Photoluminescence from electron-hole plasmas confined in $Si/Si_{1-x}Ge_x/Si$ quantum wells

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We report the first observation of photoluminescence from electron-hole plasmas in $Si/Si_{0.8}Ge_{0.2}/Si$ quantum wells. While at liquid helium temperature, luminescence due to shallow bound excitons is observed. At 77 K electron-hole plasma (EHP) luminescence dominates the spectra over a wide range of pump powers. Convolution of the occupied electron and hole densities of states gives an excellent fit to the photoluminescence line shape. A band-gap reduction of up to 15 meV at high carrier densities is observed for wide quantum wells, but no such shift is detected for narrow quantum wells.

A detailed understanding of the physical processes underlying luminescence in $Si_{1-x}Ge_x$ alloys, especially at high carrier densities, is a prerequisite for any attempt to develop light-emitting devices. At high carrier densities, the degenerate electron-hole system, in both pure Si and pure Ge, has been studied extensively and is well understood, but it has not at all been studied in $Si_{1-x}Ge_x$ strained layers. Such a study requires a growth technique capable of yielding well resolved excitonic features, which have only recently been observed.[1] We report the observation of luminescence from electron-hole plasmas at high carrier densities at 77 K from both two-dimensional (2-D) and three-dimensional (3-D) $Si_{1-x}Ge_x$ strained layer systems.

The samples consisted of a single strained epitaxial $Si_{1-x}Ge_x$ quantum well nominally undoped between undoped epitaxial silicon layers. The samples were grown by a combination of rapid thermal processing and chemical vapor deposition[2] on (100) silicon substrates. Sample 728 has an atomic germanium fraction of $x=0.20\pm0.02$ and a well width of 33 ± 3 Å, determined by a combination of high resolution cross-section transmission electron microscopy (HRTEM) and Rutherford backscattering spectroscopy (RBS). Sample 729, grown under identical conditions, also has $x=0.20$, confirmed by x-ray diffraction, and an expected $Si_{0.8}Ge_{0.2}$ width of 500 Å. Based on previous experiments, the misfit dislocation density is expected to be much less than $10^2\ cm^{-1}$. The photoluminescence (PL) spectra were taken using excitation from an argon-ion laser. The 2 and 77 K spectra were taken with samples immersed in liquid helium and liquid nitrogen baths, respectively.

At low temperature, PL spectra (Fig. 1) are dominated by luminescence due to shallow bound excitons, similar to what has been reported in Ref. 1. The $Si_{1-x}Ge_x$ contribution to the spectra has a no-phonon (NP) transition as the highest energy component, and four phonon-assisted replicas at lower energies, which are attributed to the transverse acoustical (TA) phonon replicas and to three distinct transverse optical (TO) replicas due to Ge—Ge, Ge—Si, Si—Si. The spectrum of the 500 Å sample appears similar, although at lower energies due to reduced quantum confinement effects and with a somewhat wider linewidth (full width half maximum = 10 meV). The relative intensities of four replicas, which will be used in the fitting of high-temperature spectra, are obtained from these low-temperature spectra. The relative intensities of the Si-Si, Si-Ge, and Ge-Ge TO phonon replicas agree very well with the statistical bond-counting model of Weber and Alonso.[3]

At 77 K the PL spectra (Fig. 2) show two very broad peaks, which are strongly dependent on the excitation power. In the 33 Å well sample, with increasing excitation power density the lower edge of the luminescence lines hardly changes while the high energy tail moves upwards. Because the sample is immersed in liquid nitrogen, it is thought that the high energy tail is not due to local heating. This was confirmed by fitting the luminescence of the TO replicas from the silicon substrate to a classical free-exciton line shape,[4] which gives sample temperatures differing from the bath temperature by 3° in the worst case. The power dependence of the line shape of the $Si_{1-x}Ge_x$ emission precludes the possibilities of its origin as being due to free excitons or due to an electron-hole liquid (EHL).[5]

We identify this signal as luminescence from an electron-hole plasma confined in a $Si_{0.8}Ge_{0.2}$ quantum well.

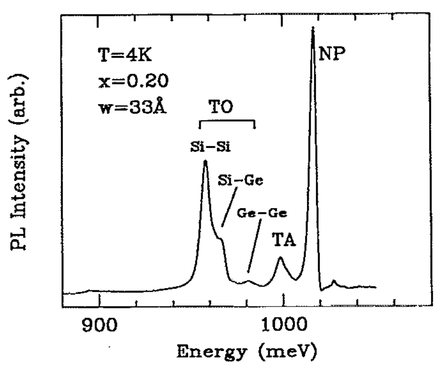

FIG. 1. PL spectrum of sample 728 at 2 K at a power density of 1 W/cm², illustrating NP and phonon replica emission.

