Recombination Enhancement due to Carrier Localization in Quantum Well Structures

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Picosecond luminescence experiments on GaAs/$Al_xGa_{1-x}As$ quantum well structures reveal a significant influence of localization on the transition probabilities of photoexcited carriers. The spontaneous lifetime of electrons and holes within the quantum well decreases with well thickness L_z from 1 ns for $L_z = 14$ nm to 350 ps for $L_z = 5$ nm because of the enhanced recombination due to localization of the carriers.

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Quantum well (QW) structures in $A^{III}B^{V}$ semiconductors have recently attained significant importance because of their unique electronic properties[1-3] which allow promising applications in modern device technology.[4] The intriguing features of these QW structures basically arise from the localization of electron and hole (exciton) wave functions which for small well thickness L_z results in a quasi-two-dimensional (2D) carrier system. Modified exciton transition probabilities should result from this localization because of the change in overlap of the electron and hole wave functions.[5,6] This effect of carrier localization on transition probabilities is a general problem in solid-state physics[7] and has been studied, e.g., for bound excitons[8] as well as exciton molecules.[9] Quantum well structures allow continuous variation of the localization by changing the well thickness and barrier height, and thus provide a unique tool for the investigation of localization effects.

We report the first measurements of spontaneous carrier lifetime in GaAs/$Al_xGa_{1-x}As$ QW structures. The carrier lifetime $\tau_{r,QW}$ decreases with decreasing well thickness from 1 ns for $L_z = 14$ nm to 350 ps for $L_z = 5$ nm. This decrease of $\tau_{r,QW}$ directly reflects the recombination enhancement due to localization. The decrease in $\tau_{r,QW}$ by a factor of 2.85 from $L_z = 14$ nm to $L_z = 5$ nm corresponds to a shrinkage of the exciton Bohr radius a_0 by 1.7, which is close to the maximum value of 2 expected for a complete transition from a three-dimensional to a two-dimensional electronic system.

The GaAs/$Al_xGa_{1-x}As$ QW structures are grown by molecular-beam epitaxy. Details of the growth process have been described elsewhere.[10] The luminescence experiments are performed at a sample temperature of about 10 K. A synchronously mode-locked cw dye laser (Rhodamine 6 G, emission wavelength ~575 nm, 4.7 ps pulse width, 76.5 MHz repetition rate) and a synchroscan streak camera (S20 spectral response) together with a 0.25-m grating spectrometer are used for excitation and detection, respectively. The usage of the synchroscan streak camera enables us to perform picosecond luminescence experiments with rather low excitation intensities. The average optical power was 10 mW and is focused to a spot of 100 μm diam, which corresponds to a photon flux per pulse of about 4×10^{12} photons/cm^2. The excitation pulses are almost completely absorbed in the 1-μm-thick top $Al_xGa_{1-x}As$ layers used in our experiments. The jitter in synchronization of the incident light pulses with the deflection of the streak camera is smaller than 8 ps. Overall time resolution of the experimental setup has been determined experimentally to be 25 ps, taking into account the temporal broadening by the spectrometer.

Results for a GaAs/$Al_xGa_{1-x}As$ double-QW structure with $L_z = 14$ nm are depicted in Fig. 1. The double-QW configuration ($x = 0.21$ for barrier and cladding layers) was chosen to demonstrate the absence of effects due to alloy clustering in the cw photoluminescence measurements.[10] The sample parameters and the respective energies for the optical transitions are shown in the inset. The time-integrated luminescence spectrum reveals the recombination of carriers within the $Al_xGa_{1-x}As$ cladding layers at 1.78 eV, and two bands at 1.513 and 1.49 eV resulting from transitions within the GaAs substrate.[11] The narrow and broad features with maxima at about 1.54 and 1.63 eV, respectively, are attributed to the $n = 1$ and $n = 2$ subband transitions within the QW. The $n = 2$ transition is only observed under picosecond excitation because of the higher initial carrier concentrations and effective carrier temperatures, which both favor population of the $n = 2$ states.[12]

