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Effect of growth conditions on optical properties of Ge submonolayer nanoiclusions in a Si matrix grown by molecular beam epitaxy

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Abstract. Optical properties of the Ge submonolayer (SML) nanoinclusions in a Si matrix grown by molecular beam epitaxy (MBE) are investigated. It is shown that at relatively high growth temperatures (> 600°C) there are new features appear in a PL spectra. These features correspond to formation of the germanium nanoobjects in a silicon media.

Introduction

Nowadays silicon is the base material in a microelectronics market but it is hardly applicable in optoelectronics due to its indirect band gap nature. Nevertheless, the integration on-the-same-wafer of the well-developed silicon microelectronics technology with optical devices is one of the most actual task which attracts significant efforts. There are proposed several approaches to realise silicon–based light-emission (SBLE) devices: using of porous silicon [1], using of Ge/Si and GeSiC/Si quantum dots fabricated by MBE [2], rare-earth doping of silicon [3], insertion of InAs nanoiclusions in a Si matrix [4], etc. Despite of progress in this area these approaches doesn't find industrial applications, yet. This point stimulates us to find out other ways allowing to get SBLE. In this paper we propose to insert Ge SML in a Si matrix and investigate optical properties of these heterostructures. Our approach is based on the following assumption: Ge submonolayers may lead to the formation of the ensemble of relatively small islands (with lateral sizes less than hole Bohr radius). This can result in a partial lifting of the selection rule for radiative recombination and possibility of exciton formation (which may be stable up to room temperature) via electron and localised–hole interaction. This situation is possible if Coulomb attraction energy will be higher to localise electrons near potential barrier which is produced by Ge SML inclusions in the conductivity band. In the case of relatively large sizes of quantum dots or large widths of quantum wells this barrier also may lead to spatial separation of the electrons and holes [5]. Furthermore, for the SML in other heteroepitaxial systems [6, 7] narrow PL line leads to the increasing of the absorption (gain). The PL intensity will increase if multiple vertical stacking of the layers with Ge SML separated by Si spacers is used. Due to relatively small strain accumulation in such system a low probability of dislocations and structural defects formation is expected.

Experiment

All structures are grown by molecular beam epitaxy using Riber SIVA 45 setup on a Si(100) n–type substrates (conductivity 3 $\Omega \cdot$ cm). Substrates are 5 inches in a diameter manufactured by OKMETIC. After chemical preparation by the method described in [13] the substrates are transferred in the MBE setup loading chamber. This method of preparation allows to remove oxide layer from silicon surface at 840°C in the growth chamber by direct radiating heating. During the growth process the rotation of the samples is used, temperature field inhomogenity across the surface is about $\sim$ 5%. In order to grow Ge SML we use submonolayer epitaxy technique, which we also used to grow SML insertions in A^3B^5 and A^2B^6 systems [8, 9, 10].

Structures consist of Si 1000 Å buffer layer, Ge(0.7–1.3 Å)/Si(44 Å) superlattice (20 pairs) and silicon 200 Å capping layer. Growth rates for Si and Ge are 0.5 Å/s and 0.05 Å/s, respectively. The growth rates is controlled by 2 mass-spectrometers with feedback which are set to 28 (Si) and 74 (Ge) masses. The substrate temperatures are varied from 600°C to 750°C. Total pressure during growth is better than $5 \cdot 10^{-9}$ Torr. Surface is monitored *in situ* by the reflection high–energy electron diffraction (RHEED).

Photoluminescence (PL) is excited by an Ar^+–laser ($\lambda = 514.5$ nm, maximal excitation density is $\sim$ 500W/cm^2). PL is detected by Ge cooled photodiode. During the growth process the RHEED patterns show independent behaviour on the growth temperature in comparison with initial surface reconstruction (2×2). Thus, even on the upper layers of the superlattice the surface remains atomically smooth and 3D–structure formation due to strain relaxation doesn't occur.

Results and discussion

As it was shown in [13] there is a new set of PL lines appear if the structure with SML Ge inclusions (structure consisted of 99 pairs of 0.7 Å Ge SML separated with 35 Å Si spacers) was grown at relatively high temperature ($>$ 750°C). These PL lines exist both at high and low levels of excitation density and correspond to emission from excitons localised at germanium islands in the SML superlattice. We have found that there are no such lines (marked in following as SL lines) in PL spectra if the growth temperature is below 600°C and above $\sim$ 800°C (due to effective silicon and germanium intermixing and formation of the solid solution). Increase in growth temperature leads to narrowing of SL PL line. A maximal PL intensity from SL is observed for the substrate temperature of 650°C (for active region) at other equal growth conditions.