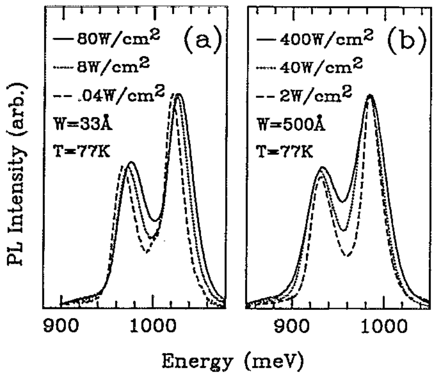

FIG. 2. PL spectra at 77 K at various excitation levels of (a) a 33 Å well, and (b) a 500 Å well.

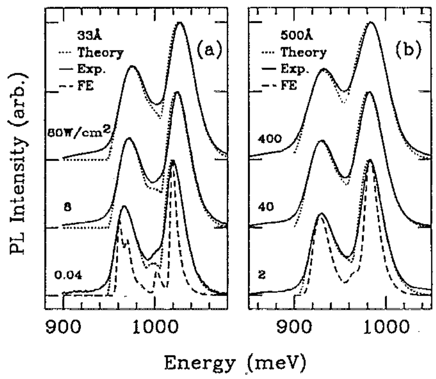

FIG. 3. Theoretical fitting of PL spectra by plasma model for (a) 33 Å well sample, and (b) 500 Å well sample. For comparison, also plotted here are (a) 2-D and (b) 3-D free exciton line shapes.

From other experiments in our lab we have found that at 77 K carriers generated in the silicon can travel long distances ($>0.1\ \mu$m) to be trapped in the $Si_{1-x}Ge_x$ quantum wells. While it is known that for Si/strained $Si_{0.8}Ge_{0.2}$ a substantial valence band offset (~ 150 meV)[6] exits to trap holes, it is not known whether electrons are trapped in the $Si_{0.8}Ge_{0.2}$ by a small conduction band offset[7] or by Coulomb attraction to the holes. The resulting carrier densities in the well, will then be much higher than that in the Si. The quasi-Fermi levels in the $Si_{1-x}Ge_x$ then move into the conduction and valence bands, resulting in the broadening line shape. To confirm this assignment, we fit the no-phonon line shape to the following expression for luminescence from EHPs:[8]

$$I(h\nu) = I_0 \int_0^{h\nu - E_{BB}} dE D_e(E) D_h(h\nu - E_{BB} - E) f(E,F_e,T) f(h\nu - E_{BB} - E, F_h, T) \quad (1)$$

where D_e and D_h are densities of states of electron and hole, respectively, F_e and F_h are the respective quasi-Fermi energies, $h\nu$ is the energy of photon emitted, and E_{BB} is the renormalized band gap due to the high carrier densities for the EHP. The f's are the Fermi functions for electrons and holes. The electron Fermi energy F_e and hole Fermi energy F_h are related through charge neutrality. The line shape given by the above expression does not depend directly on the values of the density-of-states effective masses of electrons and holes. Rather, it depends only on the dimensionality of the carrier distribution and quasi-Fermi levels. For a 3-D distribution (sample 729) $D(E) \propto \sqrt{E}$, while $D(E)$ is a constant for a 2-D distribution (sample 728).

Another issue, which needs to be addressed before attempting to fit the whole spectrum is the severe overlap among the NP PL line and its phonon replicas at 77 K. This is overcome by noting that, assuming constant phonon energies and matrix elements, these direct-allowed indirect transitions all have the same line shape no matter whether they are alloy-scattering-assisted as the NP line or phonon-assisted as for other phonon replicas. To obtain the final spectrum, which is a simple summation of the no-phonon line and all the phonon replicas, one then needs to know the relative intensities of these various lines. For phonon replicas, the relative ratio is extracted from the 2 K PL spectra, where they are well resolved. Due to the fact that at low temperature PL is dominated by bound excitons, while at high temperature it is from electron-hole plasmas, the relative intensity of the NP line with respect to the phonon-assisted replicas is left as an adjustable parameter, which should only depend on the composition of film. It should be pointed out that in doing so we have neglected 2-phonon processes. There are, in total, three adjustable parameters for fitting: the relative weight of the NP line r_{NP}, the renormalized band gap for the EHP E_{BB}, and the sum of electron and hole Fermi energies $F = F_e + E_h$. While r_{NP} determines relative height of the two peaks in high-temperature spectra, E_{BB} moves the spectrum along energy axis, Fermi energy F determines the line shape. The ratio of the Fermi energies F_e/F_h has little effect on the line shape.

