A Comparison of Photoluminescence Imaging and Confocal Photoluminescence Microscopy in the Study of Diffusion near Isolated Extended Defects in GaAs

T. H. Gfroerer,[1,2] Yong Zhang,[2] and M. W. Wanlass[3]

[1]Davidson College, Davidson, NC, 28036, USA
[2]University of North Carolina at Charlotte, Charlotte, NC, 28223, USA
[3]National Renewable Energy Laboratory, Golden, CO, 80401, USA

***Abstract* — Extended defects like dislocations augment recombination and reduce the local density of photogenerated carriers. We use photoluminescence imaging and confocal photoluminescence microscopy to study the diffusion of free carriers toward these defect-related depletion regions in GaAs. Both techniques reveal important changes in the size of the darkened region as the photoexcitation is reduced. Under lower illumination, the affected region generally expands, reflecting the impact of longer carrier lifetimes. But the physics of the confocal measurement, with highly localized excitation and detection, is very different from the physics involved in far-field imaging of photoluminescence under spatially uniform illumination. In particular, the radiative efficiency falls precipitously as the confocal excitation is reduced below a threshold of approximately 1 KW/cm^2, and we observe a simultaneous transformation in the confocal contrast profile. We attribute this dramatic change to trapping by bulk point defects, which are saturated when the effective photogenerated carrier density is sufficiently high.**

***Index Terms* — charge carrier density, diffusion processes, photoluminescence, semiconductor epitaxial layers**

I. Introduction

The performance of photovoltaic devices depends, in part, on the nature and quantity of defects in the constituent materials. Imperfections in the crystal lattice introduce localized states with energy levels that can lie within the forbidden gap of a semiconductor. These levels augment nonradiative recombination by capturing photogenerated carriers and providing them with new recombination pathways. [1] Thus, carriers are lost and the conversion efficiency of the device is reduced. Native point defects, like vacancies and unintentional impurities, are present throughout the bulk and cause a modest reduction in efficiency throughout the device. But extended defects like dislocations furnish a large concentration of defect states in a highly localized region. Here the recombination rate can be orders of magnitude higher, depleting the local carrier density and creating a concentration gradient that draws nearby carriers in via diffusion.

The effects of point and extended defects depend strongly on illumination. In high quality materials like the MOVPE-grown GaAs studied here, point defects are relatively sparse and saturate under moderate photoexcitation. But extended defects, with their high concentration of states throughout the gap, can continue to deplete the local carrier density even when the photoexcitation is extremely high. The impact of extended defects on the adjacent material depends on the effective diffusion length of the photogenerated carriers. As the photoexcitation is increased, carrier lifetimes and travel distances are reduced, so the range of influence decreases. These changes in the extent of the dislocation-induced dead-zone (where extra carriers are lost to defect-related recombination) have important implications for concentrated photovoltaics. In particular, changing the concentration factor alters the underlying dynamics in ways that have a direct bearing on the charge collection efficiency of a photovoltaic device.

II. Experimental Details

We use photoexcitation-dependent luminescence measurements to explore the impact of isolated dislocations on the surrounding material. When defect-related recombination and diffusion deplete the local carrier density, the photoluminescence (PL) signal is reduced. In this report, we compare complementary results using two very different experimental arrangements. In the first approach, we use an unfocused laser (λ = 532 nm) centered on the dislocation to provide relatively uniform illumination along with a CCD camera to image the photoluminescence profile. The optics in this configuration yield ~ 1X magnification, so image pixels have the same dimensions as detector pixels, which are 13 μm x 13 μm. In the second experiment, we use confocal microscopy to obtain coincident photoexcitation (λ = 633 nm) and detection in a sharply focused spot (~ 1 μm^2), which is scanned across the dislocation to generate a confocal PL map. All measurements are conducted at room temperature.

The test structure for this comparison is a nominally undoped and lattice-matched GaInP/GaAs/GaInP double heterostructure grown by metal organic vapor phase epitaxy (MOVPE) on a semi-insulating GaAs substrate. The approximately 2 μm thick GaAs active layer is designed to absorb most of the incident laser light while minimizing photon recycling. This configuration leads to very strong band-to-band PL emission with a peak energy of 1.43 eV. However, dark spots are also observed in low-magnification,

