Photoluminescence from ordered and disordered Si–SiGe superlattices

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Ordered (Ord-SL) and disordered (Dis-SL) Si–SiGe superlattices are grown using ultrahigh vacuum chemical vapour deposition (UHVCVD). The results of cross-sectional transmission electron microscopy (XTEM) and high-resolution double crystal x-ray diffraction (HRXRD) indicate that high quality Si–SiGe superlattices can be achieved. Well-defined band-edge excitonic luminescence is observed for the $\text{Si}_{0.86}\text{Ge}_{0.14}$ –Si superlattice. Stronger photoluminescence (PL) is observed for the Si–SiGe disordered superlattice compared with the corresponding Si–SiGe ordered superlattice. Furthermore, PL peak energy of the Dis-SL shifts to lower value with respect to the peak position of the corresponding Ord-SL. The stronger intensity of the no-phonon (NP) peak and the red shift of the PL peak are possibly a result of two probable mechanisms: (i) the tunnelling effect and (ii) the formation of localized states.

1. Introduction

Photo emission from Si-related materials has attracted much interest recently. Integration of light-emitting devices into Si circuitry is one of the final goals that researchers [1–4] are trying to achieve. Early attempts were based on the well-known zone-folding scheme proposed by Gnutzmann and Clausecker [5]: in which the indirect nature of Si was expected to change into a direct one by artificial folding of the conduction band minimum of the Δ symmetry back to the Γ point in k -space using $\text{Si}_m\text{--Ge}_n$ superlattices [6]. However, no direct evidence of zone-folded electronic transitions has yet been obtained. On the other hand, Sturm *et al.* [7] have recently reported photoluminescence (PL) from $\text{Si}_{1-x}\text{Ge}_x$ –Si quantum well structures grown by rapid-thermal chemical vapour deposition (RTCVD), and Robbins *et al.* have separately demonstrated electroluminescence from a similar $\text{Si}_{0.8}\text{Ge}_{0.2}$ –Si confining structure using a vapour phase epitaxy system [4]. These results seem to show that the $\text{Si}_{1-x}\text{Ge}_x$ –Si heterostructure is a promising candidate for replacing $\text{Si}_m\text{--Ge}_n$ superlattice systems [6]. In addition,

Sasaki has proposed a new class of semiconductor that is able to exhibit anomalously strong luminescence intensity in the indirect band $\text{Al}_{1-x}\text{Ga}_x\text{As}-\text{AlAs}$ ($x \geq 0.5$) system [8], and it is referred to as a 'disorder crystalline semiconductor'. Disordered crystalline semiconductors are constructed artificially in such a manner that the constituent atoms are sufficiently ordered to allow epitaxial growth, but the chemical composition is disordered. The improvement in luminescence capability is attributed to localized states created by artificial disordering. The concept of a disordered crystalline semiconductor can be applied to the $\text{Si}-\text{Si}_{1-x}\text{Ge}_x$ system.

In this work, ordered and disordered $\text{Si}-\text{Si}_{1-x}\text{Ge}_x$ superlattices are grown in an ultrahigh vacuum chemical vapour deposition system (UHVCVD). In order to determine and to verify the quality of the crystallinity, including layer thickness and compositional uniformity, high-resolution double crystal x-ray diffraction (HRXRD) and cross-sectional transmission electron microscopy (XTEM) are used. A dynamical x-ray simulation program based on the solution of Takagi-Taupin equations is employed to analyse the experimental rocking curves [9]. In addition, we report photoluminescence properties of these ordered and disordered superlattices.

2. Experimental procedure

Ordered and disordered $\text{Si}-\text{Si}_{1-x}\text{Ge}_x$ superlattices were grown on 7.6 cm (001) Si substrates using UHVCVD. The UHVCVD technique was first described and developed by Meyerson [10]: the construction and operation of our UHVCVD system was similar to that reported. Our system, designed and built at National Chiao Tung University, has been successfully used to grow epitaxial layers of Si and $\text{Si}_{1-x}\text{Ge}_x$ at temperatures as low as 550°C: details of our UHVCVD system are described elsewhere [11–13]. In this work, the growth temperature was kept constant at 550°C. Prior to growth, the substrate was subjected to an $\text{H}_2\text{SO}_4:\text{H}_2\text{O}_2 = 3:1$ clean and a 10% HF dip. Silane (SiH_4) and 10% germane (GeH_4) in hydrogen were used as reactant gases. The base pressure of the system was maintained at about 2.7×10^{-6} Pa in the growth chamber. During growth, the system was operated at about 0.133 Pa.

