Recombination dynamics in pseudomorphic and partially relaxed $In_{0.23}Ga_{0.77}As/GaAs$ quantum wells

M. Grundmann and D. Bimberg
Institut für Festkörperphysik I, Technische Universität Berlin, Hardenbergstrasse 36, D-1000 Berlin 12, Federal Republic of Germany

A. Fischer-Colbrie and J. N. Miller
Hewlett Packard Laboratories, Palo Alto, California 94304
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A systematic investigation of the recombination dynamics of excess charge carriers in $In_{0.23}Ga_{0.77}As/GaAs$ single quantum wells as a function of well width covering the range from the pseudomorphic to the partially relaxed regime reveals for the first time the impact of misfit dislocations on carrier decay times. At dislocation densities $n_L = 5\times10^5\ m^{-1}$ the recombination changes its character from predominantly radiative (excitonic) to nonradiative. A diffusion model describes quantitatively the observations and full agreement is obtained with independent determination of the quantum efficiency. Excitonic, (e, A^0), and (D^0, A^0) recombination times are determined. The intrinsic ambipolar diffusion length is observed to be 0.5 μm, indicating a huge hole mobility of 1400 $cm^2/V\,s$ at 10 K. Time-resolved cathodoluminescence provides the experimental results.

The $Al_xGa_{1-x}As/GaAs$ material system has played a key role in the development of semiconductor heteroepitaxy because it is nearly lattice matched for all x. Fundamental breakthroughs in two areas were achieved with this system.

(1) A large number of novel electronic and photonic heterostructure devices has been designed with performance superior to homoepitaxial devices.[1,2]

(2) A basic understanding of heteroepitaxial growth processes and the physics of quantum wells has been obtained.[3,4]

Presently $In_xGa_{1-x}As/GaAs$ plays a role similar to that of a model system for the heteroepitaxy of lattice-mismatched materials. By means of this system the performance of a variety of devices can be dramatically improved[5–7] and we learn about the basic mechanisms ruling the growth of lattice-mismatched layers[8,9] and the physics of strain relaxation.[10–12] Actual device structures include layers with thickness in the pseudomorphic regime as well as layers exceeding the critical thickness.

In particular for *lattice-matched* $Al_xGa_{1-x}As/GaAs$ (Refs. 13 and 14) and to a lesser extent for $In_{0.53}Ga_{0.47}As/InP$ (Refs. 15 and 16) *quantum wells* (QW's) the *recombination dynamics* has been carefully investigated; the excitonic lifetime shows a strong increase upon increasing QW width L_z due to decreased overlap of the excitonic wave function. For the $In_xGa_{1-x}As/GaAs$ system, to our knowledge, no reports on this subject exist although the recombination dynamics is of essential importance for the understanding of optical device performance. Only the dynamics of carrier *capture into* $In_xGa_{1-x}As/GaAs$ QW's has been investigated in Ref. 17. In this paper we present the first investigation of recombination dynamics in *pseudomorphic* and *partially relaxed* QW's. Low-temperature recombination lifetimes for $In_xGa_{1-x}As/GaAs$ single QW's are reported and the large influence of misfit dislocations on the recombination dynamics is assessed. With increasing dislocation density the character of the recombination is observed to change from purely radiative to increasingly nonradiative.

Our samples are grown by molecular-beam epitaxy (MBE) and consist of a semi-insulating GaAs substrate, a 1500-Å undoped GaAs buffer layer, a single QW of $In_xGa_{1-x}As$ ($x = 0.23$) with thickness L_z [$L_z = 16, 22, 27, 32, 38$ nm for samples No. 1 (B1309), No. 2 (B1310), No. 3 (B1311), No. 4 (B1312), and No. 5 (B1313), respectively], and a 50-nm cap layer of GaAs. The growth temperature of the $In_xGa_{1-x}As$ layer was 515 °C. Further details on the growth procedure can be found elsewhere.[18,19]

In our time-resolved cathodoluminescence (CL) experiment the sample is excited by 50-ns electron pulses, long enough so that a quasiequilibrium of the carriers is reached. The electron beam is switched off with a decay time of less than 200 ps (Ref. 20) and the luminescence decay is measured by means of a temperature-stabilized and Peltier-cooled Si avalanche photodiode (RCA C30902S, $\lambda \le 1100$ nm) and time-correlated single-photon counting. The time resolution of the whole setup is 350 ps. The acceleration voltage is $U = 30$ kV and a typical beam current is 1 nA. At low dislocation densities ($n_L \le 10^5\ m^{-1}$) the transients are measured with the electron beam focused between single dislocations (spot mode). In samples with higher n_L the excitation volume of about 4 μm (Ref. 21) averages over clusters of dark line defects and brighter areas in between.

