Superlattices and Microstructures, Vol. 23, No. 6, 1998
Article No. sm960258

Magnetostatic modulation of nonlinear refractive index and absorption in quantum wires

A. BALANDIN, S. BANDYOPADHYAY†
Department of Electrical Engineering, University of Nebraska, Lincoln, NE 68588, U.S.A.

(Received 15 July 1996)

The magnetic-field dependence of the nonlinear differential refractive index Δn and absorption $\Delta\alpha$ in quantum wires—measured by non-degenerate pump and probe spectroscopy—is investigated theoretically. The nonlinearities arise from population saturation of the excitonic state under optical pumping and the formation of biexcitons (excitonic molecules). Both Δn and $\Delta\alpha$ exhibit positive and negative peaks at certain pump and probe detuning frequencies associated with the formation of biexcitons and bleaching of excitons, respectively. The amplitude, lineshape and the frequency at which these peaks occur can be modulated by a magnetic field which opens up the possibility of realizing novel magneto-optical devices. Additionally, the magnetic field may allow us to realize a relatively large variation in the differential refractive index over a range of frequencies without significant accompanying absorption, thereby allowing the observation of optical bistability.

Key words: quantum wires, nonlinear optics, excitons.

1. Introduction

It is well known that quantum-confined structures exhibit pronounced optical nonlinearities of excitonic origin [1]. The enhanced nonlinearities arise from one-dimensional quantum confinement which increases the binding energy of all excitonic complexes and the oscillator strengths for excitonic transitions. In this paper, we report how a magnetic field influences the nonlinear differential refractive index Δn and absorption $\Delta\alpha$ in a quantum wire. This study is motivated by the realization that any significant modulation of these quantities by a magnetic field can lead to novel device applications, as well as provide a tool for probing the origin of optical nonlinearity in a quantum structure.

The physical processes associated with nonlinear refraction and absorption in quantum confined systems is a well-researched topic. In reference [2, 3], the authors reported room-temperature measurements of Δn in GaAs multiquantum well structures and found it to be 0.01 at low levels of excitation and 0.05 at high levels. They attributed the nonlinearity to band filling. In quantum wires (as opposed to wells), we can expect Δn and $\Delta\alpha$ to be much larger because of the additional degree of confinement and the much higher density of states at the subband edges. Indeed, our theoretical calculations indicate that Δn can be an order of magnitude larger in quantum wires than that found in quantum wells.

†The authors are on leave from the University of Notre Dame, Notre Dame, IN 46556, U.S.A.

