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Low-temperature formation of self-assembled Ge quantum dots on Si(100) under high carbon mediation via solid-phase epitaxy

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ABSTRACT

CMOS-compatible low-temperature formation of self-assembled Ge quantum dots (QDs) by carbon (C) mediation via a solid-phase epitaxy (SPE) has been demonstrated. The samples were prepared by a solid-source molecular beam epitaxy (MBE) system. C and Ge were successively deposited on Si(100) at 200 °C and Ge/C/Si heterostructure was annealed in the MBE chamber. Sparse Volmer-Weber mode Ge dots without a wetting layer were formed for C coverage (θ_C) of 0.25 and 0.5 ML by lowering SPE temperature (T_S) to 450 °C, but small and dense Stranski-Krastanov (SK)-mode Ge QDs with the wetting layer were obtained with increasing C coverage of 0.75 ML even at 450 °C. From the investigation of SPE temperature effect on Ge QD formation for θ_C of 0.75 ML, SK-mode Ge QDs of about 10 nm in diameter and of about 4.5×10^{11} cm^{-2} in density were formed at $T_S\geq400$ °C. The wetting layer of SK-mode QDs was almost constant 0.2-nm thick at $T_S\geq450$ °C. Measurements of chemical binding states of C in Ge QDs and at Ge/Si interface revealed that a large amount of C–Ge bonds were formed in the wetting layer for high C coverage, and the formation of C–Ge bonds, together with the formation of C–Si bonds, enabled the low-temperature formation of small and dense Ge QDs. These results suggest that the C-mediated solid-phase epitaxy is effective to form small and dense SK-mode QDs at low temperature.

1. Introduction

For optical devices, such as light emitting diodes [1,2], photodetectors [3], and solar cells [4], formation of Ge quantum dots (QDs) on a Si substrate is one of the promising candidates and is very effective in enhancing functionality of Si-ULSIs. It gives advantages such as weak phonon scattering, short carrier lifetime [1], and low detection noise [5] due to the low-dimensional confinement effect. In order to confine electrons and holes independently, diameter of Ge QDs is required to be smaller than $4a_B$, where a_B is the exciton Bohr radius (3.1 nm) [6,7]. In the Ge/Si heterostructure, the self-assemble formation of Ge dots occurs in the Stranski-Krastanov (SK) growth mode [8], in which the three-dimensional islands grow to reduce the accumulated strain energy due to the lattice mismatch after the layer-by-layer growth. However, in general, the dots are not small enough to confine carriers. Therefore, several methods to form small Ge dots, such as the growth of Ge dots on ultra-thin SiO_2 [9], sub-monolayer (ML) Sb-mediated Ge dot growth [10], and the growth of Ge dots on patterned substrate [11] were reported. In addition, it has been reported that sub-ML carbon (C) mediation is beneficial to form Ge QDs as a result of Si(100)-c(4×4) surface reconstruction [12–14]. In our previous studies, small (~20 nm in mean diameter) and dense (~10^{11} cm^{-2} in density) Ge QDs were obtained via a solid-phase epitaxy (SPE) of Ge/C/Si(100) heterostructure at 650 °C [15,16]. However, it is desirable to reduce processing temperature as low as 400 °C to embed Ge QDs in Si-CMOS LSIs. In this study, we investigated a low-temperature formation of Ge QDs via C-mediated SPE to enable embedding in Si-CMOS LSIs.

2. Experimental

The samples were prepared by a solid-source molecular beam epitaxy (MBE) system equipped with an electron beam gun for C and a Knudsen cell for Ge. After a wet cleaning and a H-termination of the Si(100) substrate, it was loaded into a chamber. Good surface morphology of the Si surface was confirmed by reflection high-energy electron diffraction (RHEED) patterns after loading. Ge(1 nm)/C/Si structure was formed at 200 °C and in-situ annealing for 5 min was carried out at solid-phase epitaxy temperature (T_S) of 300–750 °C. The

