that the energy separation between the peaks is not seriously affected by the increasing damping (see Fig. 4) or by the detuning of the pump (see Fig. 3). As a result, this technique of modulation with a magnetic field cannot only be used to extract the mechanism responsible for nonlinearity, but also to measure biexciton binding energies and their dependences on a magnetic field.

Figure 5 shows $\mathrm{Im}\chi^{(3)}$ for four values of a magnetic field and a small longitudinal damping $\gamma=0.1\Gamma$. The difference between this case and the one presented in Fig. 2 is that now the pump beam is permanently tuned to the exciton resonance at zero magnetic field, and not retuned every time the magnetic field changes. Since the ground-state exciton energy is a function of magnetic field, the negative peak in Fig. 5 is now shifted by the applied magnetic field. Another im-

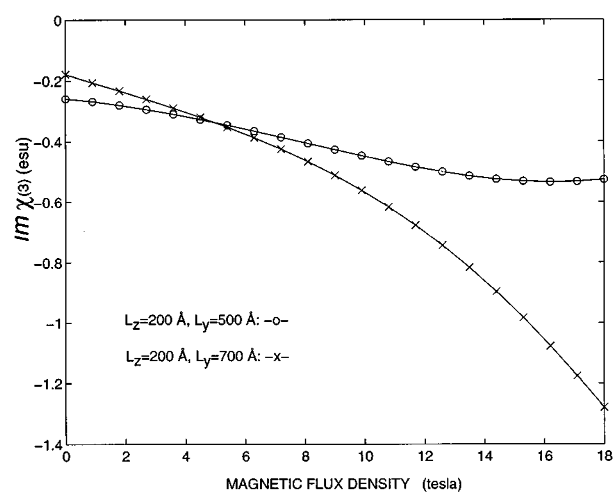

FIG. 6. The imaginary part of the third-order susceptibility as a function of an applied magnetic field for two different wire widths. The thickness of the wire is 200 Å . The effect of a magnetic field is more pronounced for a wider wire since the wave functions of the electrons and holes are ‘‘softer’’ and more ‘‘squeezable’’ by a magnetic field if the wire is wider.

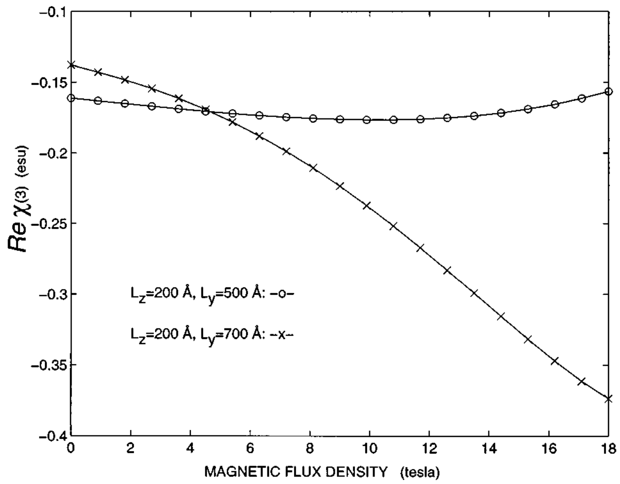

FIG. 7. The real part of the third-order susceptibility as a function of an applied magnetic field for two different wire widths. The thickness of the wire is 200 Å .

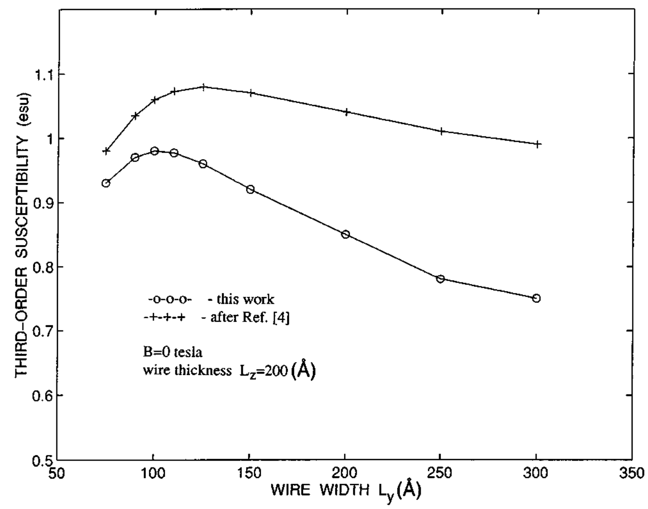

FIG. 8. Comparison of the wire-width dependence of the absolute value of the third-order susceptibility (resonant excitation) with the result given in Ref. 4. The discrepancy (15–20 %) is an aftermath of different values of material parameters used in the two studies. The thickness of the wire is 200 Å in both cases.

portant feature to notice is that when the pump is *not tuned* to an exciton resonance at a particular magnetic field, the magnitudes of both the positive and negative peaks are reduced by the magnetic field, leading to a quenching of both absorption and transmission. This effect is opposite to what is observed when the pump is tuned to the exciton resonance. However, this effect has immediate device applications such as the quantum-confined Lorentz effect.[15]

In Figs. 6 and 7 we present the imaginary and real parts of the third-order susceptibility as functions of an applied magnetic field for two different wire widths. The effect of a magnetic field is quite strong. At a magnetic flux density of 10 T, $|\mathrm{Im}\chi^{(3)}|$ is approximately three times larger than at zero field for the 700 Å -wide wire. Again, the effect of a magnetic field is more pronounced for wider wires, since in wider wires the magnetostatic localization is stronger.[11]

We have compared our results for zero magnetic field with those given in Ref. 4. The ground-state binding energies for both excitons and biexcitons are in excellent qualitative agreement. Some discrepancy can be attributed to different values of electron and hole effective masses used in our calculations and the calculations of Ref. 4. We also compare our binding energy results with the experimental observations of Refs. 16–18. Since Ref. 16 employed *T*-shaped edge quantum wires whose geometries are very different from ours, a direct quantitative comparison is not possible. Nonetheless, we find that our numerical results are within the same order of magnitude as theirs, and that their data are in excellent qualitative agreement with ours. The $\mathrm{Im}\chi^{(3)}$ curves for zero magnetic field are also consistent with those given in Refs. 4, 14, and 19. In Fig. 8, we present a direct comparison of the wire-width dependence of $\chi^{(3)}$ obtained with zero magnetic field with the result given in Ref. 4. The slight discrepancy of 15–20 % is a result of using different electron and hole effective masses in the calculation of exciton and biexciton binding energies. Other factors, such as the exact choice of the variational wave function, also contribute to the difference.

Again, in most cases, a complete quantitative comparison of the data is not possible because of the different geometry of the wires used. In our model calculations, we utilized wire sizes that are typical for structures delineated by lithography, and that correspond to the regime of moderate quantum confinement.

IV. CONCLUSION

We have investigated the effects of a magnetic field on the third-order nonlinear susceptibility in quantum wires. The magnetic field modulates the frequency shift between the transmission peak associated with the bleaching of excitonic transitions and the absorption peak associated with the formation of excitonic molecules (biexcitons). Additionally, the field also affects the magnitudes of the peaks. These effects can be utilized for magneto-optical devices, and can also be used as a tool to probe the precise mechanism responsible for optical nonlinearity in quantum wires.

ACKNOWLEDGMENTS

The authors are indebted to Dr. F. Madarasz and A. Ivanov for insightful discussions. They would like to thank the University of Nebraska for its hospitality. This work was partially supported by the Army Research Office under Grant No. DAAH04-95-1-0586.

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