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Microsecond lifetimes and low interface recombination velocities in moderately doped *n*-GaAs thin films

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We have observed lifetimes greater than 1 μs in moderately doped, thin film, *n*-GaAs/$Al_{0.3}Ga_{0.7}As$ double heterostructure membranes formed by etching away the substrate. We attribute these ultralong lifetimes to enhanced photon recycling caused by the removal of the substrate. Nonradiative recombination in the bulk and at the interfaces is very low; the upper limit of the interface recombination velocity is 25 cm/s. Such long lifetimes in GaAs doped at $N_D = 1.3 \times 10^{17}$ cm^{-3} suggest that thin-film solar cells offer a potential option for achieving very high efficiencies.

Thin-film GaAs solar cells, where the active volume is separated from the substrate, offer a potential method of achieving a 35% efficient solar cell,[1] a priority of the photovoltaic community. Such high efficiencies in GaAs require ultralong lifetimes which have thus far been achieved only in nominally undoped material unsuitable for bipolar devices.[2–4] For moderately doped GaAs, however, lifetimes on the order of 1 μs are theoretically possible if photon recycling is enhanced by removing the light-absorbing substrate.[5]

We report here our observation of lifetimes greater than 1 μs in moderately doped, thin film, *n*-GaAs double heterostructure membranes obtained by removing the substrate by chemical etching. We attribute our ultralong lifetimes to enhanced photon recycling.[3,5–8] We also demonstrate high-quality interfaces, with the upper limit of the recombination velocity being 25 cm/s and a more accurate estimate being $S < 12$ cm/s.

Figure 1 shows the basic structure of the double heterostructures (DHs) grown for this study. Five DHs with GaAs active layer thicknesses ranging from 0.25 to 10 μm were grown at 740 °C by metalorganic chemical vapor deposition in a horizontal reactor. The *n*-GaAs active layers were doped with selenium to a concentration of $N_D = 1.3 \times 10^{17}$ cm^{-3}. $Al_{0.3}Ga_{0.7}As$ cladding layers, doped to $N_D = 2 \times 10^{18}$ cm^{-3}, provide surface passivation and carrier confinement, and the $Al_{0.85}Ga_{0.15}As$ layer acts as an etch stop.[9] An area of the substrate typically 2–3 mm in diameter was removed by wet etching using granular citric acid dissolved in deionized water and H_2O_2.[9,10] The $Al_{0.85}Ga_{0.15}As$ was then removed with hydrofluoric acid diluted to 5% by volume with deionized water. The DH membrane was therefore still attached at its periphery to the substrate which was easily handled by tweezers. As depicted in Fig. 1, photoluminescence (PL) decay was observed both on the membrane area and on the adjacent region where the substrate remained intact. PL decays were observed using time-correlated single-photon counting techniques described in more detail previously.[11,12] The illumination source was a 10 ps full width at half maximum, mode locked laser focused to a 0.8 mm diam circle at a continuous incident power of 0.2 mW, with the exception of the 0.25 μm thick DH for which the incident power was 0.05 mW. With a repetition rate of 80 kHz, the laser generated $< 10^{16}$ carriers/cm^3, so the DHs were always in low injection. The PL spectra were observed in the same system using standard photon counting techniques.

The longest lifetime observed in these samples was 1.2 μs for the 10 μm DH. This decay constant, τ_{DH}, is fully 31 times the expected radiative lifetime computed using $\tau_r = 1/BN_D$, where B is 2×10^{-10} cm^3/s.[7] (Although published values of the B-coefficient vary over an order of magnitude,[7,13] $B = 2 \times 10^{-10}$ cm^3/s is the most commonly used value.[14])

Figure 2 shows the measured decays for the 5 μm DH. Without the substrate backing [curve (a)], τ_{DH} is 1.05 μs for the initial half of the decay time. Curve (a) in Fig. 2

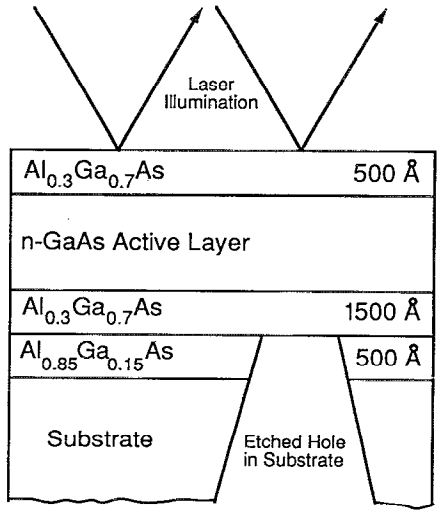

FIG. 1. Structure of double heterostructure (DH) showing hole etched in substrate and the two locations illuminated individually by the laser.

