PHYSICAL REVIEW B VOLUME 40, NUMBER 9 15 SEPTEMBER 1989-II

Rapid Communications

The Rapid Communications section is intended for the accelerated publication of important new results. Since manuscripts submitted to this section are given priority treatment both in the editorial office and in production, authors should explain in their submittal letter why the work justifies this special handling. A Rapid Communication should be no longer than 3½ printed pages and must be accompanied by an abstract. Page proofs are sent to authors, but, because of the accelerated schedule, publication is not delayed for receipt of corrections unless requested by the author or noted by the editor.

Collision broadening of two-dimensional excitons in a GaAs single quantum well

A. Honold*
Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-7000 Stuttgart 80, Federal Republic of Germany

L. Schultheis
Asea Brown Boveri Corporate Research, CH-5405 Baden, Switzerland

J. Kuhl
Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-7000 Stuttgart 80, Federal Republic of Germany

C. W. Tu†
AT&T Bell Laboratories, Murray Hill, New Jersey 07974
(Received 21 February 1989)

The phase relaxation of two-dimensional (2D) heavy-hole excitons in a 12-nm GaAs single quantum well subjected to collisions with either free carriers or incoherent heavy-hole excitons is investigated by time-resolved degenerate four-wave mixing. The homogeneous linewidth corresponding to the phase coherence time reveals a collisional broadening due to exciton-exciton scattering and an 8 times stronger exciton-free carrier scattering. Both scattering processes are enhanced for 2D excitons as compared to 3D excitons.

The optical properties of direct-gap semiconductors near the band edge at low temperatures and moderate excitation densities are determined by excitons.[1] The exciton transitions are characterized by the eigenenergy, the oscillator strength, and the homogeneous linewidth. According to theoretical models these characteristic quantities are expected to be influenced by both exciton-exciton and exciton-free carrier interaction owing to the screening of the Coulomb interaction as well as fermion correlation and exchange effects.[2,3]

In fact, three-dimensional (3D) excitons exhibit a small blue shift of the eigenenergy[4] due to exciton-exciton interaction. Additionally, a bleaching of the absorption[5] and a distinct broadening of the homogeneous linewidth,[4] due to both exciton-exciton and exciton-free carrier interactions, have been observed.

For 2D excitons in GaAs it is well known that the exciton-exciton interaction shifts the exciton eigenenergy to higher energies (blue shift) and reduces the oscillator strength (bleaching), whereas exciton-free carrier interaction only causes a bleaching of the transition.[3,6] Up to now no experimental data on the influence of these interactions on the homogeneous linewidth of the transition are available in the literature.

In this paper, we study the broadening of 2D excitons subjected to exciton-exciton and exciton-free carrier collisions. The dephasing time of the excitons is determined by a backward self-diffraction experiment in a pump-and-probe configuration while varying the density of additionally injected free carriers or incoherent excitons. The corresponding homogeneous linewidth reveals a pronounced collisional broadening for both excitons and free carriers as scatterers, the latter being 8 times more efficient. A comparison with 3D excitons indicates that interaction of 2D excitons is stronger in both cases.

The experiments are performed on a 12-nm-thick GaAs single quantum well grown on an n^+-type GaAs substrate. During the experiments the sample is immersed in superfluid helium. Photoluminescence, photoluminescence excitation, and transmission experiments reveal the transition energy of the free $1s$ heavy-hole excitons at 1.539 32 eV with a linewidth of 0.59 ± 0.02 meV and no Stokes shift between absorption and emission. The absorption of the excitons at the center of the resonance is 7.5% corresponding to an absorption coefficient of $\alpha = 6.4 \times 10^4$ cm^{-1}. The emission of free excitons from regions of the quantum well one monolayer of GaAs thicker than the major part of the sample is observed in the photoluminescence spectrum at 1.538 18 eV. The assignment of this line has been substantiated by the linear dependence of the luminescence intensity on the excitation density over more than 3 orders of magnitude.

