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Photoluminescence study of vertical transport in $Si_{1-x}Ge_x$/Si heterostructures

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We have observed band-edge photoluminescence from excitons in the center $Si_{0.75}Ge_{0.25}$ well of a triple-quantum-well structure, due to tunneling through the Si barriers from the two outer $Si_{0.87}Ge_{0.13}$ wells. Most of the photogenerated carriers originated in the Si substrate and cap, and migrated to the nearest $Si_{1-x}Ge_x$ wells (i.e., the outer wells), where in the absence of tunneling they became trapped. For Si barrier widths of 9 nm or less, center-well luminescence was observed, indicating the occurrence of tunneling. Time-decay measurements of the outer-well luminescence gave an estimated tunneling time of $\simeq 325\pm 100$ ns for 9-nm barriers, at which point luminescence was seen from both the center and outer wells. We also observed thermal hopping of carriers from the outer wells over the Si barriers and into the center well at high temperatures.

Semiconductor heterostructures have been a rich source of innovations in applied and basic research for the past twenty years. While Si-based heterostructure technology is relatively undeveloped compared to that of the III-V semiconductor systems, it has recently become the subject of intense scientific and technical interest. The advantages offered by heterostructures have already been realized in heterojunction bipolar transistors having $Si_{1-x}Ge_x$ bases.[1] *p*-type $Si_{1-x}Ge_x$/Si resonant tunneling diodes have also been demonstrated,[2-4] but because of the complicated valence-band structure for strained $Si_{1-x}Ge_x$ on Si, convincing identification of the observed resonance peaks with calculated bound-state energies has not yet been made. Magnetotunneling experiments have suggested the importance of band mixing and the strain-induced splitting of the light- and heavy-hole states in these structures.[2-4] Recently, hole band mixing has also been proposed to be essential in determining hole transfer in GaAs/Al_xGa_{1-x}As heterostructures.[5,6]

Photoluminescence (PL) studies of tunneling have proven invaluable in other materials systems and should similarly provide a means of better understanding the tunneling process in $Si_{1-x}Ge_x$/Si. In this paper we present evidence by PL spectroscopy of tunneling through Si barriers separating a deeper $Si_{1-x}Ge_x$ central well from two shallower $Si_{1-x}Ge_x$ outer wells. The photogenerated carriers were found to originate in the Si substrate and migrate to the nearest $Si_{1-x}Ge_x$ well[7] where, in the absence of tunneling, they became trapped. At high temperatures we observed thermal hopping of excitons over the Si barriers to reach the inner quantum well.[7]

The $Si_{1-x}Ge_x$/Si tunneling structures were grown epitaxially on ⟨100⟩ Si substrates by rapid thermal chemical-vapor deposition, as described previously.[7-9] The samples were made up of three $Si_{1-x}Ge_x$ quantum wells, each 10 nm in thickness, and consisting of two shallow wells with a nominal Ge fraction of $x \simeq 0.13$ on either side of a deep well having $x \simeq 0.25$. The Si barriers between each of the wells were varied in thickness from 4 to 25 nm. All samples had 15-nm Si caps on top of the upper well, and the structures were fully strained. An energy level diagram for hole states in the shallow and deep wells, assuming no coupling between them, is given in Fig. 1. We have used the 6×6 Luttinger-Kohn Hamiltonian described by Xiao *et al.*[9] The conduction-band offsets were taken to be zero, while the valence-band offsets were determined from the observed PL peaks, assuming recombination from the heavy-hole ground state.

The PL measurements were taken using a Fourier transform interferometer (Bomem DA8.02) with an

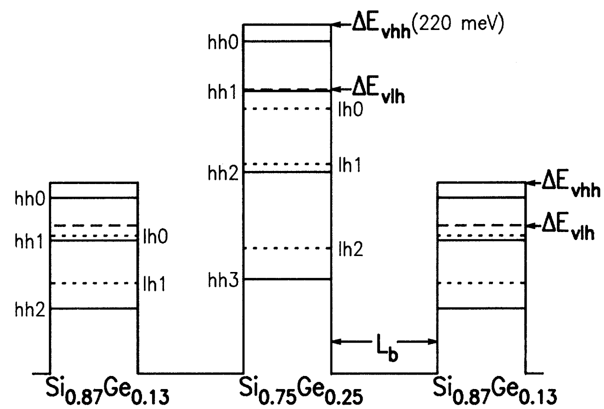

FIG. 1. Energy level diagram for heavy-hole (solid lines) and light-hole (dashed lines) states relative to the excitonic band edges in the triple-well structures, assuming no coupling between the wells. Solutions were found using the 6×6 Luttinger-Kohn Hamiltonian describe by Xiao *et al.* (Ref. 9). The valence-band offsets were determined from the observed PL peak energies for recombination from the heavy-hole ground state and conduction-band offsets of zero. The Si barrier width is indicated by L_b.

