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Time-resolved photoluminescence of pseudomorphic SiGe quantum wells

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We report low-temperature time-resolved photoluminescence experiments on a pseudomorphic SiGe quantum-well structure. Under the condition of optical absorption in the Si buffer layers, the decay time of the SiGe quantum-well luminescence is controlled by the capture of excitons and electron-hole droplets. From the onset of the SiGe luminescence, the exciton lifetime in the investigated 59-Å-wide $\mathrm{Si}_{0.72}\mathrm{Ge}_{0.28}$ quantum wells is found to be about 100 ns.

For some years SiGe/Si quantum-well (QW) structures have been the subject of intense experimental and theoretical investigations. Part of these were concerned with the optical properties of this material system, bearing in mind optoelectronic applications in the wavelength regime between 1.3 and 1.5 μm. Pseudomorphic SiGe QW's on the Si substrate are therefore of particular interest, since they are more easy to combine with existing Si technologies as compared to Si/SiGe structures on relaxed SiGe buffers.

The optical properties of SiGe QW structures have been found to depend strongly on the growth conditions, in particular the growth temperature T_G. Structures grown by molecular-beam epitaxy (MBE) at $T_G=400\,^\circ\mathrm{C}$ showed mainly defect-related photoluminescence (PL) below the expected band gap.[1] Band gap-related PL was reported for SiGe QW structures with low Ge content (up to 15%) grown at $T_G=650\,^\circ\mathrm{C}$.[2] After that, excitonic QW PL was observed in samples grown by chemical-vapor deposition at $T_G=700\,^\circ\mathrm{C}$.[3] Since then SiGe QW structures grown at enhanced growth temperatures between $T_G=600$ and $700\,^\circ\mathrm{C}$ are regarded to be state of the art, when optical recombination properties are concerned.[4] The appearance of excitonic QW PL, however, seems not to be strictly linked to high growth temperatures. Even at $T_G=400\,^\circ\mathrm{C}$, QW PL was obtained in MBE-grown samples later on.[5] The optical recombination properties of SiGe QW structures therefore seem to be controlled by the presence of defects, which may, depending on their nature, cause defect-related PL or eventually act as nonradiative recombination centers. The degree of incorporation of such defects obviously depends on the growth conditions and on residual contaminations in the particular growth chamber. Currently the quality of SiGe QW structures is characterized mainly by analysis of the PL linewidth and the intensity of the SiGe-related emission lines. Nothing is known so far about the exciton lifetime in state-of-the-art SiGe QW's. The determination of the exciton lifetime in pseudomorphic SiGe QW structures is difficult. Since the optical absorption of SiGe QW's is very weak, the optical excitation has to be performed in the Si substrate. Under such conditions, the rate of recombination in the SiGe QW will depend on the efficiency of exciton transport to the SiGe QW.

In the present paper we report about time-resolved PL experiments on a pseudomorphic SiGe QW structure. For our studies we used the structure shown in Fig. 1, which was grown by MBE on a n^- Si-(100) substrate at a growth temperature of $T_G=720\,^\circ\mathrm{C}$ (see Ref. 5 for more details). After a 1700-Å-wide Si buffer layer, five $\mathrm{Si}_{0.72}\mathrm{Ge}_{0.28}$ QW's with a nominal width L_z of 58 Å have been grown. The QW's are separated by 300-Å Si buffer layers. Finally a 2000-Å-thick Si cap layer has been grown. All layers are nominally undoped. The ground-state exciton in the SiGe QW is formed between the four-fold-degenerate electron subband $XE_0(4)$ and the heavy-hole subband HH_0. The band diagram shown in Fig. 1 is based on deformation potentials and band gaps given by Van der Walle and Martin,[6] and on valence-band offsets given by Colombo, Resta, and Baroni.[7]

Optical excitation is provided by an Ar^+ laser ($\lambda_L=458$ nm) with incidence normal to the sample surface (see Fig. 1). The penetration depth of the incident light is about 5000 Å. Consequently, the optical excitation of electron-hole pairs takes place predominantly in the MBE-grown part of the structure, in close proximity to the SiGe QW's. For the time-resolved optical studies we use an acousto-optic modulator to generate light pulses with tunable length and a transient switching time of about 30 ns. The detector is a liquid-nitrogen-cooled

