

RECOMBINATION IN LOW-BANDGAP InGaAs

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ABSTRACT

We review our investigation of recombination in $\text{In}_x\text{Ga}_{1-x}\text{As}$ with indium concentrations ranging between $x = 0.53$ (i.e., lattice-matched to InP) and $x = 0.78$. External radiative efficiency measurements were used to study how defect-related and Auger mechanisms compete with radiative recombination. The results indicated that deep mid-gap levels facilitate defect-related recombination in lattice-matched InGaAs while shallower levels play a more important role in the indium-rich alloys. Subsequent sub-bandgap photoluminescence measurements confirmed the presence of deep levels in the lattice-matched InGaAs. The superlinear excitation dependence of the sub-gap emission led to a defect-related deep-donor/shallow-acceptor pair model. Recent cathodoluminescence measurements of the subgap transitions show no spatial contrast, supporting the assignment of this mechanism to evenly distributed point defects. We hypothesize that the deep states observed in lattice-matched InGaAs are related to imperfections in the incorporation of indium or gallium, which become less likely as the indium concentration is increased.

INTRODUCTION

Indium-rich InGaAs alloys are useful for high-efficiency thermophotovoltaic cells, devices that convert thermal radiation into electricity. When lattice-matched to InP, the InGaAs bandgap of 0.74 eV is larger than ideal for this technology, but mismatched indium-rich alloys have smaller gaps. Thus, low-energy blackbody photons are more readily absorbed. For this application, recombination of carriers is detrimental because it reduces the conversion efficiency of the cells. Ordinarily, lattice-mismatched epistructures contain a high density of extended defects, which usually augment deleterious defect-related recombination. Yet we have learned how to prepare lattice-mismatched $\text{In}_x\text{Ga}_{1-x}\text{As}$ -based photovoltaic devices that have performance characteristics comparable to the lattice-matched case. [1] Even when a higher density of misfit dislocations is presumably present (as evidenced by cross-hatching in the surface morphology), our severely mismatched photovoltaic devices perform exceptionally well.

The lattice-matched $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{InAs}_y\text{P}_{1-y}$ double heterostructures are grown by atmospheric-pressure metalorganic vapor-phase epitaxy on InP substrates. The lattice-matched InAsP barriers adjacent to the InGaAs

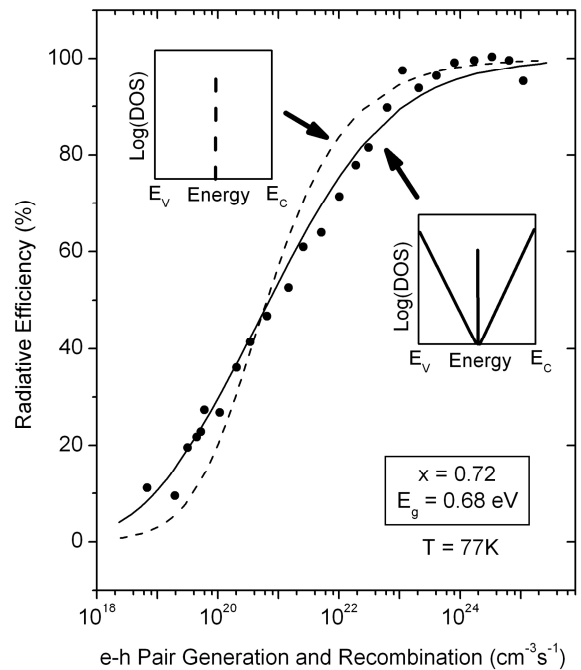


Figure 1: Radiative efficiency vs. the steady-state rate of electron-hole pair generation and recombination in a moderately indium-rich structure. The curves are theoretical fits assuming the defect-related density of states distributions shown.

layer confine carriers and passivate the interfaces. When $x > 0.53$, a carefully designed buffer of InAsP compositional step-grading layers is incorporated between the substrate and the heterostructure to accommodate the lattice mismatch. Further details on the sample structure can be found in Ref. 2. In this paper, we review our investigation of the physical and electrical properties of defects in this material system that contribute to superior lattice-mismatched photovoltaic performance. In particular, we focus on how defect-related recombination changes with alloy composition for $0.53 < x < 0.78$. We also discuss our analysis of Auger recombination in $\text{In}_x\text{Ga}_{1-x}\text{As}$ and its dependence on the bandgap energy.

