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Publisher's version / Version de l'éditeur:

<https://doi.org/10.1002/pssc.201084032>

Physica Status Solidi C: Current Topics in Solid State Physics, 8, 9, pp. 2870-
2874, 2011-05-02

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Self-assembled silicon-germanium nanostructures for CMOS compatible light emitters

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Received 3 October 2010, accepted 4 February 2011

Published online 28 April 2011

Keywords silicon, germanium, nanostructures, photoluminescence

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To be commercially valuable, light emitters based on SiGe nanostructures should be efficient, fast, operational at room temperature, and be compatible with the CMOS technology. Also, the emission wavelength should match the optical waveguide low-loss spectral region of 1.3–1.6 μm . Among other approaches, epitaxially-grown Si/SiGe quantum dot/quantum well complexes produce efficient photoluminescence and electroluminescence in the re-

quired spectral range. Until recently, the major road-blocks for practical applications of these devices were strong thermal quenching of the luminescence quantum efficiency and a long carrier radiative lifetime. The latest progress in the understanding of physics of carrier recombination in Si/SiGe nanostructures is reviewed, and a new route toward CMOS compatible light emitters for on-chip optical interconnects is proposed.

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1 Introduction Optical interconnects in the form of fiber optics have been used for many years in different long-distance communication applications. With the microprocessor clock speed approaching 10 Gbps, optical interconnects are now being considered for on-chip interconnects as an alternative to metal wires with their unavoidable RC delay, significant signal degradation, high power dissipation, and electromagnetic interference [1]. Two major avenues toward optical interconnects on a chip include a hybrid approach with densely packaged III-V optoelectronic components [2, 3] and the all-group-IV approach (mainly Si, Ge and SiGe), where all the major components, e.g., light emitters, modulators, waveguides and photodetectors, are monolithically integrated into the CMOS environment [1–3]. For several decades, the efforts on obtaining light emission from group-IV semiconductors have been mainly focused on porous Si, Si/SiO₂ superlattices, Si nanoprecipitates in SiO₂, erbium in Si, Si/Ge quantum wells, and, more recently, iron disilicide [2, 4]. However, no approach has so far been applied commercially. There are several reasons, including the lack of a genuine or perceived compatibility with conventional CMOS technology, the long carrier radiative lifetime in Si-

based nanostructures (NSs), and, especially in the case of near-infrared emitters, the significant thermal quenching of the luminescence quantum efficiency [4].

By the 1990s, a different form of SiGe NS, namely the three-dimensional (3D) self-assembled system produced by the Stranski-Krastanov growth mode in lattice mismatched materials, had been demonstrated [5]. It has been shown that dislocation-free SiGe growth can be achieved using a higher temperature ($\geq 600^\circ\text{C}$), and that the non-planar geometry is mainly responsible for the significant increase of the SiGe critical layer thickness [5]. It has also been found that, compared to two-dimensional (2D) Si/SiGe NSs, the PL and electroluminescence (EL) quantum efficiency in 3D Si/SiGe NSs is higher (up to $\sim 1\%$), especially for $T > 50\text{ K}$ [6–9]. Despite many successful demonstrations of PL and EL in the spectral range of 1.3–1.6 μm , which is important for optical fiber communications, the proposed further development of 3D Si/SiGe based light emitters was discouraged by several studies indicating a type II energy band alignment at Si/SiGe heterointerfaces [10], where the spatial separation of electrons (located in Si) and holes (localized in SiGe) was thought to make carrier radiative recombination very inefficient. Later, it was also

shown that 3D Si/SiGe NSs exhibit an extremely long ($\sim 10^{-2}$ s) luminescence lifetime [11], which is of the order of a million times longer than in III-V semiconductors and their NSs. Thus, according to this analysis, 3D Si/SiGe NSs cannot be used to achieve efficient and commercially valuable light emitting devices. Here, we show that despite these facts it is still possible to obtain conditions favorable for efficient carrier radiative recombination in SiGe NSs.