The phase relaxation of the heavy-hole excitons corresponding to the homogeneous linewidth of the optical transition is studied by a self-diffraction experiment in a new backward geometry[7] [see Fig. 1(a)] which enables the study of the exciton dynamics in thin layers without being affected by the optical properties of the substrate material. Two weak picosecond pulses of a synchronously pumped dye laser tuned into resonance with the exciton transition are used to examine the phase relaxation of the excitons. The first pulse No. 1 generates coherent excitons within the quantum well and the second pulse No. 2 tests the phase coherence of these excitons via formation of a grating and subsequent self-diffraction. Formation of a grating is only possible with those excitons created by pulse No. 1 which have conserved their phase coherence between the generation and the interaction with pulse No. 2. The intensity of the pulses is adjusted to a level where no influence of the exciton density on the measured phase coherence time is detectable. A photomultiplier measures the whole time-integrated diffracted intensity as a function of the time delay τ_{12} between the pulses. The phase coherence time T_2 is determined by fitting the numerical solution of the optical Bloch equations in the small signal regime of the corresponding two level system to the measured diffraction curves.

The phase relaxation of the low-density, coherent ensemble of excitons will be strongly modified in the presence of either free carriers or incoherent excitons. Thus, the self-diffraction experiment can be used as a probe to study the exciton-exciton and exciton-free carrier scattering. For this purpose free carriers or heavy-hole excitons are additionally created within the quantum well by a third stronger optical pulse from a second synchronously pumped dye laser the wavelength of which can be tuned independently. The heavy-hole excitons are created 20 ps before pulse No. 1 of the self-diffraction experiment arrives in the sample in order to ensure that all excitons created by pulse No. 3 have lost their phase coherence and act only as a background of scattering excitons. The free carriers are created in temporal overlap with pulse No. 1 to prevent the condensation of these free carriers to excitons. In both configurations the phase coherence time T_2 is determined while varying the density of the additionally created incoherent heavy-hole excitons or the free carriers.

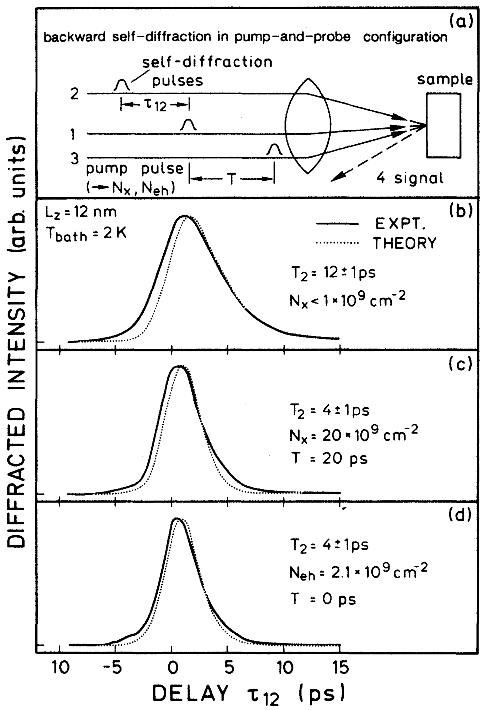

FIG. 1. Geometry (a) and diffraction curves (b)–(d) of the backward self-diffraction in pump-and-probe configuration on the lowest heavy-hole excitons in a 12-nm GaAs single quantum well at a temperature of 2 K. The experimental curves are shown as solid lines, the best-fitting solution of the optical Bloch equations of the two-level system with the corresponding phase coherence time T_2 as a dotted line. (b) depicts the low excitation case, (c) additional pumping of heavy-hole excitons, and (d) pumping of free carriers.

The optical pulses used in the experiments have a duration of 2.6 ps, a spectral width of 0.9 meV, and a repetition rate of 76.55 MHz. The jitter between the pulses of the two different dye lasers is smaller than 2 ps (cross correlation width 6.4 ps). The free carriers are created by the third pulse 9.5 meV above the heavy-hole exciton transition. The density of the heavy-hole excitons is determined with the measured absorption coefficient of $\alpha = 6.4 \times 10^4$ cm^{-1}. The direct determination of the free-carrier density is prevented by the strong background absorption of the sample substrate. From the heavy-hole exciton absorption we calculate, with the help of the measured absorption coefficients of Masumoto *et al.*,[8] the absorption at our free-carrier pump wavelength to be 2.2%.

