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Well-resolved photoluminescence excitation (PLE) spectra are reported for self-assembled GeSi dots grown on Si(001) by molecular beam epitaxy. The observation of two excitation resonance peaks is attributed to two different excitation/de-excitation routes of interband optical transitions connected to the spatially direct and indirect recombination processes. It is concluded that two dot populations are addressed by each monitored luminescence energy for the PLE acquisition. © *2010 American Institute of Physics*. [doi:10.1063/1.3424789]

Detailed knowledge on the electronic band structures, such as sublevels and related optical transitions, is very important when using self-assembled GeSi quantum dots (QDs) embedded in Si for some applications in Si-based photonic devices.[1] During the past years, GeSi dot materials have been extensively studied by photoluminescence (PL) measurements.[2–5] Due to inhomogeneous distribution of the size, alloy composition, as well as local strain in the incorporated GeSi/Si dots, the observed dot-related luminescent emission was however rather broad, which thus hampered detailed studies of the electronic transitions involved in the luminescence.[6,7]

Based on careful analyses on the peak shape of temperature- and excitation power-dependent PL of GeSi dots, Larsson *et al.*[8,9] concluded that there were two different recombination processes related to the GeSi QDs. One was spatially direct within the dot, and the other one was spatially indirect across the interface between the dot and the surrounding Si in the type-II band alignment. Understanding and control of these effects are crucial for the improvement of the optical efficiency of this material system.

PL excitation (PLE) is known as a versatile tool useful in characterization of electronic states involved in the optical transitions with a better energy resolution. However, up to date, no PLE results from any GeSi/Si nanostructures have been published. In this letter, we report a study using PLE measurements with well-resolved excitation resonance peaks from self-assembled GeSi dots grown by molecular beam epitaxy (MBE) on Si. By comparing with model calculations, our PLE results were directly related to the coexistence of the spatially direct and indirect recombination processes, which therefore provided a more precise energy scheme of the subband states involved in these optical transitions.

The sample used for the optical studies was grown by solid-source MBE (Balzers UMS-630). One Ge dot layer was formed via the Stranski–Krastanov growth mode by depositing eight monolayers of Ge on a Si(001) substrate at a temperature of 530 °C, and then capped with a 140 nm thick Si layer at 600 °C in order to minimize incorporation of point defects. The average dot diameter was ~50 nm and

the typical height was ~3 nm as measured by TEM, while the dot density was estimated to be about 4×10^{10} cm^{-2} by atomic force microscopy measurements[9] on a reference sample without Si capping. The Ge content in the dots was determined to be at a level of ~75% (Ref. 10) by means of energy dispersive x-ray microanalysis.

The grown sample was characterized by both PL and PLE experiments. The PL was measured by a Fourier transform infrared spectrometer (Bomem DA8) at 4 K using a 514 nm argon ion laser as the excitation source. The PLE measurements were excited by a picosecond optical parametric oscillator (OPO) synchronously pumped by a mode-locked Ti:sapphire laser, resulting in the spectrum-dependent output oscillation power with a maximum value at ~500 mW. The emission was spectrally dispersed by a 1 m double-grating monochromator, and measured by a liquid nitrogen-cooled Ge detector (North Coast EO-817S) in the range of 0.7–1.4 eV using standard lock-in techniques. All PLE experiments were performed at low temperatures in a helium-bath cryostat.

A typical PL spectrum of the GeSi dot sample is depicted in Fig. 1, dominated by broadband emissions peaked at ~790 meV with a full width at half maximum (FWHM) of ~60 meV. The spectral resolution of Fourier transform PL is high, allowing observation of the sharp boron-related TO replica PL at 1092 meV together with the multiple excitonic
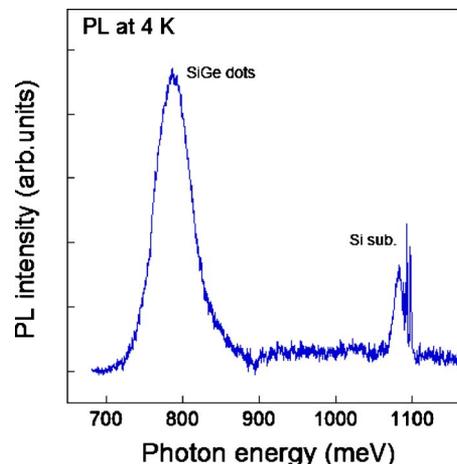

