# Influence of the kinetic energy of electrons on the formation of excitons in a shallow $In_xGa_{1-x}As/GaAs$ quantum well

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Time-resolved investigations were performed with systematic variation of the excitation energy in order to study the formation of excitons. In a proper designed shallow $In_xGa_{1-x}As/GaAs$ single quantum well heterostructure an electron system with well-defined kinetic excess energy relative to the excitonic ground state of the quantum well was prepared by photoexcitation and subsequent relaxation via cascaded LO-phonon scattering. Oscillations of the photoluminescence rise time and, in counter phase, a modulation of the decay transient are observed as function of the excitation energy. While the decay time reflects the electron transfer process into the ground state due to different phonon relaxation mechanisms, the rise time reveals an exciton formation process, which depends on the kinetic energy of the electrons. [S0163-1829(96)03640-5]

Carrier dynamics in quantum well (QW) heterostructures has gained a lot of interest during the past years. This is due to the applicability of appropriate structures for the development of optoelectronic devices as well as due to the interesting basic physics. Excellent sample quality and the application of ultrafast laser spectroscopy allow the study of intrinsic dynamic processes. Depending on the experimental condition and the geometry of the structure, carrier relaxation processes in phase,[1] momentum,[2,3] and energy,[4–6] the transport to the well,[7–9] the capture process,[10–12] and the thermal emission,[13,14] the formation of excitons,[15–17] and the recombination process[18] have to be taken into account for a complete understanding of carrier dynamics. In this paper, we present results on the dynamics of exciton formation in a shallow $In_xGa_{1-x}As/GaAs$ QW heterostructure. A dependence of the exciton formation process on electron motion in real space is revealed. An electron system with a well-defined kinetic energy was prepared after photoexcitation of carriers in barrier states and the transfer into the QW by cascaded LO-phonon scattering.

The sample under investigation was an especially designed pseudomorphic $In_xGa_{1-x}As/GaAs$ QW heterostructure. A single, 9 nm-wide $In_{0.2}Ga_{0.8}As$ QW is embedded in 30 nm-wide GaAs barriers. To complete a graded index separate confinement heterostructure (GRINSCH), 165 nm $Al_xGa_{1-x}As$ with increasing Al content from 20–50% were grown on both sides. Details of the growth process of the high-quality[2] pseudomorphic sample are described in Ref. 19. This structure enables the observation of pure energy relaxation effects, because parasitic recombination and transport processes vanish: due to the GRINSCH, electrons and holes excited in the GaAs barrier can only be captured into the $In_xGa_{1-x}As$ single QW since the higher $Al_xGa_{1-x}As$ barriers prevent carriers from reaching the sample surface. Also carrier diffusion across the barrier is negligible due to the use of only 30-nm-thick GaAs barriers.[8] This has been verified by temperature-dependent time-resolved measurements.[20]

Sample excitation was performed with a Ti-sapphire laser, yielding pulses with a full width at half maximum of 2 ps. For a high time resolution of ~2 ps the technique of frequency up conversion was used.[21] The up-converted signal was spectrally dispersed by a monochromator and the spectrum was detected by an optical multichannel analyzer. Carrier density was kept below $10^9$ $cm^{-2}$, where excitonic recombination is dominant. To get a sufficient exciton luminescence signal to noise ratio, integration times of a few minutes per transient spectrum were necessary. The sample temperature was at $T=2$ K.

In Fig. 1 the time-resolved photoluminescence (PL) intensity of the $1e$-1hh exciton in the QW ($E=1.33$ eV) is depicted for different excitation energies $E_{\mathrm{exc}}$ above the GaAs band gap. The lines are least-square fits using a simple three-level model, where carriers are generated in an excited energy level and the population rate into the QW ground state is given by the inverse of the time constant $\tau_r$. Radiative recombination takes place and depopulates the ground state with the decay time $\tau_{\mathrm{dec}}$.

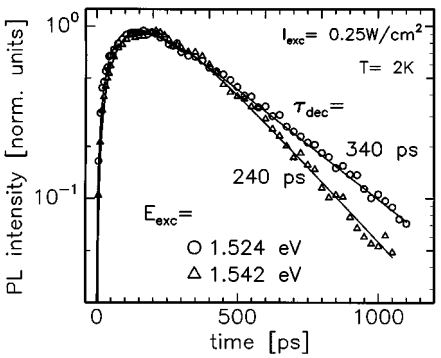


FIG. 1. The time evolution of the PL signal of the $1e$-1hh exciton in the shallow $In_xGa_{1-x}As/GaAs$ single QW heterostructure depends on excitation energy $E_{\mathrm{exc}}$. The solid lines are least-square fits yielding the rise time $\tau_r$ and the decay time $\tau_{\mathrm{dec}}$ of the PL intensity. $\tau_{\mathrm{dec}}$ shows a variation of 100 ps for the depicted time-resolved measurement at two different excitation energies.