0749–6036/98/061197 + 07 $25.00/0

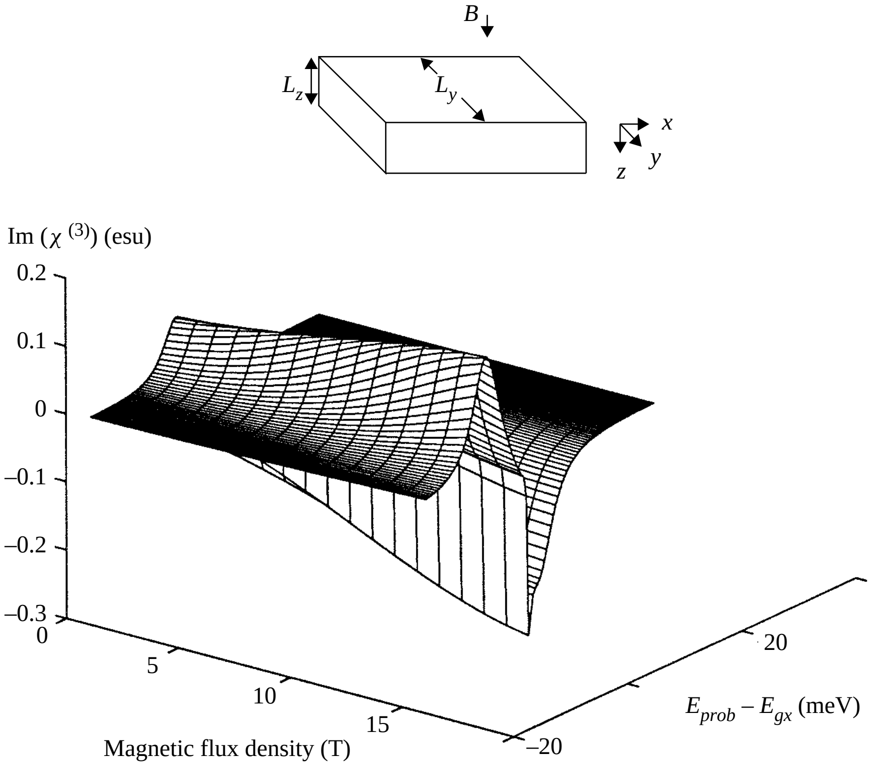

Fig. 1. The imaginary part of the third-order nonlinear susceptibility $\chi^{(3)}$ as a function of pump and probe detuning energy and magnetic-flux density. The pump is tuned slightly below the exciton resonance at each value of the magnetic field and the longitudinal broadening parameter is assumed to be one-tenth that of the transverse broadening parameter. The wire dimension is $L_z = 200$ Å and $L_y = 500$ Å. The inset shows the wire geometry and orientation of the magnetic field.

2. Theory

We wish to calculate the differential refractive index and absorption associated with the third-order nonlinear susceptibility $\chi^{(3)}$ in quantum wires. For this, we consider a rectangular quantum wire of the geometry shown in the inset of Fig. 1. An external magnetic field is applied perpendicular to the wire axis. We assume near-resonant pumping of the excitonic state in a non-degenerate pump and probe spectroscopy experiment and calculate the changes in refractive index Δn and absorption $\Delta\alpha$ relevant to this situation. The actual measurable quantities in such an experiment are usually the transmission in the absence (T_0) and in the presence (T) of the pump. The differential transmission spectra can be found from these quantities as $D = (T - T_0)/T_0$. For small values of the differential transmission (well below unity), D is proportional to the differential absorption $\Delta\alpha$. In fact, $D \approx -\Delta\alpha d$, where d is the wire thickness along the direction of the optical beam.

The nonlinear differential refractive index and absorption can be evaluated theoretically as in [4]. These quantities are given by

$$\Delta n = \frac{2\pi}{\sqrt{\epsilon_r}} \mathrm{Re}\chi^{(3)}, \tag{1}$$

and

$$\Delta\alpha = \frac{4\pi\omega}{c\sqrt{\epsilon}} \mathrm{Im}\chi^{(3)}, \tag{2}$$

Table 1: Values of the various parameters for GaAs used to calculate the nonlinear susceptibility $\chi^{(3)}$.

$E_{g_0} = 1.519$ eV
$\hbar\Gamma = 3$ meV
$E_p = 23$ eV
$N_0 = 7.89 \times 10^{14}$ cm^{-2}

where c is the speed of light, ϵ_r is a relative dielectric constant of the material, ω is a near-resonant frequency of the pump beam, and $\mathrm{Im}\chi^{(3)}$, $\mathrm{Re}\chi^{(3)}$ are the imaginary and real parts of the nonlinear third-order susceptibility $\chi^{(3)}$ which need to be calculated.

The general derivation of $\chi^{(3)}$ for low density of excitonic complexes can be found in [5]. This derivation is based on summation over 16 double Feynman diagrams. In the frequency range of interest, the lowest-lying states are the major contributors to $\chi^{(3)}$ and this allows us to reduce the expression for $\chi^{(3)}$ to a simplified form given by

$$\chi^{(3)} = \frac{-2}{\pi\sqrt{2\pi}}\frac{\tau}{\eta^2}\frac{N_0 e^4}{m_0^2\omega_{g_0}^4}E_p^2\left[\frac{1}{(\omega_1-\omega_{g_0}+i\Gamma_{g0})}-\frac{1}{(\omega_1-\omega_{g_0}+\omega_b+i\Gamma_{bg})}\right]$$
$$\times\sum_{r=1}^{2}\left\{\frac{1}{\hbar^3(\omega_r-\omega_2+i\gamma)}\left[\frac{1}{(\omega_{g_0}-\omega_2+i\Gamma_{g0})}+\frac{1}{(\omega_r-\omega_{g_0}+i\Gamma_{g0})}\right]\right\}$$
$$+\frac{1}{(\omega_1+\omega_2-2\omega_{g_0}+\omega_b+i\Gamma_{b0})}\left[\frac{1}{(\omega_1-\omega_{g_0}+i\Gamma_{g0})}+\frac{1}{(\omega_2-\omega_{g_0}+i\Gamma_{g0})}\right], \quad (3)$$

where ω_2 and ω_1 are the pump and probe frequencies, $\hbar\omega_{g_0}$ 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 element. The index i or j indicates system ground state (0), exciton ground state (g), and biexciton ground state (b). Numerical values of the various quantities used in our calculations are given in Table 1. 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 [6, 7].

