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Influence of exciton ionization on recombination dynamics in $In_{0.53}Ga_{0.47}As/InP$ quantum wells

P. Michler, A. Hangleiter, A. Moritz, V. Härle, and F. Scholz

4. Physikalisches Institut, Universität Stuttgart, Pfaffenwaldring 57, D-7000 Stuttgart 80, Germany

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We have investigated the carrier dynamics in $In_{0.53}Ga_{0.47}As/InP$ quantum wells with various well widths as a function of temperature and carrier density using time-resolved photoluminescence spectroscopy. At low temperatures ($T<50$ K), we find pure excitonic recombination. At elevated temperatures, two radiative recombination paths are present, and the proportion of free-carrier recombination to excitonic recombination increases as the temperature is raised. We observe a nonlinear increase of the radiative lifetime caused by thermal ionization of excitons. Exciton binding energies in the range between 9 and 15 meV and majority carrier concentrations of about 1.2×10^{10} cm^{-2} are deduced from the ratio between the high and low injection lifetimes. The exciton binding energy increases with decreasing well thicknesses, in good agreement with theoretical predictions.

The recombination of excess charge carriers in quantum wells has attracted much interest in the past.[1–7] It was found that, because of the increase of the exciton binding energy due to an increased electron-hole wavefunction overlap, exciton recombination plays a much more significant role in quantum wells (QW's) than in three-dimensional semiconductors.[8] On one hand, quite a few studies dealing with radiative recombination have been performed at temperatures below 60 K.[1,2,9] In these studies a monoexponential decay is observed and it is concluded that exciton recombination is the main decay channel under the assumption that nonradiative processes play a negligible role.

On the other hand, some experimental results[3,10,11] have been obtained at elevated temperatures ($T>200$ K). The experiments also show a monoexponetial decay of the photoluminescence (PL) intensity. The conclusion has been drawn,[12] therefore, that the lifetime is controlled by nonradiative channels at such high temperatures.

In principle, however, one expects a gradual thermal ionization of excitons with increasing temperature and consequently free-carrier recombination between quantized subbands should play an important role and possibly dominate the recombination mechanism at high temperatures. For a detailed investigation of the effect of exciton ionization on radiative recombination one has to examine the recombination kinetics in the temperature region between 50 and 150 K, since there exciton ionization processes mainly take place. Furthermore, the influence of exciton ionization on recombination in quantum wells can only be tested in samples where thermal emission of carriers into the barriers[13,14] and nonradiative processes[3] plays a negligible role even at elevated temperatures.

In this paper, we present a systematic study of the kinetics of radiative recombination in $In_{0.53}Ga_{0.47}As/InP$ quantum wells grown by metal-organic vapor phase epitaxy (MOVPE). We performed density-dependent time-resolved experiments to investigate the influence of exciton ionization on the recombination dynamics in the temperature range between 4 and 160 K. We show that a recently developed model by Ridley,[4] which takes into account the thermal equilibrium between excitons and free carriers, successfully describes the kinetics and the temperature dependence of the PL decay.

All of our samples investigated here were nominally undoped $In_{0.53}Ga_{0.47}As/InP$ multiple quantum wells (MQW's) grown by MOVPE on a (100) oriented semi-insulating InP:Fe substrate kept at a temperature of about 620 °C, with growth interruption (GRI) at both interfaces. The MQW's with 5- and 10-nm well widths consist of five periods of $In_{0.53}Ga_{0.47}As$ wells and the 3-nm MQW has ten identical QW's. All wells are separated by 30-nm InP barriers to avoid quantum-mechanical coupling between the wells.

Time-resolved measurements in the large-signal regime were performed using a synchronously mode-locked cavity-dumped dye laser (R6G) pumped by a mode-locked frequency-doubled Nd:YAG (yttrium-aluminum-garnet) laser. Excitation would therefore take place principally in the InP barriers. After excitation, a rapid and an efficient transfer of the photogenerated carriers occurs from the barriers into the wells. The carrier collection times are in the subnanosecond regime,[15] considerably shorter than the typical minority carrier lifetime in the barrier and in the quantum well. Therefore the collection process has no influence on the measurements reported below.