The measured diffraction curves are depicted as solid lines in Figs. 1(b)–1(d). Figure 1(b) shows the curve recorded without pumping by pulse No. 3, which yields the low-density limit of the phase coherence time T_2. Analysis of this signal under the assumption of homogeneous broadening would yield $T_2 = 6$ ps corresponding to a homogeneous linewidth $\Gamma_h = 2/T_2 = 0.22$ meV. Comparison of this value with the spectral width observed in the frequency domain reveals substantial inhomogeneous broadening of the excitonic transition in our sample. Model calculations using the third order density matrix theory[9] predict a decay of the degenerate four-wave mixing signal with time delay τ between the two pulses as $I(\tau) \sim \exp(-4\tau/T_2)$ if the inhomogeneous width $\Gamma_{inh} \geq \Gamma_h$. Taking into account the inhomogeneous broadening,[10] best fit of the optical Bloch equations (depicted as a dashed line) to the experimental diffraction curve in Fig. 1(b) results in $T_2 = 12 \pm 1$ ps corresponding to $\Gamma_h = 0.11 \pm 0.01$ meV in the low-density excitation limit. Deconvolution of Γ_h from the spectral line profile yields $\Gamma_{inh} = 0.55 \pm 0.02$ meV.

Excitation of heavy-hole excitons at a density of $N_x = 2 \times 10^{10}$ cm^{-2} changes the diffraction curve remarkably as demonstrated in Fig. 1(c) and reduces the phase coherence time to $T_2=4\pm1$ ps. Similarly, the presence of free carriers at a density of $N_{eh}=2.1\times10^9$ cm^{-2} decreases the phase coherence time to $T_2=4\pm1$ ps [Fig. 1(d)].

The homogeneous linewidths corresponding to the experimentally determined phase coherence times T_2 are depicted in Fig. 2 as a function of the density of additionally created scatterers. The homogeneous linewidth is influenced by the interaction of the heavy-hole excitons with free carriers at densities as small as $N_{eh}=2\times10^8$ cm^{-2} and with incoherent excitons around $N_x=1\times10^9$ cm^{-2}.

For both types of scatterers, the broadening of the homogeneous linewidth can be well described by a linear density dependence as expected in the low-density regime.[11] The function

$$\Gamma(N)=\Gamma(0)+\gamma a_B^2 E_B N \quad (1)$$

is fitted to the experimental data and depicted in Fig. 2 as straight lines. In this relation Γ is the homogeneous linewidth, N the density of the scatterers, a_B the exciton Bohr radius, and E_B the exciton binding energy. γ is a dimensionless parameter of the line broadening and gives a measure of the interaction strength of the excitons. The binding energy of the excitons is determined by excitation spectroscopy to be 8.5 meV in good agreement with published values.[12] The exciton Bohr radius of 9.5 nm is taken from the variational calculations of Shinozuka and Matsuura.[13] With these values the best fits to the experimental data points yield line broadening parameters of $\gamma_{xx}=1.5\pm0.3$ (exciton-exciton interaction) and $\gamma_{xeh}=11.5\pm2.0$ (exciton-free carrier interaction).

The density-independent contribution to the homogeneous linewidth $\Gamma(0)$ includes all residual interactions of the excitons with acoustic phonons, impurities, the interfaces of the quantum well, and Al concentration fluctuations within the $Al_xGa_{1-x}As$ barrier material. Collisions of excitons with acoustic phonons or impurities would result in a purely homogeneous broadening of the excitonic transition. Interface roughness and alloy disorder scattering contribute to homogeneous as well as inhomogeneous broadening, depending on the lateral extension (mean diameter Λ) of the irregularities. Well width or alloy composition fluctuations with $\Lambda \ll 2a_B$ and $\Lambda \gg 2a_B$ will cause homogeneous and inhomogeneous broadening, respectively.

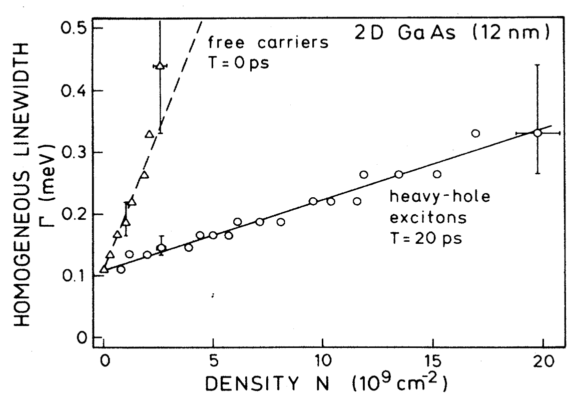

FIG. 2. Calculated homogeneous linewidth corresponding to the experimentally determined phase coherence time while varying the particle density of additionally created heavy-hole excitons (circles) and free carriers (triangles). The data are fitted by a linear density dependence which is shown as a solid line.

