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DOUBLE HETEROSTRUCTURES FOR CHARACTERIZATION OF BULK LIFETIME AND INTERFACE RECOMBINATION VELOCITY IN III-V MULTIJUNCTION SOLAR CELLS

Richard R. King, James H. Ermer, David E. Joslin, Moran Haddad, Jack W. Eldredge, N. H. Karam,
Brian Keyes *, and Richard K. Ahrenkiel *

Spectrolab, Inc., 12500 Gladstone Ave., Sylmar, CA 91342 U.S.A.
* National Renewable Energy Laboratory, 1617 Cole Blvd., Golden, CO 80401 U.S.A.

ABSTRACT: Time-resolved photoluminescence measurements are applied to double heterostructures grown with semiconductor layers and interfaces relevant to multijunction solar cells and other III-V devices, to characterize surface and bulk recombination in these layers. Experimental results include measurement of surface recombination velocity s > 10^5 cm/s at the $Al_{0.25}Ga_{0.25}In_{0.5}P/Ga_{0.5}In_{0.5}P$ interface. A number of practical considerations associated with this characterization method are discussed, such as growth of multiple DHs in a single metal-organic vapor phase epitaxy run, surface band bending, and the effect of low minority-carrier diffusion coefficient on the separation of s and bulk lifetime.
Keywords: Recombination - 1: III-V - 2: Photoluminescence - 3

1. INTRODUCTION

In recent years, $Ga_{0.5}In_{0.5}P$/GaAs dual-junction solar cells [1] have reached efficiencies of 30.2% for terrestrial concentration in the 140 to 180-sun range (AM1.5D) [2], 30.3% at one-sun (AM1.5G) [3], and 26.9% at AM0 for space applications[3]. Triple-junction $Ga_{0.5}In_{0.5}P$/GaAs/Ge cells fabricated at Spectrolab in a large-scale manufacturing environment have achieved 25.8% efficiency [4] (GaInP designates $Ga_{0.5}In_{0.5}P$ unless otherwise noted). In spite of these high efficiencies, the recombination parameters of many structures in these cells are not yet well-characterized.

The performance of high-efficiency tandem solar cells based on III-V compounds is dependent on recombination in the bulk and at the interfaces of the many layers that make up the multijunction cell, such as the window, emitter, base, and back-surface field regions in each subcell. Bulk and interface recombination are determined by a wide range of material parameters, among them semiconductor composition, doping concentration, bulk and interface crystal quality, ordering phenomena, etc. In a multijunction solar cell with 10-20 semiconductor layers, *e.g.*, GaInP/GaAs/Ge triple-junction solar cells processed at Spectrolab [4], measurement of fundamental material parameters such as bulk lifetime and interface recombination velocity becomes complicated by the interactions between the numerous layers in the device. The purpose of this study is to determine experimentally the recombination parameters of several critical layers and interfaces that control the performance of multijunction solar cells, using time-resolved photoluminescence (TRPL) measurements on double heterostructures constructed with the same layers and interfaces. The measurements of bulk lifetime and interface recombination velocity have general utility, however, and can be applied to a wide range of other devices, such as heterojunction bipolar transistors and double heterostructure lasers.

2. EXPERIMENT

In order to separate surface and bulk components of minority-carrier recombination, isotype double heterostructures (DHs) with varying base thickness w were grown by metal-organic vapor phase epitaxy (MOVPE) [5]. All growths were done on a GaAs buffer grown on a Ge substrate, consistent with the fabrication of commercial space photovoltaic cells. The choice of substrate material and orientation can have an effect on the recombination properties of layers grown on top. A method to conserve the required number of MOVPE runs was tried for many of the DH structures, in which multiple DHs of varying base thickness were grown in the same stack of MOVPE layers. Fig. 1 shows an example of a growth stack with three AlGaInP/GaInP/AlGaInP DHs separated by GaAs spacer layers. Using the same base thickness for the first and the last DHs grown provides a measure of the reproducibility of the effective lifetime at different positions in the stack. After growth, the individual DHs are exposed by a series of selective etches: undiluted HCl for phosphide layers, H_2O:H_2O_2:NH_4OH for arsenides. A more standard method of growing a single DH in several sequential MOVPE runs, with varying base thickness in each run, was also tried.