The time behavior of the photoluminescence corresponding to transitions within the $Al_xGa_{1-x}As$ (curve 1) and within the QW (curves 2–4) is dis-

played by the original streak-camera traces in the lower part of Fig. 1. The luminescence from $Al_xGa_{1-x}As$ shows an immediate response, whereas the QW recombination is delayed and the intensity increase corresponds approximately to the decrease of the $Al_xGa_{1-x}As$ luminescence. This time response directly reflects the trapping dynamics of the photoexcited carriers. The decay of the $Al_xGa_{1-x}As$ luminescence would be determined by the spontaneous carrier lifetime, if no trapping exists. This carrier lifetime is expected to be of the order of 1 ns similar to that in 3D GaAs.[13] The observed rapid decay of the $Al_xGa_{1-x}As$ luminescence is thus caused by the fast trapping of the carriers. Consequently, the delayed increase of the QW luminescence is due to accumulation of photoexcited carriers within the QW by very efficient trapping. This fast carrier trapping is consistent with the experimental values for the ratio of the integrated luminescence intensity of the QW to that of the $Al_xGa_{1-x}As$, $I_{QW}/I_{Al_xGa_{1-x}As}$. This intensity ratio would be determined by the respective volumes without trapping, and thus the QW luminescence would be negligible. The strong QW intensity observed even for double- or single-QW structures in stationary as well as in picosecond luminescence experiments ($I_{QW}/I_{Al_xGa_{1-x}As} \simeq 1$ for the time-integrated spectrum of Fig. 1) is due to the effective trapping. The decay of the QW luminescence is

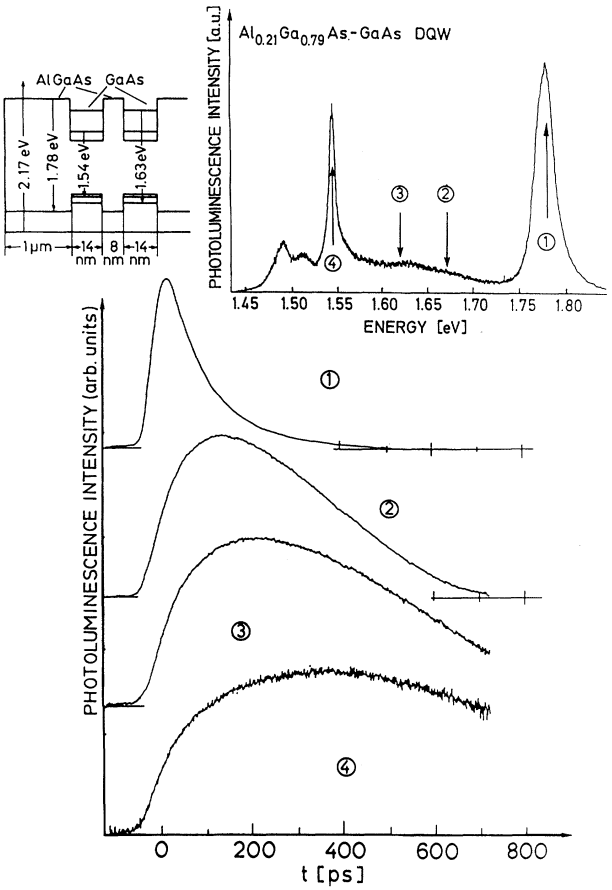

FIG. 1. Experimental luminescence data for a GaAs/$Al_xGa_{1-x}As$ double-QW structure. The sample parameters and the energy of the exciting photons (2.17 eV) as well as of the optical transitions in the $Al_xGa_{1-x}As$ (1.78 eV) and the $n=1$ (1.54 eV) and $n=2$ (1.63 eV) QW are shown at the upper left. The upper right part depicts the time-integrated luminescence spectrum. The time behavior of the emission at the spectral positions indicated by the arrows is plotted in the lower part.

