PHYSICAL REVIEW B VOLUME 44, NUMBER 23 15 DECEMBER 1991-I

Nonlinear absorption of two-dimensional magnetoexcitons in $In_xGa_{1-x}As/In_yAl_{1-y}As$ quantum wells

T. Rappen, J. Schröder, A. Leisse, and M. Wegener
Institut für Physik, Universität Dortmund, 4600 Dortmund 50, Federal Republic of Germany

W. Schäfer
Forschungszentrum Jülich, HLRZ, 5170 Jülich, Federal Republic of Germany

N. J. Sauer and T. Y. Chang
AT&T Bell Laboratories, Holmdel, New Jersey 07733

(Received 2 July 1991)

Applying a perpendicular magnetic field to a semiconductor quantum well results in a transition from a two-dimensional into a zero-dimensional system. The mutual interaction of the magnetoexciton levels is studied by populating the 1*s* magnetoexciton under steady-state conditions and measuring the related changes in absorption. We find redshifts of all energetically higher magnetoexcitons and a blueshift of the 1*s* exciton due to repulsive exciton-exciton interaction. The experimental results are compared with solutions of the semiconductor Bloch equations. Good qualitative agreement is found.

INTRODUCTION

The physics of semiconductors of reduced dimensionality has stimulated a large number of theoretical as well as experimental studies.[1] Besides the well-established technology for epitaxial growth of semiconductor quantum wells (QW's), several groups have started to fabricate one-dimensional and zero-dimensional structures by combinations of epitaxy and etching.[2] Another approach has been the investigation of (quasi-)zero-dimensional semiconductor microcrystallites embedded in glass matrices or other host materials.[3]

An interesting alternative to direct fabrication of such systems is the reduction of dimensionality by confining the wave functions via an external magnetic field. This approach has recently been subject to theoretical studies on the nonlinear optical properties of quantum wells in a perpendicular magnetic field.[4,5] Here one obtains a well-controlled transition from a two-dimensional to a zero-dimensional system. The nonlinear optical properties are of particular interest, since they provide detailed information on the microscopic interactions of the quasiparticles. The aim of this paper is to present respective experimental data under steady-state conditions, that allow a direct comparison to a stationary theory without any additional coherent effects.

The linear optical properties of two-dimensional magnetoexcitons have been studied theoretically years ago.[6] Experiments were performed on GaAs QW's (Ref. 7) in more recent years. In fact, one finds an absorption spectrum that is governed by discrete lines with splitting proportional to the strength of the magnetic field. A good measure for the latter is the parameter λ given by the ratio of the cyclotron energy $\hbar\omega_c$ and twice the (three-dimensional) exciton binding energy E_0, $\lambda=\hbar\omega_c/(2E_0)=(a_B/l)^2$. a_B is the three-dimensional Bohr radius and l is the magnetic length. Thus λ is proportional to the inverse square of the reduced effective mass. We choose the $In_xGa_{1-x}As$ material system that exhibits a smaller mass than GaAs. For a magnetic field of, e.g., $B=7$ T we estimate $\lambda\approx 7$, whereas in GaAs $\lambda\approx 2$ holds.

Experimental data on the nonlinear absorption of $In_xGa_{1-x}As/In_yAl_{1-y}As$ QW's and a brief discussion of the linear absorption is presented in Sec. I. In Sec. II these results are compared with the numerical solution of the semiconductor Bloch equations (steady state) for a two-dimensional system with a perpendicular magnetic field.

I. EXPERIMENT

A. Linear absorption

For our experiments we use high-quality $In_xGa_{1-x}As/In_yAl_{1-y}As$ multiple-quantum-well structures of three different well widths lattice-matched to InP substrates and grown by molecular-beam epitaxy. The well thicknesses are 50 Å (50 layers), 100 Å (100 layers), and 200 Å (50 layers), and the barrier widths are 75, 100, and 100 Å, respectively.

For the linear absorption measurements we image a tungsten lamp (spectrally integrated power density ≈ 0.6 W/cm^2 and the electric-field vector in the plane of the QW's) onto the samples. The transmitted light is collected by a multimode optical fiber and guided into a 0.5-m spectrometer. The signal at the output slit is detected with a Ge diode and a standard lock-in amplifier and is transferred to a computer. Magnetic fields up to 7 T are available in a superconducting split coil magnet that, together with the samples, is immersed in liquid helium at $T=2$ K.

