Radiatively controlled lifetimes in $\text{Al}_x\text{Ga}_{1-x}\text{As}$ grown by metalorganic vapor phase epitaxy

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We report long minority charge carrier lifetimes in $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{Al}_y\text{Ga}_{1-y}\text{As}$ ($x < y$) double heterostructures grown by metalorganic vapor phase epitaxy. The lifetime ($\tau_{\text{bulk}} = 8.8 \mu\text{s}$) is found to be controlled by radiative processes in samples with aluminum concentrations of $x = 0.10$ for the active regime and $y = 0.20$ for the cladding layers; an extremely low interface recombination velocity of $S \approx 6.5 \text{ cm/s}$ is found. At higher aluminum concentrations both the bulk and interface recombination rate increase rapidly.

The role of $\text{Al}_x\text{Ga}_{1-x}\text{As}$ in minority-carrier devices such as light emitting diodes¹ and multijunction solar cells² is well established nowadays. The minority-carrier lifetime is an important parameter in the optimization of the performance of these optoelectronic devices. Previous studies³ indicated that the lifetime in $\text{Al}_x\text{Ga}_{1-x}\text{As}$ was governed by nonradiative Shockley–Read–Hall (SRH)^{4,5} processes both in the bulk material and at the interfaces. In these processes carriers are captured at deep levels such as impurities and defects. It was found that both the bulk lifetime and the interface recombination velocity (S) increased with increasing aluminum concentration x .^{6–8} Oxygen is the dominating recombination center in $\text{Al}_x\text{Ga}_{1-x}\text{As}$ and, because it strongly binds to aluminum, an increase in x is believed to cause a higher concentration of the recombination centers.⁹

The minority-carrier lifetime in DHs is calculated via the time-dependent continuity equation. If it is assumed that the recombination velocity is the same at both interfaces, the measured PL lifetime (τ_{PL}) can be well approximated by¹⁰

$$\frac{1}{\tau_{\text{PL}}} = \frac{1}{\tau_{\text{rad}}} + \frac{1}{\tau_{\text{SRH}}} + \frac{2S}{d}, \quad (1)$$

where τ_{rad} is the radiative lifetime, τ_{SRH} the lifetime formed by the recombinations at the deep levels, and d the thickness of the active layer. The radiative lifetime is inversely proportional to the majority carrier density N_{maj} under conditions of low excitation density,¹¹

$$\tau_{\text{rad}} = \frac{1}{BN_{\text{maj}}}, \quad (2)$$

where B is the radiative recombination coefficient. By measuring the decay time for DHs with different active layer thicknesses both τ_{SRH} and S can be obtained.

Time-resolved photoluminescence (TRPL) experiments are usually carried out at low-level excitation conditions ($n_{\text{exc}} \ll N_{\text{maj}}$) to avoid bimolecular recombination and to ensure that only the minority carrier lifetime is determined.¹²

All photoluminescence decay times were measured by the time-correlated single photon counting technique (TCSPC).¹³ A dye laser on Rhodamine 6G was pumped by a mode-locked Ar^+ laser and produced 6 ps excitation pulses ($\lambda = 600 \text{ nm}$) at a variable repetition frequency. The recom-

binning photons were counted by a Hamamatsu R943-02 photomultiplier, the signal of which was fed into a multichannel analyzer (MCA).

The DHs were grown on Si-doped (100) 2° off towards (110) oriented GaAs substrates by metalorganic vapor phase epitaxy at a total gas flow of seven standard liters per minute and at a pressure of 20 mbar. The source gases were trimethylgallium, arsine, and trimethylaluminum. Hydrogen, which was purified by a palladium cell, was used as a carrier gas. The flow ratio of group III to group V species was 125 and the growth rate was $1.7 \mu\text{m/h}$. The samples were grown at a fairly high temperature of $T = 720^\circ\text{C}$ because the concentration of oxygen-related defects is strongly reduced at this temperature, whereas a further increase in temperature would cause interdiffusion problems.^{9,14,15}

Two series of DHs with different values for the thickness of the active layer were grown: one with $x = 0.10$ and $y = 0.20$, and one with $x = 0.23$ and $y = 0.60$. All barrier layers were $0.6 \mu\text{m}$ thick. The quality of the active and barrier layers was checked by photoluminescence (PL) measurements, performed at $T = 4 \text{ K}$.

All layers were nominally undoped. Residual doping concentrations in the active $\text{Al}_x\text{Ga}_{1-x}\text{As}$ layers were $N_D = 5 \times 10^{15} \text{ cm}^{-3}$ as determined from capacitance-voltage measurements.

