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Photoluminescence from trapped excitons in $Si_{1-x}Ge_x$/Si quantum well structures

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Abstract. Photoluminescence spectra from $Si_{1-x}Ge_x$/Si quantum well structures grown at high temperatures are investigated. The luminescence properties are found to be very different from those of free excitons. To describe correctly the spectral lineshape, the radiative recombination from excitons trapped on the local potential fluctuations in $Si_{1-x}Ge_x$ quantum wells must be considered. At low sample temperatures, the luminescence is mainly from the trapped excitons. With increasing temperature, trapped excitons are thermally activated into free excitons and luminescence peaks shift to higher energies. By comparing measured spectra with the calculated spectra, the trap density and the trap energy are derived. The origin of the trap and its relation with the crystal growth are discussed.

1. Introduction

$Si_{1-x}Ge_x$ is an indirect-band-gap semiconductor. The electron–hole recombination must involve scattering centres to conserve momentum. Considering the continuous dispersion relation for scattering centres such as phonons, the luminescence spectrum of free excitons is given by [1, 2]

$$I(E) \sim D(E)\exp[-(E-E_G)/kT] \tag{1}$$

where $D(E)$ is the free-exciton density of states. For three-dimensional structures, $D(E) \propto (E-E_G)^{1/2}$ and, for two-dimensional structures, $D(E)$ is a step function. For $Si_{1-x}Ge_x$/Si quantum well structure (QWSs),

$$E_G(T) = E_{G0}(T) + E_{1e} + E_{1h} - E_{XB} - \hbar\omega_s \tag{2}$$

where $E_{G0}(T)$, E_{1e}, E_{1h}, E_{XB} and $\hbar\omega_s$ are the band gap of strained $Si_{1-x}Ge_x$ alloy, the confinement energies of electrons and holes in a $Si_{1-x}Ge_x$/Si QWS, the exciton binding energy and the energy transferred from scattering centres, respectively. If the scattering centre is an optical phonon, then $\hbar\omega_s = \hbar\omega_p$. If the scattering centre is alloy fluctuation and disorder, then $\hbar\omega_s \approx 0$. The lineshape equation (1) for indirect excitons is very different from that for direct excitons. For the latter, since no scattering centres are involved for the first-order direct optical transitions, the energy spectrum is discrete and the luminescence lineshape is given by a symmetric Lorentzian function (i.e. a broadened delta function) [2]. For indirect excitons, due to the Boltzmann factor $\exp[-(E-E_G)/kT]$, the lineshape is asymmetric and strongly depends on temperature. With the increase in temperature, the linewidth (full width at half-maximum (FWHM)) and the asymmetry increase. Equation (2)

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indicates that the peak energy is mainly determined by the band gap of bulk $Si_{1-x}Ge_x$ alloy and decreases with increasing temperature [3].

The properties of indirect excitons described above have been observed in a bulk Si crystal [4] as well as in $Si_{1-x}Ge_x$/Si alloys and QWSs [5–7]. For a $Si_{1-x}Ge_x$/Si QWS, the photoluminescence (PL) spectra are dominated by exciton recombination with a transverse optical (TO) phonon $\hbar\omega_s = \hbar\omega_{TO} = 58$ meV) and alloy scattering (no phonon (NP)) ($\hbar\omega_s \approx 0$) involved. At a low temperature (about 10 K), the exciton may be bound to shallow impurities, forming bound excitons. Since the bound excitons have a binding energy of a few millielectronvolts, they are thermally ionized into free excitons at temperatures higher than about 30 K. In this case, the PL spectra can be well described by equation (1) [6].

In this paper, we report PL spectra showing different characters from those of free excitons. It is shown that the observed luminescence can be explained only by considering contributions from both free and trapped excitons. The comparison between the calculated and the measured PL spectra at various temperatures gives the trap density and energy.

2. $Si_{1-x}Ge_x$/Si quantum well structures

The $Si_{1-x}Ge_x$/Si QWSs used in this study were grown on Si(100) substrates by solid source molecular beam epitaxy. A typical sample includes a Si buffer layer (about 100 nm), N-period $Si_{1-x}Ge_x$/Si quantum wells and a 50 nm cap layer. The growth temperature is 800–900 °C. The thickness of the $Si_{1-x}Ge_x$ layers is below the critical value to avoid strain relaxation. All layers were undoped.