 

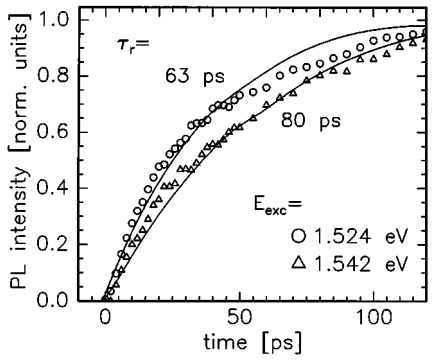


FIG. 2. To demonstrate the modification of the increase of the $1e$-1hh exciton PL intensity with $E_{\mathrm{exc}}$ the first 120 ps of the measurements depicted in Fig. 1 are magnified. The slower increase at the excitation level $E_{\mathrm{exc}}=1.542$ eV is correlated with a faster decay in comparison with the measurements at $E_{\mathrm{exc}}=1.524$ eV.

For the decrease of the PL intensity, we observe a distinct modification as the excitation energy is changed. This is expressed by different decay times $\tau_{\mathrm{dec}}$, which show a variation of 100 ps. To see the increase of the PL intensity in more detail, the first 120 ps after the excitation pulse are depicted in Fig. 2. We observe a distinct change of the slope of the PL intensity from the origin at $t=0$ ps for the two different excitation energies. Subsequently the increase of the PL intensity at the higher excitation energy ($E_{\mathrm{exc}}=1.542$ eV) takes longer compared to the lower one ($E_{\mathrm{exc}}=1.524$ eV). This increase behavior is in contrast to the intensity decrease in Fig. 1, where the decay time for $E_{\mathrm{exc}}=1.542$ eV is shorter. From the fits (solid lines) we deduce a difference of the rise time $\tau_r$ of 17 ps. Furthermore, the discrepancy between the measured increase of the PL intensity and the least-square fits indicates that the three-level model taking into account an exponential population rate is too simple for a theoretical description. Therefore, we present a more detailed investigation below.

In Fig. 3 the decay time $\tau_{\mathrm{dec}}$ and the rise time $\tau_r$ are plotted as function of excitation energy. The value of $\tau_{\mathrm{dec}}$ vary between 220 and 390 ps. For excitation above the GaAs band-gap periodic oscillations are observed. These variations were reported recently for shallow $In_xGa_{1-x}As/GaAs$ QW structures as a function of well width and In content.[22] The maxima and minima of $\tau_{\mathrm{dec}}$ are equidistant in energy and are described by

$$E_{\max}=E_0+(n+0.5)\hbar\omega_{\mathrm{LO}}\left(1+\frac{m_e}{m_{\mathrm{hh}}}\right), \qquad n=4,5,\ldots,$$
$$E_{\min}=E_0+n\hbar\omega_{\mathrm{LO}}\left(1+\frac{m_e}{m_{\mathrm{hh}}}\right), \qquad n=5,6,\ldots, \tag{1}$$

where $m_e=0.0665m_0$ and $m_{\mathrm{hh}}=0.54m_0$ are the masses of the electron and the heavy hole and $\hbar\omega_{\mathrm{LO}}=36.4$ meV corresponds to the energy of the LO-phonon in GaAs. $E_0$ is the ground-state energy in the QW. We measured the first period around the maxima of $\tau_{\mathrm{dec}}$ with $n=4$ and $n=5$ and two extreme points at higher excitation levels, the minimum at $n=6$, and the maximum at $n=8$. An excellent agreement is found with the interpolated curve according to Eq. (1), which is indicated by the solid line. The oscillations with a period of $\hbar\omega_{\mathrm{LO}}(1+m_e/m_{\mathrm{hh}})$ are explained as follows. In the case of optical excitation, the excess energy of the exciting laser is shared between electron and hole under energy and momentum conservation. Electrons lose their excess energy $\Delta E_e$ rapidly due to cascaded LO-phonon scattering in $n$ steps until $0\leqslant\Delta E_{e/s}\leqslant\hbar\omega_{\mathrm{LO}}$ ($\Delta E_{e/s}$: excess energy of electrons after cascaded LO-phonon-scattering process). Thus an electron system is prepared, where two cases are possible: First, if $\Delta E_e=n\hbar\omega_{\mathrm{LO}}$ electrons reached the ground state in the quantum well via LO-phonon emission, and, after forming an exciton, recombine radiatively. In the case $\Delta E_e\neq n\hbar\omega_{\mathrm{LO}}$ kinetic energy is left in the electron system. To reach the radiative ground state within the homogeneous linewidth with $\mathbf{K}\simeq\mathbf{0}$,[18] further cooling via slower acoustic-phonon scattering is necessary. Consequently, the radiative decay of excitons is prolonged, whereas the first case leads to a faster decay time. For time limiting processes holes need not be considered, because they lose their excess energy on a faster subpicosecond time scale due to strong phonon coupling.[23] The excess energy distribution of photoexcited holes and electrons is taken into account by the ratio of the masses in Eq. (1).

