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Enhanced energy and phase relaxation of excitons in the presence of bare electrons

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Using transient four-wave-mixing and time-resolved photoluminescence experiments we investigate the influence of bare electrons on the phase and energy relaxation of excitons in a mixed type-I–type-II GaAs/AlAs double-quantum-well structure. We find that exciton-electron scattering is twice as efficient as exciton-exciton scattering. As a consequence, the rates for exciton formation and for energy relaxation of free excitons to $K \approx 0$ are enhanced. Additionally, localized excitons become mobile in the presence of bare electrons leading to a more complete relaxation of excitons into the energetically lowest quantum-well states.

Carrier scattering in semiconductors is a subject of great technological importance owing to its relevance for modern electronic devices. Some of the processes that have been intensively studied are the cooling and thermalization of hot carriers, the formation and energy relaxation of excitons as well as their loss of coherence. A variety of experimental techniques, including transport measurements,[1–5] recombination studies of hot electrons and excitons,[6,7] and coherent laser spectroscopy have been applied to study the underlying scattering rates of carriers with other carriers, phonons, and crystal imperfections.[8–14] In particular, transient four-wave-mixing (FWM) experiments have proven to be a powerful tool to monitor the ultrafast scattering events in semiconductors.[14]

Among the various scattering processes, carrier-carrier scattering is one of the most complicated phenomena, since it represents a many-body problem, where exchange-, screening-, collective plasma-, and plasmon-phonon coupling effects play a crucial role.[14,15] In the context of carrier-carrier Coulomb interactions, the scattering of bound electron-hole states (excitons) with other carriers is particularly interesting. Schultheis *et al.*[16] have determined the rates for exciton-exciton (X-X) scattering and for scattering of excitons with unbound electron-hole pairs (X-eh) by performing transient FWM experiments with a frequency-tunable prepulse. The authors find that X-eh scattering is approximately 10 times more efficient than X-X scattering. In addition, both scattering processes are more efficient in two dimensions (2D) as compared to 3D, since screening of the Coulomb interaction is weaker in 2D. The scattering of excitons with unbound electron-hole pairs is already a quite complicated process, since holes screen the exciton-electron and electrons the exciton-hole interaction. Accordingly, it is desirable to investigate a less complicated system, where, e.g., electrons scatter with excitons in the absence of any unbound holes (X-e scattering).

One possibility would be to perform transient FWM experiments on n-modulation-doped quantum-well structures, where the electron density can be controlled by a gate voltage.[2] Kim *et al.*[17] have performed time-resolved FWM experiments on n-modulation-doped quantum wells. However, in their experiments the electron density was fixed to two very high values of 2×10^{11} cm^{-2} and 6×10^{11} cm^{-2} leading to degenerate electron systems, i.e., to Fermi seas with 7 meV and 20 meV depth. The authors find a strong inhibition of scattering near the Fermi edge due to Pauli blocking. At these high densities, where the Fermi energy is comparable to or larger than the exciton binding energy, bound exciton states do not exist and thus the concept of exciton-electron scattering makes no sense.

In this paper we report optical experiments on mixed type-I–type-II GaAs/AlAs double-quantum-well structures.[18,19] Optical excitation of electron-hole pairs in the narrow well (NW) and the successive Γ-X-Γ electron transfer to the wide well (WW) offers the unique opportunity to optically control the electron density in the WW. Transient FWM experiments, which monitor the coherent phase relaxation, show that X-e dipole-monopole scattering is twice as efficient as X-X dipole-dipole scattering. Accordingly, the presence of electrons in the WW drastically changes the relaxation dynamics of excitons in the WW. This can be inferred from time-resolved photoluminescence (PL) experiments. The enhanced scattering leads to a faster onset of the luminescence, i.e., to a faster exciton formation and energy relaxation. In addition, the presence of bare electrons leads to an energy transfer from a high-energetic quantum-well island to a low-energetic quantum-well island and an overall partial quenching of the PL. In the absence of any additional electrons excitons are localized in shallow states of the quantum-well islands, but then become mobile due to X-e scattering. Being mobile, excitons on higher-energetic islands diffuse to and are trapped into lower-energetic islands or reach nonradiative recombination centers. This leads to an energetic redistribution and a partial quenching of the luminescence.

