Photoluminescence and electroluminescence processes in $Si_{1-x}Ge_x$/Si heterostructures grown by chemical vapor deposition

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Abstract

Well-resolved band-edge exciton photoluminescence has been observed in strained $Si_{1-x}Ge_x$ quantum wells on Si(100). A growth technique which provides material with a low density of non-radiative centers and a uniform microstructure is necessary for observing such luminescence. The luminescence exhibits a characteristic no-phonon line due to the alloy randomness, and a threefold splitting of the transverse optical phonon mode due to different nearest neighbor interactions. The luminescence has been observed from 2 to 300 K, and can also be electrically pumped (electroluminescence) to over 300 K, with peak emission from 1.3 to 1.5 μm.

1. Introduction

Very-large-scale integrated circuits are made in silicon because of the excellent manufacturability of silicon devices and resulting low defect density over large areas. The intrinsic electrical and optical properties of silicon are rather poor, however (low mobilities, indirect band gap, etc.). Therefore considerable effort has been focused over the last 10 years on developing a silicon-based heterojunction technology for overcoming these inherent limitations of silicon integrated circuits, with the most effective approach being that of commensurately strained $Si_{1-x}Ge_x$ alloys on Si(100) substrates. Heterojunction bipolar transistors made in this material system at present exhibit f_T values in excess of 90 GHz at room temperature [1].

Although the growth of strained $Si_{1-x}Ge_x$ on Si(100) has been actively pursued by many groups for nearly a decade, it is only within the past 3 years that well-resolved band-edge exciton luminescence has been observed in this material. In 1990, exciton photoluminescence (PL) was reported from a fairly thick strained layer with only 4 at.% Ge ($x = 0.04$) [2]. In 1991, such PL was clearly observed for the first time in quantum wells (QWs) and superlattices (with $x \approx 0.2$) [3]. These latter samples were grown not by molecular beam epitaxy, as in most of the previous Si–Ge luminescence work, but rather by rapid thermal chemical vapor deposition (RTCVD). This paper reviews the RTCVD growth technique and the resulting Si–Ge layer PL and electroluminescence (EL) from 2 to 300 K

and then finally suggests some future directions for continuing research.

2. Rapid thermal chemical vapor deposition

RTCVD is a straightforward combination of rapid thermal processing with conventional chemical vapor deposition (CVD) (Fig. 1). A single 100 mm silicon wafer is suspended on quartz pins inside a load-locked quartz reaction tube and is heated by a bank of tungsten–halogen lamps located outside the reactor chamber. The typical source gases are dichlorosilane, germane, diborane and phosphine in a hydrogen carrier, and growth temperatures vary from 500 to 1200 °C. (Unless otherwise noted, all results in this paper are for $Si_{1-x}Ge_x$ grown at 625 °C and Si at 700 °C, at a pressure of 6 Torr.) The reactor has several unusual features which are important to the growth of high quality films. First, although the reactor is not of an "ultrahigh vacuum" type (O-ring seals are used and only a mechanical rotary vane pump is used in normal operation), through the use of the load-lock and ultrahigh purity gas handling, high quality epitaxial layers with low oxygen concentrations (10^{18} cm^{-3} or less) can be achieved at growth temperatures as low as 550 °C. (Oxygen contamination (concentrations levels in excess of 10^{18} cm^{-3}) and related defects are a common problem in silicon-based CVD at temperatures below 900 °C.) Second, there is no possible source of extrinsic contamination (such as metal wafer heaters
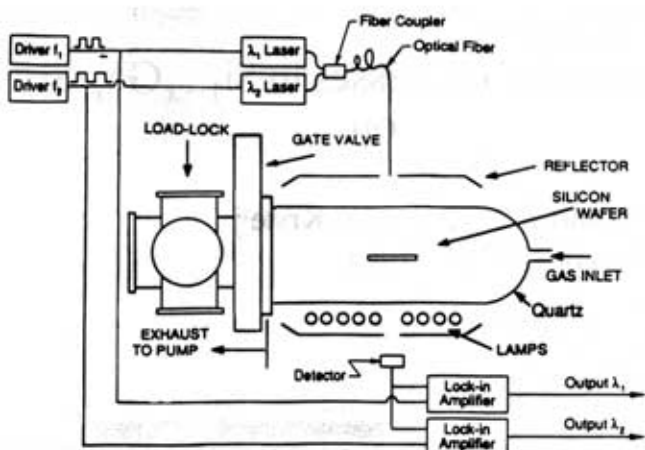
Fig. 1. Schematic cross-section of the RTCVD reactor with temperature measurement by IR transmission.
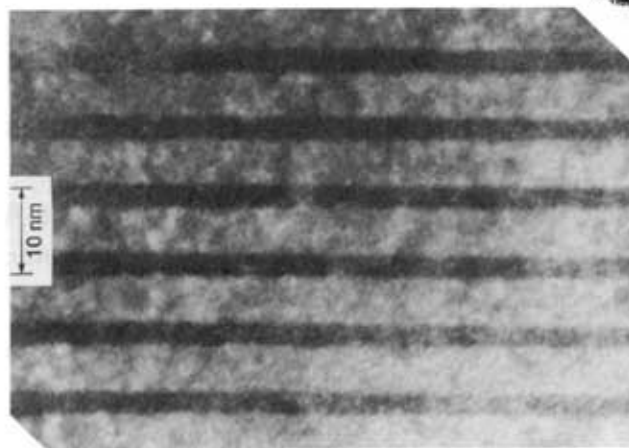