$In_xGa_{1-x}As$ detector. Above-band-gap excitation at 488 nm was provided by an Ar-ion laser. For the time decay studies the Ar laser was pulsed by an acousto-optic modulator, with the luminescence dispersed by a double spectrometer and detected with a photomultiplier tube (Varian VPM159A3) operated in the photon-counting mode. The samples were usually immersed in liquid helium, except in the elevated temperature studies where they were attached to a temperature-controlled Si block and placed in a flowing helium gap Varitemp Dewar.

Figure 2 shows PL spectra for triple-quantum-well structures with Si barriers of (a) 15, (b) 12, (c) 9, and (d) 4 nm. The PL features can be identified with shallow bound exciton luminescence from excitons confined in either the deep ($x=0.25$) or shallow ($x=0.13$) quantum wells. In each case the no-phonon (NP) transition is accompanied by the usual $Si_{1-x}Ge_x$ TO and TA phonon replicas to lower energy.[8] The observed NP peaks at about 1044 and 946 meV agree with the excitonic band gaps expected for the shallow ($x=0.13$) and deep ($x=0.25$) quantum wells, respectively.[10] Note that for barriers thicker than 9 nm there is no deep (center) well luminescence, while for barriers thinner than 9 nm there is no shallow (outer) well luminescence. Luminescence from both the deep and shallow wells can be seen in the sample having 9-nm barriers.

Our $Si_{1-x}Ge_x$/Si structures consist of only three thin quantum wells, so very little of the Ar excitation is absorbed within the $Si_{1-x}Ge_x$ layers themselves. Instead, most of the photocreated carriers originate in the Si substrate and cap layers, and some of these carriers subsequently migrate to the $Si_{1-x}Ge_x$ well regions. The integrated intensity of the $Si_{1-x}Ge_x$ PL signal is typically greater at 4.2 K than that of the Si even for a single 10-nm $Si_{1-x}Ge_x$ well. As the bound excitons in the Si dissociate from the impurities at high temperatures more free excitons find their way to the $Si_{1-x}Ge_x$ regions, so that while the Si signal drops, the $Si_{1-x}Ge_x$ PL increases by a factor of nearly 5 between 4.2 and 77 K. The $Si_{1-x}Ge_x$ PL is observed at temperatures up to 170 K for $x=0.25$.

The deep well PL in the spectra of Figs. 2(c) and 2(d) arises from carriers which have tunneled from the shallow wells through the Si barriers. As discussed above, too few excitons are directly generated by the Ar excitation within the center $Si_{1-x}Ge_x$ well to expect a PL signal in the absence of transport from the outer wells. This contrasts with direct-gap semiconductors where the stronger above-gap absorption results in a large number of carriers created near the surface, within the wells and adjacent barrier regions. In such cases all the wells luminesce, so that usually a bias is applied to create conditions for resonant tunneling to enhance the PL signal from individual wells. We expect that in our tunneling structures the carriers effectively equilibrate in the shallow outer wells, and any reaching the deep center well can arrive there only by tunneling at low temperature or by thermal excitation over the Si barriers at higher temperatures. This means that the PL intensity of the deep well is a direct indication of the tunneling or thermal hopping rate. Note that at low temperatures the photoexcited carriers quickly relax to the band edges, eliminating the possibility of resonant scattering by the outer wells.

The presence of the deep well in the samples having barriers too thick to allow tunneling can be investigated by thermally exciting carriers out of the shallow wells. Figure 3 shows the evolution from shallow to deep well PL as the temperature is raised from 77 to 120 K for a sample having no observed tunneling (Si barriers of 25 nm). Here the excitons trapped in the shallow wells acquire enough energy to thermally hop into the Si barriers and subsequently become captured by the deep well. The Arrhenius plot in the inset indicates an activation energy of the deep well PL of 145 meV, where we have neglected any variations in the density of states with temperature.