The substantial interface roughness on a 5-nm scale, which has been detected by transport measurements[14] or sophisticated imaging and pattern recognition techniques,[15] may be the origin of $\Gamma(0)$. The experimentally observed inhomogeneous linewidth of our sample $\Gamma_{\text{inh}}=0.55\pm0.02$ meV can be explained by variation of the Al mole fraction by $\pm3\%$ in the barrier layers on a length scale larger than 20 nm.

The experimentally determined collision efficiencies can be compared with theoretically calculated values. Manzke, Henneberger, and May[11] calculated the broadening constant in the case of exciton-exciton interaction to be $\gamma_{xx}=0.42$, which is smaller by a factor of 4 than the experimental value. The line broadening due to the free carriers is assumed to be mainly caused by exciton-electron collisions.[4] The calculations of Feng and Spector[16] yield a broadening constant for exciton-free carrier interaction of $\gamma_{xeh}=0.16$ at a temperature of 10 K, which is nearly 2 orders of magnitude smaller than the experimental value. The large discrepancies between experiment and theory may be explained by the difficulties associated with the quantitative theoretical treatment of many-body effects.[3] Recently published results of transmission measurements on modulation-doped GaAs multiple quantum wells by Huang *et al.*[17] at densities higher than $N_e=2\times10^{10}$ cm^{-2} show a smaller broadening of the absorption line than expected from our experiments. Equation (1), describing well our experimental results, is valid only in the low-density range and, therefore, deviations are not surprising at densities as high as those used by Huang and co-workers (mean particle distance smaller than the exciton Bohr radius). Owing to the large inhomogeneous broadening of the samples in the order of 1.5 meV and uncontrollable changes of the inhomogeneous broadening from sample to sample, the transmission experiments of Ref. 17 provide no information about the collision broadening of excitons at low densities.

The line broadening due to exciton-exciton collisions shows the same linear dependence on the density as the previously observed blue shift.[3,6] The numerical prefactor amounts for the broadening to $\gamma_{xx}=1.5$ and for the blue shift to $\sigma_{xx}=12$. The small value of γ_{xx} and the relatively broad absorption lines observed in the blue-shift experiments may be responsible for the fact that the line broadening was not detected by Peyghambarian *et al.*[6]

Finally, we compare the collision broadening of the 2D excitons in the presence of additional free carriers or incoherent excitons to corresponding data measured earlier for 3D excitons.[4] Such a comparison becomes available if the particle distance is used as the parameter for the excitation density. The interparticle distance, normalized to the respective exciton Bohr radius, can be easily calculated from the particle density[18]

$$\text{3D:}\quad r_b=(4\pi a_B^3 N/3)^{-1/3}, \quad (2)$$

$$\text{2D:}\quad r_b=(\pi a_B^2 N)^{-1/2}. \quad (3)$$

Figure 3 depicts the line broadening of 2D and 3D exci-

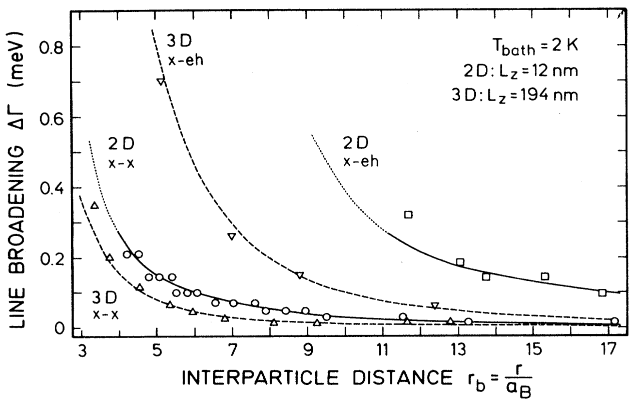

FIG. 3. Line broadening of two-dimensional (circles and squares) and three-dimensional excitons (triangles) subjected to collisions with excitons (x-x) or free carriers (x-eh) as a function of the normalized interparticle distance.

tons due to exciton-exciton (x-x) and exciton-free carrier (x-eh) collisions versus the interparticle distance r_b. It is evident that 2D excitons are much more efficient in their interactions compared with 3D excitons at the same normalized interparticle distance. The line broadening of the 2D exciton transition measured at an interparticle distance of 5–10 Bohr radii amounts to about 2–4 times the value found at the same distance for the 3D exciton for both exciton-exciton as well as exciton-free carrier collisions. This result can be understood as a consequence of the considerably weaker screening of the Coulomb interaction in 2D compared with 3D.[3] The relative unimportance of screening in 2D systems leads to an increasing interaction of the colliding excitons via repulsive forces which originate from the Pauli exclusion principle for identical fermions.[3]