2 Light-emitting properties In semiconductor materials with strong selection rule relaxation (e.g., bulk alloys), carrier recombination provides a higher PL quantum efficiency compared to that in an indirect band gap semiconductor such as bulk Si and Ge. PL measurements in Si/SiGe NSs reveal a significantly enhanced intensity ratio between no-phonon (NP) luminescence and phonon-assisted luminescence compared to bulk Si [5]. Thus, systematic studies of the PL spectra in 3D Si/Si_{1-x}Ge_x NSs with control over the average Ge atomic concentration x provide very important information regarding changes in the carrier recombination mechanism (e.g., selection rule relaxation, conduction and valence band alignment, etc.) as x increases from 0 (bulk Si) to $\sim 55\%$, which is the highest Ge composition in structurally defect-free SiGe clusters [5].

The PL spectrum in samples grown by molecular beam epitaxy (MBE) with an average Ge composition close to 50% depicts a broad feature with a major PL peak centered at 0.75 eV; this peak energy is close to the bandgap of bulk Ge at 4 K. A second PL peak is found at ~ 0.85 eV. Both PL bands are quite broad, most likely due to compositional disorder, which is in agreement with the Raman scattering measurements [9]. There are no characteristic phonons in the Si/SiGe system with an energy of ~ 100 meV, and it is thus reasonable to assume that the observed PL bands are associated with carrier recombination within two different regions of the 3D SiGe NSs. The PL band peaked at 0.85 eV has a PL quenching activation energy of ~ 20 meV while the PL intensity as function of excitation intensity is linear over a wide range of excitation intensities. The second PL band peaked at 0.75 eV has a sublinear excitation dependence and an activation energy of ~ 60 meV and is nearly temperature independent up to 100 K. Because of its high quantum efficiency, it can be monitored almost up to room temperature. This data suggests that 3D Si/Si_{1-x}Ge_x NSs with $x \approx 0.50$ contain coupled subsystems with different (lower and higher) Ge concentrations.

In contrast to MBE grown samples, chemical vapour deposition (CVD) growth of 3D Si/SiGe NSs does not provide precise control over the atomic composition, and can produce more Si/SiGe interdiffusion at heterointerfaces [5]. This is reflected in PL spectra, where no fine structure has been found. However, the broad and asymmetric PL peak is well fitted by two Gaussian bands, often identified as the NP and TO phonon lines [12] and separated by an energy of ~ 43 meV, which is close to the energy of characteristic SiGe phonons (see Fig. 1). Thus, there is at least a qualitative similarity between PL spectra in MBE and CVD

grown 3D Si/SiGe NSs. The PL efficiencies in MBE and CVD grown samples are comparable ($\geq 1\%$ at low excitation intensity). The same sub-linear (close to square root) dependence on excitation intensity (on a log-log plot) for PL associated with Ge-rich SiGe clusters has been found in nearly all 3D Si/SiGe NSs grown by both CVD and MBE [13].

Figure 1 (a) Normalized PL spectra in CVD grown Si/SiGe 3D NSs showing the PL spectral shift to higher photon energy under increasing excitation intensity. Each spectrum can be fitted with two (NP and TO) Gaussian spectral bands, as shown, for example, by the dashed lines under trace (1). (b) Summary of PL spectra changes as a function of excitation intensity.

The PL spectra in 3D Si/SiGe NSs, which is similar to that in III-V quantum wells with type II energy band alignment, exhibit a blue shift as the excitation intensity increases. This effect is found in both MBE and CVD grown samples. Figure 1 shows PL spectra in a CVD grown sample measured under different excitation intensities. At the lowest excitation intensity used (0.1 W/cm^2), the PL peaks at ~ 0.8 eV. With increasing excitation intensity, a continuous almost-parallel PL blue shift of 30–40 meV per decade of excitation intensity increase is observed. At an excitation intensity of 10 W/cm^2 , the PL peak reaches ~ 0.92 eV. Under photo-excitation of $1\text{--}10 \text{ kW/cm}^2$, the low energy part of the PL spectrum does not shift further, while the high energy part continues shifting towards higher energy. A modified Arrhenius plot of the normalized integrated PL intensity as a function of temperature for the different excitation intensities can be fitted with two thermal quenching activation energies E_1 and E_2 (Fig. 2). In all measurements of all samples, the PL thermal quenching activation energy $E_1 \sim 15$ meV and is independent of excitation intensity. In contrast, the activation energy E_2 depends significantly on the excitation intensity: the PL temperature dependence shows a step-like behavior, and E_2 increases dramatically from ~ 120 to 340 meV as the excitation intensity increases from 0.1 to 10 W/cm^2 .