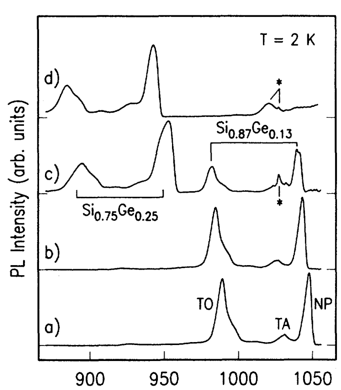

FIG. 2. Shift from outer-well to center-well luminescence with variation in Si barrier width. The outer-well PL, labeled $Si_{0.87}Ge_{0.13}$, is seen for (a) 15, (b) 12, and (c) 9 nm. In (d) 4 nm and (c) 9 nm, the barriers are thin enough to allow tunneling so that the center-well PL (labeled $Si_{0.75}Ge_{0.25}$) is observed. In each case the NP peak is accompanied by TO and TA phonon replicas. The starred features originate from the Si substrate.

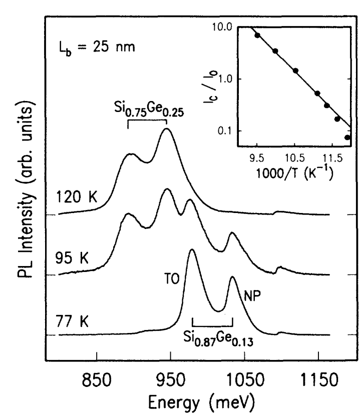

FIG. 3. Evolution of center-well ($Si_{0.75}Ge_{0.25}$) PL as the outer well ($Si_{0.87}Ge_{0.13}$) is depopulated by increasing the temperature from 77 to 95 K, then 120 K in a sample with no observed tunneling (25-nm Si barriers). The inset shows an Arrhenius plot of the center-well TO intensity (I_c) normalized by the outer-well NP peak (I_0). A fit to the data gives an activation energy of 145 meV, in agreement with the depth of the outer well as observed in the PL (132 meV).

This energy agrees roughly with the depth of the heavy-hole ground state in the shallow well relative to the Si barriers, as measured by the PL shift of 132 meV for this sample (the shallow well PL here is somewhat deeper than that of previous samples, probably due to an increase in the Ge fraction from the nominal 13% value). This confirms that the photoexcited carriers are initially trapped in the shallow outer wells at low temperatures.

The temporal decay of the shallow well luminescence at low temperatures was faster in structures having narrow Si barriers, since in addition to radiative and nonradiative channels, carriers escape the wells by tunneling. Lifetimes of $\tau_{9\ \mathrm{nm}} \simeq 210 \pm 40$ ns and $\tau_{12\ \mathrm{nm}} \simeq 600 \pm 100$ ns were obtained as shown in Fig. 4 for samples at 4.2 K with nominal Si barriers of (a) 9 and (b) 12 nm, which correspond to the cases discussed above of some tunneling [Fig. 2(c)] and no observed tunneling [Fig. 2(b)], respectively. The errors in the lifetime values reflect variations in the decay curves with excitation power. The PL decay from the sample with the 15-nm barriers [Fig. 2(a)] was comparable to that observed for the 12-nm barriers.

As evident by the less than 50-ns rise times in Fig. 4, the transport of carriers from the Si into the outer well occurs on a much faster time scale than the outer-well PL decay. This is expected, given that in Si the free exciton capture rate by impurities is very fast at liquid-He temperatures, on the order of ns even in relatively pure samples. In addition, the Si free exciton lifetime is further reduced in our structures by their capture by the $Si_{1-x}Ge_x$ wells. Very quickly after the laser pulse the free excitons either successfully diffuse the short distance to the $Si_{1-x}Ge_x$ wells or become trapped in the substrate as bound excitons. The observed lifetime for the samples with the thick barriers (600 ns) is therefore associated only with the dynamics of nonradiative and radiative processes intrinsic to the outer wells. Assuming that these same processes occur in the outer wells of the 9-nm barrier samples, the outer-well PL decay time is simply reduced by the tunneling through the barrier: $1/\tau_{9\ \mathrm{nm}} = 1/\tau_t + 1/\tau_{12\ \mathrm{nm}}$. Substituting our measured values we get a tunneling escape rate from the shallow wells of $\tau_t \simeq 325 \pm 100$ ns for the 9-nm Si barriers. These rates suggest that approximately twice as many excitons escape to the center well by tunneling ($\tau_t \simeq 325$ ns) as remain in the outer well ($\tau_{12\ \mathrm{nm}} \simeq 600$ ns). This is roughly in agreement with the observed relative intensities of the outer- and center-well PL [Fig. 2(c)], assuming that the radiative efficiencies of the wells are similar. Such slow tunneling rates have not been measured in quantum wells of direct-gap semiconductors,[5,11] due to the much shorter radiative lifetimes. The long exciton lifetime in $Si_{1-x}Ge_x/Si$ allows for a larger number of tunneling attempts so that we observe tunneling despite conditions, as discussed below, of very low transmission probability.