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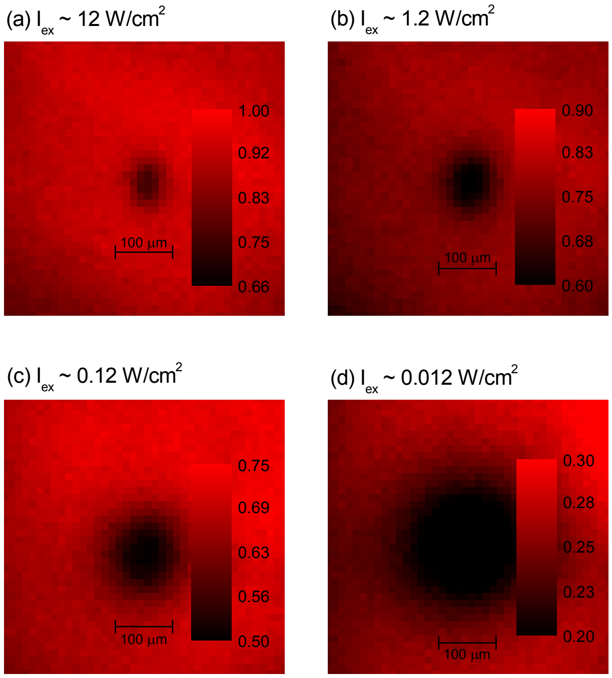

Fig. 1. PL images of an isolated dislocation in GaAs under the uniform illumination conditions cited above each image.

plan-view PL images of the sample. These features are presumably affiliated with extended defects that originate in the substrate and propagate up through the active layer. The imperfections appear to be randomly distributed with an estimated density of ~ 400 cm^{-2}.

III. Experimental Results

PL images of an isolated dislocation under a wide range of photoexcitation intensities I_{ex} are shown in Fig. 1. Even allowing for the important distinction between monochromatic above-gap illumination and sunlight, the photoexcitation used to acquire image (c) would be closest to terrestrial one sun operation (~ 0.1 W/cm^2). Greyscale values are estimates of radiative efficiency, assuming that the efficiency approaches unity far from the dislocation at high excitation, where dislocation-driven diffusion is negligible and bulk defect states are saturated. These images can be simulated by an algorithm that allows for Laplacian diffusion and SRH recombination [1] via two distinct defect level distributions: one for the dislocation pixel and another for the bulk. [2]

Complementary confocal microscopy maps of the PL signal in the vicinity of a dislocation are shown in Fig. 2. In these maps, radiative efficiency is computed by assuming that the efficiency approaches unity when the signal divided by the laser power reaches its maximum value, which occurs when I_{ex} ~ 42 KW/cm^2. In this experiment, when the photoexcitation is increased further, local heating and/or Auger recombination reduce the measured radiative efficiency. We should note that these highly concentrated excitation conditions cannot be easily compared with the spatially uniform excitation intensities used in PL imaging or the normal operating conditions of a photovoltaic cell. Since carriers will diffuse rapidly away from the high density confocal excitation spot, the effective photogenerated carrier density will be substantially smaller than the extreme excitation rate might suggest.

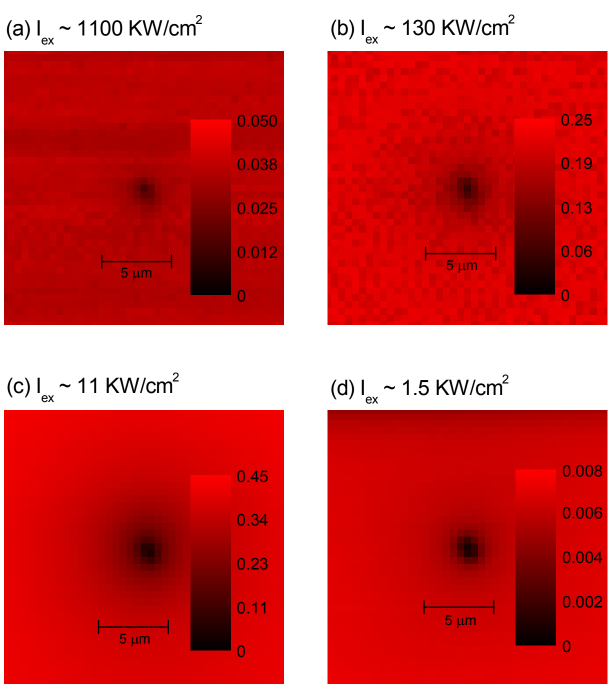

Fig. 2. Confocal PL maps of an isolated dislocation in GaAs with estimates of the photoexcitation intensity I_{ex} indicated above.