In conclusion, we studied the collision broadening of 2D excitons and demonstrated the importance of the interaction of excitons with free carriers and incoherent excitons for the line broadening. In high-quality samples their influence on the homogeneous linewidth can be observed at surprisingly low excitation densities. Exciton-free carrier collisions due to the Coulomb interaction prove to be stronger by a factor of 8 than exciton-exciton collisions which are mainly due to a hard-core repulsion force originating from the Pauli exclusion principle for fermions. 2D excitons are much more efficient in their interactions than 3D excitons in agreement with the weaker screening of the Coulomb interaction in 2D systems.

We gratefully thank H. J. Queisser for critical reading of the manuscript. We acknowledge expert technical assistance from A. Jonietz, K. Battling, and H. Kühne and help with the computer software from W. König, as well as assistance with the molecular-beam epitaxy from R. F. Kopf.

*Present address: Nippon Telegraph and Telephone Corporation, Basic Research Laboratories, 3-9-11 Midori-cho, Musashino-shi, Tokyo 180, Japan.

†Present address: Department of Electrical and Computer Engineering, University of California, San Diego, La Jolla, CA 92093.

[1]R. J. Elliott, Phys. Rev. **108**, 1384 (1957).

[2]H. Haug and S. Schmitt-Rink, J. Opt. Soc. Am. B **2**, 1135 (1985).

[3]S. Schmitt-Rink, D. S. Chemla, and D. A. B. Miller, Phys. Rev. B **32**, 6601 (1985).

[4]L. Schultheis, J. Kuhl, A. Honold, and C. W. Tu, Phys. Rev. Lett. **57**, 1635 (1986).

[5]G. W. Fehrenbach, W. Schäfer, J. Treusch, and R. G. Ulbrich, Phys. Rev. Lett. **49**, 1281 (1982).

[6]N. Peyghambarian, H. M. Gibbs, J. L. Jewell, A. Antonetti, A. Migus, D. Hulin, and A. Mysyrowicz, Phys. Rev. Lett. **53**, 2433 (1984).

[7]A. Honold, L. Schultheis, J. Kuhl, and C. W. Tu, Appl. Phys. Lett. **52**, 2105 (1988).

[8]Y. Masumoto, M. Matsuura, S. Tarucha, and H. Okamoto, Phys. Rev. B **32**, 4275 (1985).

[9]T. Yajima and Y. Taira, J. Phys. Soc. Jpn. **47**, 1620 (1979).

[10]The inhomogeneous broadening has been modeled by a Gaussian distribution function $g(\omega_0)$ for the exciton frequency ω_0 with full width at half maximum of 0.55 meV; see also L. Schultheis and J. Hegarty, J. Phys. (Paris) Colloq. **46**, Suppl. 10, C7-167 (1985).

[11]G. Manzke, K. Henneberger, and V. May, Phys. Status Solidi (b) **130**, 233 (1987).

[12]R. C. Miller, D. A. Kleinman, W. T. Tsang, and A. C. Gossard, Phys. Rev. B **24**, 1134 (1981); U. Ekenberg and M. Altarelli, *ibid.* **35**, 7585 (1987); E. S. Koteles and J. Y. Chi, *ibid.* **37**, 6332 (1988).

[13]Y. Shinozuka and M. Matsuura, Phys. Rev. B **28**, 4878 (1983); **29**, 3717 (1984).

[14]H. Sakaki, T. Noda, K. Hirakawa, M. Tanaka, and T. Matsusue, Appl. Phys. Lett. **51**, 1934 (1987); R. Gottinger, A. Gold, G. Abstreiter, G. Weimann, and W. Schlapp, Europhys. Lett. **6**, 183 (1988).

[15]A. Ourmazd, D. W. Taylor, J. Cunningham, and C. W. Tu, Phys. Rev. Lett. **62**, 933 (1989).

[16]Y. Feng and H. N. Spector, Superlattices Microstruc. **3**, 459 (1987).

[17]D. Huang, H. Y. Chu, Y. C. Chang, R. Houdré, and H. Morkoc, Phys. Rev. B **38**, 1246 (1988).

[18]G. Tränkle, H. Leier, A. Forchel, H. Haug, C. Ell, and G. Weimann, Phys. Rev. Lett. **58**, 419 (1987).