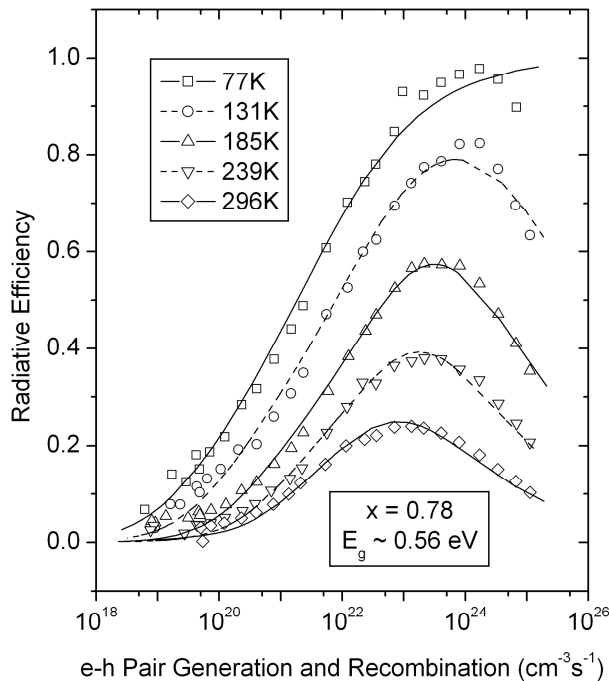


Figure 2: Radiative quantum efficiency vs. the steady-state rate of electron-hole pair generation and recombination in our lowest bandgap, indium-rich epistructure at the temperatures indicated. The solid and dashed curves are theoretical fits.

RADIATIVE EFFICIENCY

Initial experimental characterization was accomplished via external radiative quantum efficiency measurements. [3] We measure the radiative efficiency as a function of excitation power at 77K to study the changeover between defect-related (nonradiative) and radiative recombination in the structures. Since Auger recombination is negligible at low temperatures and defect recombination saturates at high carrier densities, we can usually obtain 100% internal quantum efficiency in our samples at 77K when the excitation rate is sufficiently high. The low-temperature, high-excitation result provides a baseline for calibrating measurements at other temperatures and excitation intensities.

In general, increasing lattice-mismatch is expected to generate a larger dislocation density, which typically augments the nonradiative process. However, we find that the overall rate of defect-related recombination in the indium-rich epistructures shows little change from the lattice-matched case. This result is very unusual, but it may be explained by another unique feature of our measurements: the shape of the efficiency curve changes dramatically as the lattice mismatch in the structures is increased. A simple defect recombination model [4] assuming defect levels concentrated near the middle of the bandgap fits well in the lattice-matched case but the model does not fit the shape of the efficiency curve for the mismatched samples (see the dashed curve in Fig. 1).

We have developed a rigorous computer simulation based on the more detailed theory [5] that allows for other distributions of energy levels at defects. For the mismatched structures, we find that addition of band-edge exponential tails to the defect-related density of states gives a much better theoretical fit (see the solid curve in Fig. 1). The inset graphs in Fig. 1 show the corresponding distribution of defect levels between the valence and conduction band edges (E_V and E_C) that were used to generate the theoretical fits. The results suggest that defect states in the indium-rich system are fundamentally different from those in the lattice-matched material, and that their energetic positions are less likely to facilitate nonradiative recombination.

When the temperature exceeds 77K, Auger recombination becomes more important at high carrier concentration. Fig. 2 shows how the radiative efficiency varies with excitation in this regime. The temperature-dependent Auger rate can be extracted from theoretical fits to these results (see the solid and dashed curves in Fig. 2) and compared with that of other InGaAs compositions. [2] As shown in Fig. 3, we find a strong exponential dependence on the reciprocal of the temperature (and a similar dependence on bandgap energy), which generally points to band-to-band Auger processes. In addition, by comparing the temperature- and bandgap-dependent results, we are able to rule out band-to-

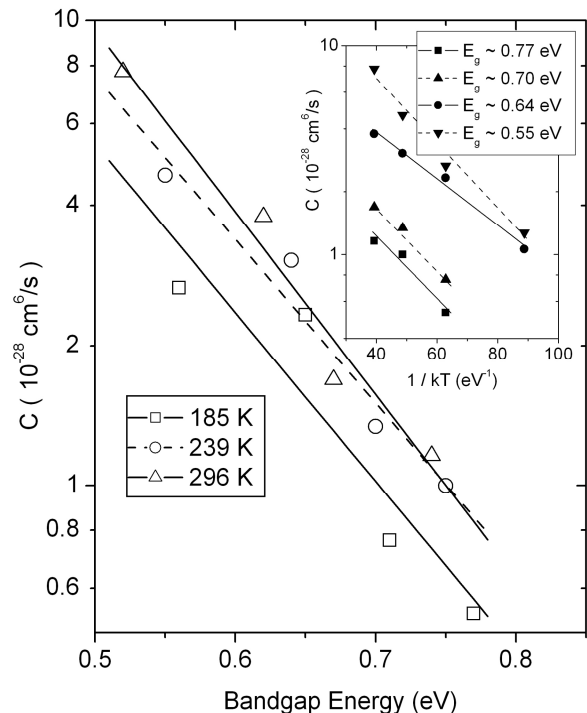


Figure 3: Auger recombination coefficient C vs. the bandgap energy at several temperatures. Inset: Arrhenius plot of C for several different alloys (identified by their bandgap energy averaged over temperature).

band mechanisms that do not include the spin-orbit splitoff band. We deduce that the conduction to heavy hole band – splitoff to heavy hole band mechanism dominates Auger recombination in undoped low-band gap $\text{In}_x\text{Ga}_{1-x}\text{As}$. The augmented Auger rate in the lower gap alloys can be expected to hinder photovoltaic performance under intense illumination.