HRXRD was used to determine the structural quality of these superlattices. X-ray rocking curves were obtained using a Philips DCD-3 double-crystal diffractometer. The system was operated in parallel (+, –) diffraction geometry using CuK_α radiation. A single crystal of Si with an (004) symmetric reflection was used for the first reflection. A dynamical x-ray simulation program was used to determine the perfection, interfacial abruptness, layer thicknesses and Ge compositions of these superlattices. Cross-sectional transmission electron microscopy was performed to examine crystal quality and interfacial abruptness. For the photoluminescence measurement, the sample was placed in a Janis cryostat and the temperature varied from 10 to 300 K. An argon ion laser with 2.54 eV was used as the pumping source. The luminescence signal was analysed by a 0.75 m double spectrometer and detected by a liquid-nitrogen-cooled Ge detector (North Coast 817L) connected to a lock-in amplifier.

3. Results and discussion

Figure 1a shows the HRXRD rocking curve obtained for an ordered $\text{Si}_{1-x}\text{Ge}_x$ -Si superlattice of 40 periods with $x = 0.14$, $\text{Si} = 6.8$ nm and $\text{SiGe} = 4.0$ nm. In this figure, peak labelled SU represents the reflection from the Si substrate, the peak labelled P0 represents the zeroth-order superlattice reflection, and the other main peaks are satellite peaks (+1, –1, –2, –3) resulting from periodicity of the ordered superlattice. As well as the main peaks, a number of secondary peaks are observed. The secondary peaks occurring between the main peaks are interference reflections corresponding to the number of periods in the ordered superlattice. The Ge fraction, x , and the thicknesses of the Si, t_{Si} , and $\text{Si}_{1-x}\text{Ge}_x$, t_{SiGe} , layers in this

Figure 1 (a) HRXRD rocking curve obtained for an ordered superlattice with an initial Si crystal; and (b) the simulated rocking curve for a full-strained $\text{Si}_{1-x}\text{Ge}_x$ –Si superlattice of 40 periods with $x = 0.14$, Si = 6.8 nm and SiGe = 4.0 nm.

Figure 2 XTEM micrograph of ordered Si–SiGe superlattices with SiGe well widths of 4.0 nm.

superlattice were determined from a comparison of stimulated and experimental double-crystal rocking curves. Figure 1b shows the simulated rocking curve for a fully strained $\text{Si}_{1-x}\text{Ge}_x$ -Si superlattice of 40 periods with $x = 0.14$, Si = 6.8 nm and SiGe = 4.0 nm. In this simulated rocking curve, we have considered the effect of diffused scattering from the first crystal and the substrate [14]. As compared with Fig. 1a, excellent matches between experiment and simulation in terms of peak position, peak intensity and full-width at half-maximum (FWHM) of each main peak are clearly observed. Because the simulated rocking curve is based on an assumption that all layers in the multiple quantum wells are perfect with regard to crystal quality, layer thickness and uniform alloy composition, it can be concluded that the Si-SiGe superlattice does not have any detectable layer variation. This point has been further confirmed by conventional and high-resolution cross-sectional transmission electron microscopic examination of this sample [15].

Figure 2 shows an XTEM micrograph of an ordered Si-SiGe superlattice with an SiGe well width of 4.0 nm. In this picture, each layer of the superlattice is clearly resolved: a good periodic structure with excellent uniform layer thickness is observed. The thicknesses of the Si and SiGe layers were found to be 6.8 ± 0.5 and 4.0 ± 0.5 nm, respectively. These values agree well with the results of x-ray simulation.

Figure 3 shows a typical band-edge PL spectrum of an $\text{Si}_{0.86}\text{Ge}_{0.14}$ -Si superlattice with an SiGe well width of 4.0 nm at 10 K. To identify the origin of the emission peaks, we follow the assignment made by Weber and Alonso [16]. The emission at 1098 meV is due to excitonic radiative recombination of excitons, created in the Si substrate, with the assistance of transverse optical (TO) photons (58 meV). Intense luminescence lines, labelled NP and $\text{TO}^{\text{Si-Si}}$, are no-phonon and transverse optical-phonon-assisted emissions, respectively, originating from the

Figure 3 Typical band-edge PL spectrum of an $\text{Si}_{0.86}\text{Ge}_{0.14}$ -Si superlattice with an SiGe well width of 4.0 nm at 10 K.