Fig. 2. Cross-section TEM of a Si(5 nm)/$Si_{0.8}Ge_{0.2}$(3 nm) MQW sample grown by RTCVD. All Si layers were grown at 700 °C and all $Si_{1-x}Ge_x$ layers at 625 °C.

and susceptors) inside the reactor chamber, so that films with very low densities of midgap states and high carrier lifetimes (*i.e.* low non-radiative recombination rates) can be grown. Third, the ability to change the sample temperature rapidly (about 100 K s^{-1}) gives the ability to optimize the growth temperature for each layer and interface. For example, substrate cleaning can be done at 1000 °C in H_2, $Si_{1-x}Ge_x$ can be grown at 625 °C or less (to avoid strain relaxation and island effects), and Si layers can be grown at a higher temperature (700 °C) to achieve a faster growth rate. (A catalytic reaction during $Si_{1-x}Ge_x$ growth allows a much higher growth rate (10–100 times) than with Si alone [4, 5].) Finally, the wafer temperature is measured *in situ* with an accuracy of a few kelvins, without the need for any adjustable parameters such as emissivity, by measuring the transmission of the substrate at 1.3 and 1.5 μm [6]. Figure 2 is a cross-section transmission electron micrograph (TEM) of a multiple quantum well (MQW) consisting of alternating layers of 5 nm Si and 3 nm $Si_{0.8}Ge_{0.2}$, where all Si layers were grown at 700 °C and all $Si_{1-x}Ge_x$ layers were grown at 625 °C. From this TEM it is evident that the interface abruptness is 1 nm or less. Furthermore, it is clear that repeatable temperatures can be achieved, as reflected in the constant thicknesses (and hence growth rates) of the multiple Si and $Si_{0.8}Ge_{0.2}$ layers, with the IR transmission technique for temperature measurement. Further growth details have been given in ref. 7.

3. Photoluminescence

Characteristic 2 and 77 K PL spectra of a single Si/$Si_{0.8}Ge_{0.2}$(33 Å)/Si QW is shown in Fig. 3. Although most of the carriers are generated deep in the substrate,
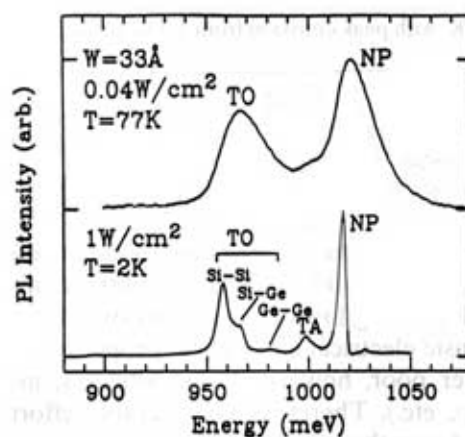

