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Photoluminescence Processes in $Si_{1-x}Ge_x/Si$ Heterostructures

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Over the past three years we have witnessed a rapid improvement in the optical quality of $Si_{1-x}Ge_x/Si$ heterostructures, so that it has now become almost routine to observe strong and well-resolved near-band-gap photoluminescence from structures grown by both CVD and MBE. As a result, PL has become an important characterization tool for these systems, and has also opened the way for new studies of the physics of indirect excitons in quantum wells of disordered alloys, which are the subject of this presentation.

Following the first report of near-gap photoluminescence (PL) from a thick, relaxed layer of Si_{1-x} $Ge_x/Si(100)$ having x = 0.15 and a thick, possibly partially relaxed x = 0.04 layer by Terashima *et al.*[1], and our subsequent observation[2] of well resolved PL from fully strained thin multi quantum wells (MQW) with x ~ 0.20, there have been many reports of similar PL from SiGe layers grown both by molecular beam epitaxy (MBE) and by chemical vapor deposition (CVD). This presentation will not attempt to review all of these contributions, but rather emphasizes new results which clarify the physical processes involved in these PL spectra.

A typical near-gap PL spectrum begins on the high-energy side with a no-phonon (NP) line, followed by momentum-conserving phonon replicas including the TA and three distinct TO replicas characteristic of Ge-Ge, Si-Ge and Si-Si vibrations, similar to what had earlier been observed in bulk, relaxed SiGe alloys.[3] The origin of the PL observed at ℓHe temperatures in these early studies[1,2] was impurity bound excitons (BE), as was also the case for bulk, relaxed alloys[3]. This was most clearly demonstrated by temperature dependent PL studies, as shown in Fig. 1. Here the NP BE line dominates at ℓHe temperature, but the BE becomes thermally

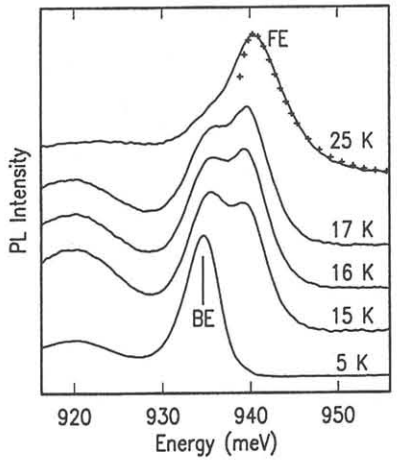

Fig. 1: NP PL of a 10 nm x = 0.25 single well sample *vs.* temperature. The crosses are a fit using a 25 K free exciton lineshape.

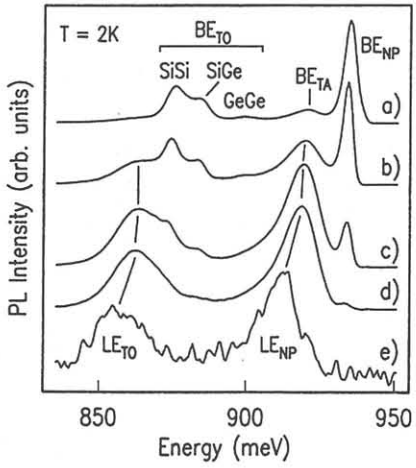

Fig. 2: PL spectrum of a 10 nm x = 0.25 QW at excitation levels of a) 3.2 Wcm^{-2}, b) 0.32 Wcm^{-2}, c) 30 cm Wcm^{-2}, d) 2.3 Wcm^{-2} and e) 2.4μWcm^{-2}.

dissociated at higher temperatures, so that the free exciton (FE), with its characteristic Maxwell-Boltzmann lineshape dominates. The BE binding energy relative to the FE is ~ 4 meV, both from the spectral displacement and from the activation energy of the thermal dissociation, which is close to the 4.5-5 meV binding energies seen in Si. Note that it is possible to see a NP FE line here (but not in pure Si) because the random alloy scattering relaxes the k-selection rule.