SUB-BANDGAP PHOTOLUMINESCENCE

If defect states within the gap are radiative, sub-bandgap features may be observed in photoluminescence measurements. Hence, we have used Fourier transform infrared spectroscopy to obtain spectra down to 0.3eV and test the modified density of states interpretation of the radiative efficiency results presented above. [6] Representative spectra for the nominally lattice-matched and nearly lattice-matched ($x = 0.60$) epistructures are shown in Fig. 4. In addition to the strong band-to-band (B-B) emission, we note a sub-bandgap peak (labeled SBG) more than 0.2eV below the B-B luminescence. The SBG peak loses intensity with increasing indium concentration and is not observed in the more indium-rich ($x = 0.72$ and $x = 0.78$) low-bandgap epistructures. The observation of a deep level in the lattice-matched structure that abates in the $x = 0.60$ alloy and does not appear when $x > 0.60$ supports the suggestion that the density of deep states is reduced with increasing indium concentration in this material system.

CATHODOLUMINESCENCE

A detailed excitation-dependent study [6] of the SBG emission described above indicates that a deep-donor/shallow-acceptor pair recombination mechanism may be involved. This result is consistent with recent cathodoluminescence measurements on these structures. Plan-view spatial maps of the SBG peak intensity do not reveal any correlation with extended defects. Indeed, the distribution of this emission appears to be very uniform throughout the epilayer, confirming that point defects are more likely to be responsible for these transitions. Since the intensity of the SBG peak depends strongly on the $\text{In}_x\text{Ga}_{1-x}\text{As}$ composition, we hypothesize that it is related to an imperfection in the incorporation of indium or gallium atoms, yielding a localized defect like a vacancy or an interstitial.

CONCLUSION

External quantum efficiency, sub-bandgap photoluminescence, and cathodoluminescence measurements have been used to explore recombination mechanisms in indium-rich InGaAs grown on InP substrates. Results of the experiments help explain why photovoltaic devices based on lattice-mismatched InGaAs perform so well. While increasing mismatch presumably generates a higher density of extended defects, a simultaneous reduction in deep levels associated with composition-dependent point defects appears to balance the overall rate of defect-related recombination.

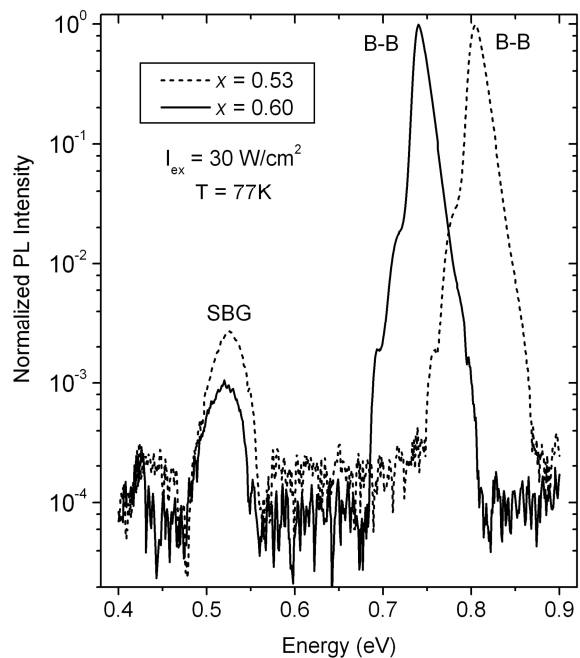


Figure 4: Photoluminescence spectra of the nominally lattice-matched and nearly lattice-matched epistructures at 77K. The spectra are plotted with a logarithmic vertical scale to highlight weak, sub-bandgap features.

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REFERENCES

- [1] M.W. Wanlass, S.P. Ahrenkiel, R.K. Ahrenkiel, J.J. Carapella, R.J. Wehrer, and B. Wernsman, *AIP Conf. Proc.* **738**, 427 (2004).
- [2] T.H. Gfroerer, L.P. Priestley, M.F. Fairley, and M.W. Wanlass, *J. Appl. Phys.* **94**, 1738 (2003).
- [3] T.H. Gfroerer, L.P. Priestley, F.E. Weindruch, and M.W. Wanlass, *Appl. Phys. Lett.* **80**, 4570 (2002).
- [4] R.N. Hall, *Phys. Rev.* **87**, 387 (1952).
- [5] W. Shockley and W.T. Read, Jr., *ibid.* **87**, 835 (1952).
- [6] T.H. Gfroerer, C.E. Gillespie, J.P. Campbell, and M.W. Wanlass, *J. Appl. Phys.* **98**, 093708 (2005).