Fig. 3. 2 and 77 K PL spectra of an Si/strained $Si_{1-x}Ge_x/Si$ QW of width 33 Å.

they can travel about 1 μm even at 2 K to the quantum well before generating luminescence [8]. By comparison with previous work in bulk unstrained $Si_{1-x}Ge_x$ [9], the PL features observed at 2 K may be easily identified. The highest energy signal is a no-phonon (NP) signal despite the fact that the alloy has an indirect band gap. This is due to the alloy randomness which breaks the translational symmetry of an ideal lattice, relaxing the requirement for conservation of crystal momentum. The lower energy features are momentum-conserving phonon replicas, the most significant of which is the transverse optical (TO). While a single TO line is seen in silicon, in the alloy three lines are seen because of the different nearest-neighbor modes (Si–Si, Si–Ge and Ge–Ge). The relative height of the different TO replicas depends on the composition as expected [9] but is different in narrow QWs compared with the bulk because of wavefunction pene-

tration into the barriers [3]. Except for thermal broadening, similar features are observed at 77 K. This shows that the NP signal is indeed an intrinsic property of the alloy and not due to any localization effects at low temperature (such as a bound exciton).

Similar spectra (except for a red shift due to an absence of quantum confinement) have been measured in wide structures with wider wells (e.g. 500 Å) [8]. This shows that the NP line is not due to any confinement effects. Similar spectra have also been obtained from ten isolated QWs (about 30 Å, $Si_{0.8}Ge_{0.2}$) and in $Si_{0.8}Ge_{0.2}/Si$ superlattices (period, about 40 Å), showing that superlattice effects are clearly not required to achieve luminescence (Fig. 4).

The temperature dependence is characterized first by a transition from a bound exciton to a free exciton in the range 10–20 K [3]. Above 30 K, there is little intensity change until over 100 K. Above 100 K, the PL decays exponentially with the onset of the decay, increasing with increasing valence band offset. Figure 5 shows the peak NP PL signal vs. temperature for two single QW samples, with $x = 0.2$ and $x = 0.35$. The decay at high temperatures is governed by the
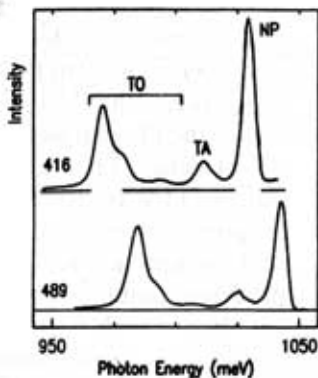
increased non-radiative recombination of carriers and has been found to be controlled by the thermal escape of carriers from the QW to a region of extremely low lifetime in the cladding Si layers, such as the top surface [10]. Therefore a large Ge fraction (about 0.3) is required to achieve room temperature luminescence.

4. Electroluminescence

The band-edge recombination mechanism can also be electrically pumped by putting the $Si_{1-x}Ge_x$ QWs inside the i region of a p–i–n diode. Early results with $x \approx 0.2$ showed that this EL decayed sharply with increasing temperature above 100 K, consistent with the PL results in Fig. 5 [11]. By increasing x to 0.35 (ten QWs), the high temperature performance can be improved. Figure 6 shows the PL of such a structure before processing and EL of the finished device at a heat sink temperature of 80 K. The 4 K PL is not as well resolved as that presented in the earlier figures, possibly owing to some differences between each of the ten wells, some inhomogeneity, or electric field effects. Nevertheless the clear NP and TO replica signature of band-edge recombination is evident, and the NP energy corresponds approximately to that expected for the given composition. The 77 K PL is similar to that expected (e.g. Fig. 3), although the blue shift is not entirely understood [12]. The EL observed at a heat sink temperature of 80 K (junction temperature somewhat higher) also shows an NP peak together with a low energy TO shoulder, providing evidence that the EL and PL generation mechanisms are the same (i.e. band-edge recombination). At a heat sink temperature of 300 K, the NP peak is still clearly evident, with a peak at 1.3 μm (Fig. 7). Some emission from the Si cladding is also evident at this higher temperature, but it accounts for less than 10% of the emitted power. Above a threshold of about 10 mA, the light emission

Fig. 4. Comparison of the PL spectra of a 50-period $Si_{0.8}Ge_{0.2}/Si$ (40 Å period) superlattice with that of ten isolated $Si_{0.8}Ge_{0.2}$ QWs (width, about 30 Å), $T = 2$ K.