The investigated mixed type-I–type-II sample consists of twenty periods of 2.5 nm narrow GaAs wells and 6.8 nm wide GaAs wells separated by a 10.3 nm AlAs barrier.[19] As shown in the upper part of Fig. 1, the lowest $n=1$ Γ electron state in the NW is energetically higher than the lowest X state in the AlAs barrier. The lowest $n=1$ Γ electron state in the WW, however, is lower than the AlAs X state. This characteristic band scheme of a mixed type-I–type-II double-

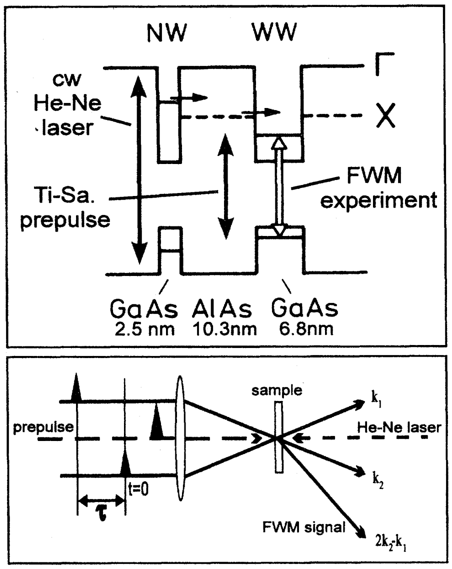

FIG. 1. Upper part: Schematic band scheme of the mixed type-I–type-II GaAs/AlAs double-quantum-well sample. The FWM experiment is performed at the heavy-hole exciton transition of the WW and is disturbed either by a prepulse from the Ti:sapphire laser (*X-X* scattering) or by a cw He-Ne laser (*X-e* scattering). Lower part: Geometrical setup for the two-beam FWM experiment with an additional prepulse or with an additional He-Ne laser.

quantum-well structure leads to an efficient Γ-*X*-Γ transfer of electrons initially photoexcited in the NW.[18,19,21] Since the lifetime of the spatially separated electron-hole system lies in the μs regime, optical excitation of electron-hole pairs in the NW at very low densities is sufficient to create high densities of cold electrons in the WW and of cold holes in the NW.

As schematically illustrated in the lower and upper parts of Fig. 1, the FWM experiments are performed at $T = 5$ K on the energetically lowest excitonic $n = 1$ transition of the WW using 110 fs pulses from a mode-locked Ti:sapphire laser. We use a so-called two-beam self-diffraction geometry, i.e., two laser pulses impinge onto the sample with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$. As a result, a FWM signal is emitted into the phase-matched direction $2\mathbf{k}_2 - \mathbf{k}_1$ and can be measured as a function of time delay between the two laser pulses (for details see, e.g., Ref. 16). We choose the excitation intensity as low as possible to obtain a long dephasing time T_2 mainly determined by scattering of excitons with acoustic phonons, and disorder potentials. We now perform two kinds of experiments. First, a prepulse from the same Ti:sapphire laser is used to create an incoherent density of excitons in the WW in order to measure the dephasing rate due to exciton-exciton (*X-X*) scattering as it was done in Ref. 16. Second, in the absense of the third Ti:sapphire pulse a cw He-Ne laser is used to photocreate electron-hole pairs in the NW and thus a density of cold electrons in the WW after the Γ-*X*-Γ transfer.

The phase relaxation time T_2, which can be determined from the decay constant τ of the FWM signal according to $T_2 = 4\tau$,[22] is related to the homogeneous linewidth by $\Gamma_{\rm hom} = h/\pi T_2$. In the presence of additional excitons or electrons $\Gamma_{\rm hom}$ increases. This is a direct consequence of the *X-X* or *X-e* scattering, respectively.[23] Thus by changing the laser intensities, the dependences of the homogeneous linewidth on the densities n of excitons or electrons can be measured. Special care is necessary for the experimental determination of n. We carefully measured the optical transmission of the sample, using pinholes to exactly determine the laser spot size. Furthermore, the lifetime of the spatially separated electron-hole system is obtained from the data reported in Refs. 19 and 20. Figure 2 shows the dependence of $\Gamma_{\rm hom}$ on the density of excitons (circles) and electrons (triangles) for densities far below the Mott density. In both cases $\Gamma_{\rm hom}$ increases linearly with density. The slopes directly reflect the interaction strengths of *X-X* and *X-e* scattering. We find 6.5×10^{-12} meV cm^2 for *X-X* scattering and 13.2×10^{-12} meV cm^2 for *X-e* scattering. *This means that exciton-electron scattering is twice as efficient as exciton-exciton scattering.*

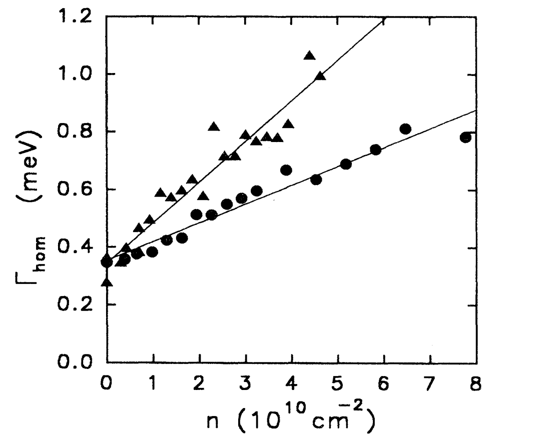

FIG. 2. Dependence of $\Gamma_{\rm hom}$ on the exciton density (circles) and on the electron density (triangles).

