Photoluminescence from pseudomorphically strained $Si/Si_{1-x}Ge_x$ multiple quantum wells grown on silicon

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ABSTRACT

We have grown $Si/Si_{1-x}Ge_x$ multiple quantum wells ($x \simeq 8\%$) lattice-matched to silicon with well thicknesses between 3 and 20 nm using UHV-CVD. The sample parameters were obtained accurately with high-resolution X-ray diffraction (rocking curves) and transmission electron microscopy. From an analysis of the band-edge related photoluminescence energies we find a blue-shift due to confinement for thin wells.

1. INTRODUCTION

The concept of confinement in a quantum well sandwiched between two barriers (with larger band gaps than the well) is well established in the case of III/V compound semiconductors.[1,2] In the last few years, however, heterostructures involving silicon and germanium (or their alloys) as the barrier and well materials have also been grown with improving perfection.[3–9] It is expected that these novel materials may have better electronic and optoelectronic properties than silicon by itself, partly due to quantum effects like zone folding[3] and confinement,[5] but also due to disorder, the influence of strain on the band structure, and trapping of carriers. Therefore, it seems useful to perform the same studies of confinement in the Si/Ge system that have been performed in compound semiconductors many years ago. This is the purpose of this report. We also want to evaluate the applicability of X-ray scattering and transmission electron microscopy (TEM) as characterization tools for such $Si/Si_{1-x}Ge_x$ strained-layer MQWs and test their power to predict optical properties such as the band gap and the energies of the observed photoluminescence features.

To our knowledge, no systematic study of confinement energies as a function of well width and barrier height has been published for this material system, although other groups are currently working on the same problem. Since there is a considerable difference between the lattice constants of silicon and germanium (4%), such a study of confinement (using pseudomorphic structures on silicon substrates) is only possible if the germanium content is below about 30%. For higher germanium concentrations, the critical thickness of $Si_{1-x}Ge_x$ alloys on (100) silicon subtrates is less than 200 Å, and no comparison between thin and thick (bulk-like) wells can be made. We note that strained-layer $Si/Si_{1-x}Ge_x$ heterostructures grown lattice-matched on (100) silicon have a peculiar band alignment: The conduction band offset is very small and its sign is not known. Most of the offset occurs in the valence band. In this work we assume that the conduction band offset is exactly zero. The electrons are therefore free to move about between the layers, whereas the holes are confined to the $Si_{1-x}Ge_x$ wells. Such structures are therefore neither type I nor type II heterostructures.

Further investigations of luminescence from $Si_{1-x}Ge_x$ alloys are also interesting for another reason: Several reports of photoluminescene (PL) from Si_mGe_n short-period superlattices[3,4] or $Si/Si_{1-x}Ge_x$ quantum wells[5–8] have recently appeared in the literature. However, the PL data from these structures are often difficult to interpret.[3,10,11] In the case of short-period Si_mGe_n superlattices, the observed features are rather broad for intrinsic transitions and could also be due to defect-related recombination observed by a number of groups[12,13] or originate from the $Si_{1-x}Ge_x$ strain-relief buffer layer. Recent PL data from quantum wells in which a $Si_{1-x}Ge_x$ alloy makes up the well (typically $x < 40\%$) have been easier to understand, since comparison with well established bulk alloy data[14,15] can be made. Band-edge excitonic luminescence from such wells has clearly been observed in this work and is in good agreement with the features expected from the TEM and X-ray analysis.

2. SAMPLE GROWTH AND TEM CHARACTERIZATION

In this paper, we discuss four $Si/Si_{1-x}Ge_x$ multiple quantum wells with nominal periods of 50–400 Å, total thickness of 4000 Å, and a nominal germanium concentration of 8% grown at 520° C on 5-inch silicon (100) lightly boron-doped substrates, 2° off towards (110), using ultra-low pressure chemical vapor deposition (UHV-CVD) as described elsewhere.[9] Transmission electron micrographs (taken for the two samples with the smallest and largest periods) show uniform layers