Time-resolved photoluminescence was used to measure the decay of photogenerated minority-carrier concentration in the DHs. All TRPL measurements in this study were conducted at room temperature. The excitation source was a rhodamine 6G dye laser with a wavelength of 580-620 nm, pumped by a frequency-doubled Nd:YAG laser, and the sample photoluminescence was measured by a microchannel plate detector. This TRPL system is capable of measuring decay lifetimes of less than 0.05 ns, and is described in greater depth in [6].

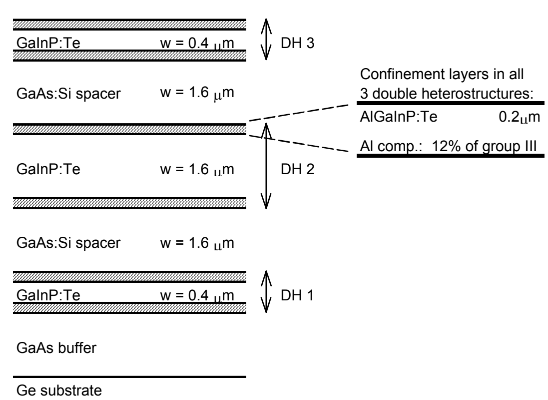

Fig. 1: Example of MOVPE stack containing three AlGaInP/GaInP/AlGaInP double heterostructures.

The effective lifetime τ_{eff} was found from the TRPL decay curves, an example of which is shown in Fig. 2 for a p-type AlGaInP/GaInP/AlGaInP DH with an effective lifetime of 0.26 ns in the single-exponential part of the decay curve. The effective lifetime τ_{eff} of the DH is dependent on the bulk lifetime τ_{bulk} in the base, and the surface recombination velocity s resulting from the wider-bandgap confinment layers on both sides of the base. The surface recombination velocity s is primarily due to states in the gap at the interface between the base and confinement layers, but in cases for which the barrier to minority carriers is small, can be due to injection into the confinement layer and recombination in the bulk or at the surface of those layers. The effective lifetime depends on the base thickness w through [9]:

$$\frac{1}{\tau_{eff}} = \frac{1}{\tau_{bulk}} + \frac{1}{\frac{w^2}{\pi^2 D} + \frac{w}{2s}} . \qquad \text{(Eq. 1)}$$

where D is the minority-carrier diffusion coefficient. In many cases, $2sw/\pi^2 D << 1$, so that:

$$\frac{1}{\tau_{eff}} = \frac{1}{\tau_{bulk}} + \frac{2}{w}s \qquad \text{(Eq. 2)}$$

and s can be extracted from the slope of a linear fit of $1/\tau_{eff}$ *vs.* 2/w, while the bulk lifetime can be found from the reciprocal of the y-intercept of this plot. However, the combination of low D and high s requires the use of the full expression in one case in this study as presented below.

Electrochemical capacitance-voltage (ECV) profiling was used to measure the electrically-active dopant concentration in the samples. The lattice match of the phosphide materials to GaAs and Ge is a sensitive function of composition, and is of critical importance for reducing defects in the bulk and at the interfaces of the grown layers. Double-crystal x-ray diffraction (DCXRD) rocking curves were measured to characterize the crystal quality of the epitaxial layers.

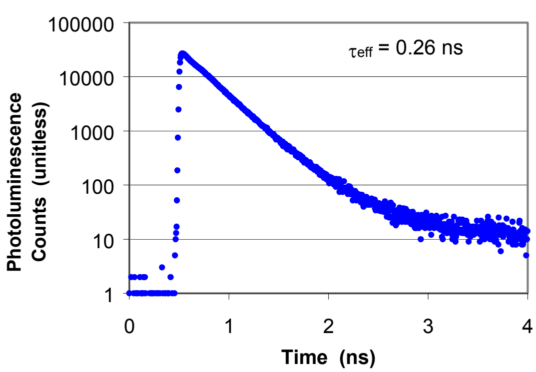

Fig. 2: Photoluminescence decay curve for a p-type AlGaInP/GaInP/AlGaInP DH with a 0.7-µm base.