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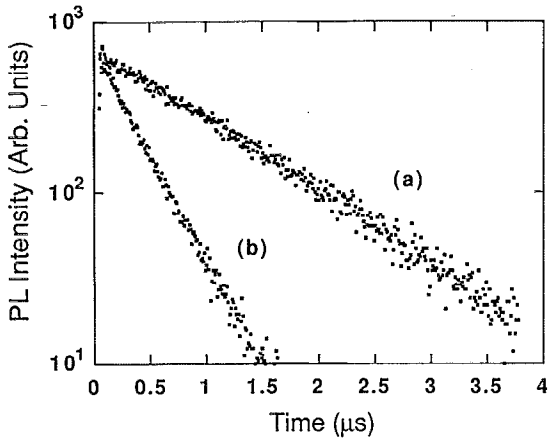

FIG. 2. Photoluminescence decays for 5 μm DH illuminated in the two locations depicted in Fig. 1. Without the substrate [curve (a)], the decay constant, τ_{DH}, is 1.05 μs, 27 times the expected radiative lifetime. Even the DH still backed by the substrate [curve (b)] has a decay constant enhanced nearly ten times over the supposed radiative limit.

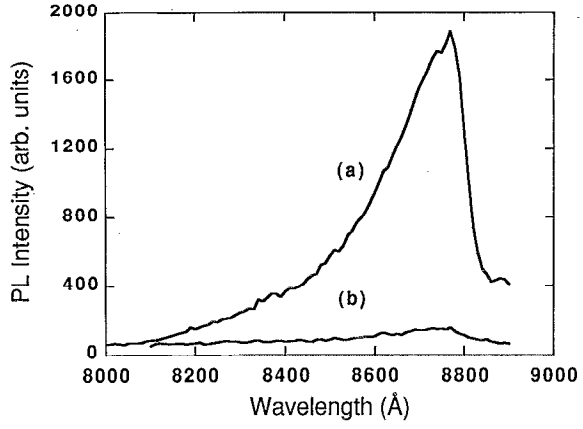

FIG. 3. PL spectra showing the 12-fold increase in intensity exhibited by the GaAs membrane [curve (a)] over that of the region [curve (b)] still backed by the absorbing substrate.

shows evidence of the saturation of Shockley–Read–Hall centers,[15] as initially the PL decays more slowly than it does toward the end of the plot. All the lifetimes reported here are for the initial parts of the curves; this more closely corresponds to the injection level appropriate for devices of interest, such as solar cells. Even with the substrate intact [curve (b)], τ_{DH} is 360 ns, nearly ten times the supposed radiative limit. These ultralong decay constants are due to photon recycling, the reabsorption of photons emitted during radiative recombination events.[5] Self-absorption generates new electron-hole pairs which increases the observed lifetime. The effective radiative lifetime in a DH can then be written as $\tau_{eff}=\phi_r\tau_r$, where ϕ_r Asbeck's recycling cofactor,[6] is the inverse of the average probability that an isotropically emitted photon escapes the DH. Previous workers[3,16] have computed $\phi_r \simeq 11$ for a 5 μm DH still attached to its substrate, which is consistent with our result, assuming that some nonradiative recombination occurs. The effective radiative lifetime of the 5 μm membrane is further enhanced over the region still attached to the substrate because the back side of the DH is now more reflective. The critical angle, Θ_{cr}, of the back interface of the DHs still attached to their substrates is about 72°, whereas for the back of the membranes $\Theta_{cr}=17°$.[16] Thus, simply removing the substrate increases ϕ_r because the isotropically emitted photons are better trapped, increasing their probability of being reabsorbed. This method of enhancing τ_{eff} has been proposed as a means of increasing GaAs solar cell efficiency.[1] Our results demonstrate that if one is able to control nonradiative recombination, ultralong lifetimes are possible, even at this moderately high doping concentration.

Figure 3 shows the PL spectra for the 5 μm DH with and without the substrate intact. The intensity of the peak without the substrate is [curve (a)] 12 times that of the peak observed with the substrate [curve (b)]. The increase in PL peak intensity is due to the decrease in Θ_{cr} for photons escaping out the back of the DH. Many photons emitted that might normally pass into the substrate are now totally internally reflected and eventually reabsorbed. The resulting electron-hole pair might later emit a photon toward the front interface during a radiative recombination event. The result of this recycling process is that there are more "opportunities" for photons to exit the front face and be detected.[17]

Figure 4 plots τ_{DH} versus DH thickness for all the samples, with and without substrates. There is a stronger thickness dependence to τ_{DH} for the standard DHs over τ_{DH} for the membranes because the highly reflective interfaces in the membranes trap most of the photons, so the probability of being reabsorbed is essentially independent of DH thickness. Therefore, ϕ_r is more strongly dependent on the critical angle of the interfaces than on DH thickness w. For the standard DHs, ϕ_r is a stronger function of w because the back interface is less reflective so an average photon in a thicker DH is more likely to be reabsorbed