Parameter	Unit	Silicon	Germanium	Reference
a	Å	5.4309	5.6575	Ref. 20
c_{11}	GPa	165.77	128.53	Ref. 20
c_{12}	GPa	63.93	48.26	Ref. 20
σ_0	1	0.278	0.271	Ref. 20
Δ_0	eV	0.045	0.297	Ref. 20
$m_{hh}(100)$	m_0	0.274	0.211	Ref. 20
a_X	eV	1.65	1.2	Ref. 10
b	eV	−2.3	−2.6	Ref. 10
E_2	eV	8.8	9.3	Ref. 10

Table 1: Material constants for silicon and germanium used in the calculations. All constants were assumed to take their virtual-crystal value in the alloy.

parallel to the substrates and confirm the actual periods to be within 10% of the intended ones. TEM also fails to observe dislocations within the field of view of about 1 μm. Therefore, we can determine the upper limit for the dislocation density to be 10^8 cm^{-2}. We conclude that no more than the 10% of the strain in the samples can be relaxed, confirming that the the samples are pseudomorphic.

3. X-RAY CHARACTERIZATION

A systematic study of confinement effects requires an accurate knowledge of the sample structure. We therefore performed X-ray measurements as described in the following paragraphs.[9] This is complicated by the fact that there is a small tilt (usually between 20 and 30 arcsec) between the epilayer and the substrate, which is caused by the strain field created by the growth on a slightly misoriented substrate.

In order to find the sample parameters, in particular, the barrier and well widths L_B and L_W and the germanium content x, we had to use the following procedure: Two high-resolution X-ray rocking curves (using a Bede Scientific Instruments QC1 double crystal diffractometer) were taken for each sample. Between the two measurements, the sample was rotated by 180° around the surface normal to average out the effect of the tilt. One spectrum for each sample is shown in Fig. 1. In addition to the dominant substrate peak (marked 'S'), we see a zero-order superlattice peak (marked '0') and satellite peaks on both sides (marked '±1' and '±2'). The width of the substrate peak is about 12" and limited by the resolution of our X-ray beam conditioner (first crystal). This confirms the results obtained from TEM that there are no dislocations penetrating into the substrate, in contrast to the anomalous strain relaxation observed in compositionally graded thin films and superlattices.[16,17] The splitting between the substrate and zero-order peaks is determined by the average lattice constant of the superlattice in the growth direction, the width of the zero-order peak is consistent with the total thickness of the superlattice. The periodic *Pendellösung* fringes on both sides of the zero-order peak indicate well-defined smooth and parallel interfaces (substrate/superlattice/air) and also yield the total thickness of the superlattice. The spacing between the zero-order and satellite superlattice peaks are determined by the total period of the superlattice (i. e., the sum of barrier and well widths). The individual barrier and well widths are more difficult to obtain and involve an analysis of the amplitudes of the zero-order and satellite peaks.

Both measured spectra for each sample were compared with calculated spectra obtained from a commercial simulation program.[18] This program assumes that the layers are pseudomorphically strained, i. e., that they take the lattice constant of the substrate in the plane and expand in the growth direction according to Poisson's ratio. (As stated earlier, this assumption is well justified by the TEM results for the two samples with the smallest and largest periods.) The lattice constant of the alloy was calculated according to Vegard's law.[19] The material constants entering into the simulation are given in Table 1. The sample parameters were varied, until a satisfactory agreement between simulation and data was found. The results of this procedure are given in Table 2. The parameters of both scans for each sample were then averaged to cancel the influence of the tilt between the epilayer and the substrate.

4. PHOTOLUMINESCENCE RESULTS

Photoluminescence measurements of the four samples were performed at 3–5 K using the 488 nm line of an Ar^+-ion laser with a power density of about 4 W/cm^2 (100 mW total power, 2 mm spot size) for excitation and a Mattson Instruments Cygnus 100 Fourier transform spectrometer. The luminescence was detected with a Northcoast EO-817S germanium detector or a cooled germanium diode. We carefully avoided hitting the edge of the samples in order to