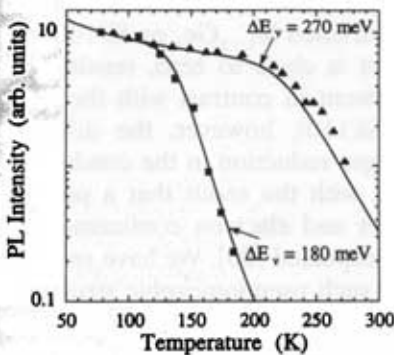Fig. 5. NP luminescence vs. temperature for single $x = 0.2$ (■) and $x = 0.35$ (▲) QWs, —— model.
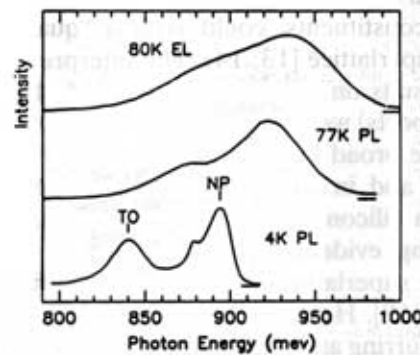

Fig. 6. 4 and 77 K PL spectra of the EL sample before processing, and the EL spectrum for 10 mA drive current and a heat sink temperature of 80 K.
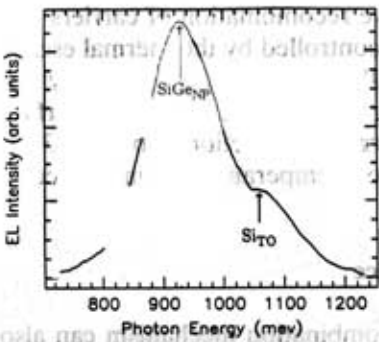
Fig. 7. EL spectra for a drive current of 10 mA at a heat sink temperature of 300 K.

Fig. 8. PL (4 and 77 K) and EL (90 mA drive current; 80 and 300 K heat sink) for the 10 Å Ge layer.

from a 60 μm × 60 μm diode increases linearly with increasing current, with an estimated lower limit to the internal quantum efficiency at 60 mA of 0.03% [12].

We have also achieved a peak room temperature EL at 1.5 μm in a structure which consists of a single layer of Ge of 10 Å thickness, surrounded by Si cladding (Fig. 8). The exact microstructure of the sample is not yet known, and the layer may contain islands or have interdiffused with the silicon. Although the PL is of comparable strength with the Si$_{1-x}$Ge$_x$ ($x \leqslant 0.35$) shown earlier, the EL is an order of magnitude weaker. Furthermore, the broad linewidth of the 4 K PL (full width at half-maximum, 80 meV) prevents clear identification of the PL mechanism. It is possible that the EL and PL in this sample are due to defects and not to band-edge carriers.

5. Future directions

Further increases in light emission from Si$_{1-x}$Ge$_x$/Si structures will require increasing the radiative recombination rate or decreasing the competing (and dominant) non-radiative recombination. One proposed approach for increasing the radiative rate is the use of short-period "zone-folded" Si/Ge superlattices. It has been proposed that such structures, created out of indirect-band-gap constituents, could yield a "quasi-direct" band gap superlattice [13, 14]. The interpretation of early PL results on such superlattices [15, 16] (and many other reports) were controversial [17], however, because of the broad linewidths, energies below expected band gaps and, in some cases, energies corresponding to known silicon dislocation luminescence. Recently, convincing evidence of band-edge PL in short-period Si/Ge superlattices of improved quality has been observed [18]. However, whether zone-folding effects were occurring and were responsible for the PL is not clear.