Figure 1 summarizes the linear absorption of the 100-Å

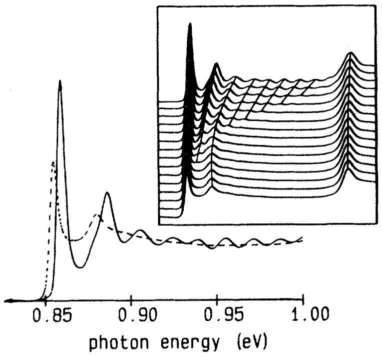

FIG. 1. Linear absorption spectra of a 100-Å $In_xGa_{1-x}As/In_yAl_{1-y}As$ quantum well under perpendicular magnetic fields up to 7 T (in 0.5-T steps). Sample temperature is $T=2$ K. For better comparison the absorption for 0 T (dashed curve) and 7 T (solid curve) are depicted separately.

QW for magnetic fields between 0 and 7 T in 0.5-T steps. In the absence of a magnetic field, the absorption is governed by the usual two-dimensional steplike density of states and the associated quantum confined excitonic levels. With the increasing magnetic field, successively more magnetoexciton levels are resolved. Simultaneously the oscillator strength of the 1*s* exciton increases dramatically and a diamagnetic shift is observed. The 1*s* light-hole magnetoexciton and the 2*s* heavy-hole magnetoexciton exhibit an anticrossing around $B=3.5$ T due to strong valence-band mixing. This is qualitatively similar to results previously obtained on GaAs QW's.[7] As another consequence of this, the higher magnetoexciton levels are not strictly equidistant. Up to eight magnetoexciton levels are observed in the 100-Å QW. This number, however, is found to depend on the well width. As the well width increases, more Landau levels are resolved in the spectrum. In a 200-Å QW (not shown) we find even more (up to 18) levels. This trend can be correlated with decreasing exciton binding energy (for $B=0$ T) and thus increasing λ as the well width increases.[8]

B. Nonlinear absorption

From the linear absorption spectra at high fields, it has already become clear that Coulomb correlations still have a strong influence on the line shape. In order to get more detailed information on the interaction between the magnetoexcitons, nonlinear absorption spectroscopy is employed. We study the absorption as a function of the population of the 1*s* exciton. This is done in a pump-probe configuration with a cw yttrium-aluminum-garnet (YAG):Nd laser (1.165-eV photon energy, polarized with the electric-field vector in the plane of the QW's) as the pump and a tungsten lamp as the weak broad-band probe. The cw YAG:Nd laser excites electron-hole pairs high in the band that rapidly (in a few tens of ps) relax into a cold population of only 1*s* excitons. Their radiative lifetime amounts to several nanoseconds. The excited population is well within the low density limit, because an average power of about 100 mW (already corrected for the reflection losses of the sample) and a Gaussian intensity diameter of ≈ 1 mm of the cw YAG:Nd laser are used. This results in an incident power density of about 13 W/cm^2. Thus the expected densities N_{1s} are on the order of 10^{-2} times the saturation density. By chopping the pump beam (at around 1 kHz) and employing standard lock-in techniques, we directly measure the change in transmission ΔT after careful subtraction of the luminescence of the 1*s* exciton. From this we compute $\Delta T/T$ which is proportional to the nonlinear absorption $\Delta\alpha=\alpha(N_{1s}=0)-\alpha(N_{1s})$.

For the measurements we use QW's of different well widths characterized above. Figure 2 depicts the nonlinear absorption spectra of a 100-Å QW at various magnetic fields up to 7 T. In the absence of a magnetic field, we find a decrease in absorption energetically below the 1*s* exciton that reflects a bleaching and a blueshift. This is the well-known blueshift due to repulsive exciton-exciton interaction.[9] In the case of $In_xGa_{1-x}As$ the bleaching part is more dominant than, e.g., in GaAs, because the excitons are larger and thus the saturation density is smaller. Moreover a slight redshift of the $n_z=2$ quantum confined level is observed (Fig. 2).