For measurements in the small-signal regime we used infrared pulses which have been generated in the 1.2–1.6-μm region by mixing a 1.064-μm Nd:YAG laser pulse and a visible pulse from the cavity-dumped, synchronously pumped dye laser. This made it possible to tune the excitation wavelength below the InP barrier to generate carriers only in the $In_xGa_{1-x}As$ wells. The carrier density was estimated from the excitation power density and the absorption coefficient of $In_xGa_{1-x}As$ to be about 8×10^9 cm^{-2} in the wells. Detection of the decaying PL was obtained by a Ge avalanche photodiode in a photon counting mode.[16] There is no difference in the transient behavior of the PL decay between resonant

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and nonresonant excitation, confirming the much shorter carrier collection times in comparison to the radiative recombination times.

Typical decay curves from the time-resolved PL measurements are shown in Fig. 1 for the temperature range 30–100 K. At low temperatures (T<50 K) we find in all samples an exponential decay of the photoluminescence intensity in good agreement with the kinetics of purely excitonic recombination. At higher temperatures we observe a carrier density-dependent nonexponential decay, which can be approximately resolved into two exponentials.

At least at high temperatures, one expects to find free carriers in addition to excitons. The excitons are formed from and dissociated into unbound electrons and holes by interaction with acoustic and optical phonons. Exciton formation and dissociation are extremely fast processes. Recently, the exciton ionization times have been directly measured by Wegener *et al.*,[17] using time-resolved absorption experiments. They found values of the order of 350 fs (T =150 K) for the exciton ionization time in $In_xGa_{1-x}As$ quantum wells. These exciton ionization times are much faster than those associated with radiative recombination (ns).

Therefore we can consider a thermal equilibrium between excitons and free carriers during recombination. The decay curves can be fitted extremely well by the expression (n-type material)

$$\frac{n(t)}{[n(t)+n_0]^{1-r}}=\frac{n(0)}{[n(0)+n_0]^{1-r}}\exp(-t/\tau) \qquad (1)$$

first given by Ridley[4] which follows from that. Therein, $n(t)$ is the electron density at time t, n_0 is the backround electron density, r is the exciton factor, and τ is the small-signal radiative recombination time constant in the nondegenerate regime. The factor r is given by[18]

$$r=\frac{4\phi_x n_0}{N_{cv}+2\phi_x n_0}, \qquad (2)$$

where N_{cv} is the reduced effective density of states. The exciton trapping factor ϕ_x is approximately given in the nondegenerate regime by[18]

$$\phi_x = e^{E_x/kT}, \qquad (3)$$

where E_x is the exciton binding energy, assumed to be carrier density independent in the density range under consideration below 1×10^{11} cm^{-2}.

The factor r varies between 0 and 2. At low temperatures, when $kT \ll E_x$, the factor r is nearly 2 and the decay is purely excitonic. With increasing temperature the r factor decreases which means increasing excitonic ionization processes and the PL decay can be resolved approximately into two exponentials with a low injection lifetime τ and a high injection lifetime $r\tau/2$.[4] At high temperatures, when $kT \gg E_x$, then $r\approx 0$, and the decay is predominantly via free-carrier recombination. The measured high and low injection lifetimes are shown in Fig. 2 for the 3- and 5-nm QW. At low temperatures (T<20 K), the measured lifetimes are nearly independent of temperature. This behavior of the measured lifetime has previously been reported,[19] and was explained with the dominance of localized excitons at low temperatures, since they have a temperature-independent transition probability. Another alternative interpretation is to analyze our measured lifetimes with the help of the theory developed by Feldmann, Peter, and Göbel.[1] This model