As an alternative to increasing the radiative rate, one can attempt to reduce the non-radiative rate. In prin-
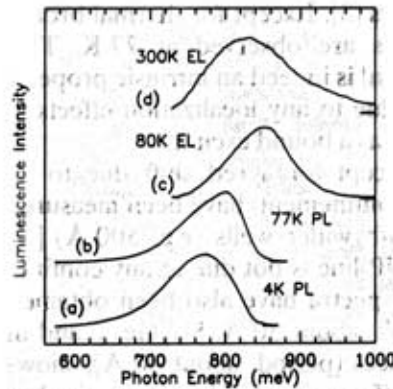
ciple this can be done by localizing excitons to prevent their finding non-radiative sites. Localization of bound excitons at shallow impurities (at 2 K, for example) is not acceptable because a bound exciton contains three carriers, and hence non-radiative Auger recombination occurs. Excitons may also be localized at areas where there are local fluctuations in band gap (locally higher Ge concentration), and the probability of such a region (about (50 Å)^3) containing impurities or defects is very low. At low pump densities we have obtained evidence of this localization, such as the low energy signal in Fig. 9. Note that the energy of the signal is less than that of the average band gap represented by the PL energies at higher pump powers (bound exciton signal). The radiative efficiency is indeed much larger (11% vs. 10^{-2}%) and the lifetime is much slower (greater than 1 ms vs. about 1 μs) compared with bound excitons at 2 K [19]. This feature saturates at only moderate pump powers, however, because of the finite number of such fluctuations. Intentionally creating such localizing regions with higher confining potential may be an option towards even higher efficiencies and higher temperatures.

Yet another approach involves growth on different substrate orientations. Because of the biaxial strain component in pseudomorphic layers, the crystal orientation strongly affects the band structure of the strained layer. For example, in strained Si$_{1-x}$Ge$_x$ on Si(100) the conduction band offset is close to zero, resulting in poor electron confinement in contrast with the well-confined holes. On Si(110), however, the different symmetry causes a larger reduction in the conduction band in the Si$_{1-x}$Ge$_x$, with the result that a positive conduction band offset and electron confinement to the Si$_{1-x}$Ge$_x$ well are expected [20]. We have recently succeeded in growing such pseudomorphic structures on Si(110) by RTCVD at 625 °C, and strong PL was observed (Fig. 10). Most significantly, a substantially larger NP line (compared with the TO line) is observed compared with that seen on (100) substrates (e.g. Figs.
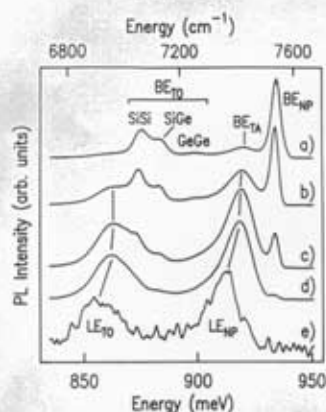
Fig. 9. 2 K luminescence for different pump powers for a single Si$_{0.8}$Ge$_{0.2}$ QW. At low pump power, a low energy (LE) feature (with a NP line and TO replica) due to excitons localized by alloy fluctuations is present, with an efficiency about 10^3 times higher than the regular band edge signal. BE represents the bound exciton signal, T = 2 K.
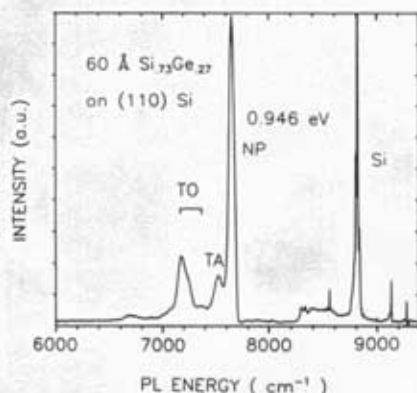

Fig. 10. 2 K luminescence from a single 60 Å quantum well of Si$_{0.7}$Ge$_{0.3}$ on Si(110) (a.u., arbitrary units).