For bulk semiconductors, i.e., for three-dimensional electron systems Haug[24] calculated the matrix elements for *X-X* (dipole-dipole) and *X-e* (dipole-monopole) interaction. He found that the *X-e* matrix element is 2.8 times larger than that for the *X-X* interaction, which is in reasonable agreement with our results. However, since the calculations in Ref. 24 are performed for the three-dimensional case and consider only matrix elements they might not be directly comparable to our experimental findings. To our knowledge, a comprehensive theory concerning the problem of incoherent *X-e* scattering in quasi-two-dimensional quantum structures has not been developed so far.

In order to show that our density calculations are correct, we compare our results on *X-X* scattering with those of Kuhl *et al.*[25] The homogeneous linewidth due to *X-X* scattering can be written as

$$\Gamma_{\rm hom}(n) = \gamma_{XX} a_B^2 E_B n_X , \tag{1}$$

where a_B is the exciton Bohr radius (7 nm for our sample), E_B is the exciton binding energy (13 meV for our sample), and n_X is the exciton density.[16] From the slope of 6.5×10^{-12} meV cm^2 for *X-X* scattering we then obtain a γ_{XX} value of 1.0 for the 6.8 nm thick GaAs quantum-well structure in good agreement with Ref. 25.

We now investigate how the addition of electrons changes the energetic relaxation and recombination dynamics of ex-

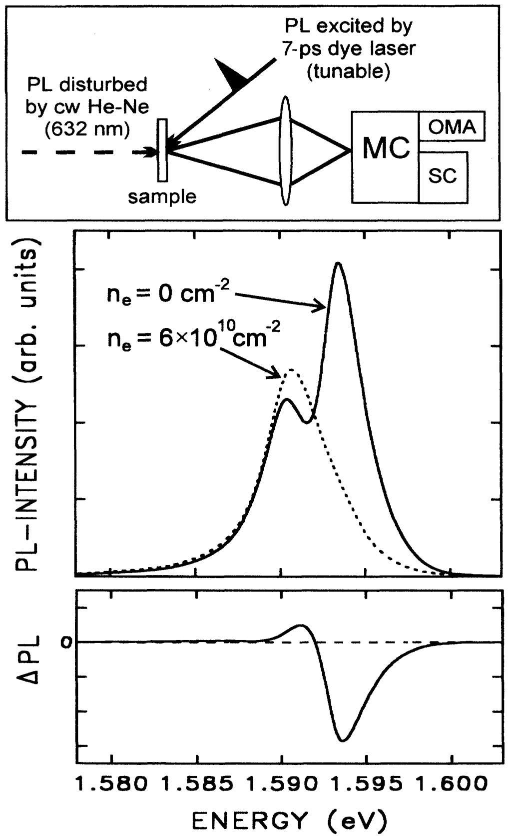

FIG. 3. Upper part: Experimental setup for the PL experiments using a dye and a He-Ne laser. Lower part: PL spectra of the WW without (solid curve) and with He-Ne laser excitation (dashed curve). The lower part shows the difference ΔPL between the two PL spectra.

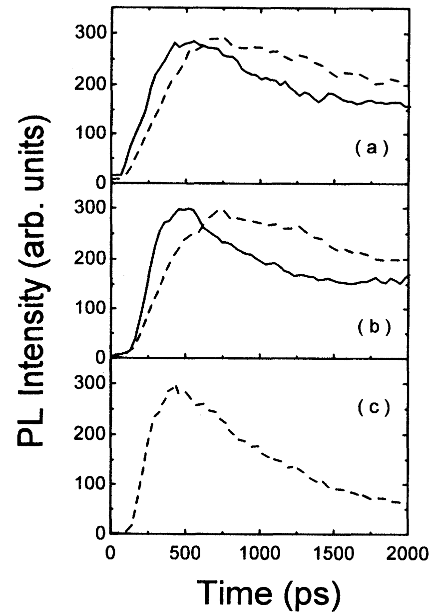

FIG. 4. Time-resolved photoluminescence of the low (dashed line) and high (solid line) energy peak of the WW for different electron densities as described in the text ($T=5$ K).

citons in the wide GaAs well. As an experimental technique we employ low-temperature PL measurements. The experimental setup is shown in the upper part of Fig. 3. A synchronously pumped dye laser emitting 7 ps pulses at 1.63 eV is used to optically excite *only* the wide GaAs well. As in the FWM experiments a He-Ne laser is used to photocreate electron-hole pairs in the NW and thus a cold electron density in the wide GaAs well. Again the electron density is controlled by the intensity of the He-Ne laser. For the time-resolved detection of the PL we apply a streak camera system with a time resolution of about 30 ps.