Table 1. Sample parameters.

Sample	x	L_w (nm)	L_b (nm)	N
A	0.035	20	50	20
B	0.20	4.0	15	10

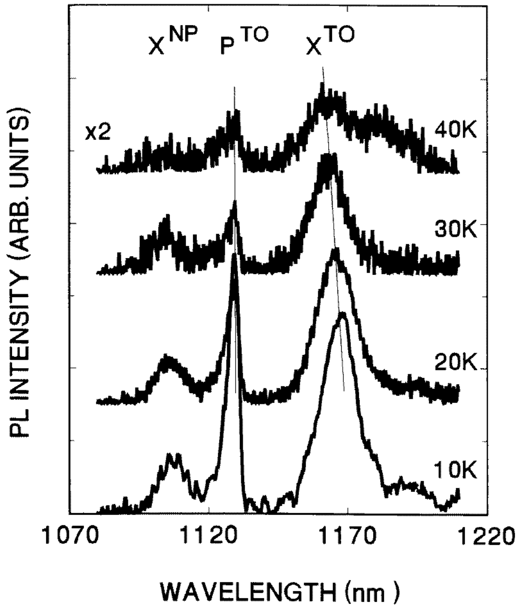
The structure parameters for two samples discussed in this article are listed in table 1, where L_w and L_b are the thicknesses of the $Si_{1-x}Ge_x$ (well) and Si (barrier) layer, respectively, and x is the Ge concentration. These parameters were determined from the growth condition, x-ray diffraction, Rutherford back scattering and PL measurements. For sample A, due to the small x-value and large L_w, the carriers in the $Si_{1-x}Ge_x$ layers are similar to those in bulk alloys and the quantum confinement effects are negligible. For sample B, the x-value is relatively large and L_w is small. The quantum confinement effects are important (quasi-two-dimensional structure).

3. Photoluminescence spectra

PL measurements were carried out using a Jobin–Yvon U1000 monochromator. The 488 nm line from an Ar-ion laser was used as the excitation source. The PL signal was detected with a 77 K cold Ge photodiode and recorded with a computer using a conventional lock-in technique. The samples were fixed in a closed-cycle cryostat and the measurement temperature was adjusted from 10 to 300 K.

Figures 1(*a*) and 1(*b*) give the PL spectra measured at different temperatures for samples A and B, respectively. At a low temperature (10 K, for example), the spectra are dominated by three main lines labelled P^{TO}, X^{NP} and X^{TO}. P^{TO} (peak at 1130 nm or 1.097 eV) is

(*a*)



(*b*)

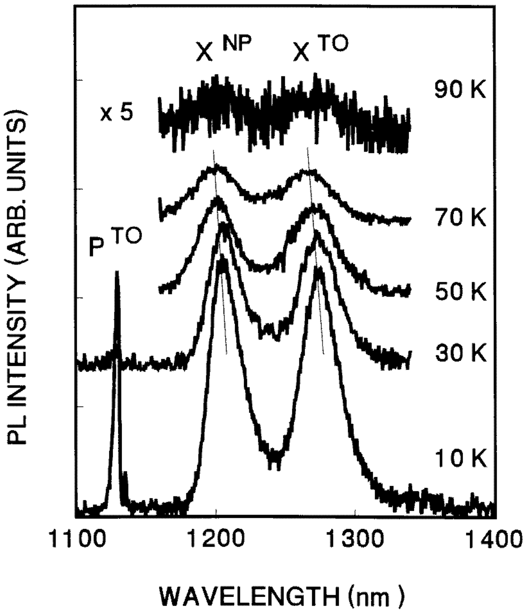


Figure 1. PL spectra from (*a*) sample A and (*b*) sample B at different temperatures.

structure independent. It is the TO-phonon-involved exciton peak from the Si substrate. Two X lines are structure dependent and arise from the $Si_{1-x}Ge_x$ QWS. The energy difference between the two X peaks is 57 meV (TO phonon energy). They are the exciton peaks with

(X^{TO}) and without (X^{NP}) phonon involvement from $Si_{1-x}Ge_x$ layers [5–7].