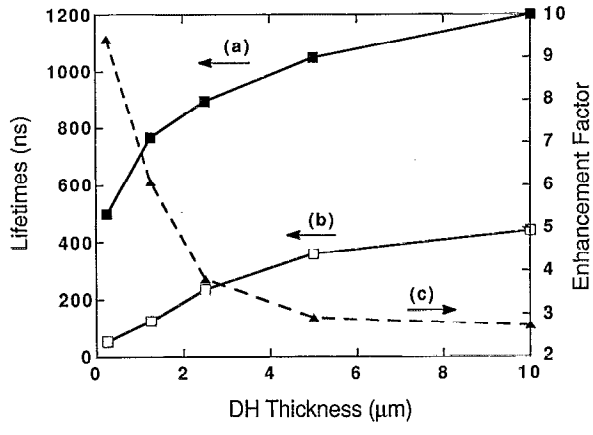

FIG. 4. DH decay constants vs DH thickness, with [curve (b)] and without [curve (a)] the substrate. Under both circumstances, τ_{DH} increases with increasing thickness due both to stronger self-absorption in thicker DHs and to the decreasing contribution from the interfaces to the total recombination. Also shown [curve (c)] is the enhancement of τ_{DH}, defined as the thin-film decay constant divided by the decay constant observed with the substrate intact.

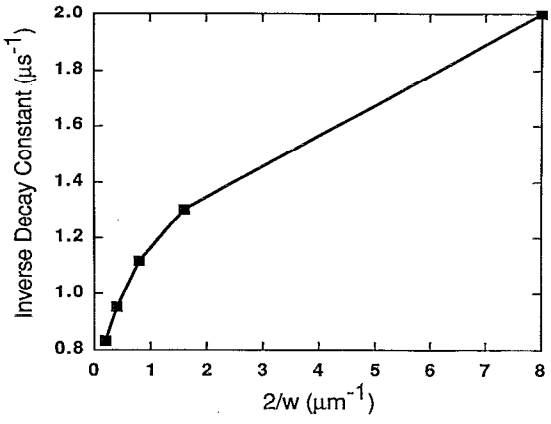

FIG. 5. Plot of $1/\tau_{DH}$ vs $2/w$ for the DHs membranes. The slope of the plot yields an interface recombination velocity of less than 12 cm/s.

before reaching the largely transparent back interface, as compared to a photon in a thinner DH. These considerations explain why the enhancement factor, defined as τ_{DH} without the substrate divided by τ_{DH} with the substrate for the same thickness, steadily increases with decreasing DH thickness.

The long lifetimes observed in thin films make it easy to deduce the interface recombination velocity because interface recombination is more important than it was with the substrate intact. If $Sw/D<1$, where D is the minority carrier diffusivity, recombination in a DH can be described by[18,19]

$$\frac{1}{\tau_{DH}}=\frac{1}{\tau_{bulk}}+\frac{2S}{w}, \tag{1}$$

where τ_{bulk} is the bulk lifetime. One can obtain an upper limit to the interface recombination velocity S by attributing all the recombination of the thinnest DH membrane to the interfaces. In this case $S=w/2\tau_{DH}$, and since for the 0.25 μm DH, $\tau_{DH}=500$ ns, the upper limit of S is 25 cm/s. One can obtain a more accurate value of S by plotting $1/\tau_{DH}$ vs $2/w$, as shown in Fig. 5. Equation (1) suggests that the data in such a plot should lie on a straight line with slope S. However, τ_{bulk} changes with w due to photon recycling so plots of $1/\tau_{DH}$ vs $2/w$ always display negative curvature when the material is dominated by radiative recombination and photon recycling.[20] A least-squares fit to the data points of the three thinnest DH membranes yields $S=12$ cm/s, while a line drawn through the points representing the two thinnest DH membranes has a slope of $S<11$ cm/s. We believe these numbers to be more reasonable values for the interface recombination velocity.

To conclude, we have measured lifetimes longer than 1 μs in thin-film GaAs double heterostructure membranes with active regions doped to $N_D=1.3\times10^{17}$ cm^{-3}. These ultralong lifetimes are possible because of a combination of enhanced photon recycling produced by the removal of the backing substrate and minimal nonradiative recombination in the bulk and at the interfaces. We deduced an absolute upper limit of $S=25$ cm/s for the interface recombination velocity and a more reasonable estimate of $S<12$ cm/s. Such long lifetimes suggest that thin-film GaAs solar cells designed to enhance photon recycling may represent a viable way of achieving higher cell efficiencies.

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