Exciton absorption saturation by phase-space filling: influence of carrier temperature and density

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We investigate the temporal evolution and the density dependence of exciton bleaching in $GaAs/Al_xGa_{1-x}As$ quantum wells. We find that the carrier-induced reduction of exciton oscillator strength is not influenced by carrier cooling after creation of hot carriers with femtosecond laser pulses. Excitation density experiments suggest that either phase-space filling or Coulomb screening dominates the exciton bleaching, depending on the carrier density.

A characteristic feature of semiconductor quantum wells is the strong enhancement of excitonic optical transitions due to the confinement of electrons and holes. This results in sharp peaks in the linear absorption even at room temperature and provides strong optical nonlinearities that can be used for all-optical or electro-optic switching devices.[1] One fundamental nonlinearity is the absorption saturation, or bleaching of the excitons, by photoexcited free or bound electron-hole pairs.[2] Understanding the basic physics of this saturation is essential for optimizing the performance of electroabsorptive photonics devices, such as self-electro-optic effect devices (SEED's).[3]

Carrier-induced excitonic nonlinearities are principally based on the variation of the binding energy, oscillator strength, and lifetime of the lowest ($1s$) exciton state.[4,5] The corresponding optical transition may show a shift of the transition frequency, a reduction of the oscillator strength (ROS), and an increase of the linewidth. These effects may be experimentally distinguished by spectrally resolved nonlinear transmission measurements. However, all three effects may lead to a reduced absorption at the original excitonic peak, which will be denoted as "exciton bleaching" throughout this paper. The different changes of the excitonic absorption line can be (at least roughly) attributed to different physical origins. Phase-space filling (PSF) causes directly a reduction of the exciton oscillator strength by reducing the number of single-particle states that contribute to the bound electron-hole state. Exchange effects lead to renormalization of the single-particle states and the exciton states, and may contribute to ROS, too. These two physical mechanisms are due to the fermion character of the carriers and reflect the occupation of single-particle states. On the other hand, long-range Coulomb screening (CS) is related to the charge and the density of the carriers, and is independent of their spin. The contribution of CS to the ROS can be neglected compared to PSF and exchange effects,[5] but CS will cause a broadening of the exciton absorption line.

Exciton bleaching has been addressed in many experimental and theoretical studies, in particular for quantum wells. The first experimental data obtained in subpicosecond time-resolved saturation measurements were interpreted in terms of PSF only.[6] It was concluded that ROS due to the occupation of excitonic states is about twice as effective as that due to a thermalized electron-hole (e-h) plasma at room temperature. Theoretical calculations[4] including PSF and exchange were able to explain these experimental results and the saturation densities observed in other experiments.[7,8] This theory is based on the assumption of strictly two-dimensional excitons, arguing that long-range Coulomb screening should be negligible in quantum wells. The efficiency of the ROS is given in terms of a saturation density N_S, which is derived as a function of carrier temperature for the case of exciton bleaching by hot free carriers in Ref. 4. This theory predicts a linear increase of the saturation density with the carrier temperature. As a consequence, a pronounced increase of the ROS efficiency with decreasing carrier temperature should be observed.

Several other experimental studies, are in disagreement with the predictions of Ref. 4. For instance, it has been found that CS bleaches the exciton as effectively as PSF,[9] or that pure broadening may be the dominant bleaching mechanism.[10,11] A recent time-resolved study in II-VI quantum wells[12] has shown that the bleaching due to excitons is weaker than the bleaching due to a cold e-h plasma, in contrast to the prediction of Ref. 4. More recent theoretical calculations predict a comparable bleaching efficiency of excitons and free carriers.[5] In addition, the predicted temperature dependence for both mechanisms is less pronounced than in Ref. 4.