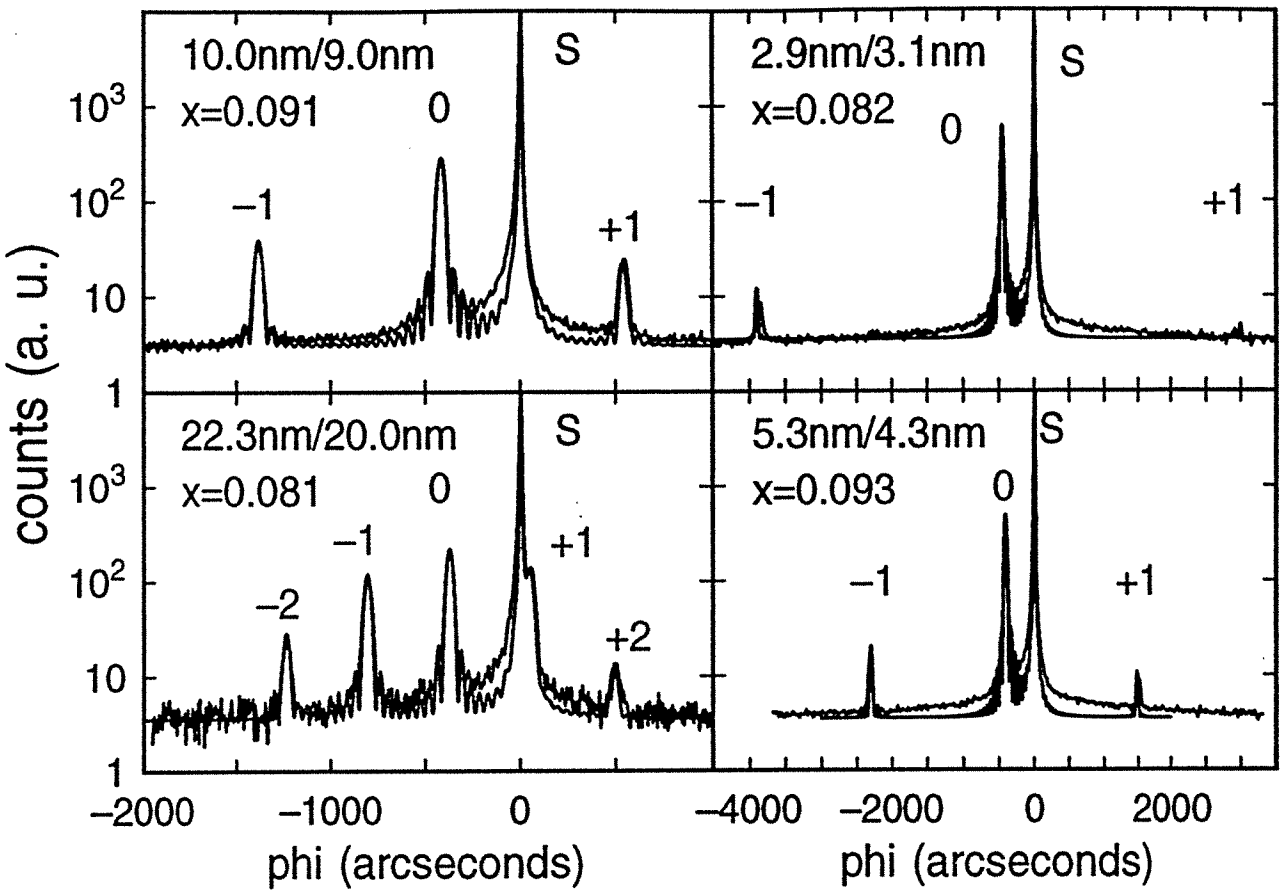

Figure 1: High-resolution x-ray rocking curves from four pseudomorphically strained $Si/Si_{1-x}Ge_x$ multiple quantum wells. Apart from the silicon substrate peak marked 'S', superlattice peaks up to second order (labelled '0', '±1' and '±2') are visible. The sample parameters were obtained by comparison with simulated spectra and are given in Table 2.

minimize the signal from the substrate, i. e. the lines labelled FE (free exciton TO-phonon replica), BE (boron-relaxed bound exciton, TO replica), and BMEC (boron-related bound multiexciton complex, TO replica) in Fig. 2. On the other hand, the substrate lines provide a convenient energy reference since their position is the same for all samples, see Refs. 22,23.