3. RESULTS AND DISCUSSION

3.1 AlGaInP/GaInP/AlGaInP p-type DHs

The interfaces in this double heterostructure are relevant for the base/BSF (back-surface field) interface in a GaInP top cell of n-on-p multijunction GaInP/GaAs solar cells, and perhaps for emitter passivation of the GaInP emitter in a p-on-n configuration. Fig. 3 shows the band diagram of these structures grown on a p-type GaAs buffer layer, using the program AMPS-1D developed at The Pennsylvania State University. The GaInP base and AlGaInP cladding layers are Zn-doped, with N_A of 9×10^{16} cm^{-3} in the base, and $\sim 3 \times 10^{17}$ cm^{-3} in the AlGaInP. A single DH was grown in each of the three runs in this series, with a different base thickness in each run, rather than growing several DHs in a stack. The Al composition in the AlGaInP was ~25% of the group III elements ($Al_{0.25}Ga_{0.25}In_{0.5}P$) with an estimated bandgap E_g of 2.17 eV, compared to a nominal value of 1.85 eV used for GaInP. This is about midway in the 1.8-1.9 eV range possible for GaInP lattice-matched to GaAs due to group III sublattice ordering.

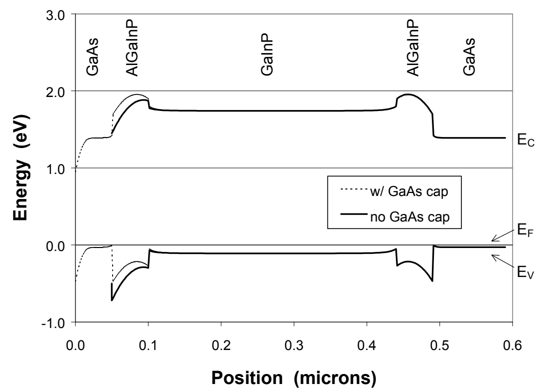

Fig. 3: Band diagram of an AlGaInP/GaInP/AlGaInP double heterostructrure with and without a GaAs cap.

As shown in Fig. 3, DH samples were prepared with and without a thin, p-type GaAs cap over the top AlGaInP barrier layer. For lightly-doped and/or thin barrier layers, pinning of the Fermi level at the semiconductor surface can cause depletion of the layer and sufficient band bending to significantly lower the barrier to minority carriers in the base. When this is the case, a heavily-doped GaAs cap on the surface can accommodate most of the band bending, and result in more symmetrical boundary conditions for carrier transport on both sides of the base. The GaAs cap must, however, be thin enough to allow enough of the excitation wavelengths to reach the DH, and to allow the luminescence emitted by the DH to escape through the cap. To construct the diagram in Fig. 3, the doping levels and layer thicknesses for the samples in the present study were used, and the Fermi level was assumed to be pinned at a point one-third of the bandgap up from the valence-band edge of the surface semiconductor material. The presence of the GaAs cap causes a clear shift in the bands in the top AlGaInP layer. But the thickness and doping in the AlGaInP layers are fairly high in these samples, so the barrier to minority carriers remains high enough that little effect on s is expected.

Steady-state, energy-resolved photoluminescence is shown as a function of emitted wavelength in Fig. 4, for the three base thicknesses grown. The photoluminescence intensity increases rapidly with base thickness, due to the higher effective lifetime of thick samples, and the greater amount of excitation light absorbed in the thicker bases.

The peak intensities occur at fairly low photon energies of 1.817-1.827 eV, indicating that the GaInP bases of these samples have substantial group III ordering. There is a trend of decreasing peak energy with increasing base thickness, possibly due to greater reabsorption of the emitted light on its way out of the thicker structures.