In Fig. 1 we compare transients of samples No. 1, No.

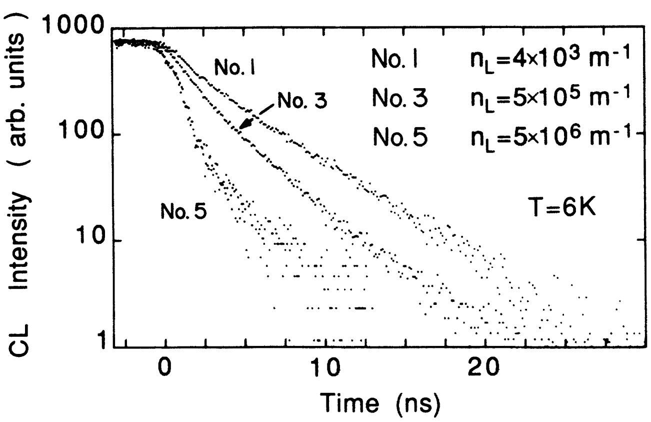

FIG. 1. CL transients at low temperature for samples with different dislocation density n_L. Increasing n_L leads to faster decay.

3, and No. 5 with $L_z = 16$, 27, and 38 nm, respectively, at $T = 6$ K. The results for samples No. 2 and No. 4, with $L_z = 22$ and 32 nm are not shown, for clarity. It is evident that with increasing L_z, i.e., with increasing dislocation density n_L, the decay becomes much faster. The transients can be almost perfectly fitted by a sum of two exponentials with time constants τ_1 and τ_2, which are plotted in Fig. 2(c) versus L_z. The misfit dislocations generated at the heterointerface when the layer thickness exceeds the critical thickness are centers of *locally* reduced quantum efficiency and *locally* reduced excess charge carrier lifetime.[19] With increasing dislocation density the *local* reduction of quantum efficiency causes a reduction of the *total* quantum efficiency η. A diffusion model[19] gives the theoretical dependence

$$\eta = 1 - 2Ln_L \tanh[1/(2Ln_L)] , \quad (1)$$

L being the ambipolar diffusion length. The measured misfit dislocation density n_L is plotted in Fig. 2(a) as a function of L_z. In Fig. 2(b) the experimental total quantum efficiency and its prediction according to Eq. (1) and a diffusion length $L = 0.5$ μm are compared and found to agree perfectly. The decrease of quantum efficiency with increasing L_z predominates the L_z dependence of the transients at larger well widths. The high quantum efficiency on the 16- and 22-nm QW's makes it straightforward to identify the decay constant τ_1 with the radiative excitonic $[X(e,\mathrm{hh}), n = 1]$ lifetime τ_r. The excitonic lifetime in the thicker well is slightly larger, as expected from the theory of excitonic oscillator strength in QW's.[22] The values of about 2 ns compare well to values found for lattice-matched systems like $Al_xGa_{1-x}As$/GaAs (Refs. 13 and 14) and $In_{0.53}Ga_{0.47}As$/InP (Ref. 16) at low temperatures and provide further evidence for the high quality of our samples in addition to the large diffusion length. The time constant τ_2 is given by (e, A^0) recombination as will be discussed below. Figure 2(c) shows that for thicker wells ($L_z \geq 27$ nm) the decay constants decrease strongly due to the increasing importance