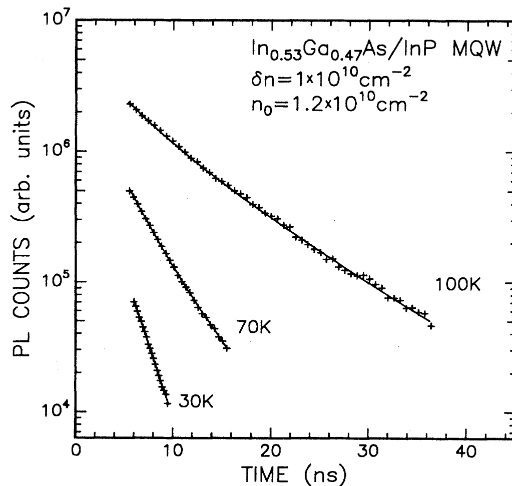

FIG. 1. Typical PL decay curves at three different temperatures for the 5-nm MQW. The solid curves represent fits as described in the text. The curves are vertically shifted and shown on a logarithmic scale.

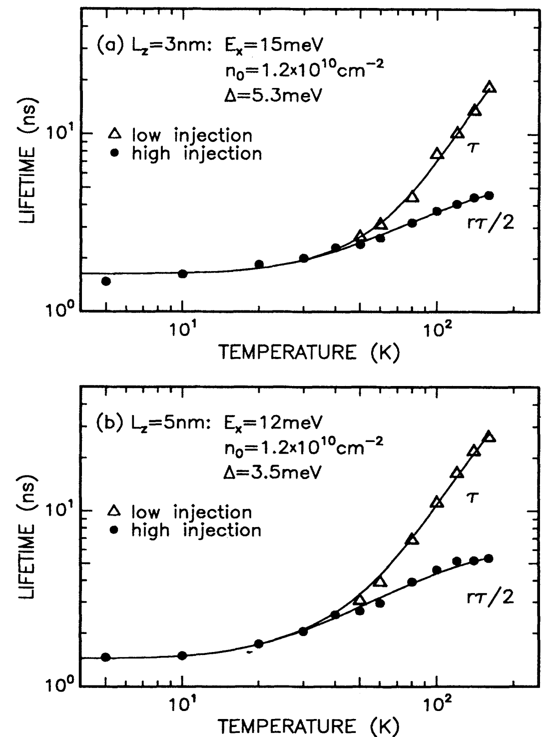

FIG. 2. Temperature dependence of the low injection lifetime τ and the high injection lifetime $r\tau/2$ for the (a) 3- and the (b) 5-nm MQW. The solid lines represent fits as described in the text.

also predicts for low temperatures $kT \ll \Delta$ an almost temperature-independent lifetime due to the homogeneous linewidth Δ. At elevated temperature ($kT \gg \Delta$), a linear increase of the excitonic lifetime is expected.

Increasing the temperature above 40 K we observe a carrier density-dependent nonexponential decay which can be approximately resolved into two exponentials with a low and a high injection lifetime. For the low injection lifetime we find a nonlinear increase with temperature caused by thermal ionization processes of excitons. The ratio between low and high injection lifetimes increases with increasing temperature in agreement with the theory mentioned above. By plotting $\ln[T/(2/r-1)]$ versus $1/T$ according to Eq. (2) the values are expected to follow a linear relationship. Nearly straight lines are obtained in the temperature range between 50 and 160 K. From the intercept with the $\ln[T/(2/r-1)]$ axis of the extrapolated straight lines we deduce the majority carrier concentration n_0, and from the slopes the exciton binding energies are calculated. It is important to note that the exciton binding energies and the majority carrier concentrations are directly derived from the measured high and low injection lifetimes with only the effective electron- and heavy-hole masses as material parameters. We use the values m_e=0.041 and m_h=0.465 for the $In_{0.53}Ga_{0.47}As$ material.[20] Furthermore, no other parameters are necessary. We deduce exciton binding energies E_x=15, 13, and 9 meV and majority carrier concentrations of about 1.2×10^{10} cm^{-2} for the 3-, 5-, and 10-nm nominal thick well widths. The value of 1.2×10^{10} cm^{-2} for the majority carrier concentration n_0 in the well seems to be too high with respect to Hall measurements on $In_{0.53}Ga_{0.47}As$ bulk material [$n_0 \approx (5-8) \times 10^{14}$ cm^{-3}]. We therefore assume that the high majority carrier concentration is caused through a spillover of carriers from the InP barriers to the well. As expected and observed earlier,[21,22] the exciton binding energy increases with decreasing well width. Our values, derived from a thermodynamic consideration based on the two-dimensional law of mass action, are somewhat larger than the calculations of Tran *et al.*,[21] who predicted maximum values less than 8 meV. However, for the thickest well, having a nominal well width of 10 nm, the exciton binding energy of approximately 9 meV corresponds closely with the value observed from thermally modulated PL (TMPL) measurements on 10-nm $In_xGa_{1-x}As$/InP wells reported by Gal *et al.*,[22] who measured a value of 7 meV. Furthermore, the maximum value of 15 meV for the nominal 3-nm QW agrees quite well with TMPL measurements of Zin *et al.*,[23] who reported a maximum excitonic binding energy of 17 meV occurring at a well width of approximately 1.5–1.8 nm.