FIG. 1. (Color online) PL spectrum of the GeSi/Si QDs measured at T=4 K with an excitation power of 50 mW at λ=514 nm.

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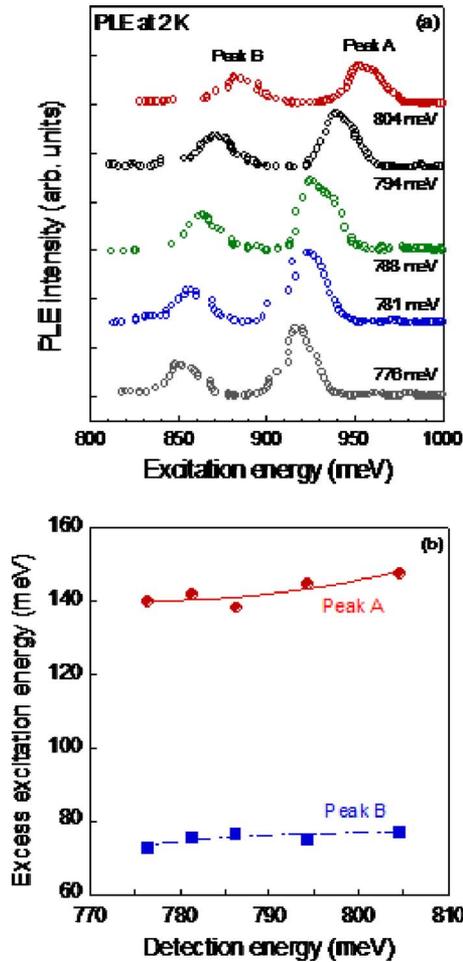
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FIG. 2. (Color online) (a) PLE spectra of GeSi/Si dots acquired at different detection energies at T=2 K. The spectra are shifted vertically for clarity. (b) Variation in the excess excitation energy (ΔE) as a function of the detection energy for the two resonance peaks in the PLE spectra.

emissions at 1090 meV and 1088 meV,[11] respectively, from the Si substrate. Besides, the broad emission around 1080 meV was due to recombination of electron-hole droplets, and the luminescence intensity dip at ~0.9 eV was attributed to the quartz beamsplitter used in the measurement setup. No PL could be detected from the pseudomorphic Ge wetting layer (WL) probably because of the carrier depletion effect associated with the high dot density that captured most of the carriers from the WL.[12]

The PLE experiments were carried out by monitoring one section of the PL peak (e.g., 788 ± 2 meV) of the GeSi/Si QDs, and then scanning the excitation energies (E_{exc}) with the OPO in the range of 800–1040 meV. Two PLE peaks were observed at 928 meV and 862 meV, respectively, as marked by peak A and peak B in Fig. 2(a). The FWHM of the PLE peaks was ~20 meV, which is much smaller than that of the PL peak, meaning that only the dots with rather coherent properties (size, composition, and strain, etc.) could be monitored with the selected detection condition.

The PLE experiments were also performed at four different detection energies along the up- and down-slope of the GeSi-dot PL peak, in order to probe the variation in transition energies related to the electronic structure of different dot ensembles. One can see in Fig. 2(a) that the appearance

of the PLE spectra for different detection energies was very similar, while both peaks revealed a blueshift as the detection energy increased. This can be seen more evidently by plotting the excess excitation energy ($\Delta E = E_{exc} - E_{det}$) as a function of the detection energy (E_{det}) in Fig. 2(b) for all five PLE measurements.