Figure 2 Typical integrated PL intensity for CVD grown samples as a function of the reciprocal temperature measured under different excitation intensities, as indicated. The activation energies obtained from fits (lines) to the data (points) are also given.

The PL dynamics under pulsed laser excitation reveal important information regarding the carrier recombination mechanism. Figure 3 shows the normalized low-temperature PL decays collected from a CVD grown Si/SiGe 3D sample. The initial PL decay is fast, close to the resolution of our detection system (<20 ns). The longer-lived PL shows a strong dependence on the detection photon energy: the PL lifetime at photon energies below 0.8 eV is found to be ~ 20 μ s and then drastically decreases to ~ 200 ns for the PL measured at 0.89 eV. More complicated and even faster ($\sim 10^{-7}$ s) decays are observed at very high excitation intensities [14].

Figure 3 Low temperature PL dynamics for a CVD grown sample measured at the indicated photon energies using a ~ 6 ns excitation pulse.

3 Carrier recombination In discussing carrier recombination in Si/SiGe NSs, we focus on MBE and CVD grown samples with an average Ge atomic composition $\sim 50\%$. These samples show the highest observed PL quantum efficiency with a PL peak wavelength close to 1.5–1.6 μ m. The PL spectral distribution extending well below the bandgap of pure Ge and the extremely long carrier radi-

ative lifetime of ~ 10 ms [11], as well as the ~ 30 meV per decade PL spectral shift toward higher photon energies as the excitation intensity increases, point out strong similarities between the PL in 3D Si/SiGe NSs and the PL in III-V quantum wells with type II energy band alignment [15]. Generally, a type II energy band alignment at the hetero-interface is a strong disadvantage for light emitting devices due to a weak overlap between spatially separated electron and hole wave functions. In reality, however, the critical limitation in the efficiency of light-emitting structures is rather the presence of competing non-radiative recombination channels for excess electrons and holes. The most important non-radiative mechanism is carrier recombination via defects, especially hetero-interface structural defects such as propagating dislocations and dislocation complexes. The 3D Si/SiGe NSs investigated here show an almost undetectable density of dislocations. Thus, at low excitation intensities, we find a high quantum efficiency of PL with photon energy < 0.9 eV, which is associated with carrier recombination in the vicinity of SiGe clusters.

It has already been suggested that SiGe Stranski-Krastanov (S-K) clusters with a small (3–5 nm) height and $\sim 10:1$ base-height aspect ratio can be modeled as NSs with a type II energy band alignment and possible SiGe cluster valence-band energy quantization in the direction of growth [10]. Strained Si and Si-rich SiGe alloy regions near the base of the clusters (also called SiGe wetting layers) also need to be considered. Including the effect of strain, the observed PL bands at 0.916 and 0.972 eV indicate a composition of the $\text{Si}_{1-x}\text{Ge}_x$ transition region, which is located near the bottom of the Ge/Si pyramid-like clusters, to be close to $x = 0.2$, and this conclusion is supported by recent direct analytical TEM measurements [16].

It has also been proposed that the broad PL band with peak energy ~ 0.8 – 0.9 eV is due to the recombination of carriers localized in the Ge-richest areas of the clusters, which is close to the center of “pancake” shaped SiGe clusters [16]. We suggest that at the lowest excitation intensity, the PL arises from electron-hole recombination between holes localized in the Ge-richest regions of the cluster and electrons localized in the strained SiGe alloy region near the cluster base. This immediately explains the extension of the observed PL spectrum below the pure Ge band-gap energy.

The observed excitation-independent PL thermal quenching activation energy of ~ 15 meV is close to the exciton binding energy in SiGe alloys and Si/SiGe superlattices. Thus we conclude that one of the mechanisms of PL thermal quenching is the thermal dissociation of excitons. The activation energy of ~ 15 meV can therefore be associated with exciton localization on specific regions of the clusters associated with variations of the SiGe composition. Hence, the non-uniform SiGe cluster composition and, perhaps, variations in SiGe cluster size and shape could be responsible for the observed relatively broad PL spectra.