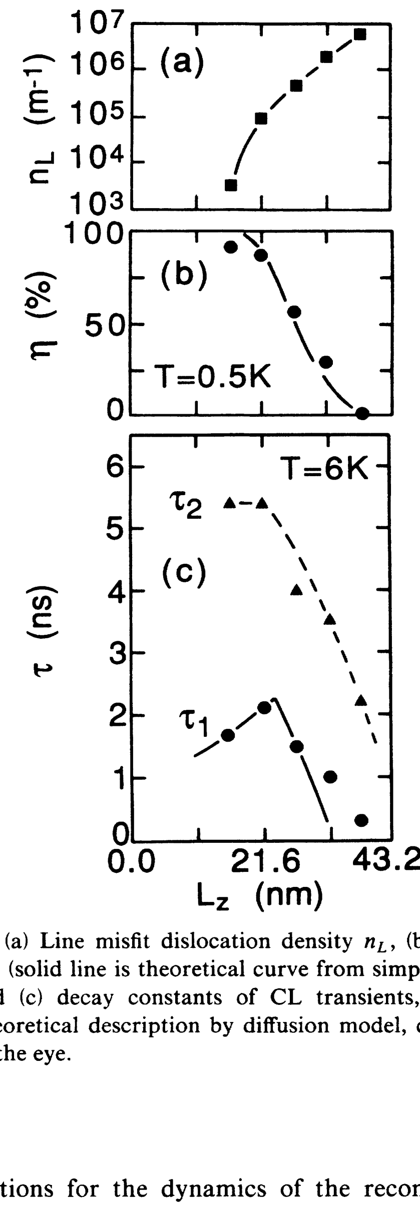

FIG. 2. (a) Line misfit dislocation density n_L, (b) quantum efficiency η (solid line is theoretical curve from simple diffusion model) and (c) decay constants of CL transients, solid line denotes theoretical description by diffusion model, dashed line is guide to the eye.

of dislocations for the dynamics of the recombination. To evaluate their influence in more detail now we use our simple diffusion model treating the dislocation lines at an average distance $d = 1/n_L$ as fast nonradiative recombination centers.[19] The rate equation including diffusion for an excess charge carrier density n is

$$\frac{\partial n(x,t)}{\partial t} = -\frac{n(x,t)}{\tau_{\mathrm{tot}}} + D\frac{\partial^2 n(x,t)}{\partial x^2} , \quad (2)$$

D being the diffusion coefficient and τ_{tot} the total recombination lifetime given by the radiative lifetime τ_r at high quantum efficiency. With $\tau_r = 2$ ns and $L = 0.5$ μm determined above, an ambipolar diffusion coefficient $D = L^2/\tau_r = 1.2$ cm^2/s is obtained. The ambipolar mobility which is dominated by the hole mobility at $T = 10$ K is $\mu = De/kT = 1.4 \times 10^3$ cm^2/V s, a surprisingly high value.

The recombination dynamics is well described by an approximate solution of Eq. (2) found by development into a Fourier series and taking only the leading term into account. The actually observed decay time τ is then given by

$$\frac{1}{\tau}=\frac{1}{\tau_r}+\frac{1}{\tau_D}=\frac{1}{\tau_r}[1+(\pi L n_L)^2] . \quad (3)$$

$\tau_D=1/[D(\pi n_L)^2]$ is a lifetime caused by the presence of misfit dislocations. Thus for low dislocation density ($Ln_L \ll 1$) the decay time is given by the radiative excitonic lifetime. At high dislocation densities ($Ln_L \gg 1$) the recombination dynamics is dominated by τ_D describing the diffusion into dislocations and fast nonradiative decay there. It is expected from Eq. (3) that τ_D is equal to τ_r (i.e., 50% quantum efficiency) for $n_L=6.4\times10^5$ m^{-1}. The 27-nm QW, which is the thinnest QW showing a drop of lifetime, has exactly 50% quantum efficiency, and the experimentally observed dislocation density is $n_L=5.1\times10^5$ m^{-1} in very good agreement with the prediction above. The dislocations in the QW's thinner than 27 nm are not expected to deteriorate the lifetime in agreement with experiment. The theoretical L_z dependence of the dominating recombination time constant τ_1 including the variation of excitonic oscillator strength and the influence of the misfit dislocations is shown in Fig. 2(c) as a solid line. The decay time changes its character from radiative ($n_L \le 10^5$ m^{-1}) to increasingly nonradiative ($n_L \ge 10^6$ m^{-1}).

Using long signal integration times and taking advantage of the high dynamics of single photon counting, we found that the transients of the almost unrelaxed QW's with $L_z=16$ and 22 nm exhibit a third nonexponential decay. As an example the transient of the 22-nm QW is shown in Fig. 3 on a much larger time scale than that of Fig. 1. This decay is governed by the (D^0,A^0) recombination. The best possible fit with a single exponential is shown in Fig. 3 to be clearly unsatisfactory. The assignment of τ_2 to the (e,A^0) and the third decay to the (D^0,A^0) process are based upon a detailed discussion of the time-delayed spectra presented below.

Observation of the thermalization of the excess carriers measured by means of time-delayed CL spectra gives further insight into the recombination dynamics. Experimentally several time windows are set into the transient at different delay times Δt with respect to the end of the electron pulse and spectra are taken for all these windows simultaneously. In Fig. 4 the time evolution of the spectrum of the $L_z=16$ nm QW is shown. The evolution includes several steps.