Apart from the substrate features, we observe two peaks labelled TO and NP as well as a shoulder (TA) in between. The energy of the TO phonon replicas is always about 58 meV below the no-phonon line, as should be expected.[14] This signal originates in the $Si_{1-x}Ge_x$ alloy wells. For x=0.08, $Si_{1-x}Ge_x$ is an indirect semiconductor alloy with a band structure similar to silicon.[14] We therefore assume most radiative recombination processes to involve a phonon to conserve the crystal momentum. This explains the TA and TO replica. See Ref. 14 for details. In addition to these phonon replicas, we also see a no-phonon peak 'NP'. This structure is disorder-activated and not observed in pure silicon. As explained in detail in Refs. 24 and 25, wave vector is not a good quantum number in a semiconductor alloy. Therefore recombination processes can occur without requiring a phonon.

These features related to the $Si_{1-x}Ge_x$ alloy layers are shifted from the bulk positions as reported in Ref. 14 because of the built-in strain. This can be taken into account by calculating the strain shifts from the most probable values of the deformation potentials compiled in Ref. 21 (see Table I) and will be discussed in more detail in the next section.

We note that the energy of the NP line for x=0.08 is in the position we would expect for an electron-hole droplet (or bound multi-exciton complex) in bulk silicon. However, this assignment is incompatible with the power and temperature dependence of this line. We observe the NP replica at temperatures in excess of 130 K. The electron-hole droplet in Si,

Table 2: Structural parameters of the samples and the respective error bars in parentheses (95% confidence limits): Barrier width L_B, well width L_W, total period $L_B + L_W$, average Ge content $xL_W/(L_W + L_B)$, and Ge content x in the well. Because of a small tilt between the epilayer and the substrate, two X-ray rocking curves were taken with the sample rotated by 180° in between. The average was used to calculate the energy E_x of the expected PL peak from the bound exciton in the strained bulk alloy. ΔE is the uncertainty calculated from the probable error of the composition x.

Sample	Scan	$L_B + L_W$ (Å)	$xL_W/(L_W + L_B)$ %	L_B (Å)	L_W (Å)	x %	E_x (meV)	ΔE (meV)
SE1	Scan 1	398(1)	4.0(1)	209(10)	189(10)	8.5(5)		
	Scan 2	446(1)	3.6(1)	236(10)	210(10)	7.7(5)		
	average	422(1)	3.8(1)	223(10)	200(10)	8.1(5)	1079	4
SE2	Scan 1	180(1)	4.6(1)	90(10)	90(10)	9.2(10)		
	Scan 2	200(1)	4.5(1)	110(10)	90(10)	9.0(10)		
	average	190(1)	4.5(1)	100(10)	90(10)	9.1(10)	1071	8
SE3	Scan 1	90(1)	4.8(1)	50(5)	40(5)	9.7(10)		
	Scan 2	102(1)	4.4(1)	56(5)	46(5)	8.8(10)		
	average	96(1)	4.6(1)	53(5)	43(5)	9.3(10)	1069	8
PL3	Scan 1	64(1)	4.0(1)	32(5)	32(5)	7.9(10)		
	Scan 2	56(1)	4.3(1)	27(5)	29(5)	8.5(10)		
	average	60(1)	4.2(1)	29(5)	31(5)	8.2(10)	1079	8

on the other hand, decreases drastically in intensity above 10 K. Furthermore, we do not observe the side bands usually associated with an electron-hole droplet. At temperature above 50 K, all $Si_{1-x}Ge_x$ related lines shift to higher energies by about 4 meV. We therefore believe that the recombination at 4 K is due to bound excitons, possibly related to shallow donors or acceptors. Since boron is the main dopant in the substrate, it seems likely that there may be some diffusion into the epilayer.

