Photoluminescence fatigue in three-dimensional silicon/silicon-germanium nanostructures

Modi, N.; Tsybeskov, L.; Baribeau, J.-M.; Wu, X.; Lockwood, D. J.

Publisher's version / Version de l'éditeur:
https://doi.org/10.1063/1.3698303
Journal of Applied Physics, 111, 6, pp. 064318-1-064318-3, 2012-03-26

Photoluminescence fatigue in three-dimensional silicon/silicon-germanium nanostructures

N. Modi, L. Tsybeskov, J.-M. Baribeau, X. Wu, and D. J. Lockwood

Citation: J. Appl. Phys. **111**, 064318 (2012); doi: 10.1063/1.3698303
View online: http://dx.doi.org/10.1063/1.3698303
View Table of Contents: http://jap.aip.org/resource/1/JAPIAU/v111/i6
Published by the American Institute of Physics.

Photoluminescence fatigue in three-dimensional silicon/silicon-germanium nanostructures

N. Modi,[1] L. Tsybeskov,[1,a)] J.-M. Baribeau,[2] X. Wu,[2] and D. J. Lockwood[2]
[1]*Department of Electrical and Computer Engineering, New Jersey Institute of Technology, Newark, New Jersey 07102, USA*
[2]*Institute for Microstructural Sciences, National Research Council, Ottawa, Ontario K1A 0R6, Canada*

(Received 17 December 2011; accepted 22 February 2012; published online 26 March 2012)

We find fatigue of low temperature photoluminescence (PL) in Si/SiGe three-dimensional island morphology nanostructures under continuous excitation. Initially, the PL intensity slowly decreases by less than 15%, and after ∼10 min it decreases rapidly by more than 80%. After the PL intensity stabilizes, a complete recovery requires heating the sample to nearly room temperature. We propose that accumulation of charge within SiGe islands is responsible for the enhancement of Auger recombination and hence the observed PL fatigue. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.3698303]

I. INTRODUCTION

Photoluminescence (PL) fatigue has been reported in porous Si,[1] amorphous Si,[2] and amorphous SiGe,[3] chalcogenide glasses[4] and some other systems. In most cases, the PL intensity degradation under continuous wave (CW) excitation is induced by significant chemical or structural changes that have been detected by infra-red spectroscopy and electron spin resonance.[5] At the same time, PL from individual nanometer size quantum dots (QDs) exhibits a sequence of "on" and "off" periods (also known as the PL intensity blinking), similar to a random telegraph signal.[6] This process is not accompanied by significant structural/chemical modification, and it is attributed to quantum dot ionization and luminescence quenching by non-radiative Auger recombination.[7]

Compared to core-shell II-VI quantum dots, which are typically a few nanometers in diameter and exhibit type I energy band alignment,[8] Si/SiGe island morphology nanostructures, also known as three-dimensional (3 D) Si/SiGe nanostructures, are much larger in size and are expected to have a type II energy band alignment at the Si/SiGe heterointerface with low conduction energy barriers for electrons in Si and much deeper valence energy wells for holes in SiGe.[9] Several experimental effects associated with Auger processes in Si/SiGe 3 D nanostructures were reported[10] (including Auger fountain and hole transfer from SiGe to Si, Ref. 11), but the PL intensity blinking on the time scale of seconds and reversible PL degradation were never reported. It is quite possible, however, that charging of the larger size SiGe islands (with a base of greater than 100×100 nm^2 and height of more than 10 nm) is controlled by slow processes, such as electron diffusion within Si spacer layers and hole redistribution between SiGe islands via tunneling.[12] In this paper, we report on low temperature PL fatigue under steady-state photoexcitation, which shows complex kinetics with a typical time constant of minutes and a significant dependence on excitation intensity and wavelength.

a)Author to whom correspondence should be addressed. Electronic mail: tsybesko@adm.njit.edu.