In the presence of an external magnetic field, this blueshift remains and all resolved higher magnetoexciton transitions are shifted to lower energies (corresponding to

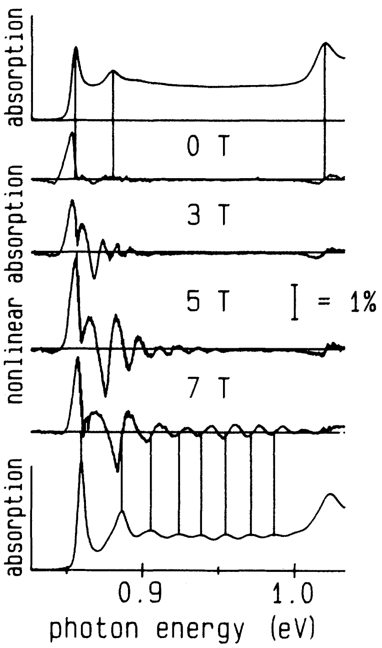

FIG. 2. Nonlinear absorption spectra of a 100-Å $In_xGa_{1-x}As/In_yAl_{1-y}As$ quantum well. These spectra reflect the changes in absorption associated with an occupation of the 1*s* magnetoexciton for various magnetic fields. The vertical bar corresponds to $\Delta T/T=10^{-2}$. One clearly sees a blueshift and bleaching of the heavy-hole exciton for zero magnetic field. The blueshift of the heavy-hole resonance remains in the presence of the magnetic field and in addition the higher magnetoexciton levels experience a pronounced redshift. For better reference the linear absorption of $B=0$ and 7 T are also shown.

the inter-Landau-level self-energy corrections), as can be seen by the derivativelike line shape of the nonlinear absorption. The zeros of the nonlinear absorption coincide with the energetic positions of the magnetoexcitons. The redshift of the second magnetoexciton level is comparable in size to the blueshift of the 1*s* exciton. The successive magnetoexciton levels exhibit a smaller but very slowly decreasing shift as their Landau index increases, which especially can be seen for the 100-Å QW (Fig. 2) and the 200-Å QW (not shown here), where many higher magnetoexcitons are resolved. The strength of bleaching is a nonmonotonic function of the magnetic field for all three samples. This might be related to effects of finite well width discussed in Ref. 4 if the radiative lifetime is not strongly influenced by the B field.

This overall scenario has recently also been observed in femtosecond pump-probe experiments on GaAs QW's.[10] However, the blueshift of the 1*s* magnetoexciton exhibits a somewhat different behavior. We still observe a blueshift at $\lambda \approx 7$, whereas the blueshift in Ref. 10 has effectively vanished at $\lambda \approx 3$. This corresponds to the fact that all shifts are most pronounced in the stationary limit and are reduced for impulsive excitation.

II. THEORY: SOLUTION OF THE SEMICONDUCTOR BLOCH EQUATIONS

The linear and nonlinear optical properties of two-dimensional magnetoexcitons were recently treated in the framework of the semiconductor Bloch equations.[5] A purely two-dimensional system and only one valence band are considered in this theory. Thus no quantitative agreement with quantum wells, where heavy-hole and light-hole transitions are present, can be expected. However, we can expect a correct qualitative description of the microscopic interactions. From Ref. 5 we get

$$[\gamma_\perp + i(\Omega_n - \omega)]\Psi_n = i(1-2n_n)\left[\mu E + \sum_{n'} V_{n,n'}\Psi_{n'}\right], \tag{1}$$

where

$$\Omega_n = \varepsilon_{cn} - \varepsilon_{vn} + \sum_{n'} V_{n,n'}(1-2n_{n'}) . \tag{2}$$

Ψ_n is the off-diagonal term of the density matrix, n_n is the corresponding inversion, Ω_n is the resonance frequency of level n, and ω is the frequency of the light field E. From this the absorption coefficient is obtained by

$$\alpha(\omega) \propto \frac{\lambda\mu}{2\pi} \operatorname{Im} \sum_n \frac{\Psi_n(\omega)}{E} . \tag{3}$$

These equations describe a set of two-level systems coupled by the Coulomb matrix elements $V_{n,n'}$. These couplings can be interpreted in terms of an effective local field for each level n.[11]

In order to apply these equations to the nonlinear absorption data, we assume a steady-state population of only the 1*s* magnetoexciton, i.e., $n_{n'} = n\delta_{n',1}$. (This is only rigorously valid if the Coulomb interaction is neglected at this point.) With known $V_{n,n'}$ (see Ref. 5) we compute the linear and nonlinear absorption by matrix inversion. The calculated linear and nonlinear absorption spectra are depicted in Fig. 3. They qualitatively reproduce the important features seen in the experiment. In particular the blueshift of the 1*s* exciton and the redshifts of the higher levels appear. The blueshift of the 1*s* exciton can immediately be seen from Eq. (1). This is caused by the same exciton-photon interaction responsible for the optical Stark effect. It is related to the sum on the right-hand side $-2n_1\sum_{n'\neq 1} V_{1,n'}\Psi_{n'}$. The corresponding terms for the higher levels vanish because of $n_{n'\neq 1} \equiv 0$. This blueshift dominates the overall redshift of all levels given by the inter-Landau-level self-energy corrections $[-2\sum_{n'} V_{n,n'}n_{n'} = -2V_{1,1}n_1$ in Eq. (2)]. An additional redshift for the higher levels originates from the sum in Eq. (1). This shift decays slowly with n because the $V_{n,n'}$ couple the levels even for large $|n-n'|$ as a result of the long-range Coulomb interaction. These features are valid for the low density regime, whereas a qualitatively different scenario is expected if the 1*s* exciton is inverted and higher magnetoexcitons are populated.