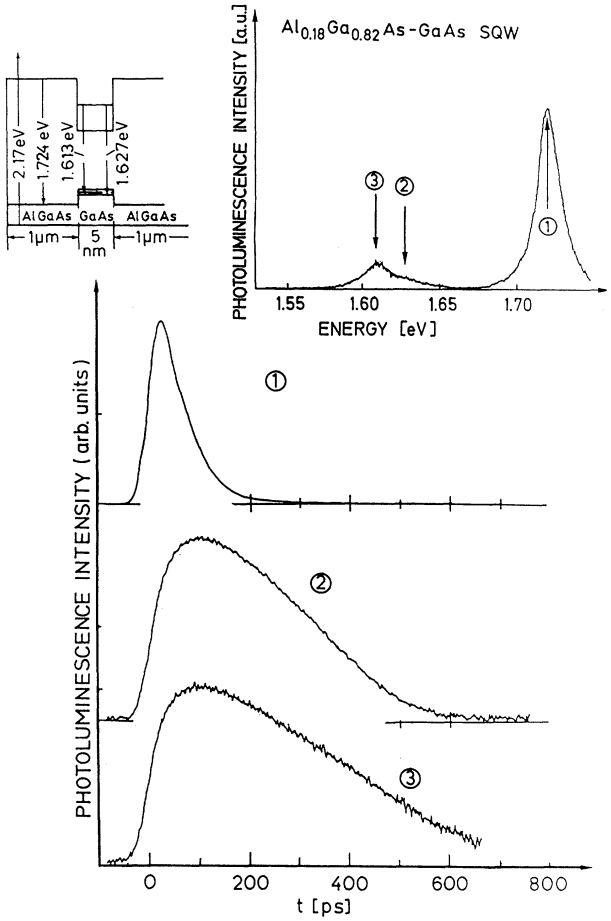

FIG. 2. Experimental luminescence data for a GaAs/$Al_xGa_{1-x}As$ single-QW structure with $L_z=5$ nm. The two arrows for the QW transitions in the schematic graph of the structure correspond to the $n=1$ transition including the heavy (1.613 eV) and light (1.627 eV) hole valence subbands.

caused by recombination. The faster decay of the $n=2$ emission with respect to the $n=1$ QW recombination reflects the faster decrease of the $n=2$ state due to relaxation and recombination.

The results obtained from a GaAs/$Al_xGa_{1-x}As$ single-QW structure with $L_z=5$ nm are shown in Fig. 2. The time-integrated spectrum reveals the $Al_xGa_{1-x}As$ luminescence at 1.724 eV as well as the $n=1$ QW transition with a maximum at 1.613 eV. The weak structure in the QW emission arises from the splitting of the $n=1$ valence subband because of heavy and light hole masses (see also the inset in Fig. 2). QW luminescence from $n=2$ states is not observed, since for $L_z=5$ nm the $n=2$ states are already located within the continuum of the $Al_xGa_{1-x}As$. The measured value of about 0.2 for the $I_{QW}/I_{Al_xGa_{1-x}As}$ ratio is again by far higher than that expected from the respective volumes for homogeneous excitation. The efficient carrier trapping required to explain this result is once more manifested in the extremely fast decay of the $Al_xGa_{1-x}As$ emission (curve 1) and in the delayed increase of the QW recombination (curves 2 and 3). The recombination in the QW itself, however, is considerably faster as compared to the QW with $L_z=14$ nm.

The experimental results are next analyzed quantitatively on the basis of a simple rate-equation model. We only consider electron kinetics because of the residual p-type doping (N_A-N_D on the order of 10^{15} cm^{-3}) of molecular-beam epitaxially grown GaAs and $Al_xGa_{1-x}As$.[10] The dynamics of the electron concentration in the $Al_xGa_{1-x}As$ cladding layers, n_{AlGaAs}, and in the QW, n_{QW}, can be described by

$$\frac{dn_{AlGaAs}}{dt}=G(t)-\frac{n_{AlGaAs}}{\tau_{tr}}-\frac{n_{AlGaAs}}{\tau_{r,AlGaAs}}, \quad (1)$$

$$\frac{dn_{QW}}{dt}=\frac{n_{AlGaAs}}{\tau_{tr}}-\frac{n_{QW}}{\tau_{r,QW}}, \quad (2)$$

where $G(t)$ is the generation rate according to the actual optical pulse and τ_{tr}, $\tau_{r,AlGaAs}$, and $\tau_{r,QW}$ are the trapping time and the carrier lifetimes in the $Al_xGa_{1-x}As$ and the QW, respectively. Direct excitation of the QW by absorption of light can be neglected because the excitation pulse is almost completely absorbed in the front $Al_xGa_{1-x}As$ layer (absorption coefficient $>10^4$ cm^{-1} in both cases) and because of the small well thickness. The trapping time τ_{tr} represents an average value and may be expressed by $\tau_{tr}\simeq d/v_d$, where d is the thickness of the front $Al_xGa_{1-x}As$ cladding layer and v_d is an average drift velocity.

Numerical solutions of Eqs. (1) and (2) are depicted in Fig. 3 together with experimental results for the samples shown in Figs. 1 and 2. The time behavior of only the low-energy QW luminescence is considered, because the high-energy emission also reflects the relaxation and cooling of the carrier system resulting in a faster decrease of the intensity (cf. Fig. 2). The increase of the respective QW luminescence can be described by the same τ_{tr} independent of photon energy. The experimental data can be well described by our model and τ_{tr} amounts to 100 and 50 ps for the QW with $L_z=14$ and 5 nm, respectively. These trapping times correspond to drift velocities of the photoexcited carriers in $Al_xGa_{1-x}As$ on the order of 10^6 cm/s, which have been observed also in other experiments.[14] The observed difference in τ_{tr} could be partly due to the different initial excess energies of the photoexcited carriers which result from the slightly different band gaps of the $Al_xGa_{1-x}As$ cladding layers in the two samples. In addition, however, the difference in the electronic levels of the QW's which are resonant with the $Al_xGa_{1-x}As$ continuum states may affect carrier trapping.