To clarify this debate, we present in this paper a time-resolved investigation of ROS in GaAs quantum wells due to the presence of a hot electron-hole plasma. In time-resolved differential transmission spectra, the ROS contribution to the exciton bleaching is separated from pure broadening (and from a possible shift of the exciton peak) by integrating over the area of the broadened exciton. We find an "instantaneous" reduction of the oscillator strength, which is not influenced by subsequent carrier cooling, in contrast to the theoretical predictions. We also find that the density dependence of exciton bleaching and ROS is quite different. Unoccupied higher sub-

bands, that can only be influenced by CS, do *not* show carrier-induced ROS.

Our experiments are performed in a pump-probe setup, using a white-light continuum probe and near-infrared (NIR) pump pulses of ≈90 fs duration. The laser system consists of a colliding pulse mode-locked dye laser oscillator operating at 620 nm, a six-pass dye amplifier, white-light generation in an ethylene glycol jet, and a second, five-pass NIR dye amplifier. Both amplifier stages are pumped by a 6.8 kHz copper vapor laser. The pump pulses obtained from the NIR amplifier are tunable between 790 nm and 860 nm. Time-resolved differential transmission spectra (DTS), showing pump-induced transmission changes $\Delta T/T_0$, are measured with a multichannel analyzer system. We measure the intrinsic chirp of the probe continuum according to Ref. 13 and correct the experimental data for this frequency-dependent time delay.

The sample investigated is a multiple quantum well (MQW) with 40 periods of 10 nm GaAs wells and 20 nm $Al_{0.35}Ga_{0.65}As$ barriers, grown by molecular beam epitaxy. The GaAs substrate was removed over an area of $\approx 1 \times 1\text{mm}^2$ by selective etching in order to allow transmission measurements. Excitonic absorption peaks are clearly resolved in the linear absorption spectrum. The positions of these peaks agree well with theoretical calculations. The pump photon energy was tuned to 1.53 eV, slightly below the transition from the second heavy-hole subband to the second electron subband ($E2H2$). This creates carriers with a total excess energy of 70 meV in the lowest subbands only, thus avoiding effects due to intersubband relaxation. We assume a homogeneous excitation density, since the probe focus is kept much smaller than the pump spot and the total thickness of the absorbing quantum well layers is about 40% of the absorption length at 820 nm. The excitation densities are given as the mean value averaged over all 40 wells.

In Fig. 1 a typical set of DTS at time delays −50 fs, 50 fs, and 1 ps is shown, taken at an excitation density of 3.9×10^{11} cm^{-2}. The spectra show induced transmission, i.e., bleaching of various excitonic transitions, which evolves during the pump pulse duration. The peaks in the DTS coincide with the excitonic peaks in the linear absorption and are identified as the electron to heavy-hole and light-hole transitions of the first subband $E1H1$ (1.46 eV) and $E1L1$ (1.47 eV); and the $E2H2$ (1.57 eV) transition. Above the $E1L1$ peak, the spectra also show some broadband bleaching due to occupation of continuum states within the lowest subbands. During the pump pulse, this bleaching peaks slightly below the pump energy, indicating a nonthermal carrier distribution. However, even at long delay times there remains a (less pronounced) broad peak at 1.525 eV that corresponds to bleaching of the $E1H3$ transition, which would be forbidden in an ideal QW with infinitely high barriers.

During the initial stage of free-carrier excitation in the quantum wells, at −50 fs, strong carrier-induced broadening of the exciton lines is evidenced by a reduced transmission above and below all exciton resonances. With increasing carrier density and beginning thermalization of the carriers, the positive DT signal becomes dominant for the $E1H1$ and the $E1L1$ transitions, indicating a net loss of exciton oscillator strength. The negative DT signal remains visible at the low energy side of the $E1H1$ exciton only, but is masked by the $E1L1$ and continuum bleaching at the high energy side. In contrast to the lowest exciton lines, the DTS around the $E2H2$ peak show an oscillatory structure at all time delays. There is a substantial broadening and a slight redshift of the exciton, but the net absorption change, integrated over the area of the broadened exciton line, remains essentially zero. This result is confirmed in Fig. 2, where we compare the temporal evolution of the integrated transmission change around the $E1H1$ and $E1L1$ excitons (which cannot be separated, since the transitions are too close), around the $E2H2$ exciton, and the continuum bleaching at 1.5 eV. The integrated transmission changes are directly related to the carrier-induced ROS, since the integration separates the effect of ROS from pure broadening ascribed to Coulomb screening. On the other hand, the continuum bleaching gives information about the free-carrier