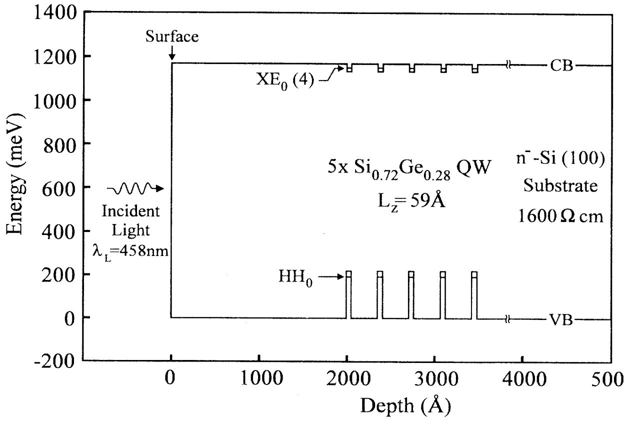

FIG. 1. Band diagram of the investigated pseudomorphic SiGe QW structure. Five 58-Å-wide $\mathrm{Si}_{0.72}\mathrm{Ge}_{0.28}$ QW's, separated by 300-Å barriers, are contained 2000 Å below the surface.

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Ge avalanche diode, which is operated as a single-photon detector in the Geiger mode (the present device is operated like related Si devices; see for example Ref. 8). The available time resolution of the detector is about 1 ns. The electrical pulses from the avalanche diode are registrated by a discriminator and analyzed by a time-to-amplitude converter with a fast analog-to-digital converter (ADC) and computer readout. The back-metalized sample was indium soldered to a copper plate and mounted in a cryogenic microscope setup. The PL light was dispersed with a 0.25-m grating spectrometer.

Time-integrated PL data are shown in Fig. 2. The data were recorded at an excitation power density P_L of 1 kW/cm^2 and a bath temperature of $T=10$ K. Four major PL lines are contained in the spectrum: Those are the TO phonon replica of the bound Si exciton at $E=1097$ meV; the TO replica of the electron-hole droplet (EHD) at $E=1081$ meV, which appears very intense because of the high excitation power density; the SiGe QW no-phonon (NP) line at $E=975$ meV; and the corresponding phonon replica at 923 meV. In Si, the condensation of EHD's is only possible at temperatures below $T_c \approx 27$ K.[9,10] The appearance of the EHD-related PL line therefore proves that the temperature within the photoexcited region of about 50-μm diameter can be kept below 27 K at a bath temperature of 10 K.

Time-resolved results for the same excitation power density are summarized in Fig. 3. For the experiments an optical pulsewidth of 2000 ns was used at a repetition frequency of 100 kHz. The turn-on and -off time of the laser pulse is about 30 ns. Time-resolved PL data are shown for all four lines labeled in Fig. 2. All curves are normalized to common peak amplitudes at the turn-off time of the laser pulse. The simplest time behavior is observed for the Si-TO line. The corresponding onset and decay of the PL signal follows an exponential decay law with a time constant of 800 ns. Time constants in this range have been observed earlier in time-resolved experiments on bulk Si.[11]

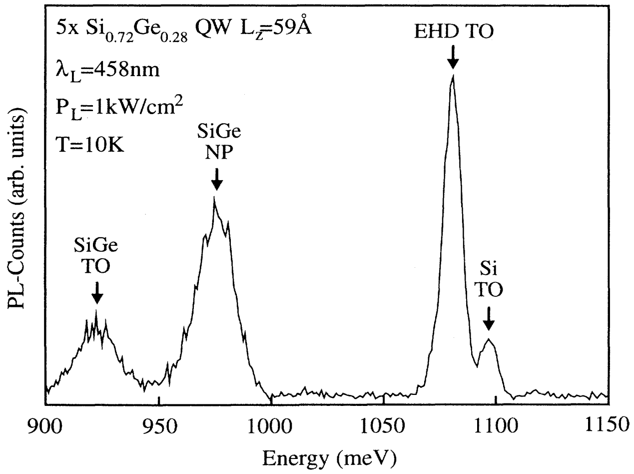

FIG. 2. Low-temperature PL spectrum recorded under the condition of continuous optical excitation. The Si- and SiGe-related PL lines are indicated.