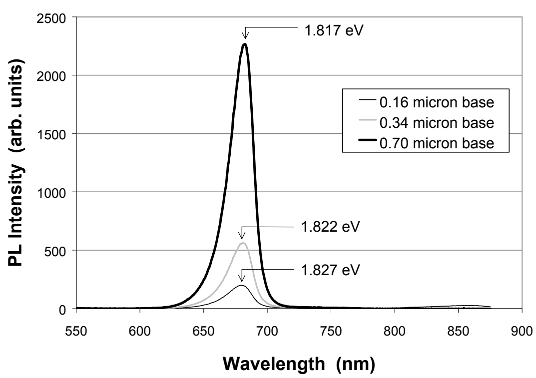

Fig. 4: Photoluminescence spectra for three base thicknesses of AlGaInP/GaInP/AlGaInP DHs.

Fig. 5 plots the lifetime data from TRPL and the base thicknesses of the samples on axes of $1/\tau_{eff}$ *vs.* 2/w. The excitation illumination is at a level which results in a quasi-Fermi-level splitting of ~1.4 V at the beginning of the decay, near the open-circuit voltage V_{oc} of the GaInP subcell of a multijunction cell under one-sun illumination. The data fall on a straight line for each of the cases with and without a GaAs cap. The value of s extracted from the measurements is very high for the AlGaInP/GaInP interfaces in these samples, at 1.3×10^5 cm/s without the GaAs cap, and 1.5×10^5 cm/s with the cap. The uncertainty in τ_{bulk} is large because the y-intercept is so close to zero, but the data indicate that $\tau_b > 2$ ns. This lower limit of τ_{bulk} is much lower than the radiative limit for the lifetime in GaInP at $N_A = 10^{17}$ cm^{-3}, estimated at 50 ns, using a value of the probability of radiative recombination B of 2×10^{-10} cm^3/s at 300 K for GaInP with $E_g = 1.85$ eV as calculated from [6]. For comparison, B for GaAs calculated using the same method results in B = 1.5 $\times 10^{-10}$ cm^3/s.

In the limiting case of a GaInP top cell in which all recombination takes place at the back surface, the saturation current density J_o of the cell is:

$$J_o = q\frac{n_i^2}{N_{A,back}}\frac{D}{w}\left[\frac{1}{1+\frac{D}{sw}}\right] \qquad \text{(Eq. 3)}$$

where w is the thickness of the top cell base and $N_{A,back}$ is the doping at the back of the GaInP base. Using a typical value of D/w of 5×10^5 cm/s, $n_i^2 = 7 \times 10^5$ cm^{-6}, and $N_{A,back} = 9 \times 10^{16}$ cm^{-3} as for the DHs that were measured, gives a value of $J_o = 1.3 \times 10^{-25}$ A/cm^2 for $s = 1.3 \times 10^5$ cm/s at the GaInP/AlGaInP interface. Using a photgenerated current density J_{ph} of 0.016 A/cm^2 in:

$$V_{oc} = \frac{kT}{q}\ln\left(\frac{J_{ph}}{J_o}\right) \qquad \text{(Eq. 4)}$$

leads to a corresponding V_{oc} of 1.37 V, with the simplifying approximations considered here. So even excluding all other sources of recombination — in the bulk, space-charge region, emitter, and perimeter region of the top cell — recombination at the back limits the top cell V_{oc} to 1.37 V. Increasing $N_{A,back}$ and modifying MOVPE growth parameters at this critical interface to reduce the value of s offer routes to reduce J_o and thus increase V_{oc} .

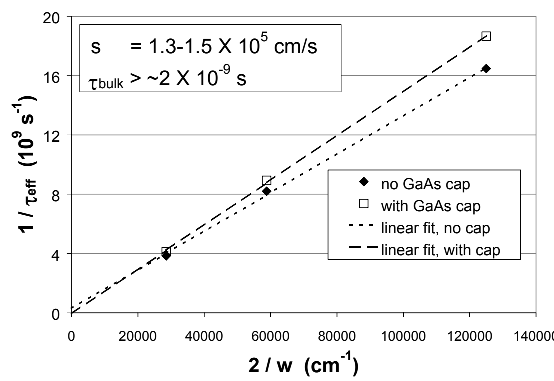

Fig. 5: Plot of measured $1/\tau_{eff}$ *vs.* 2/w to extract s and τ_b for AlGaInP/GaInP/AlGaInP DHs with and without GaAs cap.