II. EXPERIMENTAL RESULTS

A number of multilayer Si/SiGe island samples grown by molecular beam epitaxy (MBE) were studied in this work (see Fig. 1; for sample growth details, see Ref. 13). Transmission electron microscopy (TEM) measurements were performed using a 200 kV JEOL JEM 2100 F Advanced Field Emission Electron Microscope. The PL spectroscopy measurements were performed using a 0.5 m Acton Research spectrometer and a thermo-electrically cooled InGaAs photomultiplier tube. The sample was held in a vacuum chamber at temperature $T = 16$ K using a closed cycle cryostat, and excited using a collimated 405 nm high-power light emitting diode (LED), a multiline Ar+ laser and a HeCd laser at a wavelength of 325 nm.

Figure 2 shows two PL spectra recorded at $T = 16$ K under ∼0.1 W/cm^2 excitation intensity: first without prior exposure (initial) and second after ∼20 min of continuous exposure (fatigued). In the initial PL spectrum, two distinct PL features peaked at 0.82 eV and at 0.88 eV can clearly be seen. After continuous exposure, both PL features become less intense, with the 0.82 eV PL peak losing nearly 75% of its initial intensity and the 0.88 eV PL peak losing less than 50%. In the fatigued PL spectrum, the full width at half maximum (FWHM) of both PL features obtained by applying Gaussian fits (not shown) increases significantly, more than 50%. The fatigued PL spectra are not completely restored even after several hours of incubation in the dark at low temperature; a complete recovery requires warming the sample to temperature $T > 250$ K.

The evolution of the PL intensity under 405 nm excitation and two excitation intensities (150 and 15 mW/cm^2) recorded at 0.82 eV is shown in Fig. 3. When irradiated at 150 mW/cm^2, a slow PL intensity degradation is observed for ∼10 min, after which a much faster degradation reduces the PL intensity to ∼10% of its initial value. Under the lower excitation intensity of 15 mW/cm^2, any slow PL intensity degradation is essentially undetectable, and the initial period of relative stability lasts ∼15 min. This is followed by a fast PL intensity degradation, also until ∼10% of the initial

0021-8979/2012/111(6)/064318/3/$30.00

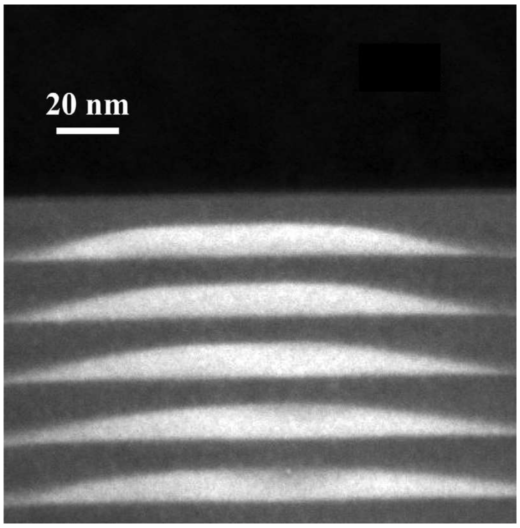

FIG. 1. Transmission electron micrograph (TEM) of a sample with five layers of SiGe islands (lighter areas) embedded into a Si matrix (darker gray area). The SiGe island base is ∼120–150 nm long, and the island height is <20 nm.

PL intensity remains. In both cases, the PL intensity stabilizes after the fast degradation period, and the rate of the fast PL intensity degradation is nearly the same. For the samples studied in this work, the PL fatigue is detectable when using excitation wavelengths $\lambda \leq 405$ nm, but not when using excitation with a wavelength longer than 457 nm.

Figure 4 shows the PL intensity degradation as a function of time under ∼50 mW/cm^2 CW excitation (using 325 nm wavelength) recorded at different temperatures. The PL fatigue is clearly observed at $T \leq 50$ K, and it is not found at $T > 65$ K for up to 30 min of excitation. A similar result was obtained using 405 nm excitation.