5. DISCUSSION AND CONFINEMENT ENERGIES

The energy positions of the no-phonon lines in the photoluminescence spectra were analysed to yield confinement energies in the following way: From the Ge content in the wells (x) as determined by X-ray scattering (see Table 2), we can calculate the band gap (energy of the free exciton) of the unstrained bulk alloy from Eq. (2) in Ref. 14. Since our data were taken at 4.2 K, the recombination is due to bound excitons. We therefore subtract 4 meV from this value.[14] From x, we also calculate the amount of strain in the wells. Using the deformation potentials compiled in Ref. 21 (see Table 1), we calculate the shift due to strain and also subtract it from the band gap. We thus obtain the energy of the bound exciton in a thick strained layer of $Si_{1-x}Ge_x$. This value E_x has several sources of errors: The largest error is related to the uncertainty in composition Δx, which causes an error ΔE in the band gap, also given in Table 2. We are not so much concerned about errors introduced by uncertainties in the material parameters of Table 1, since these do not depend on the well width and would simply cause a systematic shift between our data and the theory. Errors due to imperfections of the sample, such as interface roughness, interdiffusion of the layers, partial relaxation of the strain, or spontaneous ordering have also been neglected, since they are difficult to estimate.

For the sample with the thickest wells (200 Å), the position of the NP peak agrees within 1 meV with E_x. We therefore conclude that our determination of strain and composition using X-ray rocking curves is very accurate. For the thinner samples, we observe a blue-shift up to 17 meV of the luminescence due to confinement in the thin wells. This blue-shift E_c is given for our four samples in Table 3. We also list data obtained by other groups taken from the literature.[5–7]

For comparison with theory, we note that the confinement energies can be estimated from an envelope function model for a single quantum well with finite barrier height by matching the wave function and the current at the interface (see Table 1 for parameters). It is generally assumed that the conduction band offset for a $Si/Si_{1-x}Ge_x$ MQW is zero, if the Si barriers are unstrained. We therefore have a valence band offset of 70 meV for x=0.08. The results of this