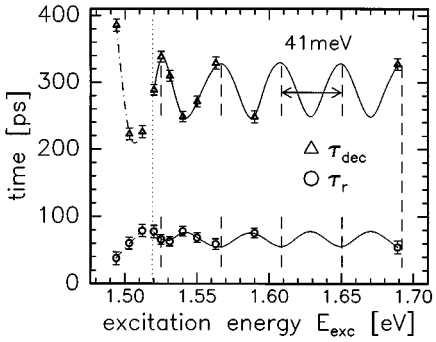


FIG. 3. The decay time $\tau_{\mathrm{dec}}$ and the rise time $\tau_r$ for the $In_{0.2}Ga_{0.8}As/GaAs$ single QW structure show a counterphase behavior as function of the excitation energy. For excitation above the GaAs barrier, indicated by a dotted line, oscillations correlated with the LO-phonon energy are observed. The period of 41 meV is marked by dashed lines. The full line is interpolated according to Eq. (1). Below the GaAs barrier light holes are excited resonantly at $E_{\mathrm{exc}}=1.492$ eV in the QW. The dash-dotted lines serve as a guide to the eye.

A significant slowing down of the relaxation process, indicated by an enhanced decay time, is observed for resonant excitation of light-hole excitons in the QW at 1.492 eV. Hot heavy-hole excitons with high excess energies are created by an efficient transition between the light-hole and the heavy-hole excitons.[15] Before recombination at $\mathbf{K}\simeq\mathbf{0}$ the excitons must lose their excess energy by emission of phonons. The long decay time is due to the weak exciton-acoustic phonon interaction.[22]

As a first result we conclude that the decay time $\tau_{\mathrm{dec}}$ is not only given by the radiative lifetime of excitons but is strongly influenced by the energy relaxation rate[24] of the carrier system due to carrier-phonon interactions. From the measured decay time $\tau_{\mathrm{dec}}$ as a function of excitation energy, we deduce an upper limit for the radiative lifetime of the exciton in the $In_{0.2}Ga_{0.8}As/GaAs$ QW heterostructure. It is given by the minimum decay time of $\tau_{\mathrm{dec}}=220$ ps.

With the picosecond time resolution of our experiment we were able to determine the rise time $\tau_r$ of the PL intensity in extension to the measurements in Ref. 22. The results are depicted in Fig. 3 by circles. The behavior of $\tau_r$ as function of $E_{\mathrm{exc}}$ is in counterphase to $\tau_{\mathrm{dec}}$ with values between 39 and 80 ps. Long decay times are correlated with short rise times and vice versa.

For a qualitative explanation of the behavior of $\tau_r$, we suggest the following: Excited above the GaAs barrier, electrons relax via LO-phonon emission. The QW ground state is reached directly if $\Delta E_e = n\hbar\omega_{\mathrm{LO}}$. No kinetic energy is left ($\Delta E_{e/s} = 0$), so electrons do not move in real space. Without kinetic energy, the probability of finding a hole in the QW layer and to form an exciton is reduced at early times. The dynamics of exciton formation is reflected in a slow intensity increase. In the case $\Delta E_e \neq n\hbar\omega_{\mathrm{LO}}$ however, kinetic energy is left for electrons ($\Delta E_{e/s} > 0$). Due to electron motion, the probability of finding a hole and of forming an exciton is enhanced. In the hot carrier system some carriers are scattered into the ground state with $\mathbf{K} \simeq \mathbf{0}$, where radiative recombination takes place. This enables a faster increase of the PL intensity. Cooling of the remaining carriers takes long due to the weak carrier-acoustic phonon interaction, and the radiative decay is delayed. From this interpretation we conclude that the decay time $\tau_{\mathrm{dec}}$ is strongly influenced by hot carrier cooling, whereas the rise time $\tau_r$ is given by the population of the ground state and the exciton formation process, which is a function of kinetic energy of the electrons. From the extreme points of the rise time $\tau_r = 55$ and $\tau_r = 80$ ps it is deduced that the exciton formation process in real space can take 25 ps in a $In_xGa_{1-x}As$ QW structure at low temperature.