The lifetime for the carriers within the QW,

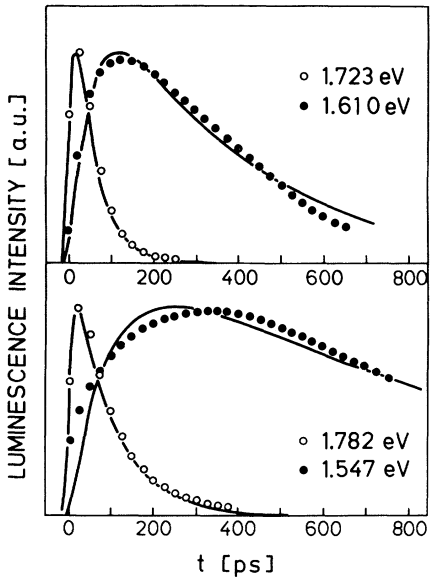

FIG. 3. Comparison of numerical data on the basis of Eqs. (1) and (2) with experimental results for the two samples shown in Figs. 1 (bottom) and 2 (top). The open and full circles correspond to the $Al_xGa_{1-x}As$ and the low-energetic QW luminescence, respectively. A value of 1 ns has been assumed for $\tau_{r,AlGaAs}$ in the calculations.

$\tau_{r,QW}$, is found to be approximately 1 ns for $L_z = 14$ nm and 350 ps for $L_z = 5$ nm. This decrease of $\tau_{r,QW}$ with decreasing L_z results from the increase of the recombination probability P_{eh} due to the increased overlap of electron and hole wave functions because of the localization. P_{eh} is proportional to a_0^{-2} for excitonic recombination in 2D systems.[5,15] The exciton radius in an ideal 2D system, i.e., $L_z \to 0$, amounts to just half of the 3D exciton radius, and a_0 increases for $L_z > 0$ as a result of the spread of the 1s exciton wave function.[6] The measured decrease in lifetime by a factor of 2.85 for the $L_z = 5$ nm QW with respect to $L_z = 14$ nm QW corresponds to a shrinkage of a_0 by a factor of 1.7,[16] which is close to the maximum value of 2, corresponding to a complete transition from a 3D to a 2D electronic system. This result provides definite experimental evidence that carriers within QW structures with $L_z \simeq 5$ nm behave almost like an ideal 2D electronic system.

In conclusion, we have directly determined the carrier trapping and the spontaneous carrier lifetimes in GaAs/$Al_xGa_{1-x}As$ QW structures by picosecond luminescence measurements. Carrier trapping into the QW is extremely efficient and requires diffusion velocities in $Al_xGa_{1-x}As$ on the order of 10^6 cm/s. The lifetime of carriers within the QW decreases with decreasing well thickness from 1 ns for $L_z = 14$ nm to 350 ps for $L_z = 5$ nm. This reduction of the lifetime reflects the increased overlap of the electron and hole wave functions in the QW and thus demonstrates the fundamental effect of carrier localization on transition probabilities.

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[11]The GaAs substrate is excited by reabsorption of the QW and $Al_xGa_{1-x}As$ luminescence, as can be concluded from its delayed temporal increase, which is determined by the decrease of the AlGaAs and QW luminescence.

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[13]For the substrate luminescence shown in Fig. 1 a decay constant of 2.5 ns is determined.

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[15]This is valid for radiative recombination, which is expected to dominate in the GaAs because the carrier concentration is rather low (the incident photon flux corresponds to an initial carrier concentration in the 1-μm-thick AlGaAs of about 5×10^{16} cm^{-3}) and furthermore Auger recombination may be even reduced with respect to 3D GaAs as a result of the modified band structure of the 2D electronic system [see, e.g., A. Sugimura, Appl. Phys. Lett. 42, 17 (1983); N. K. Dutta, J. Appl. Phys. 54, 1236 (1983)].

[16]The effect of the slightly different composition of the $Al_xGa_{1-x}As$ for the two samples on the degree of localization is of minor importance as can be shown by comparison of the respective subband energies for fixed L_z.