Using the model discussed above of type II energy band alignment at the Si/SiGe heterointerface, we focus

next on non-radiative carrier recombination and the different mechanisms of electron-hole separation. Electron transport in 3D Si/SiGe NSs is limited by a small (≤ 10 – 15 meV) conduction band energy barrier and SiGe compositional disorder [17]. Thus, the PL thermal quenching activation energy of ~ 15 meV could also be associated with a small conduction band energy barrier for electrons in Si/SiGe 3D NSs.

In contrast to a low energy barrier for electrons, hole diffusion in 3D Si/SiGe multilayer NSs with a high Ge content is controlled by large (> 100 meV) valence band energy barriers at Si/SiGe heterointerfaces. In this system, we consider two major mechanisms of hole transport: hole tunneling and hole thermionic emission. Hole tunneling in 3D Si/SiGe NSs with thin (5–7 nm) Si separating layers and nearly perfect SiGe cluster vertical self-alignment could be very efficient. These NSs are usually grown by MBE and exhibit a PL thermal quenching activation energy of ~ 60 meV. The same PL thermal quenching activation energy is found in our CVD-grown samples with 7.5 nm thick Si separating layers for the lowest excitation intensity. We suggest that in 3D Si/SiGe multilayer NSs with thin Si layers at low excitation intensity, the electron-hole separation and non-radiative carrier recombination are mainly controlled by hole tunneling between SiGe clusters. Due to significant variations in SiGe cluster size, shape and chemical composition, the process of hole tunneling could be assisted by phonon emission and/or absorption [13]. Therefore, the observed PL thermal quenching activation energy is close to the Si TO phonon energy. In 3D Si/SiGe multilayer samples with 20 nm thick Si layers, where SiGe cluster vertical self-alignment is practically absent [5, 16], the probability of hole tunneling is reduced, and hole thermionic emission over the Si/SiGe heterointerface barrier is playing a bigger role. Thus, in these samples the PL thermal quenching activation energy is expected to be greater, as has been found in our experiments.

In this simple model, efficient hole tunneling between adjacent SiGe nanoclusters requires not only reasonably low and thin Si barriers but also a low carrier concentration (i.e., a large enough number of empty adjacent SiGe clusters). By increasing the photo-excitation intensity (i.e., the number of photogenerated carriers), hole tunneling can effectively be suppressed since fewer empty adjacent SiGe clusters can be found. At high excitation intensity, assuming a negligible value of the conduction band offset compared to that in the valence band and a nearly pure Ge composition in the SiGe cluster core, the maximum anticipated PL thermal quenching activation energy should be ~ 400 meV. This value sets the upper limit of the activation energy of PL intensity thermal quenching in 3D SiGe multilayer NSs, and it is close to the activation energy of E2–340 meV that has been found under the highest excitation intensity.

The same model also explains the experimentally observed strong decrease in carrier radiative lifetime (~ 100 times) as the detection photon energy increases from 0.77

to 0.89 eV (Fig. 3). Under a low level of excitation intensity, holes are localized within the Ge-rich core of SiGe clusters, and the hole wave function does not penetrate into the Si barriers. Thus, in this quasi-type II energy band alignment, electron and hole wave functions do not overlap, causing a long carrier radiative lifetime. Under high level excitation, holes occupying the excited energy states in small size SiGe clusters, as well as holes leaking into the SiGe wetting layer with a lower Ge concentration, have their wave function significantly extended into the Si barriers. Therefore, a stronger overlap between electron and hole wave functions is responsible for a shorter radiative lifetime, i.e., a higher PL quantum efficiency at greater photon energy. Thus, this explanation is consistent with both, the decrease of the PL decay time at greater photon energy and the PL spectral blue shift as excitation intensity increases.