Figure 1 shows that the X and P lines are very different in character. The X lines are more symmetric. With increasing temperature, the broadening, the linewidth asymmetry and the intensity change more slowly for X lines than for the P line. Most importantly, within the temperature range shown in figure 1, the peaks of X lines shift to high energies while the peak of the P line changes little or decreases with increasing temperature.

As discussed earlier, peak P can be described by free-exciton luminescence with the lineshape given by equation (1). The X lines, however, cannot. The spectral analysis shows that even an inhomogeneous broadening is considered; the lineshape of X peaks as a function of temperature still cannot be explained by the free-exciton luminescence.

4. Trapped excitons

In semiconductor QWSs, the material inhomogeneity can induce spatial potential fluctuations. At low temperatures, photoexcited electrons and holes are more likely to stay in the low-potential region. If the dimension of the fluctuation is comparable with the radius of free excitons (called the short-range fluctuation), then the potential fluctuation may play the role of traps for excitons, forming localized trapped excitons. The fluctuation with the dimension much larger than that of free excitons (called the long-range fluctuation) simply induces line broadening. Figure 2 schematically shows the potential fluctuation, the trap, and the trapped excitons. We use the subscripts FX and TX to represent the free excitons and the trapped excitons, respectively. At low temperatures, the PL from trapped excitons is expected to dominate the spectra. As the temperature increases, trapped excitons are thermally ionized into free excitons, and the PL from free excitons will eventually dominate. If the line broadening (FWHM) due to the long-range fluctuation is comparable with the trap energy, the PL peaks for free and trapped excitons are no longer resolved.

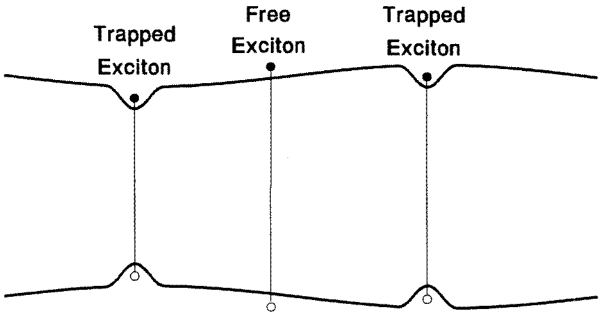


Figure 2. Schematic diagram for free and trapped excitons in inhomogeneous $Si_{1-x}Ge_x$ layers and QWSs.

Considering the contributions from both free and trapped electrons, PL spectra are given by [8]

$$I_{PL}(E) = I_{TX}(E) + I_{FX}(E) \quad (3a)$$

$$I_{TX}(E) = N_T \exp(E_b/kT) \exp[-(E + E_b)^2/\sigma^2] \quad (3b)$$

$$I_{FX}(E) = N_{FX} \exp(-E^2/\sigma^2) \quad (3c)$$

where N_T and N_{FX} are the densities of traps and free excitons, E_b is the average trap energy and σ is a factor proportional to the broadening reflecting the long-range fluctuation. For a three-dimensional system,

$$N_{FX}(3D) = (2\pi MkT/h^2)^{3/2} \tag{3d}$$

and, for a two-dimensional system,

$$N_{FX}(2D) = 2\pi MkT/h^2 \tag{3e}$$

where M is the mass of free excitons given by $M = m_e^* + m_h^*$.

There are three unknowns in equation (3): N_T, E_b and σ. These parameters can be obtained by fitting the calculated spectra to measured spectra. The linewidth is determined by σ, and the peak shift with the temperature is determined by N_T and E_b. To see that equation (3) can really describe the measured spectra, PL spectra are calculated and compared with the measured spectra for both samples at different temperatures. The results are shown in figure 3. The parameters used in the calculation are listed in table 2, where equations (3*d*) and (3*e*) are used for samples A and B, respectively, to represent the bulk and two-dimensional properties of the structure. The quantitative agreement between the calculated and the measured PL spectra is evident.

Table 2. Parameters used in the calculations of the PL spectra.