Figure 4 The temperature dependence of PL intensities of an $\text{Si}_{0.86}\text{Ge}_{0.14}$ –Si superlattice with an 8.7 nm SiGe well width.

SiGe wells of the superlattice. The 58 meV energy difference between the NP and $\text{TO}^{\text{Si-Si}}$ lines closely matches the TO phonon energy of Si. Phonon-assisted emission due to Si–Ge and Ge–Ge TO phonons and transverse acoustic (TA) phonons is also observed between NP and $\text{TO}^{\text{Si-Si}}$ emission lines. Strong NP emission is a magnification of the broken translation symmetry of the SiGe layers and is affected through momentum-conserving scattering due to compositional disorder in SiGe. The lowest energy component ($\text{TO} + \text{TO}^{\text{Si-Si}}$) in Fig. 3 can be attributed to two-phonon replica because the energy difference between this peak and the NP line is equal to twice that of the TO phonon energy of Si.

Furthermore, we measure the dependence of the PL spectra on temperature and excitation intensity. Figure 4 shows the temperature dependence of the PL intensities of an $\text{Si}_{0.86}\text{Ge}_{0.14}$ –Si superlattice with an 8.7 nm SiGe well width. It appears that the data cannot be fitted by an Arrhenius plot. Instead, a relationship similar to that used for amorphous semiconductors and disordered superlattices holds:

$$I_{\text{PL}} = I_0 / [1 + A \exp(T/T_0)] \quad (1)$$

where I_{PL} is the PL intensity, T is the measured temperature, T_0 is the characteristic temperature corresponding to the energy depth of the localized states, A is a tunnelling factor, and I_0 is the PL intensity at the low-temperature limit. Equation 1 is valid for amorphous semiconductors

Figure 5 The temperature dependence of PL intensities of an $\text{Si}_{0.86}\text{Ge}_{0.14}$ -Si superlattice with an 8.7 nm SiGe well width. The PL energy of the SLS first increases, then decreases with increasing temperature. The PL energy of the Si substrate decreases with increasing temperature. (a) 10 K, (b) 30 K, (c) 60 K, (d) 100 K, (e) 140 K.

and disordered superlattices, as opposed to a typical Arrhenius-type relationship, because of the existence of localized states [17, 18]. From this, it seems reasonable to infer that localization of carriers may occur in SiGe-Si strained-layer superlattices (SLSs). The localized states could be due to regions of higher than average Ge content that arise simply from the random nature of the alloy. Similar localized exciton luminescence peaks have been observed earlier in other semiconductor alloys [19–22]. Another piece of evidence to support the argument here is the temperature dependence of the PL energy, as shown in Fig. 5. In the figure, the PL energy of the superlattice first increases, then decreases with increasing temperature. This is different from the temperature dependence of the bandgap of a semiconductor. In Fig. 5, we also show the PL spectra due to the Si substrate. It behaves normally; that is, the PL energy decreases with increasing temperature. The anomalous temperature dependence of the PL energy can be explained easily by the localized excitons. At low temperatures, carriers occur in the sites of minimum potential energies. As the temperature increases, carriers can absorb enough thermal energy and be excited to higher energy states, thus an increase in emission energy is observed. As the temperature increases further, most carriers are in extended states, and the temperature dependence of the emission energy is determined by the bandgap of the semiconductor, which decreases with increasing temperature.

In addition, two superlattices with thinner Si barriers and SiGe wells were grown for the PL study: an ordered superlattice (Ord-SL) with $\text{Si-Si}_{0.9}\text{Ge}_{0.1} = 2.0\text{--}2.0\text{ nm}$, and a total

Figure 6 Low temperature (10 K) PL spectra for (a) Ord-SL and (b) Dis-SL.

superlattice thickness of $0.4\ \mu\text{m}$; and a disordered superlattice (Dis-SL). In the growth of $(\text{Si})_m - (\text{Si}_{0.9}\text{Ge}_{0.1})_n$ ($m, n = h, k, l$) Dis-SL, the layer thicknesses of SiGe and Si were given by a random serial, with equal probability of appearance for h, k, l . The values of h, k, l were 1.0, 2.0 and 3.0 nm, respectively. Therefore, these different thicknesses appeared randomly with equal probability of appearance. The superlattice period and the Ge composition were determined by HRXRD measurement.

