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Strain-symmetrized Si/SiGe multi-quantum well structures grown by molecular beam epitaxy for intersubband engineering

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Abstract

Three strain-symmetrized Si/SiGe multi-quantum well structures, designed for probing the carrier lifetime of intrawell intersubband transitions between heavy hole 1 (HH1) and light hole 1 (LH1) states with transition energies below the optical phonon energy, were grown by molecular beam epitaxy at low temperature on fully relaxed SiGe virtual substrates. The grown structures were characterized by using various experimental techniques, showing a high crystalline quality and very precise growth control. The lifetime of the LH1 excited state was determined directly with pump-probe spectroscopy. The measurements indicated an increase of the lifetime by a factor of ~2 due to the increasingly unconfined LH1 state, which agreed very well with the design. It also showed a very long lifetime of several hundred picoseconds for the holes excited out of the well to transit back to the well through a diagonal process.

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1. Introduction

In the past few years, a large number of unique properties of far-infrared radiation in the THz frequency range were discovered [1]. Therefore, it has attracted great interests in fabrication of semiconductor THz lasers to meet the strong need for compact and efficient THz emitting sources [2]. Adopting the quantum cascade (QC) scheme that has been successfully developed based on III–V materials [3–5], the Si/SiGe system is considered as a more promising candidate for THz lasers due to several advantages. Particularly, in contrast to a significant reduction of the intersubband non-radiative lifetime above ~40 K in III–V semiconductors [6], Si/SiGe structures showed a rather constant long intersubband lifetime up to 100 K due to the absence of strong polar optical phonon scattering [7,8]. Consequently, this potentially permits lasing from Si/SiGe THz QC devices at room temperature with much higher efficiency.

Apart from the achievement of a longer lifetime in mid-infrared Si/SiGe QC structures through an interwell intersubband transition across a Si barrier [9], it has also been proposed that the lifetime can be actually engineered via special designs of the subbands [8]. In this work, we investigated the structure dependence of intersubband lifetime with several band structure designs, and the lifetime was measured by time-resolved pump-probe spectroscopy. Due to the difficulty of precise calculations of the scattering rate, direct lifetime measurements for the intersubband transitions of interest could provide crucial information for the THz QC structure design. In addition,

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one decisive factor for successful implementations of the design is the material growth. Challenges essentially originate from the large lattice mismatch between Si and Ge, which limits the epi-layer thickness and causes misfit-introduced imperfections. Although several structures were successfully made by applying low temperature growth on SiGe virtual substrates [10–12], there is still a high demand on further studies on the growth and characterization of Si/SiGe QC structures with various complex designs.

2. Experimental procedure

In order to tailor the intersubband lifetime, three Si/SiGe multi-quantum well (MQW) structures were designed on $Si_{0.8}Ge_{0.2}$ virtual substrates with (0 0 1) orientation. Each sample consisted of 20 identical $Si_{0.67}Ge_{0.33}$ quantum wells, sandwiched between two Si barrier layers with a $Si_{0.8}Ge_{0.2}$ spacer for separation. The layer thickness of the Si barriers (L_b)/quantum wells (L_w) for samples 1, 2 and 3 are $L_b = 1.6\,nm/L_w = 5\,nm$, $L_b = 1.3\,nm/L_w = 4\,nm$, and $L_b = 1\,nm/L_w = 3\,nm$, respectively. These layer parameters were selected so that the tensile strain in Si layers was to be equal to the compressive strain in SiGe QWs, i.e., the structures were strain-symmetrized. At both sides of each QW and 2 nm away from the Si barrier, there was a 5 nm wide region inside the spacer doped with boron at $5 \times 10^{17}\,cm^{-3}$ to form modulation doping. The spacer layer was 64 nm thick for all samples in order to reduce the influence of spacer states on the pump-probe signal from the HH1–LH1 transition within the QWs. 6-band $k \cdot p$ band structure calculations showed the energy difference between the LH1 excited state and the HH1 ground state to be 35, 33 and 24 meV for samples 1, 2 and 3, respectively, which were below the Ge–Ge optical phonon energy (37 meV). The LH1 state in sample 1 was well confined inside the QW and above the spacer valance band edge by ~10 meV (in electron energy scale), while it was much less confined in sample 3, spreading significantly outside the well into the spacer and resonant with the spacer valance band edge. Therefore, the spatial extent of the LH1 state wavefunction in sample 3 was about twice wider than that in sample 1, resulting in ~1/2 probability density at the center of the well for LH1 state. Sample 2 had an intermediate situation, in which, however, there was still a considerable amplitude of the LH1 state wavefunction outside the well, i.e., more similar to the sample 3 case. The calculated band structures were detailed in Ref. [13].