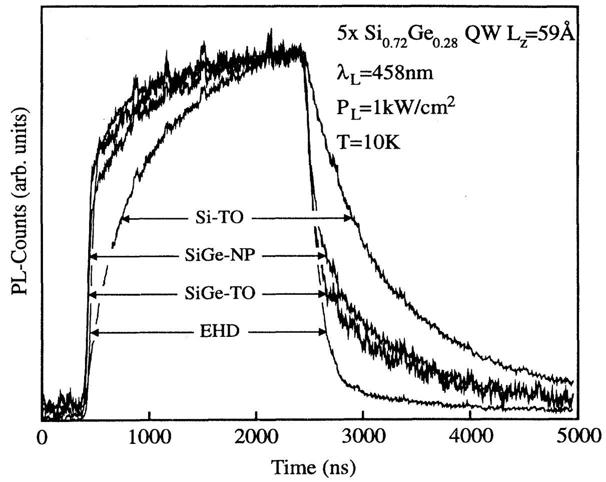

FIG. 3. Time-resolved PL data for Si- and SiGe-related lines. Shown is the time evolution of the various PL lines under the condition of an intense, pulsed optical excitation ($P_L=1$ kW/cm^2, pulse width 2000 ns) above the Si band gap.

The EHD, SiGe-NP, and SiGe-TO lines, in contrast, exhibit a more complicated behavior. The rise time of those lines is almost similar, and considerably faster than their corresponding decay times. The onset of the EHD PL is delayed by approximately 30 ns in comparison to the SiGe QW PL. In earlier work it has already been pointed out that such a behavior is presumably caused by the time delay associated with EHD condensation from a high-density exciton gas.[12] The decay of the EHD PL is approximately exponential, with a time constant of 140 ns. The SiGe QW PL first closely follows, after turn-off of the laser pulse, the decay of the EHD. After about 100 ns, however, the decay rate slows down and falls even below the decay rate of the bulk Si-TO line.

The observed dynamics of the various PL lines are controlled by a variety of different mechanisms. For discussion and interpretation of the present data, one has to keep in mind that the different PL lines originate partly from different regions in the sample. Excitons in Si have been shown to diffuse on macroscopic length scales. The average drift velocity of excitons from the photoexcited region close to the surface into the bulk has recently been found to be in the range of 5×10^4 cm/s.[13] Within a lifetime of 800 ns excitons in Si therefore are able to spread out on a length scale of typically 400 μm. EHD's, in contrast, can only condense within the region of high optical excitation within the penetration depth of the laser. EHD's therefore have high spatial overlap with the SiGe QW's. Under such conditions the dynamics of the Si EHD and the SiGe QW PL can be regarded to be closely coupled, whereas, in particularly on a short-time scale, the interactions with bulk Si excitons are not important.

The lifetime of EHD's in the present structure can be directly inferred from the decay of the EHD line. For evaluation of the EHD lifetime τ_{EHD}, the decay of the EHD line was numerically simulated with the simple rate equation $dn/dt=G(t)-n/\tau_{\mathrm{EHD}}$, which takes into account the experimentally determined turn-off time of the laser via the time-dependent generation rate $G(t)$. Good agreement between experiment and simulation is obtained for $\tau_{\text{EHD}}=140$ ns, as shown in Fig. 4. The obtained lifetime is also in good agreement with previous experiments on bulk Si, where τ_{EHD} has been found to be about 150 ns.[11,14–16] The major decay mechanism for EHD's in Si is given by exciton evaporation.[11]

The close relationship between the decay of the EHD and the SiGe QW PL after turn-off of the laser originates therefore from the close proximity between the EHD's and the SiGe QW's. The observed decay of the SiGe QW PL is determined by the capture of excitons from evaporating EHD's, and their subsequent decay in the SiGe QW's. After turn-off of the laser, within an EHD lifetime, the captured excitons are mainly supplied by evaporating EHD's. After the EHD's have ceased to exist (an EHD lifetime after laser turn-off), only bulk Si excitons can be captured, and the observed decay rate of the SiGe QW PL slows down. At delay times of 2000 ns and more, the apparent differential decay time of the SiGe QW already exceeds 1500 ns. The observed SiGe QW decay time is therefore only a consequence of exciton capture and transport, and not an intrinsic property of the SiGe QW's.