With fixed r_{NP} and E_{BB}, varying F alone gives excellent fitting to all three spectra of sample 728 (33 Å), shown in Fig. 3(a). The relative strength of the various TO peaks was not a function of pump power, and the fitting was done with a fixed relative TO intensity as described above. With increasing excitation, the line shape remains well behaved with the high energy tails shifting upwards, suggesting no phase separation occurs between an electron-hole plasma and free excitons.[9,10] In contrast to sample 728, which has a well thickness of 33 Å, sample 729 with a 500 Å well shows quite a different dependence of the line shape on the excitation level [Fig. 2(b)]. As the excitation level increases, a pronounced change is found on the low energy side of the lines, and it was necessary to vary E_{BB} (renormalized band gap) to obtain the fits of the model shown in Fig. 3(b). The renormalized band-gap energies E_{BB} of both samples obtained from best fits are plotted versus excitation power density in Fig. 4. While sample 728 (33 Å) shows less than 2 meV change in E_{BB} over a range of more than 4 decades excitation level, a reduction of 15 meV is evident for sample 729 (500 Å well). This difference in band-gap renormalization suggests that there is a substantial difference in screening effects between 2-D and 3-D systems.

In our fitting of the spectra we have entirely ignored

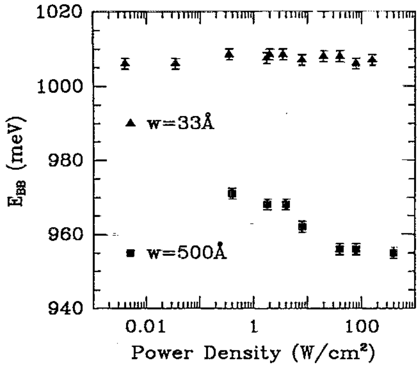

FIG. 4. Renormalized band-gap energy E_{BB} obtained by fitting vs excitation power density for both samples 728 and 729. The relative power densities are accurate within 10%, but the uncertainty in the absolute values is a factor of two.

free excitons, which usually dominate silicon PL at 77 K at low power densities. This is now examined on both theoretical and empirical grounds. Theoretically, at low carriers densities, electrons and holes form excitons, while at high carrier densities, excitons will dissociate into electron-hole plasmas. The Mott criterion[11] for such a FE-EHP transition is given by $n_c = k_B T / 16\pi E_x a_x^3$,[7] where n_c is the critical density above which an EHP is favored, E_x is the exciton Rydberg, and a_x is the exciton Bohr radius. Assuming Si-like effective masses, the critical density is 8.8×10^{16} cm^{-3} at 77 K. Using a simple model of densities-of-states for strained $Si_{0.8}Ge_{0.2}$, from our fitting we estimate carrier densities for the points of Fig. 4 from $\sim 2\times10^{17}$ to 1×10^{18} cm^{-3} in both samples 728 and 729. (The sublinear dependence of carrier density in the $Si_{0.8}Ge_{0.2}$ quantum well on pump power may be due to a reduction of the carrier collection efficiency into the quantum well because of band bending induced by charged carriers or due to increased nonradiative Auger recombination at high carrier densities.) These estimated carrier densities are well above the critical density for plasma formation at high power, but close to the transition criteria at low powers. Therefore we attempted to fit the spectra of samples 728 and 729 with 2-D and 3-D free-exciton models, respectively. As can be seen [Figs. 3(a) and 3(b)], a very poor fit was obtained. This leads us to believe that an electron-hole plasma is responsible for the luminescence at all power densities measured, except for possibly the 500 Å sample at the lowest power density (0.4 W/cm^2), which has a linewidth close to that of a FE line shape. At 60 K, pure free-exciton PL with a Boltzmann line shape[1] has been seen in $Si/Si_{0.8}Ge_{0.2}$ superlattices (not isolated quantum wells) grown in the same reactor as samples 728 and 729. Although these superlattices were wider ($\sim$2500 Å) than the quantum wells studied here, the reason for the different nature of the PL is not known and is under further investigation.

In summary, we presented the first observation of photoluminescence from electron-hole plasmas confined in strained $Si_{1-x}Ge_x$ single quantum wells. The PL spectra can be well fitted by a simple convolution of occupied electron and hole densities of states, with contributions from the NP line and the various phonon replicas included. While 2-D EHPs show little evidence of band-gap reduction, shift of the EHP renormalized band gap up to 15 meV has been observed for wide quantum wells.

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