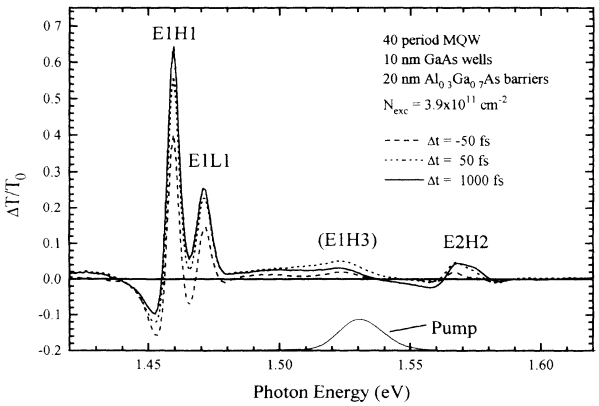

FIG. 1. Time-resolved diffferential transmission spectra (DTS), taken at time delays of −50 fs, 50 fs, and 1 ps. The peaks correspond to exciton lines in the linear absorption, which are bleached by the pump-induced carriers.

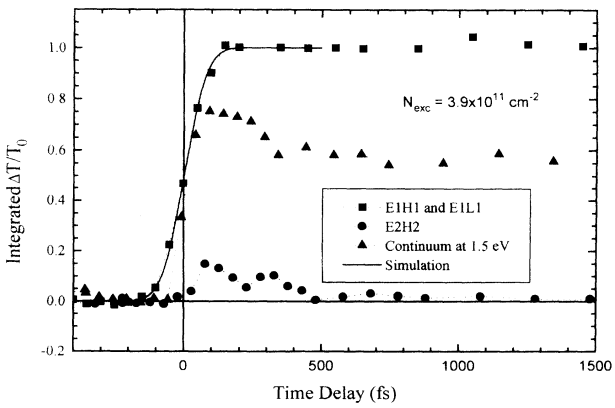

FIG. 2. Time evolution of the integrated absorption change around the $E1H1$ and $E1L1$ excitons, around the $E2H2$ exciton, and of the continuum bleaching at 1.5 eV.

relaxation within the lowest subbands, i.e., carrier thermalization and cooling. The continuum bleaching at 1.5 eV, slightly below the pump energy, shows a rapid increase with time, passes to a maximum at $\Delta t = 100$ fs, and decays to a lower quasistationary level (on a picosecond time scale), which is reached after roughly 700 fs. This indicates that the intrasubband carrier relaxation, in particular the cooling down to the lattice temperature, takes several hundreds of femtoseconds, as can be expected from previously measured energy loss rates.[14]

The data of Fig. 2 demonstrate that the broadening of the $E2H2$ exciton is not associated with a significant net absorption change, indicating that there is no carrier-induced ROS. Since the corresponding subbands are not occupied, this exciton line is not influenced by PSF or exchange, but only by Coulomb screening[15] (the weak positive signal within the first 500 fs is most probably caused by continuum bleaching of the lowest subbands due to broadening of the initial carrier distribution). In contrast to the $E2H2$ transition, the integrated transmission change around the lowest excitons ($E1H1$ and $E1L1$, squares in Fig. 2) shows an instantaneous rise during the pump pulse duration and remains at its maximum level after the pump pulse has passed. The solid line in Fig. 2 is a simulation, obtained by assuming that the ROS is proportional to the total carrier density, and neglecting any influence of carrier relaxation. The simulation fits the measured time evolution of the ROS signal almost perfectly (for the simulation, the pump pulse has been taken as a Gaussian with 110 fs full width at half maximum, slightly longer than the pump pulse duration deduced from autocorrelation measurements).