To fit the temperature dependence of the high and low injection lifetime we apply the theory of Ridley,[4] and additionally for low temperatures (T<30 K) we take into account the finite homogeneous excitonic linewidth Δ according to ($\tau(T) \sim \Delta(T)\{1-\exp[-\Delta(T)/kT]\}^{-1}$).[1] The

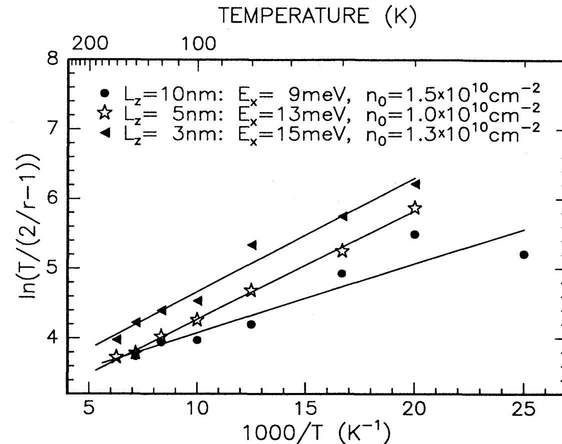

FIG. 3. Semilogarithmic plot of the factor $[T/(2/r-1)]$ as extracted from the relation between the measured high and low injection lifetimes. The straight lines are the best fits to the experimental data using Eq. (2) and correspond to values E_x =15, 13, and 9 meV.

excellent fits in Fig. 2 demonstrate the validity of this model. Taking for the electron-hole squared overlap integral $G = 1$, and ignoring photon recycling effects which is quite reasonable for our quantum wells (total thickness < 50 nm), we obtain fit parameters E_x and n_0 (see Fig. 2) which are consistent with the previously determined values (see Fig. 3). Furthermore, we obtain the following values for the homogeneous linewidth Δ=5, 3.5, and 2.5 meV for the 3-, 5-, and 10-nm-thick QW's, respectively. If we compare these values with the PL-linewidth values of 30, 18, and 9 meV for the 3-, 5-, and 10-nm-thick QW's, the conclusion can be drawn that the line broadening is mainly due to inhomogeneous effects in these samples.

In summary, we have presented time-resolved PL measurements demonstrating the importance of exciton ionization processes on recombination dynamics in $In_{0.53}Ga_{0.47}As$/InP quantum wells. Our experiments show for temperatures above 40 K a carrier density-dependent nonexponential decay, which can be approximately resolved into two exponentials. This can be explained by a recently developed model by Ridley[4] which is based on the law of mass action for describing the thermal equilibrium between excitons and free carriers. Furthermore, we have extracted exciton binding energies from the ratio of the measured high and low injection lifetimes. We find that the binding energy increases with decreasing well thickness in agreement with theoretical predictions.

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