It is unlikely that the limiting tunneling process is due to electrons since the conduction-band offset is known to be very small [$\lesssim 3$ meV (Refs. 12 and 13)] for $x \leq 0.25$. While time-resolved PL spectroscopy might prove useful in detecting whether any of the recombining excitons having spatially separated electrons and holes, for this preliminary study we simply presume that the presence of

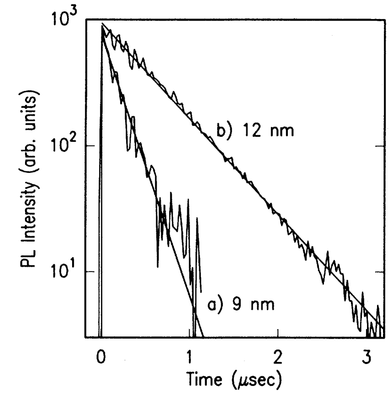

FIG. 4. Time decay of the outer-well PL ($Si_{0.87}Ge_{0.13}$) for (a) 9-nm Si barriers is faster than for (b) 12-nm Si barriers, consistent with the evidence of tunneling in the PL spectral features (Fig. 2). The solid lines show fits to the data which correspond to lifetimes of (a) 210 and (b) 575 ns. The PL intensities have been scaled to give equal values at $t=0$. The PL rise times are less than 50 ns, indicating that the dynamics of the carrier migration from the substrate to the wells is much faster than the PL decay.

the deep PL is determined by either hole or exciton tunneling. The question of whether the excitons tunnel as a whole or each particle arrives separately is still under study even in $GaAs/Al_xGa_{1-x}As$ (Ref. 11) and we will not attempt any discussion here.

Another issue is whether the tunneling occurs by a resonant or a nonresonant process. Without the added degree of freedom of an applied bias, it is unlikely that occupied eigenstates in the shallow well will be coincidentally resonant in energy with levels in the deep well. We can quantify this somewhat by calculating the transfer integrals for the two isolated well eigenstates with energy separation ΔE, to determine the strength of the well coupling constant λ_c. The condition for appreciable tunneling is then that $2\lambda_c \gg \Delta E$.[5] For simplicity we assume that the light-holes states in the shallow quantum well (initial state) are unoccupied and neglect any coupling between the hole bands. This is consistent with the observation of Fig. 3 that the deep well PL activation energy corresponds to the shallow well depth for heavy holes. For heavy-hole tunneling between the shallow well heavy-hole ground state and the two nearest deep well heavy-hole states, $n=2$ and 3, we obtain $2\lambda_c \simeq 0.003$ and 0.012 meV, respectively, for Si barriers of 9 nm. Since it is unlikely that the eigenenergies of the two wells are accidentally tuned to within this amount, we speculate that the tunneling occurs by some nonresonant scattering process. However, as discussed earlier, the situation is considerably more complicated than outlined above, with the likelihood of there also being some enhanced hole tunneling due to mixing between the light- and heavy-hole bands.

In conclusion, we have used PL spectroscopy to observe tunneling between $Si_{1-x}Ge_x$ quantum wells through Si barriers. Because most of the photogenerated carriers originate in the Si substrate, the PL intensities from the different wells provided a direct measure of the tunneling. Despite the large hole effective mass in $Si_{1-x}Ge_x/Si$ ($m_{hh} \simeq 0.3m_0$) and the unlikelihood of resonance due to the asymmetry in these structures, the tunneling occurred through remarkably thick (9 nm) Si barriers. This may be due to the very long exciton lifetime and suggests $Si_{1-x}Ge_x/Si$ is an ideal system in which to further study nonresonant tunneling processes and hole band mixing.

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