III. DISCUSSION

It is well known that the reversible PL degradation associated with photo-induced structural changes requires high

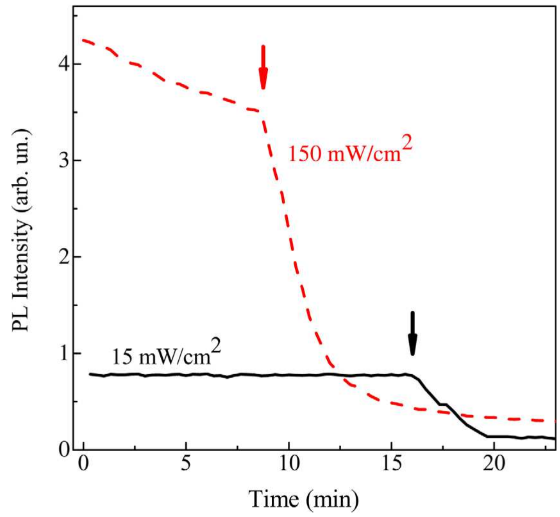

FIG. 3. The PL transients showing the onsets (arrows) of the PL fatigue recorded at 0.82 eV at different (marked) excitation intensities. The excitation wavelength is 405 nm and sample temperature is T = 16 K.

temperature annealing to restore the initial PL intensity.[1–4] Since the observed PL fatigue is a low temperature phenomenon (see Fig. 4), and it can be restored by just warming the sample to $T > 250$ K, we think that this effect is primarily associated with accumulation and retention of charge in SiGe islands. Also, degradation of the PL intensity from an ensemble of QDs is proposed to be due to Auger autoionization, and the PL intensity blinking in individual QDs has been described by a statistical difference in the lifetimes of charged and neutral states.[6] Since Si/SiGe heterointerfaces exhibit a type II energy band alignment, contribution of the Auger processes in overall carrier recombination can be significant even at relatively low carrier concentration, specifically if carriers are strongly localized.[11] Energy band offsets for holes localized in SiGe islands and electrons localized in Si layers, both increase with an increase of Ge content. In Si/SiGe nanostructures, valence energy wells for holes associated with

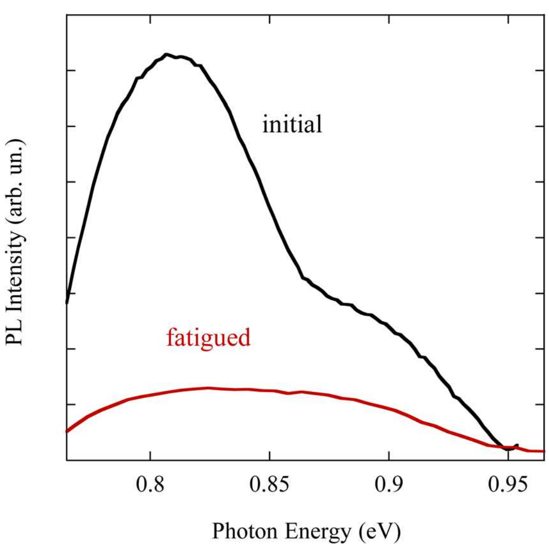

FIG. 2. The PL spectra of Si/SiGe nanostructures under ∼0.1 W/cm^2 of 325 nm excitation observed before (initial) and after (fatigued) continuous exposure for 20 min. The sample temperature is T = 16 K.

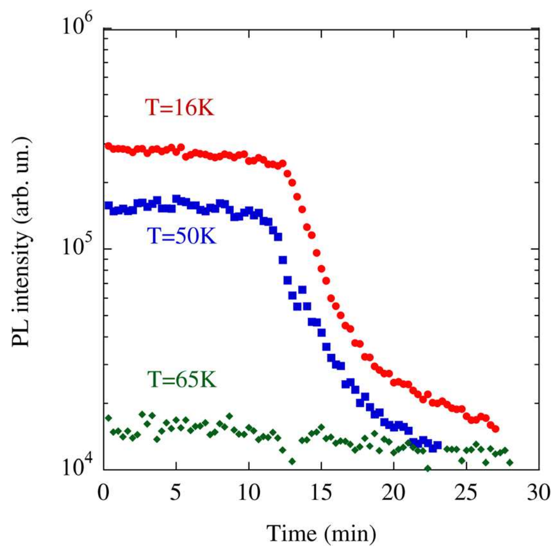

FIG. 4. The PL intensity transients recorded at 0.82 eV under 50 mW power and 325 nm wavelength CW excitation. Measurements are performed at different (indicated) temperatures.