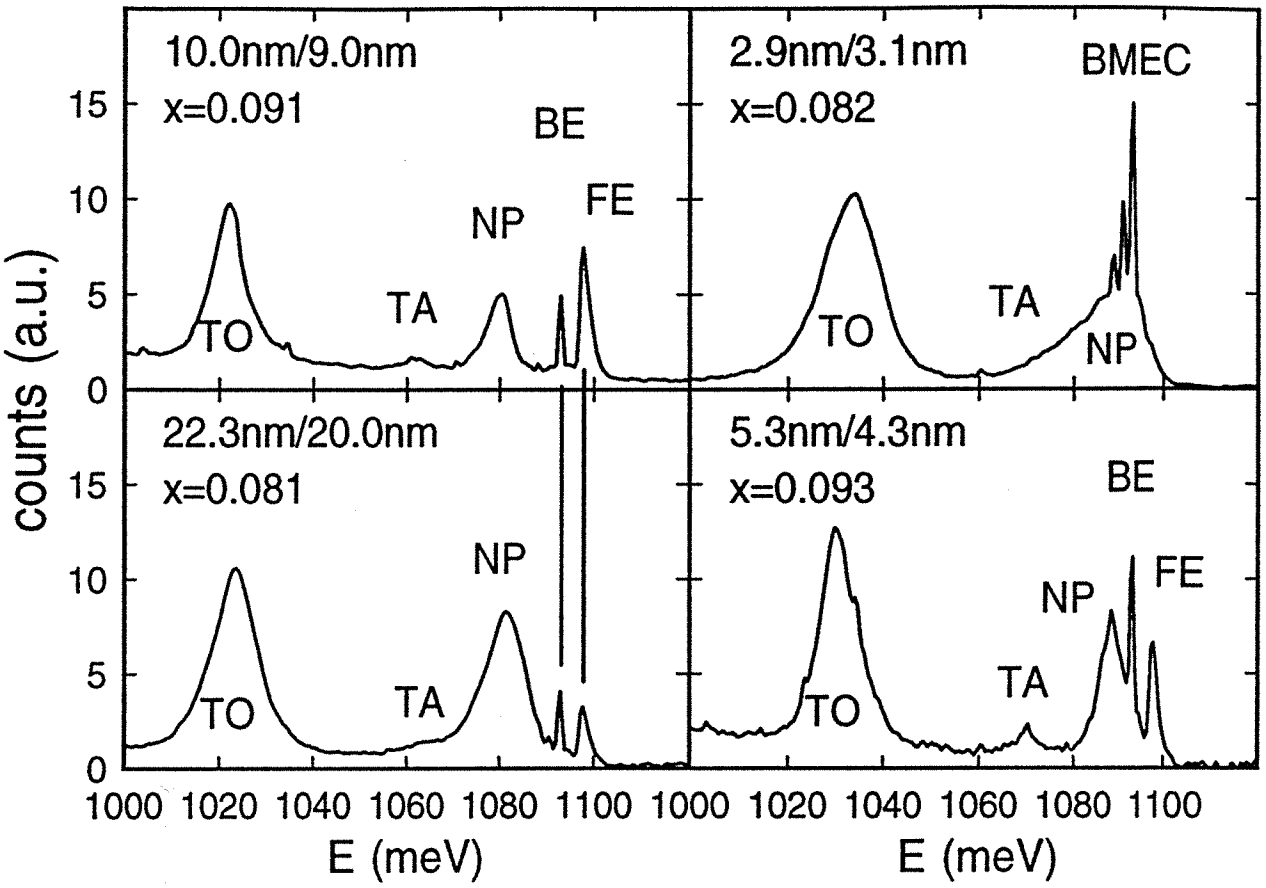

Figure 2: Photoluminescence spectra at 4 K from $Si/Si_{1-x}Ge_x$ MQWs. The peaks labelled FE, BE, and BMEC are the TO-phonon replicaa of the free and bound excitons from the boron-doped substrates, the features NP, TA, and TO are the no-phonon lines and phonon replicas originating from radiative recombination of electron-hole pairs confined in the $Si_{1-x}Ge_x$ alloy wells. They are shifted from the positions of Ref. 14 because of strain and confinement.

model are shown in Fig. 3 (solid line) in comparison with our data. Although the error bars of our data are fairly large (because of the error Δx of the Ge concentration as determined by the analysis of the X-ray rocking curve, usually about Δx=0.01), we see that our model gives good agreement with the data for the wider wells, but somewhat overestimates the confinement energies for the narrowest well. Sample imperfections, such as partial strain relaxation and interdiffusion of the layers, would tend to blue-shift the photoluminescence features and therefore cannot explain this discrepancy. Interface roughness, on the other hand, could cause the excitons to diffuse to wider regions in the well and red-shift the photoluminescence, but variations in the well widths of more than a few monolayers would be necessary to explain the small confinement energy observed for the sample with the narrowest wells. This is very unlikely and in clear contrast to the excellent uniformity of the layers observed in TEM. Interface roughness would also cause a shift to higher energies at elevated temperatures, when thermal activation reduces the red-shifting effects of diffusion, but no such shift was observed. Formation of superlattice minibands can also reduce the exciton energies, but we expect such effects to be small (less than 5 meV), because the fairly large valence band masses (compared to electrons in an $Al_{1-x}Ga_xAs/GaAs$ superlattice) do not allow a considerable leakage of the wave function into the barrier. The dependence of the hole masses on pressure and strain as well as non-parabolicity effects were also neglected in our model.[26]

6. CONCLUSIONS

We have discussed the advantages and limits of X-ray scattering in determining the sample parameters of $Si/Si_{1-x}Ge_x$

Sample	E(NP) meV	E(TO) meV	x %	L_W Å	E_c meV	E_{th} meV
SE1[a]	1082	1024	8.1	200	3(4)	3
SE2[a]	1080	1022	9.1	90	9(8)	10
SE3[a]	1088	1030	9.3	43	19(8)	26
PL3[a]	1091	1033	8.2	31	12(8)	36
489[b]	1043		18	23	43(25)	72
489-4[b]	1038		19	25	47(24)	68
416[b]	1030		17	29	22(16)	55
416−1B[b]	1015		18	34	15(17)	48
201[c]	1021		25	20	78	97
206[c]	1046		25	20	103	97
322[c]	1032		25	40	89	44
B[d]	1042	984	40	15	214	160

[a]This work. [b]Sturm *et al.*, Ref. 5.
[c]Steiner *et al.*, Ref. 6. [d]Glaser *et al.*, Ref. 7.