Figure 4 depicts the anticipated quasi-type II energy band alignment at Si/SiGe heterointerfaces, where a gradual increase of Ge concentration toward the SiGe cluster core has recently been experimentally verified [5, 16]. It also shows five major processes controlling electron-hole recombination in 3D Si/SiGe NSs with a Ge-rich core:

(1) radiative recombination between electrons localized at the Si/SiGe heterointerface and holes localized within the Ge-rich core of a SiGe cluster—this slow recombination is associated with the lower photon energy part of the PL band;

(2) radiative recombination between electrons localized at the Si/SiGe heterointerface and holes localized in excited states in small-size SiGe clusters as well as holes leaking into the Si-rich outer part of the cluster or/and SiGe wetting layer—this faster carrier recombination channel is responsible for the higher photon energy part of the PL spectrum;

(3) non-radiative carrier recombination due to electron-hole separation via hole tunneling in Si/SiGe NS samples with thin (< 10 nm) Si barriers;

(4) non-radiative carrier recombination due to electron-hole separation via hole thermionic emission in samples with thicker (> 15 – 20 nm) Si barriers;

(5) non-radiative carrier recombination due to Auger processes.

It has been pointed out that possible transformations of hole energy spectra due to quantization and/or strain might dramatically increase the rate of non-radiative Auger recombination in SiGe quantum wells by more than 100 times [18]. Thus, the ‘Auger limit’ in Si/SiGe NSs could be considerably lower compared to that in bulk Si and Ge. On the other hand, the Auger process competes with radiative processes, and their characteristic lifetimes are $\sim 10^{-3}$ s in bulk Si and Ge and just $\sim 10^{-7}$ s in the high-photon-energy part of the PL spectra in 3D Si/SiGe NSs [14]. Therefore, more work needs to be done to understand the important details of Auger processes in Si/SiGe NSs.

Figure 4 also reflects a continuous change in the Ge atomic concentration by a gradually increasing energy

band gap from the cluster center toward the cluster edge and SiGe wetting layer where the Ge atomic concentration is estimated to be $\sim 20\%$. The proposed energy band diagram and carrier recombination/diffusion mechanisms explain the experimental observations of (i) the PL spectral blue shift under increasing excitation intensity; (ii) the dramatic (~ 100 times) decrease in carrier radiative lifetime measured at photon energies from 0.77 to 0.89 eV; and (iii) the unusual PL intensity temperature dependence, which shows a different PL thermal quenching activation energy at different excitation intensities. This schematic representation also points out that moderate excitation intensity changes the overlap of electron-hole wave functions and allows a faster carrier radiative recombination. In other words, the type II energy band alignment at the Si/SiGe hetero-interface can effectively be replaced by the “dynamic type I” alignment, where the electron and hole spatial separation no longer controls the recombination rate and the quantum efficiency of PL and EL.

Figure 4 Schematic of the Si/SiGe cluster multilayer energy band diagram with different electronic transitions: 1 - slow electron-hole recombination due to spatial separation and a weak wave function overlap; 2 - a faster recombination, presumably involving holes at excited energy states in SiGe clusters; 3 - hole diffusion due to cluster-to-cluster tunneling; 4 - hole diffusion due to thermionic processes; and 5 - Auger hole excitation with possible charge transfer.

4 Conclusion Comprehensive experimental studies of the light-emitting properties of 3D Si/SiGe NSs show that these nanostructures emit light at the technologically important 1.3–1.6 μm wavelength region. The highest PL (and EL) quantum efficiency is found in SiGe clusters with a $\sim 50\%$ Ge composition near the cluster core. Using time resolved PL measurements, it has been found that within the broad PL band, the part of the PL spectra at higher photon energies exhibits a ~ 100 times faster radiative transition, which is explained by a proposed model where radiative recombination occurs between holes occupying excited states in SiGe clusters and electrons mostly localized at Si/SiGe interfaces. This fast PL is less susceptible to Auger-induced intensity saturation as the excitation intensity increases. In addition, this PL exhibits an improved intensity temperature dependence: the PL (and EL) can easily be extended up to room temperature. These experimental observations suggest that by controlling and modifying the composition of Ge-rich SiGe clusters (and possibly

other types of Si/SiGe 2D and 3D NSs) it is possible to fabricate a more efficient SiGe light-emitting device. Despite the challenges associated with their integration into the traditional CMOS environment, the proven compatibility between Si/SiGe and conventional CMOS technology suggests that this task is not an impossible one.

Acknowledgements We acknowledge the invaluable contributions from our many collaborators and especially B. Kamenev, J.-M. Baribeau, X. Wu, and T. Kamins. We thank US National Science Foundation, Intel Corporation, Semiconductor Research Corporation, and Foundation at NJIT for financial support.

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