Figure 1 shows a 4 K PL spectrum of the $\text{Al}_{0.10}\text{Ga}_{0.90}\text{As}$ active layer recorded at an excitation density of $P = 2.6 \text{ W cm}^{-2}$. At 1.63 eV a carbon related luminescence peak can be seen. The luminescence at 1.657 eV is due to bound exciton transitions. The intensity ratio of the carbon related emission to the exciton emission is $\approx \frac{1}{6}$, indicative of good material quality.¹⁶

In Fig. 2 a plot is shown of the inverse active layer thickness against the inverse lifetime for the $\text{Al}_{0.10}\text{Ga}_{0.90}\text{As}$ DHs. A similar plot for the $\text{Al}_{0.23}\text{Ga}_{0.77}\text{As}$ DHs is given in Fig. 3. From the slope of the line through the data points in Fig. 2, an interface recombination velocity of $S = 6.5 \text{ cm/s}$ can be extracted. The intercept of the slope with the τ_{PL}^{-1} axis gives a bulk lifetime of $\tau = 8.8 \mu\text{s}$.

Estimates of B for $\text{Al}_x\text{Ga}_{1-x}\text{As}$ indicate that $B_{\text{AlGaAs}} \ll B_{\text{GaAs}}$.¹⁷ Because $B_{\text{GaAs}} \approx 1.4 \times 10^{-10} \text{ cm}^3/\text{s}$ at 300 K, it follows from Eq. (2) that $\tau_{\text{rad}}(\text{AlGaAs}) \geq 10 \mu\text{s}$ at a doping level of $N_D = 5 \times 10^{15} \text{ cm}^{-3}$. Hence, the lifetime in

FIG. 1. Typical 4 K-PL spectrum of $\text{Al}_{0.10}\text{Ga}_{0.90}\text{As}$ active layer ($d=0.2\ \mu\text{m}$).

these samples is dominated by radiative transitions. To our knowledge this is the first time that radiatively controlled decay times have been reported for $\text{Al}_x\text{Ga}_{1-x}\text{As}$.³ Furthermore, the value of the interface recombination velocity $S=6.5\ \text{cm/s}$ is unprecedentedly low and comparable with the best value found for the $\text{GaAs}/\text{GaInP}_2$ interface: $S \leq 1.5\ \text{cm/s}$.¹⁸

Significantly lower lifetimes were measured for the $\text{Al}_{0.23}\text{Ga}_{0.77}\text{As}/\text{Al}_{0.60}\text{Ga}_{0.40}\text{As}$ DHs, as is shown in Fig. 3. The bulk lifetime of $\tau=17\ \text{ns}$ indicates that the decay in these samples is dominated by recombinations at deep levels. This is confirmed by the higher interface recombination velocity

FIG. 3. τ_{PL}^{-1} vs d^{-1} for the $\text{Al}_{0.23}\text{Ga}_{0.77}\text{As}/\text{Al}_{0.60}\text{Ga}_{0.40}\text{As}$ double heterostructures measured at room temperature. The solid line represents a best fit to the data points using Eq. (1).

of $S=2.3 \times 10^3\ \text{cm/s}$ and by measurements performed at higher excitation density, where a pronounced majority-charge carrier lifetime was seen in the decay curve. This higher S value is expected to be correlated to the high aluminum concentrations in the cladding layers. Because $\tau_{\text{SRH}} \propto 1/N_t$, with N_t the trap density, a decrease of a factor of approximately two in lifetime as compared to the $\text{Al}_{0.10}\text{Ga}_{0.90}\text{As}$ DHs is expected if oxygen is the only recombination center in these samples. As τ_{SRH} is about 500 times lower we attribute the decrease in lifetime to the formation of DX centers in $\text{Al}_{0.23}\text{Ga}_{0.77}\text{As}$. However, the decay times in the $\text{Al}_{0.23}\text{Ga}_{0.77}\text{As}$ DHs can still compare with the best values in literature for $\text{Al}_x\text{Ga}_{1-x}\text{As}$ ($x=0.23$) and the interface recombination velocity is comparable with typical values of S for $\text{GaAs}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ heterojunctions.^{3,19}

In conclusion, we report extremely high bulk lifetimes and low interface recombination velocities S in $\text{Al}_x\text{Ga}_{1-x}\text{As}$. In the $\text{Al}_{0.10}\text{Ga}_{0.90}\text{As}$ DHs the recombination is for the first time found to be radiatively controlled; S was as low as $6.5\ \text{cm/s}$. In the $\text{Al}_{0.23}\text{Ga}_{0.77}\text{As}$ structures the nonradiative recombination becomes more dominant. There is strong indication that not the oxygen recombination centers but the formation of DX centers is responsible for the sharp decrease in the lifetime for these samples. The bulk lifetime in $\text{Al}_{0.23}\text{Ga}_{0.77}\text{As}$ is still among the highest reported for this material. The S of the $\text{Al}_{0.23}\text{Ga}_{0.77}\text{As}-\text{Al}_{0.60}\text{Ga}_{0.40}\text{As}$ interface can compare with typical values found for $\text{GaAs}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ interfaces.

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FIG. 2. τ_{PL}^{-1} vs d^{-1} for the $\text{Al}_{0.10}\text{Ga}_{0.90}\text{As}/\text{Al}_{0.20}\text{Ga}_{0.80}\text{As}$ double heterostructures measured at room temperature. The solid line represents a best fit to the data points using Eq. (1).

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