As has been established, the electronic structure and accordingly the optical transition energies of GeSi dots are influenced by several factors such as dot size, composition, and strain.[13] In general, the PL observed at a higher energy could be caused by dots with either a smaller size or a lower Ge content. Assuming that the excitation would be mainly influenced by hole subbands in the GeSi dot, the corresponding excess excitation energy ΔE would however have a different trend. The ΔE would increase with the reduction in dot size but it would decrease when decreasing the Ge content. Therefore, our observation of a small PLE energy increase with the increasing PL detection energy would be explained mainly due to the size effect, while partly compensated by the change in the Ge composition.

Moreover, the excitation power dependence of the PLE peak intensity was quite different between the measured excitation peaks A and B. By reducing the primary power of the OPO one order of magnitude lower using a neutral density filter, the intensity of peak A decreased by about 50%, but the intensity of peak B nearly vanished. The strongly different power dependences of the two PLE features is an indication that the detected PL would originate from two different recombination processes but coincidently with the same transition energy for the detected PL originating from two separate ensembles of GeSi dots.

According to the earlier PL work by Larsson *et al.*,[8,9] there are two recombination processes in the GeSi/Si dot material system, corresponding to the spatially direct and indirect transitions, respectively, observed for specific experimental conditions. The emission corresponding to the direct transition in the dot is solely observed at elevated temperatures and higher excitation power. The PL spectra in Fig. 1 are *not* measured under these particular conditions and accordingly only the peak corresponding to the indirect transition is expected in this case. Furthermore, it is concluded that the PL and PLE peaks are dominated by no-phonon transitions, i.e., associated with a relatively modest phonon coupling strength due to local lattice disordering introduced by alloy composition and strain variation across the dot partially relaxing the k-momentum conservation.

Hence the physical origin of the PLE peaks is attributed to three routes with respect to different transition processes, as illustrated by arrows and grouped by marks of PLE peak A, PLE peak B, and PLE peak C in the potential diagram in Fig. 3 for a single GeSi dot.

The PLE peak A is proposed to correspond to a spatially direct transition in the GeSi dot between the light hole ground sublevel (lh_0) of the valence band and the Δ_4 valley of the conduction band, where the recombination between the Δ_4 electron state and the heavy hole ground sublevel (hh_0) is detected. The lower energy PLE peak B is interpreted as the spatially direct excitation from the hh_0 state to the Δ_4 valley inside the GeSi dot, when the spatially indirect recombination at the interface between a Δ_2 electron state in the notch potential of the strained Si barrier layer and the hh_0 state in the GeSi dot is detected. Therefore, within the varia-
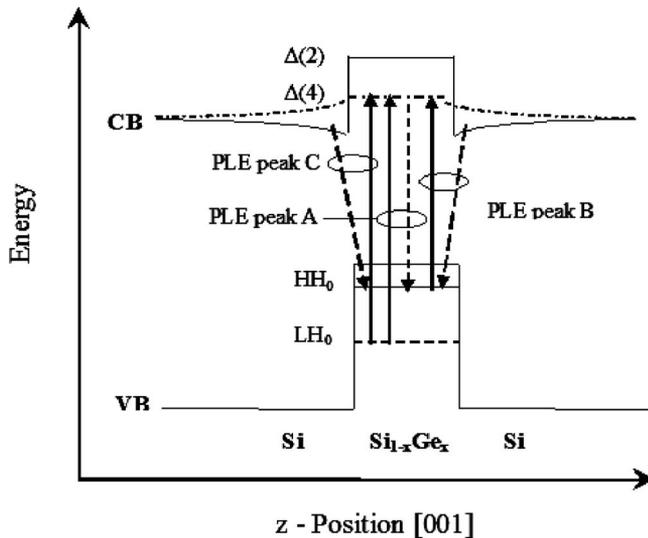
FIG. 3. Schematic band-edge diagram of the GeSi-dot/Si heterojunction along the z direction with the relevant interband transitions corresponding to the observed PLE peaks.
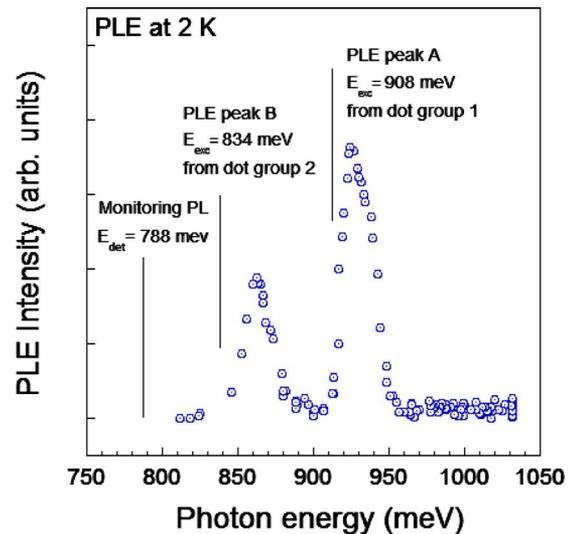