Two conclusions can be drawn from this observation. (i) There is no significant influence of the time-dependent carrier distribution during the initial phase of thermalization (as previously observed in Ref. 16). (ii) Obviously, there is *no significant influence* of carrier cooling, in contrast to the predictions of Ref. 4. After internal thermalization of the pump-induced electrons and holes, which occurs predominantly by elastic carrier-carrier scattering within about 100 fs,[16] the electron gas has an average energy per particle of roughly 60 meV corresponding to a temperature of 700 K. According to the predictions of Ref. 4, cooling of the electrons to the lattice temperature would correspond to an increase of the ROS efficiency by a factor of ≈ 2.5. The more elaborate calculations of Ref. 5 lead to much higher saturation densities and a weaker temperature dependence, but still predict an increase in saturation efficiency of more than 50% for carriers cooling from 700 K to 300 K. However, the calculations of Ref. 5 have not included correlation effects on exciton bleaching. These effects are expected to be rather temperature independent. Our data do not show a delayed increase of the ROS, which would be expected if the ROS increased during plasma cooling.

In order to obtain more information about the different contributions to the exciton bleaching, we compare the density dependence of the bleaching of the exciton absorption peak (i.e., the maximum of the DTS), which is given by ROS *and* broadening, and the integrated negative absorption change, which is given by ROS only.

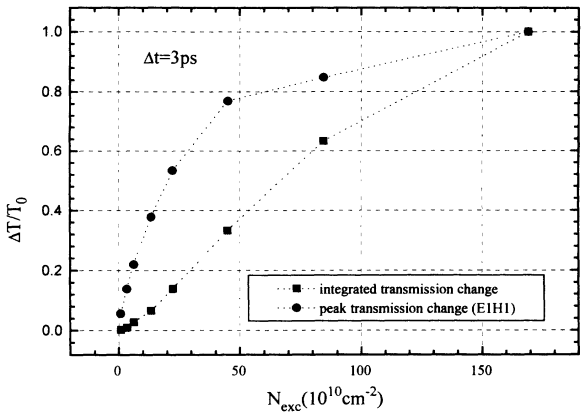

FIG. 3. Density dependence of the peak $E1H1$ exciton bleaching, which is influenced by PSF and CS, and of the integrated absorption change ($E1H1$ and $E1L1$), which is given by PSF alone.

These values are shown in Fig. 3 for excitation densities up to 1.7×10^{12} cm^{-2}, at a fixed time delay of 3 ps after excitation. While the integrated signal increases rather smoothly and roughly linearly with density up to 10^{12} cm^{-2}, the peak bleaching clearly shows some saturation already at rather low densities. Although it is difficult to quantitatively compare peak and integrated absorption change (we therefore show normalized values), the different density dependences suggest that pure broadening by CS may be dominant at very low carrier densities, while the contribution of ROS to the exciton bleaching becomes more important at higher densities. In addition, these data show that the integrated absorption change can rather well be taken as a direct measure of the carrier density within the respective subbands, justifying the simulation shown in Fig. 2.

These results might explain the contradicting reports on the dominant bleaching mechanisms that can be found in the literature. Unfortunately, the carrier density is not specified in all references. However, the data of Refs. 10 and 11 that indicate a dominance of broadening have been taken at low excitation densities.[17] These measurements were performed at room temperature and 8.5 K, respectively, indicating that there is no significant influence of the lattice temperature. Other experimental details such as pump wavelength or pulse duration can be expected to be of minor importance, since most experiments study the exciton bleaching caused by quasiequilibrium thermalized carrier distributions. It should be noted that in Refs. 7 and 8 the data analysis is based on the theory of Ref. 4, assuming that broadening of the exciton line can be neglected. Our results indicate that this assumption is generally not correct.