Sample	σ (meV)	N_T	E_b (meV)
A	7.5	5×10^{15} cm^{-3}	5.0
B	15	5×10^{10} cm^{-2}	8.0

To demonstrate further the importance of trapped excitons, the energy shift of PL peak as a function of temperature is calculated using equation (3). Results are shown in figure 4 in which the full circles represent the measured data minus the band-gap shift of $Si_{1-x}Ge_x$ alloy (data for bulk Si from [3] is used). The agreement between the calculated and the measured data is also evident.

We note that the calculated relations shown in figure 4 are sensitive to the trap density and energy, where N_T mainly determines the temperature at which the PL peak starts to shift rapidly and E_b gives the shift at high temperatures: $E_b = \Delta E_p(T = \infty)$. Since the measured PL spectra (including the linewidth, the symmetry and the peak energies which shift with temperature) are quantitatively reproduced by our calculation, the correct interpretation is demonstrated. The parameters such as N_T and E_b derived are reliable.

5. Discussion

The parameter σ in equation (3) describes the long-range fluctuation in x for $Si_{1-x}Ge_x$ quantum wells. The fluctuation may be induced by the deviation from spatial uniformity of the substrate temperature and in the molecular beam flux during the sample growth. The scale of this fluctuation is of an order larger than a micrometre and much larger than the radius of free excitons. Thus, within the region of interest, excitons can be viewed as free.

The short-range fluctuation, however, has a different origin from the long-range fluctuation. It is mainly related to the growth dynamics: three-dimensional growth at high growth temperatures. The length of the fluctuation must be comparable with that of free excitons or in the range of 10 nm. For example, in some regions the Ge concentration may

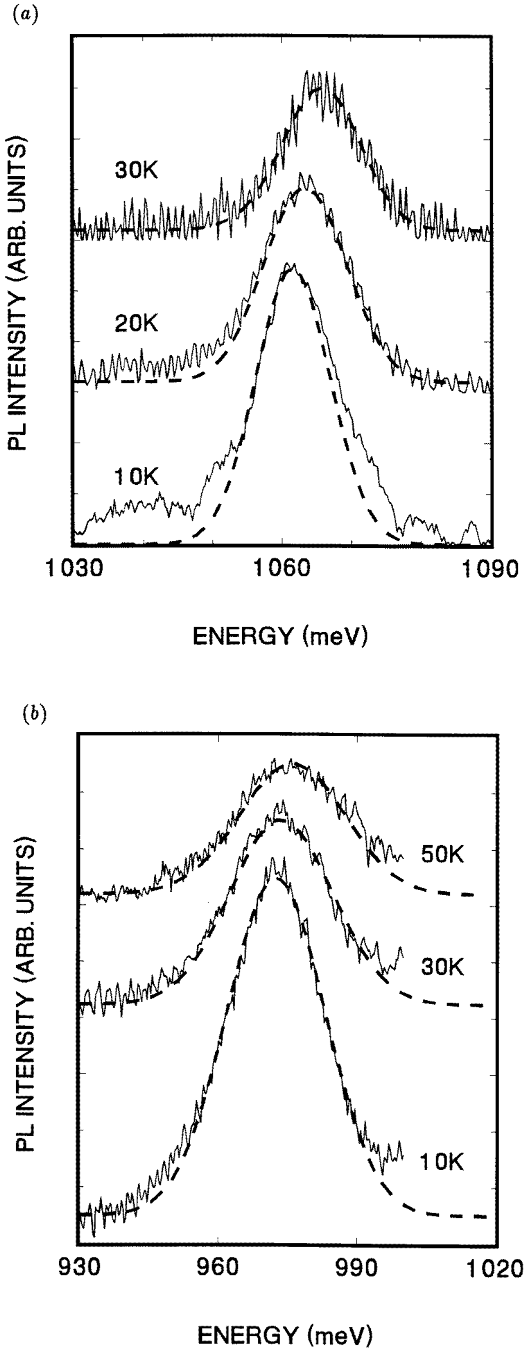


Figure 3. Calculated (– – –) and measured (——) PL spectra (X^{TO} peak) from (*a*) sample A and (*b*) sample B at different temperatures.

be higher than in the nearby region, forming a Ge-rich cluster in $Si_{1-x}Ge_x$ alloy. Reflection high-energy electron diffraction (RHEED) has been used to examine the growth dynamics.