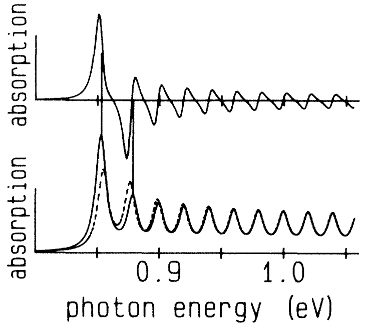

FIG. 3. Numerical solutions to the semiconductor Bloch equations. Comparison with the experimental results, e.g., on the 100-Å sample (Fig. 2), shows that all salient features are well reproduced. For the calculation a basis set of 50 Landau levels is used. A dephasing rate (for all Landau levels identical) of $\gamma_\perp = 5$ meV, an exciton binding energy of $E_0 = 2$ meV, $\lambda = 5$, and $n_1 = 0.15$, $n_{j\neq 1} = 0$ are chosen.

SUMMARY

We have studied the nonlinear absorption of $In_xGa_{1-x}As/In_yAl_{1-y}As$ quantum wells under strong perpendicular magnetic fields. Steady-state population of the 1*s* magnetoexciton leads to redshifts of all higher magnetoexciton levels and a blueshift of the 1*s* magnetoexciton itself. This is a result of strong Coulomb correlations, which is confirmed by comparison with numerical solutions of the semiconductor Bloch equations. The $In_xGa_{1-x}As$ material system proves to exhibit particularly pronounced magnetic-field effects because of its small effective mass.

ACKNOWLEDGMENTS

One of us (M.W.) acknowledges many discussions on magnetoexcitons with J. B. Stark, W. H. Knox, D. S. Chemla, and S. Schmitt-Rink during the phase of their early femtosecond experiments on GaAs magnetoexcitons. This has significantly stimulated the present work. We acknowledge financial support by the Deutsche Forschungsgemeinschaft through the Schwerpunkt "Dynamik Elementarer Anregungen im Festkörper."

[1] See, e.g., *Proceedings of the 20th International Conference on the Physics of Semiconductors, Tessaloniki, 1990* (World Scientific, Singapore, 1991).

[2] *Proceedings of the 6th International Winterschool on Localisation and Confinement of Electrons in Semiconductors,* edited by G. Bauer, H. Heinrich, and F. Kuchar (Springer, Berlin, 1990), and references cited therein.

[3] Al. L. Efros and A. L. Efros, Fiz. Tekh. Poluprovodn. **16**, 1209 (1982) [Sov. Phys. Semicond. **16**, 772 (1982)]; L. E. Brus, J. Chem. Phys. **80**, 4403 (1984).

[4] G. E. W. Bauer, Phys. Rev. Lett. **64**, 60 (1990).

[5] C. Stafford, S. Schmitt-Rink, and W. Schäfer, Phys. Rev. B **41**, 10 000 (1990).

[6] O. Akimoto and H. Hasegawa, J. Phys. Soc. Jpn. **22**, 181 (1967); M. Shinada and K. Tanaka, *ibid.* **29**, 1258 (1970).

[7] J. C. Maan, A. Fasolino, G. Belle, M. Altarelli, and K. Ploog, Phys. Rev. B **30**, 2253 (1984).

[8] R. L. Green, K. K. Bajaj, and D. E. Phelps, Phys. Rev. B **29**, 1807 (1984).

[9] N. Peyghambarian, H. M. Gibbs, and J. L. Jewell, Phys. Rev. Lett. **53**, 2433 (1984).

[10] J. B. Stark, W. H. Knox, D. S. Chemla, W. Schäfer, S. Schmitt-Rink, and C. Stafford, Phys. Rev. Lett. **65**, 3033 (1990).

[11] M. Wegener, D. S. Chemla, S. Schmitt-Rink, and W. Schäfer, Phys. Rev. A **42**, 5675 (1990).