SiGe islands are quite deep (up to ~400 meV, Ref. 10). At low temperatures, holes cannot overcome these energy barriers to diffuse, even under large concentration gradients, and transport of holes is, most likely, limited to tunneling.[12] Electrons however, are only weakly localized at the Si/SiGe heterointerface, and are more mobile compared to holes. In this model, the observed degradation of the PL intensity (or PL fatigue) could be explained due to a net accumulation of holes within SiGe islands and enhancement of Auger recombination.

It has been proposed that the two major PL spectral features observed in Si/SiGe nanoislands (similar to that shown in Fig. 2) correspond to the recombination of excitons localized within regions with different Ge compositions, presumably more Ge for the PL peaked at 0.82 eV and less for the PL peaked at 0.88 eV.[14] We suggest that charge accumulation (most likely, holes captured within SiGe islands) enhances Auger recombination, and that this process should be more effective at Ge-rich regions of SiGe islands (SiGe island core[10–13]) with correspondingly deeper energy wells for holes. The increase in FWHM of the PL peaks could also be caused by SiGe island charging and redistribution of the built-in potential at the Si/SiGe heterointerface.

Compared to holes, electrons are weakly localized, and they may have a fairly unobstructed path for diffusion through the contiguous Si spacing layers. Since we observe the PL fatigue under a short excitation wavelength, surface states can serve as major diffusion sinks for electrons. Also, electron traps at dislocations are known to have a $\sim 10^4$ times greater capture cross section than those for holes;[15] thus, dislocation related traps can capture electrons as well.[16]

The experimentally observed rate of PL degradation is proportional to the number of electron-hole pairs N, and $dN/dt = -\gamma N$. The number of electron-hole pairs is taken to decrease exponentially, $N = N_0 \exp(-\gamma t)$, where N_0 is the initial density of generated electron-hole pairs and γ is the net rate of events that cause the PL degradation. The resulting concentration of accumulating holes is $N_0\,[1 - \exp(-\gamma t)]$. The effective rate of Auger recombination, which increases as positive charge is accumulated in the islands, is given by $C \times N_0 \exp(-\gamma t) \times N_0\,[1 - \exp(-\gamma t)]$, where C is the Auger rate constant. Using the Auger rate constant of $\sim 10^{-30} - 10^{-31}$ cm^6/s (which is similar for both bulk Si and bulk Ge), the above expression is plotted in Fig. 5 with $N_0 \approx 100$ electron-hole pairs per SiGe nanoisland (not necessarily distributed uniformly), and $\gamma \approx 10^{-6}$ (similar to that in Refs. 17 and 18). The rate of radiative recombination is also shown. According to our estimation, the rate of charge accumulation (and Auger recombination enhancement) begins to compete with that of radiative recombination after ~10 min, explaining the onset of the significant PL intensity degradation.

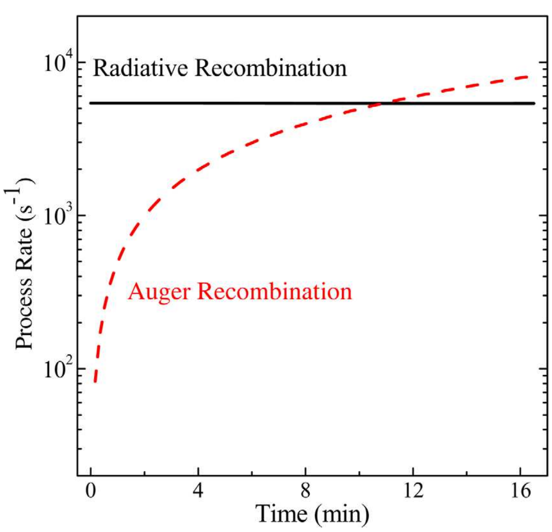

FIG. 5. Evolution of the rates of radiative recombination and Auger recombination in SiGe nanoislands with a volume of $\sim 10^{-16}$ cm^3.