Figure 6 shows the low temperature (10 K) PL spectrum of the Dis-SL compared with that of the Ord-SL, which has a Ge composition, x , and averaged period the same as for Dis-SL. The two major PL peaks from both Ord-SL and Dis-SL correspond to NP and its TO phonon replica peaks of exciton luminescence. As can be seen in Fig. 6, the intensity of TO from Dis-SL is almost the same as that of TO from Ord-SL. However, in terms of the NP peak, the PL intensity of Dis-SL is several times larger than that of Ord-SL. Furthermore, the PL peak energy of Dis-SL shifts to lower values with respect to the peak position of the corresponding Ord-SL. Since the PL peak is affected not only by the superlattice structure but also by the Ge composition in the well layer, comparison of the PL peak position should be done using carefully selected samples in which the compositional error of Ge is as small as possible. HRXRD indicates that the average Ge composition in Dis-SL equals that in Ord-SL. By comparing the PL spectra of Ord-SL with that of Dis-SL, peak shift energies of both NP and TO peaks in the Dis-SL are approximately 35 meV. Stronger intensity and a red shift of the PL peak are reported for both AlAs–GaAs and AlP–GaP disordered superlattices [23–26]. The red shift energies of AlAs–GaAs and AlP–GaAs disordered superlattices are about 100 and 67 meV, respectively.

Figure 7 The tunnelling effect on the PL experiment of the Si–SiGe disordered superlattice.

Although the peak shift energy for the Si–SiGe disordered superlattice is not very different from those of the other disordered superlattice systems, the enhancement effect of PL intensity in the SiGe–Si disordered superlattice does not seem to be so large. The stronger intensity (for the NP peak) and the red shift of the PL peak result from two possible mechanisms: (i) the tunnelling effect and (ii) the formation of localized states.

Figure 7 shows the tunnelling effect on the PL experiment of the Si–SiGe disordered superlattice. For the sample Dis-SL, the Si barrier is thin enough for the tunnelling phenomenon to occur. Holes having higher energy in the two thinner SiGe wells (width, $w = 1.0, 2.0$ nm) tunnel into the widest SiGe well ($w = 3.0$ nm), which has the lowest quantum level. Most of the recombination of the carriers occurs in the widest SiGe wells. As a result, the PL peak energy of the Dis-SL shifts to lower values with respect to the peak position of the corresponding Ord-SL. The other possible reason is the formation of a localized state in the Si–SiGe disordered superlattice. In the disordered superlattice system, wave functions are randomly reflected and are interfered with by artificially introduced spatial disordering and the stagnant wave, which means carrier localization can be formed. Thus the PL mechanism of the disordered superlattice is considered to be recombination through the localized states resulting from the introduction of artificial disordering. If relaxation of nanoscale localization of momentum occurs in the conversion Si–SiGe disordered superlattice, the intensity ratio of NP : TO must change compared with the ordered superlattice. However, the change in the NP : TO ratio is not so great after the introduction of disordering. This result suggests that the effect of localization on the PL properties in the SiGe–Si disordered superlattice is not so large. The exact mechanism causing the stronger intensity (for NP peak) and the red shift of the PL peak of the disordered Si–SiGe superlattice as compared with the ordered Si–SiGe superlattice must be clarified: further research is under way.

4. Conclusions

Ordered and disordered Si–SiGe superlattices were grown using UHV/CVD. XTEM and HRXRD indicate that high-quality Si–SiGe superlattices can be achieved. Well-defined band-edge excitonic luminescence is observed for the $\text{Si}_{0.85}\text{Ge}_{0.14}$ –Si superlattice. Stronger PL are observed for the Si–SiGe disordered superlattice as compared with the corresponding

Si–SiGe ordered superlattice. In addition, we show the anomalous temperature dependence of the PL intensity and energy. The PL energy first increases, then decreases with increasing temperature. The intensity has a temperature dependence similar to that of amorphous semiconductors and disordered superlattices. Blue shifting of the PL energy with increasing excitation intensity is also observed. These phenomena suggest localization of carriers in the SiGe wells due to the random nature of the alloy. Furthermore, the PL peak energy of the Dis-SL shifts to lower values with respect to the peak position of the corresponding Ord-SL. The stronger intensity of the NP peak and the red shift of the PL peak possibly result from two mechanisms: (i) the tunnelling effect and (ii) the formation of localized states.

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