In conclusion, we investigate the temporal evolution and the density dependence of exciton bleaching in GaAs/$Al_xGa_{1-x}As$ MQW's. We distinguish between bleaching of the exciton absorption peak and reduction of the exciton oscillator strength. We find that there is no significant influence of carrier cooling on the reduction of exciton oscillator strength, in contrast to the existing theoretical predictions. Our data further show a different density dependence of (peak) exciton bleaching and the

decrease of oscillator strength. For the lowest excitons, the reduction of oscillator strength is roughly proportional to the carrier density in the respective subbands. For higher unoccupied subbands, the oscillator strength is not affected by carrier generation in lower bands.

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[1] *Optical Nonlinearities and Instabilities in Semiconductors*, edited by H. Haug (Academic Press, Boston, 1988).
[2] D. A. B. Miller, D. S. Chemla, D. J. Eilenberger, P. W. Smith, A. C. Gossard, and W. T. Tsang, Appl. Phys. Lett. **41**, 679 (1982).
[3] D. A. B. Miller, D. S. Chemla, T. C. Damen, T. H. Wood, C. A. Burrus, Jr., A. C. Gossard, and W. Wiegmann, IEEE J. Quantum Electron. **QE-21**, 1462 (1985).
[4] S. Schmitt-Rink, D. S. Chemla, and D. A. B. Miller, Phys. Rev. B **32**, 6601 (1985).
[5] R. Zimmermann, Phys. Status Solidi B **146**, 371 (1988).
[6] W. H. Knox, R. L. Fork, M. C. Downer, D. A. B. Miller, D. S. Chemla, and C. V. Shank, Phys. Rev. Lett. **54**, 1306 (1985).
[7] K. Tai, J. Hegarty, and W. T. Tsang, Appl. Phys. Lett. **51**, 86 (1987).
[8] K.-H. Schlaad, C. Weber, J. Cunningham, C. V. Hoof, G. Borghs, G. Weimann, W. Schlapp, H. Nickel, and C. Klingshirn, Phys. Rev. B **43**, 4268 (1991).
[9] M. J. Snelling, P. Perozzo, R. Bambha, A. Miller, and D. C. Hutchings, *Phase-Space Filling and Coulomb Screening in Quantum-Well Excitonic Saturation Determined by Time-Resolved Optical Orientation*, QELS Technical Digest Series (Optical Society of America, Washington, D.C., 1993), pp. 38–39.
[10] D. S. Chemla, D. A. B. Miller, P. W. Smith, A. C. Gossard, and W. Wiegmann, IEEE J. Quantum Electron. **QE-20**, 265 (1984).
[11] D. R. Wake, H. W. Yoon, J. P. Wolfe, and H. Morkoc, Phys. Rev. B **46**, 13 452 (1992).
[12] P. C. Becker, D. Lee, A. M. Johnson, A. G. Prosser, R. D. Feldman, R. F. Austin, and R. E. Behringer, Phys. Rev. Lett. **68**, 1876 (1992).
[13] T. F. Albrecht, K. Seibert, and H. Kurz, Opt. Commun. **84**, 223 (1991).
[14] J. Shah, in *Spectroscopy of Nonequilibrium Electrons and Phonons*, edited by C. V. Shank and B. P. Zakharchenda (Elsevier, Amsterdam, 1992), pp. 57–112.
[15] E. Lach, M. Walther, G. Tränkle, A. Forchel, and G. Weimann, Phys. Status Solidi B **150**, 679 (1988).
[16] W. H. Knox, C. Hirlimann, D. A. B. Miller, J. Shah, D. S. Chemla, and C. V. Shank, Phys. Rev. Lett. **56**, 1191 (1986).
[17] For Ref. 10, we deduce this from the low signal intensity.