3.2 GaInP/GaAs/GaInP n-type DHs

Recombination at the interfaces and in the GaAs base of this type of DH is applicable to the window/emitter structure in an n-on-p GaAs solar cell, or to the base/BSF structure in a p-on-n cell. The GaAs and GaInP layers are Te-doped, with a doping level of 1-3 $\times 10^{18}$ cm^{-3} in the GaAs, and 5×10^{17} cm^{-3} in the GaInP confinement layers. The DHs were grown in a stack in a single MOVPE growth run, and exposed by selective etching as described above.

Fig. 6 plots the results of the TRPL measurements on these DHs, during which no GaAs cap was present. The two thin DHs with a 0.2 µm base, which were the first and the last DHs in the growth stack, have somewhat different measured values of τ_{eff} . This can be expected to some degree due to their different position in the growth, and correspondingly different thermal history, defect distribution, etc. The value of s at this fairly heavily-doped

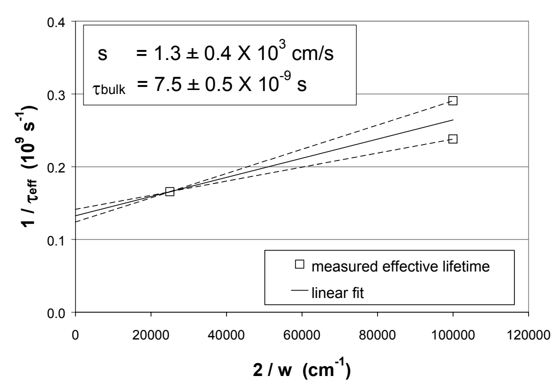

Fig. 6: Extraction of s and τ_{bulk} for GaInP/GaAs/GaInP DHs with 1-3 $\times 10^{18}$ cm^{-3} n-type base doping.

GaAs/GaInP interface is $1.3 \pm 0.4 \times 10^3$ cm/s, and τ_{bulk} in the GaAs base is 7.5 ± 0.5 ns, where the uncertainties correspond to the different τ_{eff} values measured for the thin DHs. The value of s for this heavily-doped n-type sample is much higher than has been measured for the undoped GaAs/GaInP interface [5]. It is not yet known whether this difference is due to the much higher doping levels in the present study, or to other differences in MOVPE growth parameters which affect the interface. The extracted value of bulk lifetime is consistent with the estimated radiative lifetime of 6.7 ns at $N_D = 10^{18}$ cm^{-3}, within the uncertainties in the experiment and in the radiative coefficient B.

3.3 AlGaInP/GaInP/AlGaInP n-type DHs

This structure is relevant for emitter passivation for the GaInP top cell of an n-on-p multijunction solar cell, or for the base/BSF interface of a p-on-n GaInP top cell. The three DHs in this series were grown in a stack as illustrated in Fig. 1. The Al composition in the confinement layers is ~12% of the group III elements, resulting in an estimated bandgap of 2.0 eV. Steady-state, room-temperature photoluminescence measurements on thicker layers of AlGaInP with this composition have a peak intensity corresponding to 1.96 eV. For comparison, the peak intensity from the GaInP base of these samples is at a fairly high energy of 1.89-1.90 eV, presumably due to high degree of group III sublattice disorder, so the bandgap difference ΔE_g at the interface is quite small. Both the GaInP and the AlGaInP were doped with Te, at $6\text{-}8 \times 10^{17}$ cm^{-3} in the GaInP base, and in the mid-10^{16} range for the AlGaInP confinement layers. This is significantly lower than the desired doping level for the confinement layers, which should be at least in the 10^{18} range.