The exciton ground-state energy $\hbar\omega_{g_0}$ is defined as follows

$$E_g^X = \hbar\omega_{g_0} = E_G + E_{e_1} + E_{hh_1} - E_B^X, \quad (4)$$

where E_G is the bulk band gap of the material, E_{e_1}, E_{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].

It should be noted from eqn (3) 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 $i - j$ 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 the little experimental data available, and in order to simplify the calculations, we assume that $\Gamma_{ij} = \Gamma$ for all i, j.

Since in this work we are interested in the modulation of the differential refractive index and absorption of quantum wires with a magnetic field, the influence of the field on all parameters in eqn (3) is especially important. The value of Γ in quantum wires is primarily determined by carrier–phonon interactions [8]. As shown in [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 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 eqn (3) is the following: if all the transverse relaxation parameters are assumed to be equal (as in our case) and the biexciton binding energy ($\hbar\omega_b$) approaches zero, then $\chi^{(3)}$ vanishes. This is a manifestation of the well-known fact that noninteracting ideal independent bosons do not show any nonlinearity [9]. Consequently, exciton–exciton interaction, leading to biexciton formation, is necessary for the existence of this type of the nonlinearity.

A calculation of the excitonic contribution to $\chi^{(3)}$ requires that the exciton and biexciton binding energies be obtained first. Additionally, all 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]. Once these quantities are evaluated, we can calculate $\chi^{(3)}$ from eqn (3) as a function of a magnetic field, wire width and pump and probe detuning frequencies. The differential refractive index and absorption are then computed from the real and imaginary parts of $\chi^{(3)}$ as given by eqns (1) and (2).

3. Results and discussion

All results in this paper are pertinent to GaAs quantum wires. In Fig. 1, we present a three-dimensional plot of $\mathrm{Im}\chi^{(3)}$ 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. The pump frequency is chosen to be slightly detuned from the exciton resonance by a frequency $-\frac{\sqrt{2}}{2}\Gamma/\hbar$. The quantum-wire dimensions which have been used to plot this figure are $L_y = 500$ Å, $L_z = 200$ Å. The longitudinal broadening parameter γ is chosen to be one-tenth that of the transverse broadening parameter Γ which is a physically reasonable ratio.

A pronounced negative peak is present in the spectrum for all values of a magnetic field. It represents 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. Another feature of interest is in the region of positive $\mathrm{Im}\chi^{(3)}$ that corresponds to optical *absorption*. This absorption may be attributed to the formation of an excitonic molecule (biexciton). The initial exciton population enables the probe to be more strongly absorbed when its energy matches the exciton–biexciton transition energy $\hbar(\omega_{g_0} - \omega_b)$.

The same basic features are repeated in the absorption spectrum presented in Fig. 2. Here we plot the differential absorption $\Delta\alpha$ as a function of the pump and probe detuning frequencies when the longitudinal broadening parameter γ is one-tenth of the transverse broadening parameter Γ. As we can see, when the pump frequency is nearly resonant with the excitonic absorption, the swing in the differential absorption $\Delta\alpha$ is very large (0.5×10^5 cm^{-1}–10^5 cm^{-1}). Another feature to note is that the frequency separation between the positive and negative peaks (associated with biexciton formation and exciton bleaching) is quite sensitive