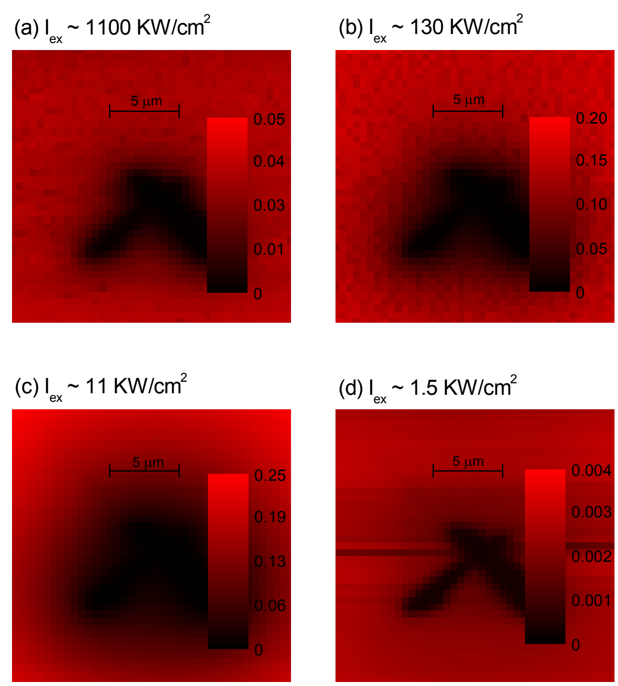

Fig. 3. Confocal PL maps of the same location mapped in Fig. 2 after laser modification. Please note the changes in greyscale.

Despite fundamental changes in experimental design and scale, we generally observe a similar phenomenon: as the photoexcitation intensity is reduced, diffusion to the dislocation becomes more pronounced. In particular, when I_{ex} ~ 11 KW/cm^2, the darkened region spreads in comparison with higher photoexcitation conditions.

However, we also observe a surprising departure from this behavior at the lowest excitation (I_{ex} ~ 1.5 KW/cm^2). In this case, the affected region and apparent diffusion length actually shrink. The departure from expectations is more evident in the sequence of maps shown in Fig. 3. These maps were obtained at the same site examined in Fig. 2 after prolonged laser exposure altered the physical character of the central dislocation. The dark features, aligned along ⟨100⟩ directions, are clearly sharper in the lowest excitation map. We note that the clarity enhancement is accompanied by a steep drop in the bulk radiative efficiency. Hence, we deduce that the abrupt change in diffusivity is related to the filling of bulk defect states. In other words, local bulk defects impede diffusion, but they are only effective at very low excitation density.

IV. Discussion

When the confocal excitation intensity is near or below ~ 1 KW/cm^2, point-like defect states in and just beyond the illuminated volume are available for occupation, and trapping at these evenly distributed sites limits long-range diffusion to the dislocation. In this regime, nonradiative point defect-related recombination dominates, and the radiative efficiency is extremely low. But the density of these states is relatively small, so at higher excitation they are saturated. As the excitation is increased above this threshold, these sites play a diminishing role in the free carrier dynamics.

An analogous low-excitation transition is not observed in the PL imaging experiment for at least two reasons. First, the confocal measurement is more sensitive because the high numerical aperture (NA) objective collects nearly all of the PL signal at each point, in contrast to lower NA imaging and distribution of the PL signal among an array of pixels. This confocal advantage enables an investigation of much lower effective carrier densities. Second, even if the density in the confocal volume is relatively high, it is too low to saturate the point defects along the path to the extended defect. In PL imaging, we generate nearly the same carrier density everywhere, so the point defects become filled simultaneously throughout.

In both experiments, the saturation of bulk defect states provides the opportunity for long-range diffusion to the high density of defect states in the vicinity of a dislocation. Diffusion to the dislocation depletes the neighboring carrier density so the PL contrast reaches deeper into the bulk. Clearly, the relative significance of bulk and extended defects depends strongly on the excitation density, or the photovoltaic device equivalent – the concentration of solar radiation.

Meanwhile, the band-to-band radiative recombination rate also increases as the photoexcitation and band occupations are raised. This mechanism, which scales as the product of electrons and holes in their respective bands [1], reduces the free carrier lifetime and the associated diffusion length. Thus, the defective features become more clearly delineated again under the highest excitation. From a photovoltaic design perspective, this recombination mechanism is less problematic because the new photons are often recycled in the device.

The dislocations in this special well-matched epistructure are spaced widely enough to enable a detailed investigation of the behavior near individual sites. Photovoltaic devices are necessarily more complex, and are likely to contain many more dislocations per unit area. Recombination loss depends on the capture cross-sections and defect level distributions at dislocations and at point defect sites throughout the bulk. These parameters dictate how diffusion operates under different illumination conditions. While defect-related recombination always becomes less important relative to competing mechanisms with increasing excitation, point-like and extended defects play different roles in carrier transport. Optimal operation will generally occur when the excitation is high enough to saturate bulk sites and minimize lifetime-regulated diffusion to dislocations, while being low enough to avert Auger recombination and excessive heat.

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References

[1] W. Shockley and W. T. Read, Jr., “Statistics of the Recombinations of Holes and Electrons,” Phys. Rev. **87**, 1952, pp. 835-842; R. N. Hall, “Electron-Hole Recombination in Germanium,” *ibid.* **87**, 1952, p.387.

[2] T.H. Gfroerer, C.M. Crowley, C.M. Read, and M.W. Wanlass, “Excitation-dependent recombination and diffusion near an isolated dislocation in GaAs”, *J. Appl. Phys.* **111**, 093712 (2012).