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

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We report long minority charge carrier lifetimes in Al$_x$Ga$_{1-x}$As/Al$_x$Ga$_{1-y}$As ($x<y$) double heterostructures grown by metalorganic vapor phase epitaxy. The lifetime ($\tau_{\text{bulk}}=8.8$ $\mu$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 $S=6.5$ cm/s is found. At higher aluminum concentrations both the bulk and interface recombination rate increase rapidly.

The role of Al$_x$Ga$_{1-x}$As in minority-carrier devices such as light emitting diodes and multijunction solar cells is well established nowadays. The minority-carrier lifetime is an important parameter in the optimization of the performance of these optoelectronic devices. Previous studies indicated that the lifetime in Al$_x$Ga$_{1-x}$As was governed by nonradiative Shockley–Read–Hall (SRH) processes both in the bulk material and at the interfaces. It was found that both the bulk lifetime and the interface recombination velocity ($S$) increased with increasing aluminum concentration $x$. Oxygen is the dominating recombination center in Al$_x$Ga$_{1-x}$As and, because it strongly binds to aluminum, an increase in $x$ is believed to cause a higher concentration of the recombination centers.

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 ($\tau_{\text{PL}}$) can be well approximated by

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

where $\tau_{\text{rad}}$ is the radiative lifetime, $\tau_{\text{SRH}}$ 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 $N_{\text{maj}}$ under conditions of low excitation density,

$$\tau_{\text{rad}} = \frac{1}{BN_{\text{maj}}},$$

where $B$ is the radiative recombination coefficient. By measuring the decay time for DIIs with different active layer thicknesses both $\tau_{\text{SRH}}$ and $S$ can be obtained.

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

All photoluminescence decay times were measured by the time-correlated single photon counting technique (TCSPC). A dye laser on Rhodamine 6G was pumped by a mode-locked Nd laser and produced 6 ps excitation pulses ($\lambda=600$ nm) at a variable repetition frequency. The recombinating 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) $2^\circ$ 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 $\mu$m/h. The samples were grown at a fairly high temperature of $T=720^\circ$C because the concentration of oxygen-related defects is strongly reduced at this temperature, whereas a further increase in temperature would cause interdiffusion problems.

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 $\mu$m thick. The quality of the active and barrier layers was checked by photoluminescence (PL) measurements, performed at $T=4$ K. All layers were nominally undoped. Residual doping concentrations in the active Al$_x$Ga$_{1-x}$As layers were $N_D=5\times10^{16}$ cm$^{-3}$ as determined from capacitance-voltage measurements.

Figure 1 shows a 4 K PL spectrum of the Al$_{0.10}$Ga$_{0.90}$As active layer recorded at an excitation density of $P=2.6$ 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 $\approx 0.5$, indicative of good material quality.

In Fig. 2 a plot is shown of the inverse active layer thickness against the inverse lifetime for the Al$_{0.10}$Ga$_{0.90}$As DHs. A similar plot for the Al$_{0.22}$Ga$_{0.78}$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 $S=6.5$ cm/s can be extracted. The intercept of the slope with the $\tau_{\text{PL}}$ axis gives a bulk lifetime of $\tau=8.8$ $\mu$s.

Estimates of $B$ for Al$_x$Ga$_{1-x}$As indicate that $B_{\text{AlGaAs}}<B_{\text{GaAs}}$. Because $B_{\text{GaAs}}=1.4\times10^{-10}$ cm$^3$/s at 300 K, it follows from Eq. (2) that $\tau_{\text{rad}}$ (AlGaAs)$\approx 10$ $\mu$s at a doping level of $N_D=5\times10^{15}$ cm$^{-3}$. Hence, the lifetime in

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

Significantly lower lifetimes were measured for the Al$_{0.23}$Ga$_{0.77}$As/Al$_{0.60}$Ga$_{0.40}$As DHs, as is shown in Fig. 3. The bulk lifetime of $\tau=17$ ns indicates that the decay in these samples is dominated by recombinations at deep levels. This is confirmed by the higher interface recombination velocity of $S=2.3\times10^3$ 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 $\tau_{\text{SRH}}\approx1/N_i$ with $N_i$ the trap density, a decrease of a factor of approximately two in lifetime as compared to the Al$_{0.10}$Ga$_{0.90}$As DHs is expected if oxygen is the only recombination center in these samples. As $\tau_{\text{SRH}}$ is about 500 times lower we attribute the decrease in lifetime to the formation of DX centers in Al$_{0.23}$Ga$_{0.77}$As. However, the decay times in the Al$_{0.25}$Ga$_{0.75}$As DHs can still compare with the best values in literature for Al$_x$Ga$_{1-x}$As ($x=0.23$) and the interface recombination velocity is comparable with typical values of $S$ for GaAs/Al$_x$Ga$_{1-x}$As heterojunctions.\(^\text{3,19}\)

In conclusion, we report extremely high bulk lifetimes and low interface recombination velocities $S$ in Al$_{0.10}$Ga$_{0.90}$As. In the Al$_{0.10}$Ga$_{0.90}$As DHs the recombination is for the first time found to be radiatively controlled; $S$ was as low as 6.5 cm/s. In the Al$_{0.23}$Ga$_{0.77}$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 Al$_{0.23}$Ga$_{0.77}$As is still among the highest reported for this material. The $S$ of the Al$_{0.23}$Ga$_{0.77}$As/Al$_{0.60}$Ga$_{0.40}$As interface can compare with typical values found for GaAs/Al$_{0.10}$Ga$_{0.90}$As interfaces.

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\begin{figure}[h]
\centering\includegraphics[width=\textwidth]{fig1.png}
\caption{Typical 4 K-PL spectrum of Al$_{0.10}$Ga$_{0.90}$As active layer ($d=0.2$ \textmu m).}
\end{figure}

\begin{figure}[h]
\centering\includegraphics[width=\textwidth]{fig2.png}
\caption{$\tau_{\text{PL}}^2$ vs $d^{-1}$ for the Al$_{0.10}$Ga$_{0.90}$As/Al$_{0.23}$Ga$_{0.77}$As double heterostructures measured at room temperature. The solid line represents a best fit to the data points using Eq. (1).}
\end{figure}

\begin{figure}[h]
\centering\includegraphics[width=\textwidth]{fig3.png}
\caption{$\tau_{\text{PL}}^2$ vs $d^{-1}$ for the Al$_{0.23}$Ga$_{0.77}$As/Al$_{0.60}$Ga$_{0.40}$As double heterostructures measured at room temperature. The solid line represents a best fit to the data points using Eq. (1).}
\end{figure}

\begin{figure}[h]
\centering\includegraphics[width=\textwidth]{fig4.png}
\caption{$\tau_{\text{PL}}^2$ vs $d^{-1}$ for the Al$_{0.23}$Ga$_{0.77}$As/Al$_{0.60}$Ga$_{0.40}$As double heterostructures measured at room temperature. The solid line represents a best fit to the data points using Eq. (1).}
\end{figure}

\begin{figure}[h]
\centering\includegraphics[width=\textwidth]{fig5.png}
\caption{$\tau_{\text{PL}}^2$ vs $d^{-1}$ for the Al$_{0.23}$Ga$_{0.77}$As/Al$_{0.60}$Ga$_{0.40}$As double heterostructures measured at room temperature. The solid line represents a best fit to the data points using Eq. (1).}
\end{figure}