However, it was soon discovered that the PL from SiGe/Si systems could be considerably more complex than the BE/FE pair of lines.[4] As shown in Fig. 2, at low excitation densities a broad NP band shifted down in energy by ~ 20 meV from the BE NP line was observed in a wide variety of SiGe samples. This "LE" band was interpreted as due to excitons localized in small regions of higher than average Ge content (and thus lower bandgap) which resulted from the random nature of the alloy. The LE band was found to have remarkably high PL quantum efficiency, >10%, and a long lifetime (≥1 ms).[4] The high efficiency of the LE band arises from the localization of the excitons in these alloy fluctuations, which prevents them from diffusing to impurities where non-radiative Auger decay dominates. The saturation of the LE PL at very low power levels results from 1) the very long radiative lifetime, 2) the very low density of such deep fluctuations, and 3) the fact that once a second exciton binds to a given fluctuation, Auger recombination becomes possible, so the radiative efficiency for the pair is ~ 10^{-3} that for a single exciton.

More recently we have found that with decreasing well width, the LE band tends to move closer to the BE, eventually becoming spectrally indistinguishable, as shown in Fig. 3. It is clear that two different PL processes are superimposed in Fig. 3e, both from the excitation level dependence of the PL as well as from the PL lifetime results shown in Fig. 4.[5] This clearly reveals that at low excitation levels the PL is dominated by LE while at high excitation it is dominated by a less efficient process with a much shorter lifetime.

We speculated[5] that this other, high excitation process need not be limited to impurity BE, but could also be due to biexcitons (or localized biexcitons). We have now demonstrated the importance of biexciton processes in SiGe PL under quite ordinary excitation conditions by observing the visible PL at ~ twice the SiGe band-gap, which is emitted when the two e-h pairs in a biexciton recombine simultaneously.[6] A typical visible PL spectrum from a sample containing 3 QW's of different thicknesses is compared with the infrared PL spectrum of the same sample in Fig. 5.

The complexity of the PL possible from a single QW is summarized in Fig. 6, obtained from a high quality MBE sample having very narrow linewidths. The NP region is shown for 3 different excitation levels at 4 different sample temperatures. Starting from the lower left, the low T, low power spectrum is dominated by very shallow LE, shallower than the BE, which lie only ~ 1 meV below the average alloy energy. While it has not been previously discussed in the literature, it is in fact quite clear that at ℓHe temperatures there should be no free excitons, even in pure SiGe, since the half-width of the alloy distribution averaged over an exciton volume is several meV, much greater than kT.

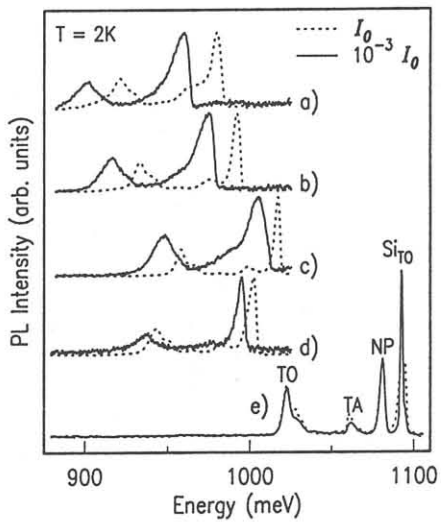

Fig. 3: PL spectra at two different excitation levels for a) an 8.3 nm x = 0.2 QW, b) a 5.8 nm x = 0.2 QW, c) a 3.3 nm x = 0.2 QW, d) a 1.5 nm x = 0.35 QW and e) a 1.2 nm x = 0.38 QW.

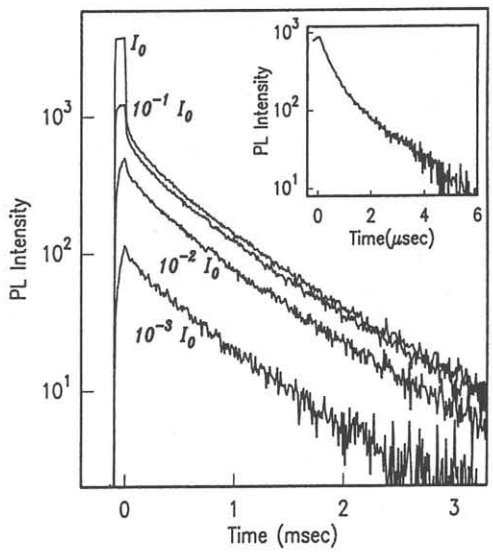

Fig. 4: Time decay of the PL from the 1.2 nm QW shown in Fig. 3e. The initial fast decay of ~ 1 μs is followed by a slow decay (750 μs).