For resonant light-hole excitation, hot heavy holes are generated efficiently. A certain quota is scattered into the $\mathbf{K} \simeq \mathbf{0}$ ground state and recombines radiatively. Again, this leads to a fast intensity increase, which is even faster compared to nonresonant excitation conditions as described above. Hot excitons relax slowly via acoustic-phonon scattering, which implies a long decay time.

We emphasize, that the process which leads to the variation of the PL rise time as a function of excitation energy in the shallow $In_xGa_{1-x}As/GaAs$ single QW is quite different than those reported so far in $GaAs/Al_xGa_{1-x}As$ QW heterostructures.[16,12,26] The oscillation period of 41 meV in the GaAs barrier is clearly correlated with the excess energy of the electron with respect to the ground state in the QW in terms of the LO-phonon energy. No indication of oscillations due to LO-phonon-assisted exciton formation is observed. They have been observed in $GaAs/Al_xGa_{1-x}As$ with a typical period of 25 meV.[16] A further process under discussion is the quantum-mechanical capture of carriers from barrier states into the QW. There the resulting time constant is related to the subband structure and not to the electron excess energy.[12] In our experiment the counterphase behavior of $\tau_{\mathrm{dec}}$ and $\tau_r$ in a shallow QW demonstrates that the kinetic energy of the electrons after the relaxation process via cascaded LO-phonon-scattering plays a dominant role for the exciton formation. At low carrier density of about $10^9$ cm$^{-2}$ an electron has to move over an average distance of ten exciton Bohr radii ($r_B = 18$ nm) to hit a hole. So, it is quite obvious, that the exciton formation process depends on electron motion and velocity. For measurements at higher carrier densities the average distance between carriers reduces and the variation of $\tau_r$ with excitation energy vanishes.

The features observed in our experiment are quite obvious and are confirmed by statistical analysis of the data, expressed by error bars in Fig. 3. The discrepancy between measurements and fits for the increase of the PL intensity (see Fig. 2) resulting in error bars of up to 19% for the rise time $\tau_r$ indicates, however, that the three-level model taking into account a population process with an exponential rate is too simple. More theoretical work, including the mutual interaction between electrons and holes, excitons, and phonons, and the formation of excitons as a function of kinetic excess energy of the electron is necessary for a detailed quantitative understanding. A nonequilibrium thermal distribution of carriers has to be considered, because scattering rates at low density and low temperature are so weak that within the radiative recombination no thermal distribution of carriers turns up.

In summary, we have investigated the transfer process of photoexcited carriers into the ground state and the formation of excitons using time-resolved PL spectroscopy. To focus on the energy relaxation and exciton formation process, a shallow $In_xGa_{1-x}As/GaAs$ single QW GRINSCH was used. As a key result for excitation energies above the GaAs band gap, we observe counterphase oscillations of the PL rise and decay time of the 1*e*-1hh exciton with a period related to the LO-phonon energy. The decay time is correlated with the energy relaxation rate of the electron system via the short cascaded LO-phonon scattering process and, in the case where kinetic energy is left, via slow acoustic-phonon scattering. In the latter case, electron motion in real space enhances the probability of finding a hole and thus forming an exciton, which results in shorter rise times. For a selective excitation of the light-hole exciton in the QW, hot heavy excitons are generated efficiently. Exciton scattering into the radiative $\mathbf{K} \simeq \mathbf{0}$ ground state leads to a fast PL rise time. In contrast, the decay time is long because of slow energy relaxation via acoustic-phonon scattering.

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[24] To obtain exclusively the processes of energy relaxation and capture into one QW, we investigated a single quantum well GRINSCH. Due to sample geometry the radiative efficiency of the QW luminescence does not depend on the excitation energy in the GaAs barrier. This was proved by continuous-wave PL excitation spectroscopy, where the spectrum does not change significantly with energy. With the absorption coefficient being nearly constant within the energy range under consideration, the transfer efficiency of carriers into the radiative QW ground state is constant. This is in contrast to the results in Ref. 22, where the PL excitation spectra of the shallow $In_xGa_{1-x}As/GaAs$ QW structures showed oscillations of the PL intensity as function of excitation energy. For these investigations, the sample consisted of four QW's with different thicknesses separated by barriers. In these samples, carriers generated in the barrier are captured by the four QW's. The transfer efficiency into the different QW's depends on excess energy of the electrons and implies a radiative efficiency for each QW, which varies periodically as a function of excitation energy (Ref. 22).