The minority carriers in the n-type base of this DH are holes and thus D is relatively small. Majority hole mobilities in GaInP have been measured to be 60-140 cm^2/Vs for Zn doping in the high 10^{17} cm^{-3} range [10], depending on the degree of group III sublattice ordering. The minority hole mobilities are likely to differ from these values, but a straightforward use of these mobilities gives D in the 1.6-3.6 cm^2/s range. The approximation of $2sw/\pi^2 D << 1$ that has worked well for the previous samples does not necessarily apply for the values of s and w of these samples, so Eq. 1 must be used to analyze these DHs. In addition, both Eqs. 1 and 2 assume that more than the diffusion transit time $w^2/D\pi^2$ [6] has elapsed since the excitation pulse before measuring τ_{eff} from the TRPL signal. If the value of D used for the minority holes is at the lower end of the range given above, the TRPL signal for the thickest DH considered here (1.6 μm) decays significantly before this time has been reached.

If we call s_∞ the surface recombination velocity extracted from the slope of the straight line in Eq. 2, since this is the limiting form as $D \to \infty$, then:

$$s_\infty = \left(\frac{1}{\tau_{eff,2}} - \frac{1}{\tau_{eff,1}}\right) \Big/ 2\left(\frac{1}{w_2} - \frac{1}{w_1}\right) \qquad \text{(Eq. 5)}$$

for a straight line between two data points, with thicknesses w_1 and w_2, and measured effective lifetimes $\tau_{eff,1}$ and $\tau_{eff,2}$. The actual surface recombination velocity s that results from fitting Eq. 1, which includes the effect of D, through the same two data points is given by:

$$s = s_\infty \left[\left(\frac{1}{2} - \frac{2s_\infty \overline{w}}{\pi^2 D}\right) + \sqrt{\left(\frac{1}{2} + \frac{2s_\infty \overline{w}}{\pi^2 D}\right)^2 - \left(\frac{2s_\infty}{\pi^2 D}\right)^2 w_1 w_2} \right]^{-1} \qquad \text{(Eq. 6)}$$

where $\overline{w} = (w_1 + w_2)/2$.

The measured TRPL data points are shown in Fig. 7. Using the measured τ_{eff} of the thick sample in Fig. 7 as one data point, and the average τ_{eff} of the thin samples for the other, yields $s_\infty = 7 \times 10^4$ cm/s and $\tau_{b,\infty} = 0.6$ ns, where $\tau_{b,\infty}$ is the bulk lifetime assuming $D \to \infty$. To illustrate the effect of a finite D, although D is not known precisely, fitting Eq. 6 to the same two data points as shown by the solid curve in Fig. 7 gives the values $s = 8 \times 10^4$ cm/s and $\tau_{bulk} = 0.47$ ns as consistent with $D \cong 2$ cm^2/s.

As pointed out above, the diffusion transit time of the thick sample may be longer than the point of the TRPL curve at which τ_{eff} was measured, depending on the value of D used. As a check, it is useful to place limits on the value of τ_{bulk} consistent with the measured TRPL lifetimes. In the limiting case of zero surface recombination, the measured τ_{eff} would be equal to the bulk lifetime. So the 0.39 ns τ_{eff} measured for the DH with the largest GaInP base thickness represents a lower limit for τ_{bulk}, close to the extracted value of 0.47 ns. It is suspected that the bulk lifetime of these samples may have been degraded by growth defects, perhaps due to imperfect lattice matching between the thick GaInP layers and the GaAs buffer layers.

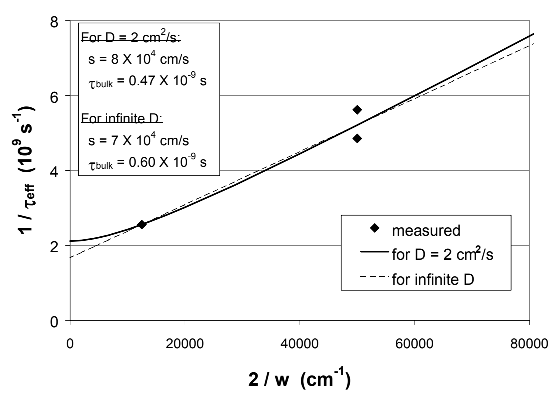

Fig. 7: Plot of $1/\tau_{eff}$ *vs.* 2/w for n-type AlGaInP/GaInP/AlGaInP DHs, showing the effect of D on the extraction of s and τ_{bulk}.