In the present experiment, however, information about the exciton lifetime in the SiGe QW's is contained in the rise time of the SiGe QW PL. As shown in Fig. 3, the rise times of the EHD and SiGe QW PL are effectively identical. The transient strength of both signals depends on the transient photoexcited electron-hole density in the direct vicinity of the SiGe QW. On time scales which are short compared to the lifetime of the bulk Si exciton, the onset of the EHD and SiGe QW PL can be described by the rate equation $dn/dt=G(t)-n/\tau_{\text{EHD}}-n/\tau_{\text{QW}}$, where the number of electron-hole pairs n is given by the time-dependent generation rate $G(t)$ and by the decay time of the EHD (τ_{EHD}) and SiGe QW (τ_{QW}). According to this rate equation the turn-on time of the EHD and SiGe QW PL is dominated by the lifetime of the fastest contributing recombination channel. For a detailed analysis the rate equation was solved numerically, using the experimentally determined, time-dependent generation rate, in order to account for the finite rise time of the laser pulse of about 30 ns. With $G(t)$ and τ_{EHD} given, τ_{QW} can be determined as the only remaining free parameter. From comparison between the experimentally observed turn-on of the SiGe-NP line and the solutions of the rate equation for different τ_{QW}, we are able to obtain $\tau_{\text{QW}}=104$ ns for the exciton lifetime in the SiGe QW (see Fig. 4 for a comparison between experiment and simulation). The experimentally observed onset of the SiGe QW lines is similar to the onset of the EHD line, which, however, is delayed by about 30 ns. In the present system the effective turn-on time is given by $\tau=\tau_{\text{EHD}}\times\tau_{\text{QW}}/(\tau_{\text{EHD}}+\tau_{\text{QW}})=60$ ns, which is more than a factor of 2 smaller than τ_{EHD}. In such a regime, the numerical evaluation of τ_{QW} can therefore be performed with high accuracy.

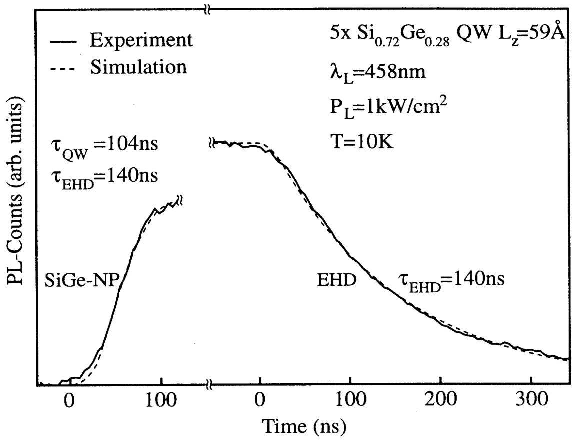

FIG. 4. Left: Experimental data for the onset of the SiGe-NP PL (solid line) and corresponding simulation (dashed line; see text). Right: the decay of the EHD PL (solid line) can be described by an exponential decay with a lifetime of $\tau_{\text{EHD}}=140$ ns (dashed line).

The recombination in the SiGe QW's is believed to be mainly nonradiative. In bulk Si the recombination of donor- and acceptor-bound excitons has been found to be controlled by a phononless Auger mechanism.[17–20] Depending on the type of impurity, bound exciton lifetimes between 1 and 300 μs have been measured,[18] which sets a lower limit on the radiative lifetime. If the radiative recombination time in SiGe QW's is significantly smaller than that, the SiGe PL intensity would be expected to be larger by orders of magnitude. Even with more than 70% of the photoexcited excitons captured by the SiGe QW's,[13] the relative intensity of the SiGe QW PL remains, however, only weakly enhanced compared to the Si-related lines. Nonradiative processes therefore dominate the recombination in SiGe QW's. The relevant mechanisms are so far unknown, however.

In summary we have performed low-temperature time-resolved experiments on a pseudomorphic SiGe QW structure under the condition of strong optical excitation above the Si band gap. The decay time of the SiGe QW PL has been found to be controlled by exciton capture from decaying EHD's and by the capture of bulk Si excitons. The exciton lifetime in the SiGe QW was determined from the onset of the SiGe QW PL. By numerical analysis on the basis of a simple rate equation, the exciton lifetime in the SiGe QW's was found to be nominally 104 ns. The observed lifetime is dominated by nonradiative recombination processes.

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