3 and 4). Whether this effect is due to a higher electron density in the alloy region or other effects is not yet understood, however.

6. Summary

Well-resolved band-edge luminescence has been observed in Si$_{1-x}$Ge$_x$/Si structures grown by RTCVD. The NP emission mechanism, which is due to alloy randomness, provides a radiative process which can be up to an order of magnitude larger than those found in silicon. Furthermore, the confining effect of a Si$_{1-x}$Ge$_x$ quantum well on carriers can prevent them from reaching non-radiative recombination sites and hence decrease the non-radiative recombination rate. Room-temperature EL at 1.3 μm can be achieved with an effi-

ciency of about 0.03%. Further improvements will depend on increasing radiative mechanisms or decreasing non-radiative processes.

References

1 A. Gruhle, H. Kibbel, U. Erben and E. Kasper, *Electron. Lett.*, *29* (1993) 415.
2 K. Terashima, M. Tajima and T. Tatsumi, *Appl. Phys. Lett.*, *57* (1990) 1925.
3 J. C. Sturm, H. Manoharan, L. C. Lenchyshyn, M. L. W. Thewalt, N. L. Rowell, J. P. Noel and D. C. Houghton, *Phys. Rev. Lett.*, *66* (1991) 1362.
4 B. S. Meyerson, K. J. Uram and F. R. Legoues, *Appl. Phys. Lett.*, *47* (1988) 721.
5 P. M. Garone, J. C. Sturm, P. V. Schwartz, S. A. Schwarz and B. J. Wilkens, *Appl. Phys. Lett.*, *56* (1990) 1275.
6 J. C. Sturm, P. M. Garone and P. V. Schwartz, *J. Appl. Phys.*, *69* (1991) 542.
7 J. C. Sturm, P. V. Schwartz, E. J. Prinz and H. Manoharan, *J. Vac. Sci. Technol. B*, *9* (1992) 2011.
8 J. C. Sturm, X. Xiao, P. V. Schwartz, C. W. Liu, L. C. Lenchyshyn and M. L. W. Thewalt, *J. Vac. Sci. Technol. B*, *10* (1992) 1998.
9 J. Weber and M. I. Alonso, *Phys. Rev. B*, *40* (1989) 5683.
10 J. C. Sturm, X. Xiao, Q. Mi, L. C. Lenchyshyn and M. L. W. Thewalt, *Materials Research Society Symp. Proc.*, Vol. 298, Materials Research Society, Pittsburgh, PA, 1993, in press.
11 D. J. Robbins, P. Calcott and W. Y. Leong, *Appl. Phys. Lett.*, *59* (1991) 1350.
12 Q. Mi, X. Xiao, J. C. Sturm, L. C. Lenchyshyn and M. L. W. Thewalt, *Appl. Phys. Lett.*, *60* (1992) 3177.
13 U. Gnutzmann and K. Clausecker, *Appl. Phys.*, *3* (1974) 9.
14 S. Satpathy, R. M. Martin and C. G. van de Walle, *Phys. Rev. B*, *38* (1988) 13 237.
15 H. Okumura, K. Miki, S. Misawa, K. Sakamoto, T. Sakamoto and S. Yoshida, *Jpn. J. Appl. Phys.*, *28* (1989) L1893.
16 R. Zachai, K. Eberl, G. Abstreiter, E. Kasper and H. Kibbel, *Phys. Rev. Lett.*, *64* (1990) 1055.
17 U. Schmid, N. E. Christensen and M. Cardona, *Phys. Rev. Lett.*, *65* (1990) 2610.
18 U. Menczigar, G. Abstreiter, J. Olajos, H. Grimmeiss, H. Kibbel, H. Presting and E. Kasper, *Phys. Rev. B*, *47* (1993) 4099.
19 L. C. Lenchyshyn, M. L. Thewalt, J. C. Sturm, P. V. Schwartz, E. J. Prinz, N. L. Powell, J. P. Noel and D. C. Houghton, *Appl. Phys. Lett.*, *60* (1992) 3174.
20 C. G. van de Walle and R. M. Martin, *Phys. Rev. B*, *34* (1986) 5621.