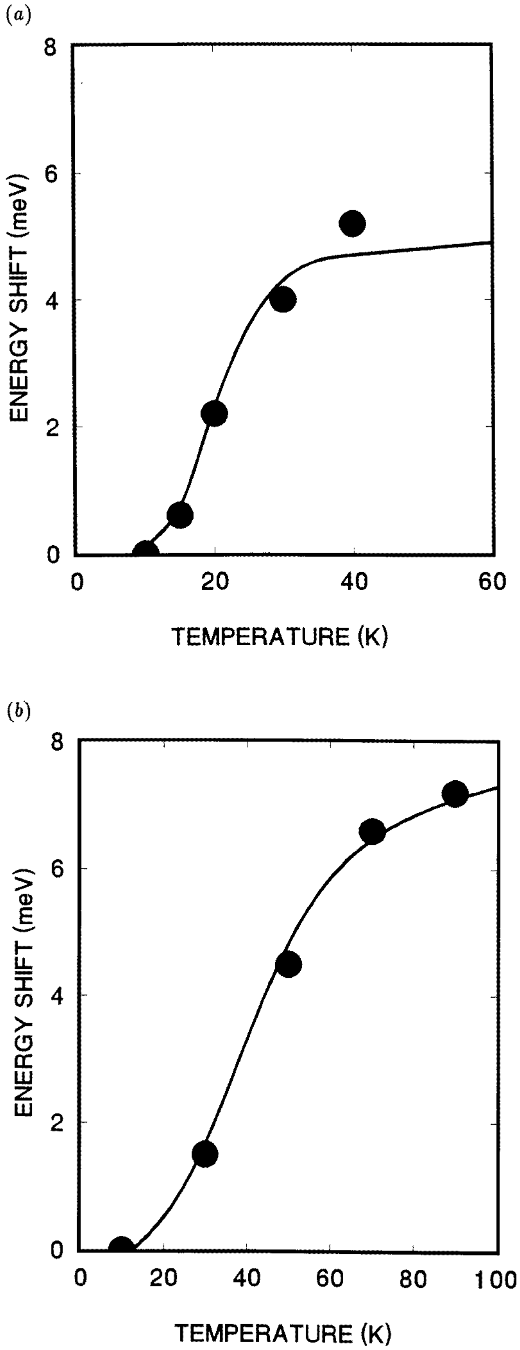


Figure 4. Calculated (——) and measured (●) X^{TO} peak shift as a function of temperature for (*a*) sample A and (*b*) sample B.

For the samples showing trapped exciton luminescence, RHEED lines with two-dimensional structure are blurred and the transition from two- to three-dimensional growth of $Si_{1-x}Ge_x$

layers is indicated.

The PL from trapped excitons has been observed previously in III–V compound semiconductors [8, 9] and in $Si_{1-x}Ge_x/Si$ heterostructures [10, 11]. In GaAs QWSs the fluctuation in well thickness (the $GaAs/Al_xGa_{1-x}As$ interface fluctuation) can induce traps for excitons [8]. In $In_xGa_{1-x}As$ QWSs the fluctuation in In concentration can also produce localized traps for excitons [9]. In $Si_{1-x}Ge_x/Si$ QWSs the blue shift of PL lines with increasing temperature similar to those shown in figure 1 can also be found in the literature [12]. The results indicate that the fluctuation in alloy composition is really important in affecting the luminescence properties of the excitons in the $Si_{1-x}Ge_x/Si$ system.

6. Conclusions

PL spectra from $Si_{1-x}Ge_x/Si$ QWSs with different character from those of free excitons were investigated. The lineshape is found to be more symmetric, the linewidth is seen to change slowly with temperature, and the peak position shifts to a high energy with increasing temperature. These properties can only be described quantitatively by considering contributions from both free and trapped excitons. By comparing calculated spectra with the experimental results, parameters relating to traps such as the density and the average energy are obtained. It is suggested that the origin of the traps is related to the material growth dynamics.

Acknowledgments

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