All the samples were grown using a Balzers UMS 630 solid-source Si/Ge molecular beam epitaxy (MBE) system [14] on SiGe virtual substrates fabricated by means of low-pressure chemical vapor deposition (LPCVD) by QinetiQ Ltd. The substrates consisted of a linearly graded i-$Si_{1-y}Ge_y$ ($y = 0$–0.2) structure of about 2 μm followed by a constant-composition i-$Si_{0.8}Ge_{0.2}$ relaxed buffer layer (RBL) of ~0.5 μm. Prior to the MBE growth, the virtual substrates were subjected to a chemical cleaning procedure, and then in situ baked at ~800 °C for 15 min [12]. The growth started with a $Si_{0.8}Ge_{0.2}$ buffer layer of 80 nm, followed by the MQW structure without growth interruption, giving the total epi-growth thickness of ~1.5 μm for each sample. During the buffer layer deposition, the substrate temperature was gradually decreased from ~440 °C to the temperature at which the MQW structures were to be grown. It was found that the quality of strain-symmetrized SiGe superlattices grown on virtual substrates was very sensitive to the growth temperature. A higher growth temperature (even just by few tens of degrees) may cause strong Ge segregation and diffusion, as well as local Ge accumulation that leads to layer undulation [10]. Based on our previous success in growing QC structures with a large number of superlattice periods at ~350 °C [12], together with the consideration of enhancing the boron dopant activation, all growths in the present work were carried out at a temperature of ~380 °C. Correspondingly, the Si growth rate was optimized at a slightly higher value of ~0.033 nm/s (constant through the whole growth), with varied Ge growth rate to achieve the desired Ge contents.

The surface and structural properties of the grown samples were characterized by atomic force microscopy (AFM), high-resolution X-ray diffraction (HR-XRD) together with reciprocal space mapping (RSM), cross-sectional transmission electron microscopy (XTEM), scanning transmission electron microscopy (STEM) and energy-dispersive X-ray (EDX) spectrometry. The intersubband lifetime measurements were performed by using time-resolved pump-probe spectroscopy with Dutch free-electron laser Felix.

3. Results and discussion

AFM measurements on the virtual substrates and the grown structures revealed that cross-hatch patterns—the characteristic of strain relaxation via formation of misfit dislocation network during the growth of virtual substrates—were dominating features (not shown here), and no growth introduced feature was observed. The measured surface r.m.s. values were in the order of 2–3 nm over $10 \times 10\,\mu m^2$ scan area for the samples both before and after MBE growth, in accordance with our previous observation [12].

For the XRD measurements, a Philips X'pert X-ray diffractometer ($\lambda = 0.15406\,nm$) was used with a hybrid monochromator. The diffracted beam was collected using an asymmetric Ge(2 2 0) collimator ($\Delta(2\theta) = 0.005^\circ$) [15]. The strain condition of grown samples was obtained based on HR-XRD RSMs around (0 0 4) and (1 1 3) reciprocal lattice points. Fig. 1 shows the (0 0 4) symmetric and (1 1 3) asymmetric RSMs of sample 1. A high crystalline quality of the grown structure was concluded based on the clear presence of a large number of MQW diffraction fringes in both RSMs. Although there was a lattice plan tilt of ~0.1° between the Si substrate and the SiGe RBL along ⟨1 1 0⟩ in-plane direction, which led to a peak shift in the ω-scan direction in (0 0 4) RSM (illustrated by the drawn lines), all