3.4 AlInP/AlGaInP/AlInP n-type DHs

This structure is interesting for general characterization of the bulk properties of the wide-bandgap AlGaInP base, which is often used as a barrier layer, as well as the ability to passivate this material with AlInP. The Al composition in the base was ~12%, and ~50% in the confinement layers. The AlGaInP base was Te-doped in approximately the mid-10^{17} range, while the AlInP cladding layers were doped with Si to $\sim 5 \times 10^{18}$ cm^{-3}.

Three double heterostructures were grown in a stack with intervening GaAs spacer layers, using the scheme shown in Fig. 1, but with different layer thicknesses and materials. The last DH to be grown (DH3) has a very short measured τ_{eff} of ~0.08 ns, and may have suffered from increasing defect concentration as the MOVPE growth progressed. The other two DHs (DH1 and DH2) may be used to place limits on s and τ_{bulk} , consistent with the measured τ_{eff}. The thickest sample, with an AlGaInP base thickness of 0.8 μm, has a measured τ_{eff} of 0.22 ns. So a lower limit for the τ_{bulk} in AlGaInP with 12% Al composition as grown in these samples is 0.22 ns. Al-containing compounds tend to incorporate high concentrations of oxygen impurities, which may be responsible for this very low lifetime. The first thin DH to be grown, with 0.2-μm base thickness, can be used to establish an upper limit for s. The measured τ_{eff} of this sample is 0.16 ns. If one assumes that all recomination in the sample is at the surface, and that diffusion transit times are short, the maximum value of surface recombination velocity s_{max} consistent with this measurement is $s_{max} = w/(2\tau_{eff}) = 6.2 \times 10^4$ cm/s for the AlInP/AlGaInP interface in this sample.

4. SUMMARY

Interface and bulk recombination components have been characterized in a number of semiconductor layers critical to the performance of high-efficiency, multijunction, III-V solar cells. Fabrication of relevant test heterostructures and their measurement by time-resolved photoluminescence offers a general and practical method to evaluate new materials, interfaces, and growth conditions for high-efficiency solar cells. Specific results obtained in this study include measurement of $s > 10^5$ cm/s at the p-type AlGaInP/GaInP interface for 25% Al composition. The value of s at the GaInP/GaAs interface was near 10^3 cm/s for the samples measured here, at low 10^{18} cm^{-3} n-type doping levels in the GaAs base. The bulk recombination in GaAs in these samples was near the radiative limit. A lower limit of 0.22 ns was measured for the bulk lifetime of n-type AlGaInP with 12% Al composition, doped in the 10^{17} range.

Several practical considerations for the use of double heterostructures in TRPL measurements were revealed in the course of this work. The method of growing several DHs of different base thicknesses in a single MOVPE run, to facilitate the preparation of DHs needed for separation of s and τ_{bulk} , was tried in a variety of material systems. An important issue in this method is the correlation of recombination properties in layers grown with the same nominal MOVPE conditions at different positions in the growth sequence, where the layers may be subject to different thermal histories, strain and dopant redistribution from nearby layers, point-defect injection, and other non-ideal effects. Such interactions can be important in the many layers that make up multijunction solar cells as well. DHs grown with the same base thickness at the top and bottom of the stack showed that in some cases recombination parameters remained similar through the growth, while in others material quality degraded significantly. The degree of lattice matching as measured by XRD is critical to maintaining similar recombination parameters through multiple layers in the same MOVPE stack. Other measurement issues include surface band bending, which can be corrected with a thin GaAs cap for the case of thin, lightly doped confinement layers in the DH. For low values of the minority-carrier diffusion coefficient, the effect of carrier diffusion toward the surfaces and transit time in the DH on the extraction of s and τ_{bulk} were considered. With these practical considerations taken into account, TRPL measurements of double heterostructures provide a direct avenue for improved understanding and performance for multijunction solar cells and other III-V devices.

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