(1) Carriers relax from higher subbands.

(2) The spectral maximum of the $X(e,hh),n=1$ transition shifts slightly to lower energies indicating the thermalization of carriers within the inhomogeneous spectral broadening.

(3) At delay times longer than 4 ns the (e,A^0) transition completely dominates the spectrum. Since the material is, however, p type in character, free electrons are captured by both free and bound holes and ionized donors.

(4) Finally the (D^0,A^0) recombination remains causing the clearly nonexponential decay as visualized in Fig. 3.

This behavior is rather three dimensional in character, similar to what has been reported by Christen *et al.*[13] for an 11-nm $Al_xGa_{1-x}As/GaAs$ multiple QW. The peak assignments and the peak positions denoted by arrows in Fig. 4 stem from photoluminescence and absorption investigations.[23] The acceptor binding energy E_b^A in the QW is 15 meV, which agrees well with the theoretical value of 16 meV.[24] The capture cross section $\sigma(e,A^0)=(\tau_2 v_{th} p)^{-1}$ of free electrons onto neutral acceptors can be calculated from the (e,A^0) recombination

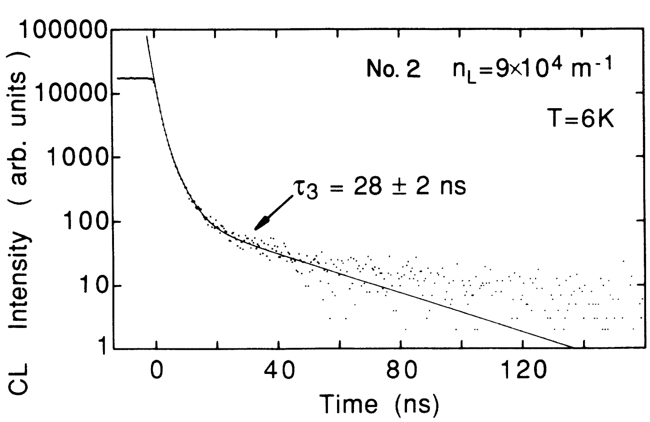

FIG. 3. CL transient of $L_z=16$ nm QW on larger time scale showing a long decay time τ_3 attributed to extrinsic (D^0,A^0) recombination. Solid line is χ^2 fit.

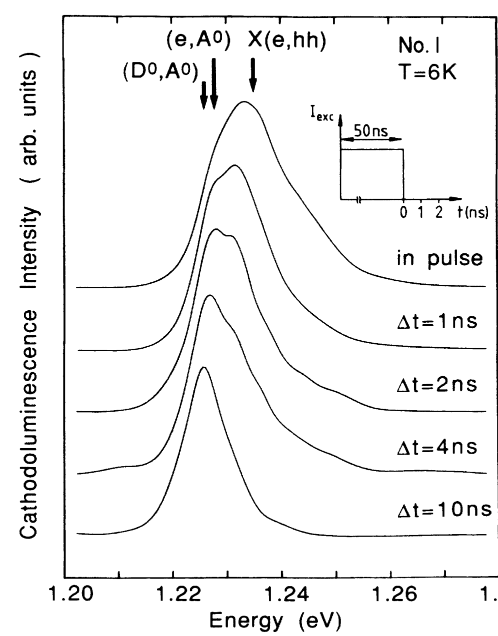

FIG. 4. Time-delayed spectra (scaled to same height) of 16-nm QW, showing thermalization of carriers. At long delay time Δt the (D^0,A^0) recombination dominates the spectrum. The inset displays schematically the excitation pulse.

time constant $\tau_2=5.4\pm0.2$ ns and the experimentally determined background (p-type) impurity concentration $p=5\times10^{14}$ cm^{-3}. $v_{\rm th}=(2kT/m_e)^{0.5}$ is the thermal electron velocity; assuming a mean electron temperature of 10 K and $m_e=0.055m_0$ we obtain $v_{\rm th}=7.4\times10^6$ cm/s. Then the capture cross section is $\sigma(e,A^0)=5\times10^{-14}$ cm^2.

In summary we have presented a systematic investigation of recombination dynamics in partially relaxed $In_{0.23}Ga_{0.77}As$/GaAs QW's. The character of recombination changes from radiative to nonradiative upon increase of misfit dislocation density. The threshold is at L_z values appreciably larger than the transition from pseudomorphic to partially relaxed QW's. The thermalization of carriers has been visualized by time-delayed spectra.

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