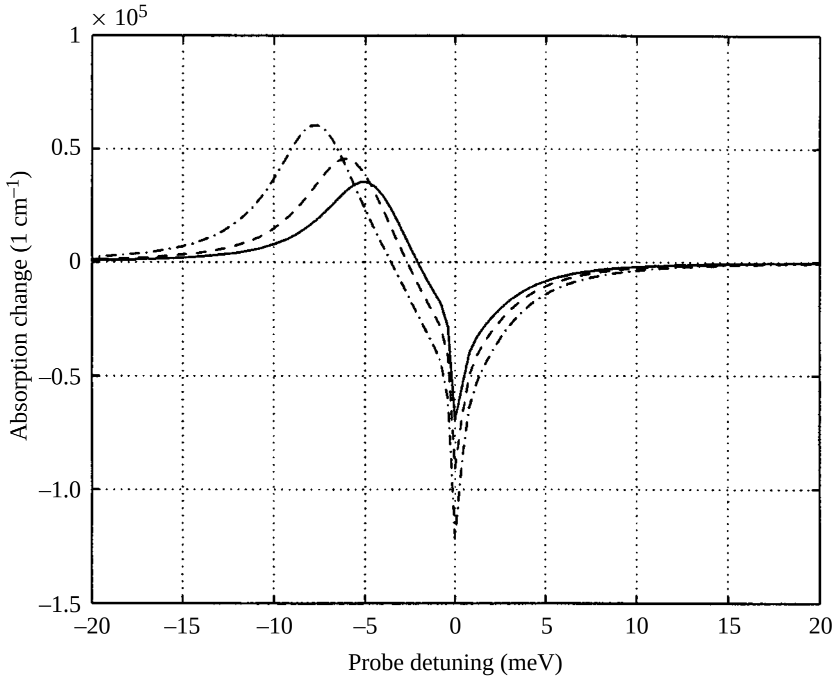

Fig. 2. The differential absorption $\Delta\alpha$ as a function of pump and probe detuning energies for different values of a magnetic field. The pump is set at exciton resonance for each value of a magnetic field. The longitudinal broadening parameter is one-tenth that of the transverse broadening parameter.

to the magnetic field. This separation is not sensitive to damping (values of γ and Γ) or slight detuning of the pump. Therefore, we can use a magnetic field to tune this separation, thus realizing magneto-optical devices.

In Fig. 3, we show the differential refractive index Δn as a function of the pump and probe detuning frequency and the magnetic field. More complicated behaviour is exhibited by Δn, with a strong negative peak occurring at the energy between the positive and negative resonances in the absorption change. The negative peak is related to the fact that $\Delta\alpha$ has a positive dispersive peak on its low-energy side.

Although not shown in this paper, we also found that damping has a deleterious effect on the nonlinearity. As the damping parameter γ increases from 0.1Γ to Γ, the swing in Δn drops from 0.4 to 0.05 when no magnetic field is present, resulting in a 20-fold reduction in the nonlinearity. However, when a magnetic flux density of 10 T is present, Δn drops by only a factor of 6. Therefore, a magnetic field makes the nonlinearity less sensitive to damping.

The strong dependence of Δn and $\Delta\alpha$ on an external magnetic field has an important consequence for device applications. One possible application of band-gap resonant optical nonlinearities in quantum-confined systems is *optical bistability* and switching devices associated with it. Miller *et al.* [2] pointed out that in order to achieve optical bistability, one should provide a *large refractive index swing* at a relatively *low absorption* level. For bistable etalons using quantum wells, the relationship between minimum index change and absorption in the material for bistability to be observable can be written as $\Delta n/\alpha\gamma > \sqrt{3}/6\pi$, where λ is the wavelength of the pump beam. Using this criterion, Miller *et al.* [2] concluded that bistability is not achievable in quantum-well etalons from excitonic mechanisms alone since in the region of large Δn, excitonic absorption is also very high. However, in quantum wires, the criterion for bistability can be met, especially in the presence of a magnetic field. This is a significant advantage.