Table 3: Observed PL energies of the no-phonon line E(NP) and the TO replica E(TO) for samples with various Ge concentrations x and well thicknesses L_W. $E_c = E(\mathrm{NP}) - E_x$ can be interpreted as the confinement energy. The error from the uncertainty in the composition is given in parentheses. Values taken from the literature were added for comparison. E_{th} is the confinement energy calculated from an envelope-function model for a single potential well with finite barrier (masses in Table 1).

multiple quantum wells. We find that X-ray scattering can predict the band gaps and the position of the observed photoluminescence features very accurately, usually to within 1 meV, when the combined thickness of the $Si_{1-x}Ge_x$ wells is larger than 1000 Å. For the narrower wells, we find a blue-shift due to confinement, in reasonable agreement with an envelope function model. For the narrowest sample, however, this agreement is not so good. We conclude that more work is necessary to correlate the optical and structural properties of $Si/Si_{1-x}Ge_x$ quantum wells with well widths of less than 50 Å.

7. ACKNOWLEDGEMENTS

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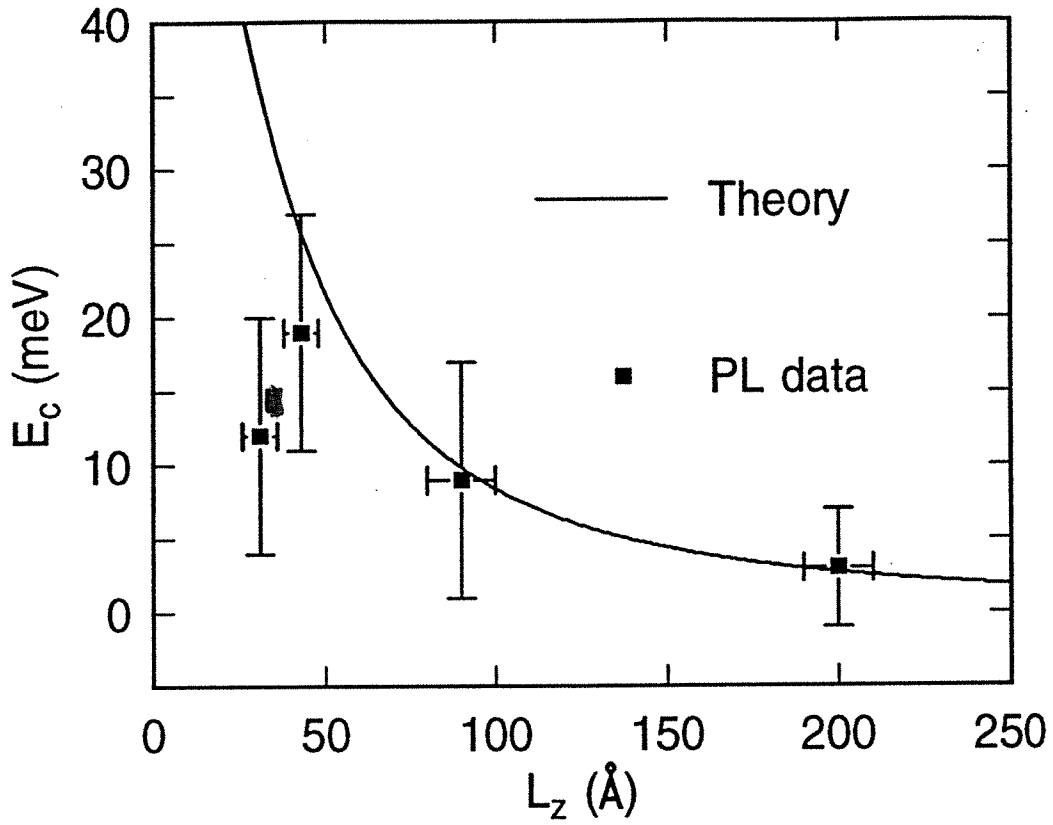

Figure 3: Confinement energies for a single $Si_{1-x}Ge_x$ quantum well with finite silicon barrier, calculated for x=0.08 (solid line) in comparison with the experimental data obtained in this work (symbols).

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