The reason that the excitons are effectively trapped in these very shallow fluctuations in these samples, rather than hopping and tunneling to the deeper fluctuations as in Fig. 2, is related to the well thickness. In very thin wells such as shown in Fig. 3e or Fig. 6, the exciton is constrained to move in only two dimensions, which increases the localization by limiting the paths by which the exciton can diffuse to deeper fluctuations (or impurities).

Returning to the 1.7 K spectra of Fig. 6, at medium and high excitation the LE PL saturates and the less efficient BE PL dominates. Upon raising the temperature the shallow LE vanish with an activation energy of ~ 1/2 meV caused by transport to donors and/or acceptors responsible for the BE PL. Thus the 4.2 K spectrum of this sample is dominated by BE at all excitation levels. At 8 K the BE begin to dissociate and we begin to see a true free exciton peak. In the low excitation 8 K spectrum we can now observe an LE band below the BE, since the excitons are now sufficiently mobile to diffuse to the much less numerous deeper alloy fluctuations.

At 15 K the BE PL has disappeared, but at high excitation levels biexciton (BI) PL is clearly observable. Biexciton visible PL is also very strong under these conditions of temperature and excitation. The 'BE' peak in the 8 K high excitation spectrum likely also has a significant biexciton component.

In conclusion, we have seen that the physics of excitons in SiGe QW's can be quite rich, and that considerable care must be taken in assigning a label to a given line. Unlike in Si, at ℓHe temperatures there is no simple relationship between BE to FE intensity ratio and impurity concentration. We have also shown the importance of localization and of biexciton processes in excitation and temperature regimes which are quite common in such studies.

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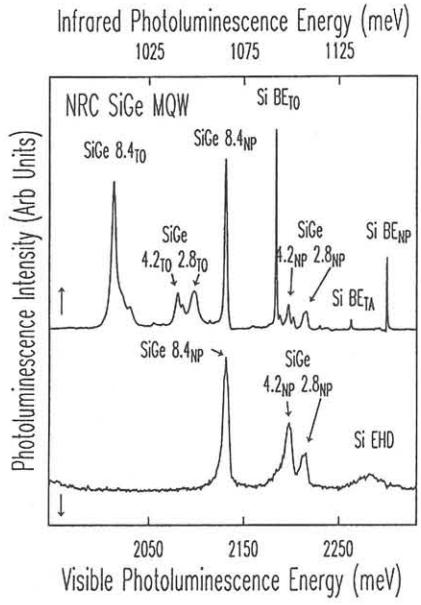

Fig. 5: Visible PL spectrum of an x = 0.14 MBE sample having 2.8, 4.2 and 8.4 nm wide wells is compared with the infrared PL on an energy scale reduced by two.

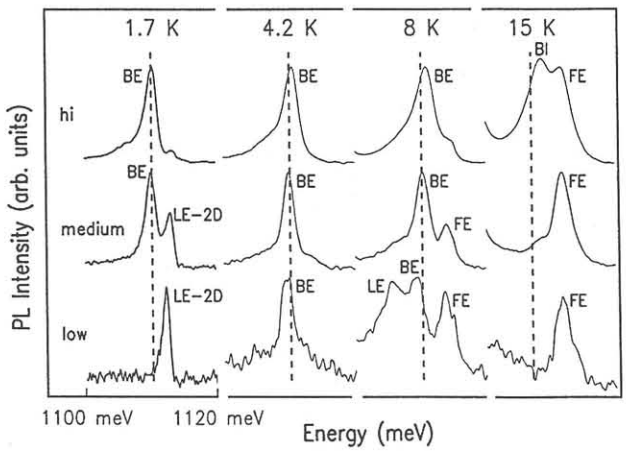

Fig. 6: NP PL of a high quality 2.8 nm x = 0.14 MBE QW at 3 different excitation levels and 4 different temperatures. The vertical dashed line in the four repeated spectral windows is at the position of the 1.7 K BE line.