IV. CONCLUSION

In conclusion, we report a low-temperature PL fatigue in Si/SiGe 3 D nanostructures. In contrast to the previously reported PL fatigue phenomena, this process begins after ~10 min of continuous photoexcitation. The observed PL intensity degradation can be fully erased by warming the sample to temperature $T > 250$ K. According to our model, significant decrease in the PL intensity can be explained due to accumulations of holes within SiGe nanoislands and enhancement of Auger recombination.

ACKNOWLEDGMENTS

We thank G. Parent for TEM sample preparation. This work is supported by the National Science Foundation, Hewlett-Packard Research Laboratories Open Innovation Program and Foundation at NJIT.

[1]P. M. Fauchet, J. von Behren, K. D. Hirschman, L. Tsybeskov, and S. P. Duttagupta, Phys. Status Solidi A **165**, 3 (1998).
[2]R. A. Street, Phys. Rev. B **17**, 3984 (1978).
[3]F. Mollot, J. Cernogora, and C. Benoit a la Guillaume, J. Non-Cryst. Solids **35–36**, 939 (1980).
[4]N. F. Mott and E. A. Davis, *Electronic Processes in Non-Crystalline Materials*, 2nd ed. (Oxford University Press, New York, 1979).
[5]P. C. Taylor, U. Strom, and S. G. Bishop, Philos. Mag. B **37**, 241 (1978).
[6]Al. L. Efros and M. Rosen, Phys. Rev. Lett. **78**, 1110 (1997).
[7]D. I. Chepic, Al. L. Efros, A. I. Ekimov, M. G. Ivanov, V. A. Kharchenko, I. A. Kudriavtsev, and T. V. Yazeva, J. Lumin. **47**, 113 (1990).
[8]B. O. Dabbousi, J. Rodriguez-Viejo, F. V. Mikulec, J. R. Heine, H. Mattoussi, R. Ober, K. F. Jensen, and M. G. Bawendi, J. Phys. Chem. B **101**, 9463 (1997).
[9]T. Baier, U. Mantz, K. Thonke, R. Sauer, F. Schaeffler, and H.-J. Herzog, Phys. Rev. B **50**, 15191 (1994).
[10]Leonid Tsybeskov and David J. Lockwood, Proc. IEEE **97**, 1284 (2009).
[11]E.-K. Lee, D. J. Lockwood, J.-M. Baribeau, A. M. Bratkovsky, T. I. Kamins, and L. Tsybeskov, Phys. Rev. B **79**, 233307 (2009).
[12]E.-K. Lee, L. Tsybeskov, and T. I. Kamins, Appl. Phys. Lett. **92**, 033110 (2008).
[13]J.-M. Baribeau, X. Wu, N. L. Rowell, and D. J. Lockwood, J Phys.: Condens. Matter **18**, R139 (2006).
[14]B. V. Kamenev, L. Tsybeskov, J.-M. Baribeau, and D. J. Lockwood, Appl. Phys. Lett. **84**, 1293 (2004).
[15]P. N. Grillot, S. A. Ringel, E. A. Fitzgerald, G. P. Watson, and Y. H. Xie, J Appl. Phys. **77**, 676 (1995).
[16]Similar experiments were performed for samples with detectible (using TEM, etc.) dislocations right underneath the SiGe island multilayers. Only in these samples was the PL fatigue found using a longer wavelength excitation (>457 nm) and where the exciting light penetrates further into the sample.
[17]M. Nirmal, B. O. Dabbousi, M. G. Bawendi, J. J. Macklin, J. K. Trautman, T. D. Harris, and L. E. Brus, Nature **383**, 802 (1996).
[18]T. Krauss and L. Brus, Phys. Rev. Lett. **83**, 4840 (1999).