When the sample is excited solely with the dye laser, i.e., no cold electron density is created in the WW, the PL spectrum of the WW displays two dinstinct peaks, as shown by the solid line in the upper part of Fig. 1. These two peaks exhibit a splitting of 3.1 meV and can be associated with the recombination of excitons localized in different quantum-well islands differing in thickness by one monolayer.[26] In the presence of a cold electron density in the WW the PL spectrum changes drastically. With increasing electron density the intensity of the high energy peak of the WW PL gradually drops. Exciting an electron density of 6×10^{10} cm^{-2}, only the low energy peak of the WW emission can be observed (dashed line in the upper part of Fig. 3). Moreover, the low energy peak has gained intensity as compared to the case without any additional electrons. This is shown more clearly in the lower part of Fig. 3, where we plot the difference between the two PL spectra shown in the upper part. Besides the energy transfer from the high energy part of the WW PL to its low energetic region, an overall decrease of the PL efficiency is obvious. Related effects induced by an electron-hole plasma were recently observed by Ashkinadze *et al.*[27] in a conventional type-I GaAs/AlGaAs quantum-well sample within one spectrally broad PL peak.

The drastic change of the PL induced by additional electrons can be understood in the following way. It is reasonable to assume that in the absence of any additional electrons excitons on any quantum well island are partially localized due to short-scale potential fluctuations. Otherwise, excitons located on islands with narrower well widths would diffuse to islands with wider well widths within the lifetime and a high-energetic PL peak would not be as pronounced as observed in Fig. 3 (solid curve). As can be inferred from Fig. 2, the presence of 6×10^{10} cm^{-2} electrons leads to a high scattering rate of excitons. This means that excitons localized in the absence of additional electrons now become mobile. When they reach a quantum-well island with wider well width they relax down to the lowest energetic states of this island (island trapping). Accordingly, with increasing electron density the low-energetic PL peak associated with islands having a wider well width gains in intensity, whereas the high-energetic peak is more and more quenched. In addition, the higher mobility of excitons in the presence of electrons leads to a higher probability that excitons reach nonradiative recombination centers. This explains the overall quenching of the PL.

The dynamics of this enhanced energy relaxation in the presence of additional electrons can be studied by time-resolved PL. The transient behavior of the PL from the high (solid line) and low (dashed line) energy peak of the WW PL is depicted in Fig. 4 for three different electron densities. For a relatively low electron density of 2×10^9 cm^{-2} [Fig. 4(a)], both peaks exhibit comparable luminescence decay times. The low energy peak, however, reveals a longer rise time, which is due to the delayed population of the wider island, i.e., to the island trapping process. For an electron density of 1.2×10^{10} cm^{-2} [Fig. 4(b)] the high-energetic part of the WW PL shows a pronounced shortening of the rise time accompanied by a faster initial decay of the PL. In contrast, the rise and decay of the low energy peak is nearly unaffected. Since the WW is excited in the continuum, i.e., we create carriers/excitons with excess energy, we attribute the shorter rise of the high energy peak PL to an enhanced exciton formation rate and energy relaxation rate, caused by enhanced exciton electron scattering.[28] The faster initial decay of the PL is related to a more efficient contribution of island trapping, which acts as a nonradiative channel for the high energy PL peak. The second component in the decay of the high energy PL peak is partly determined by the decay of excitons which are not trapped into wider wells and by the low energy peak of the PL, which energetically overlaps with the high energy peak due to inhomogeneous broadening (see Fig. 3). At the highest electron density (2.5×10^{10} cm^{-2}) [Fig. 4(c)], i.e., when the time-integrated PL spectrum is predominantly determined by the low energy peak, a shortening of the rise and decay time of the low energy peak is observed, the later being due to the stronger contribution of nonradiative recombination (overall quenching of the PL efficiency) and the former due to the enhanced energy relaxation of excitons by exciton electron scattering.

In summary, we have used the unique properties of a mixed type-I–type-II GaAs/AlAs double-quantum-well structure to optically create a pure electron density in the wide GaAs well and to investigate the effects of exciton-electron scattering on exciton dephasing and on the exciton relaxation scenario. We find that X-e scattering is twice as efficient as X-X scattering in qualitative agreement with theory. As a consequence of X-e scattering, carrier relaxation rates are enhanced. Excitons normally localized in shallow states of the quantum-well islands become mobile and migrate to islands with wider well width and to nonradiative recombination centers. This leads to drastic changes in the PL spectrum.

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