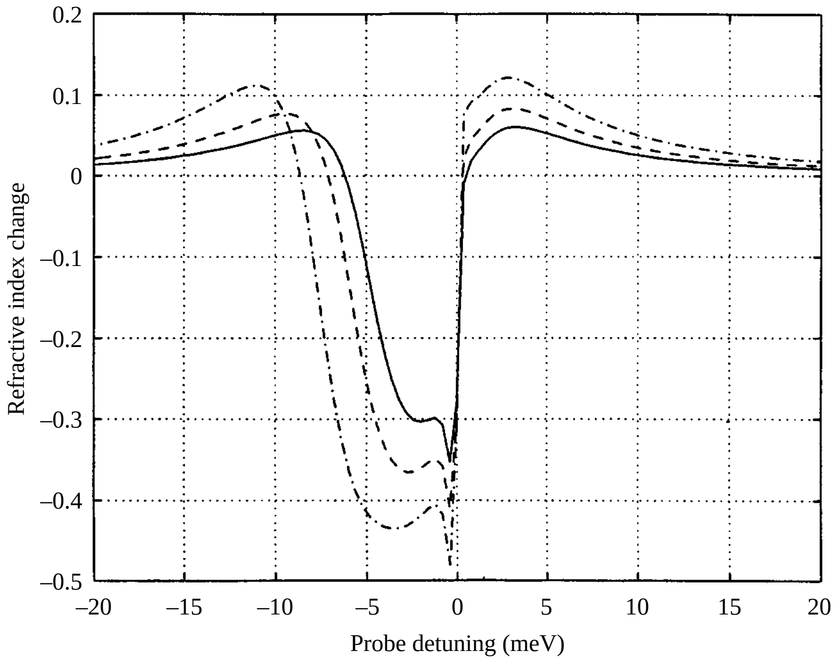

Fig. 3. The differential refractive index Δn as a function of pump and probe detuning energies for different values of a magnetic field. All parameters and conditions are the same as in Fig. 2.

4. Conclusion

In conclusion, we have investigated the dependence of Δn and $\Delta\alpha$ in a quantum wire on an external magnetic field. We found that the field makes these differential parameters less sensitive to damping and may make it possible to observe optical bistability. Additionally, the field can modulate the spectral characteristics of Δn and $\Delta\alpha$ which may have device applications.

Acknowledgements—The authors are indebted to Dr Madarasz, University of Alabama, Huntsville and Dr Dneprovskii, Moscow State University for many insightful discussions. AB acknowledges support of the University of Notre Dame Zahm Research Travel Fund. This work was partially supported by the Army Research Office under Grant number DAAH04-95-1-0586.

References

[1] See, for example, D. S. Chemla, D. A. B. Miller, and S. Schmitt-Rink, *Optical Nonlinearities and Instabilities in Semiconductors*, edited by Hartmut Haug (Academic Press, New York, 1988) and references therein; D. S. Chemla, D. A. B. Miller, and P. W. Smith, *Semiconductors and Semimetals*, edited by Raymond Dingle (Academic Press, San Diego, 1987) Vol. 24, Chap. 5.

[2] A. Miller, P. K. Milsom, and R. J. Manning, *Nonlinear Optics and Optical Computing*, edited by S. Martellucci and A. N. Chester (Plenum Press, New York, 1990) p. 119.

[3] N. Peyghambarian, S. W. Koch, H. M. Gibbs, and H. Haug, *Nonlinear Optics and Optical Computing* edited by S. Martellucci and A. N. Chester (Plenum Press, New York, 1990) p. 99.

[4] L. Banyai, Y. Z. Hu, M. Lindberg, and S. Koch, Phys. Rev. **B38**, 8142 (1988).

[5] F. L. Madarasz, F. Szmulowicz, F. K. Hopkins, and D. L. Dorsey, Phys. Rev. **B49**, 13528 (1994); F. L. Madarasz, F. Szmulowicz, F. K. Hopkins, and D. L. Dorsey, Phys. Rev. **B52**, 8964 (1995); F. L. Madarasz, F. Szmulowicz, F. K. Hopkins, and D. L. Dorsey, J. Appl. Phys. **75**, 639 (1994).
[6] A. Balandin and S. Bandyopadhyay, Phys. Rev. **B52**, 8312 (1995); A. Balandin and S. Bandyopadhyay, Phys. Rev. **B54**, 5721 (1996).
[7] A. Balandin and S. Bandyopadhyay, Superlattices Microstruct. **19**, 97 (1996). For the influence of a magnetic field on electron and hole wavefunctions see also A. Balandin, and S. Bandyopadhyay, J. Appl. Phys. **77**, 5924 (1995).
[8] N. Telang and S. Bandyopadhyay, Phys. Rev. **B48**, 18002 (1993); N. Telang and S. Bandyopadhyay, Appl. Phys. Lett. **62**, 3161 (1993).
[9] H. Ishihara and K. Cho, Phys. Rev. **B42**, 1724 (1990).