FIG. 4. (Color online) Comparison between calculated absorptions from two different dot populations (noted as dot group 1 and dot group 2) and an experimental PLE spectrum of the GeSi/Si dot with the PL detected at 788 meV. Calculated transitions are represented by bars in the figure.

tion range of dot size, composition, and connected strain in the grown GeSi/Si dot material, it is possible to observe these two PLE peaks from two different subgroups of GeSi dots, when monitoring the PL at one single detection energy. Our model predicts also a third peak, denoted peak C in Fig. 3, which could not be resolved in the PLE spectra shown in Fig. 2(a). This is probably due to the fact that this peak is below the detection limit since the combined effects of the lower OPO power in the expected energy range and the weaker matrix elements for the LH_0-Δ_4 transition compared to the HH_0-Δ_4 transition in the PLE as well as for the indirect transition monitored in PL. These combined effects are expected to cause a lowered detected intensity by about one order of magnitude of peak C relatively to peaks A and B.

To support our interpretation of the experimental results, a numerical analysis of the electronic band structure of the GeSi/Si dots, with the three-dimensional strain distribution that was calculated by assuming isotropic elastic constants,[14] was performed using a 6×6 band $\mathbf{k} \cdot \mathbf{p}$ approach for the valence band[15–20] and solving the Schrödinger equation with anisotropic effective electron masses for the conduction band. Each QD was assumed to have a truncated-pyramid shape with the base orientation along the [100] and [010] directions. The pyramid base was fixed to 30 nm and the height was varied continuously from 2 to 4 nm. Because of the small aspect ratio (height/base) of the QDs, the quantum confinement effects are mostly determined by the dot height along the [001] growth direction, which justifies that the QDs can, in a simplified picture, be modeled by local GeSi QWs embedded in Si.

The computation results are presented in Fig. 4 together with a PLE spectrum [depicted as the middle one in Fig. 2(a)] for comparison. It shows that with the detected PL energy at 788 ± 2 meV, the spatially direct PL is to be excited by a photon energy of 908 meV via route A from the GeSi dot with 78% of Ge at a dot height of 2.8 nm, while the spatially indirect PL (route B) could be observed by an excitation energy of 834 meV from the dot containing 75% Ge and 2.0 nm in height. These results are in a fairly good agreement with the experimental observations.

In summary, the first luminescence excitation spectra from self-assembled GeSi dots grown by MBE on Si are reported. By comparing with six-band $\mathbf{k} \cdot \mathbf{p}$ calculations for truncated pyramidal shape GeSi/Si QDs, our experimentally observed PLE features are assigned to be due to optical transitions of two kinds involving the two following different dot populations: the spatially direct transition inside the GeSi dot and the spatially indirect transition across the Si/Ge interface. In fact, these two processes are coexisting in any single GeSi dot, and dispersed in a wide spectral range over all dots with a certain variation in size, composition, and strain, which consequently results in a broadband PL emission.

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