For the set of the samples investigated in a frame of this work we have found that integral intensity ratio between SL–TO line and Si–TO line critically depends on the excitation density. In Fig. 1(a) are shown PL spectra taken at different excitation density for the sample with 20 layers of 1 ML of Ge separated by 44 Å Si spacers (growth temperature for the active region is 750°C). There is steady increasing of the integral intensity ratio of Si–TO/SL–TO lines with the increasing of excitation density, but even at very high excitation levels the integral intensity ratio of these lines is comparable. At low excitation levels SL–TO line dominates in spectra. Full width at half maximum is about 15 meV. There is SL–TO line shifting towards long–wavelengths with the increasing of the excitation density.

In Fig. 1(b) are shown PL spectra for the same sample taken at different temperatures and constant excitation density of $\sim$ 500 W/cm^2. There is significant intensity decreasing of the SL–TO line, which accompanies with blueshifting. Opposite situation (large redshift in compare with shift of band PL) was observed, for example, in InAs/GaAs heteroepitaxial system [12] and it is explained by both bandgap shifting and evaporation of carriers from small QDs having lower localization energy. In our case the observed phenomena cannot be explained in a frame of this simple model. and, indeed, is due to occupation of excited states on the islands with increase in temperature or redistribution of carriers between islands having different sizes.

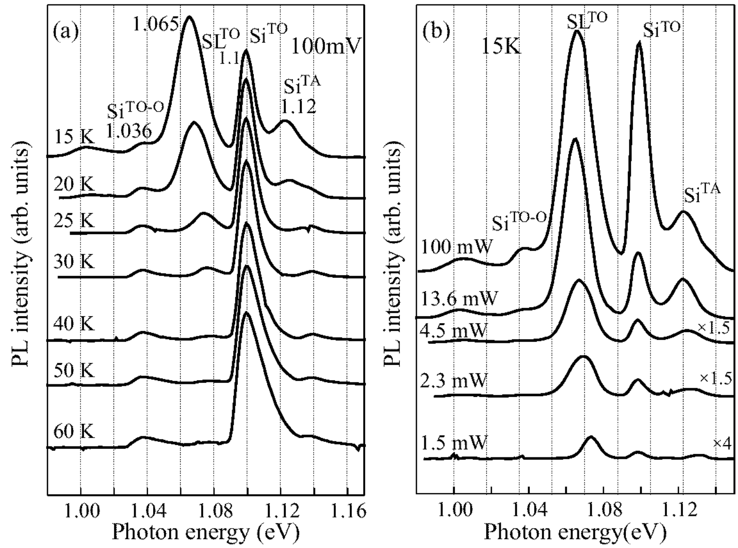

Fig. 1. PL spectra for the Ge/Si SML structure grown at 750°C taken at (a) different temperature and (b) different excitation density.

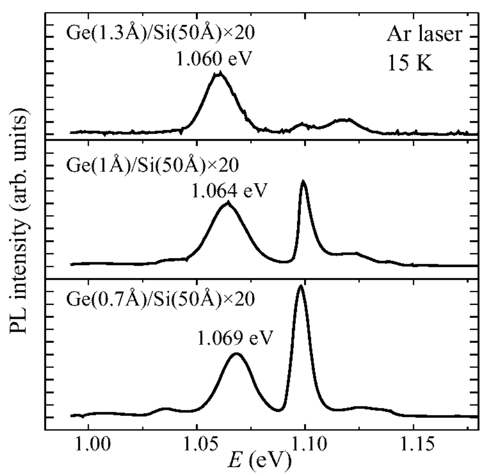

Fig. 2. Effect of the influence of Ge amount in each layer on PL peak position.

The SL–TO line exhibits shift towards long-wavelengths with increasing of the quantity of Ge in each layer of the SL. In Fig. 2 are shown PL spectra for the samples with 0.7 Å, 1.0 Å and 1.3 Å of Ge in each layer of the SL. This shift is explained by the increasing of the Ge islands lateral sizes. However, if Ge nanoislands become higher in size than the respective hole Bohr radius then holes delocalise in direction perpendicular to the growth direction and efficiency of corresponding lines in the PL spectra can be decreased. High-resolution transmission electron microscopy study shows that for the case of lower germanium amount deposited there are well pronounced Ge nanoinclusions with 7 nm in lateral size and 3.5 nm in height clearly seen [14]. For higher Ge amount (1 ML) single monolayer of Ge is observed.

In conclusion, we have investigated optical properties of the Ge SML nanoinclusions in a Si matrix. We have shown that under certain growth conditions there are new lines in the PL spectra appear. These lines correspond to formation of the Ge nanoislands with sizes lower than respective hole Bohr radius. If their sizes are higher there are no such PL lines. The investigated system may be considered as the one the possible way for the SBLE devices creating.

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