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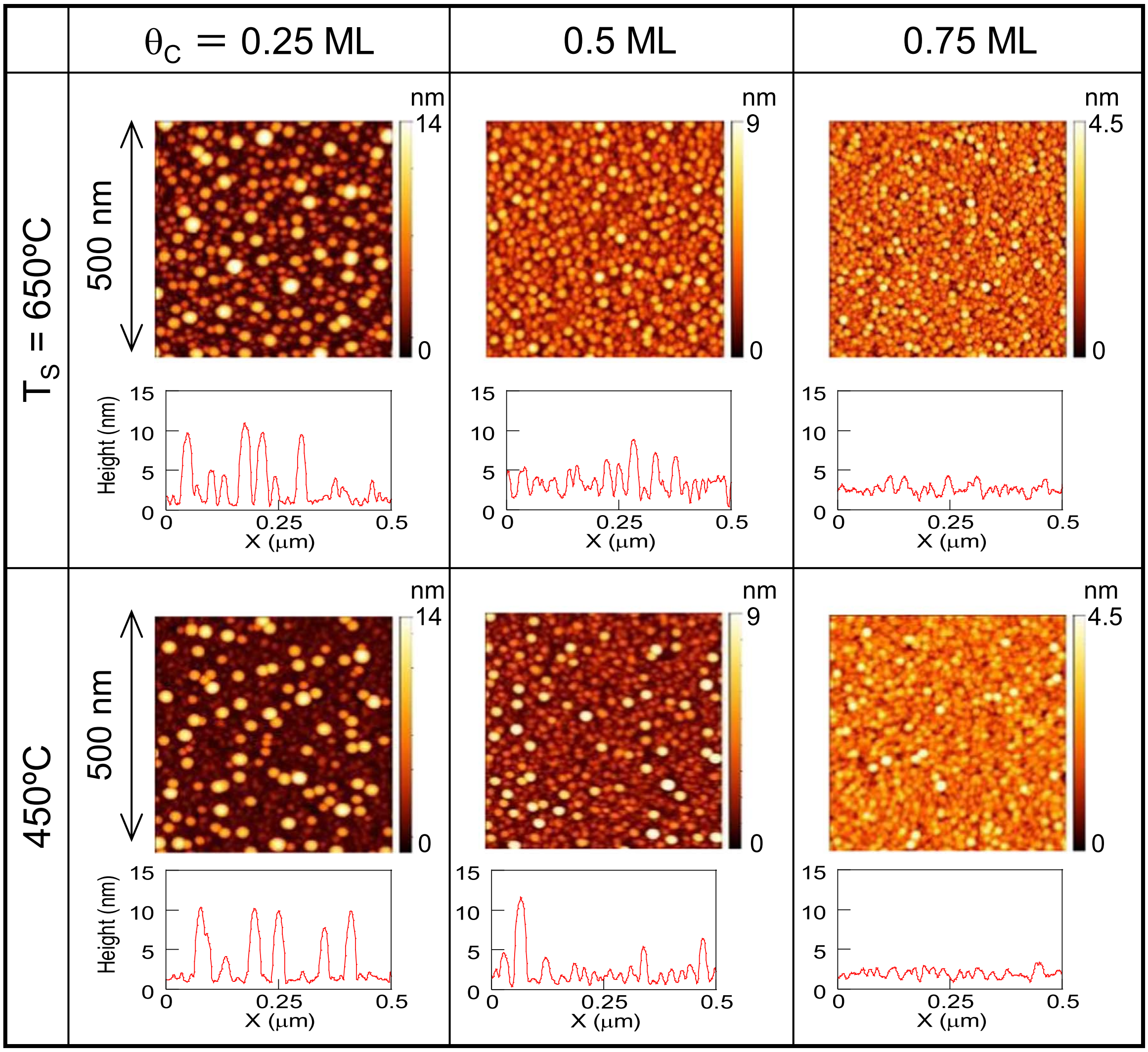

Fig. 1. AFM images and line profiles of Ge dots for θ_C=0.25, 0.5, and 0.75 ML at T_S=650 and 450 °C.

deposition rate of C and Ge are 0.03 ML/min and 1 nm/min, respectively. Morphology of Ge dots was evaluated by atomic force microscopy (AFM, Asylum, Cypher ES), and chemical binding states of C were analyzed by X-ray photoelectron spectroscopy (XPS, AXIS Nova, Kratos Analytical). XPS measurements were performed before and after etching (15-vol% H_2O_2 solution) of Ge QDs in order to evaluate the C binding states both at Ge/Si interface and in Ge QDs.

3. Results

3.1. Influence of C coverage on Ge QD formation at low temperature

The influence of C coverage on Ge QD formation under low T_S was investigated. The AFM images and line profiles of Ge dots for C coverage (θ_C) of 0.25, 0.5, and 0.75 ML at T_S=650 and 450 °C are shown in Fig. 1. Their diameter histograms are shown in Fig. 2. For θ_C=0.25 and 0.5 ML, the dot density at T_S=450 °C was lower than that at T_S=650 °C, and small dots, in particular, 8–18 nm in diameter were hardly formed at T_S=450 °C. In contrast, for θ_C=0.75 ML, almost the same diameter and density of QDs were formed at both T_S=450 and 650 °C. Here, to investigate the presence of a Ge wetting layer, dot equivalent thickness (DET), which indicates the leveled thickness of the dot part without the wetting layer, was evaluated. At both T_S, DET for θ_C=0.25 and 0.5 ML were almost equal to the deposition thickness of 1 nm while that for θ_C=0.75 ML was thinner than the deposition thickness. This means that Ge QDs grew in Volmer-Weber (VW)-mode without the wetting layer for θ_C=0.25 and 0.5 ML while SK-mode Ge QDs with the wetting layer were formed for θ_C=0.75 ML. The Si 2p XPS spectra for θ_C=0.25 and 0.75 ML at T_S=650 °C are shown in Fig. 3. The peak intensity of SiO_X for θ_C=0.25 ML was higher than that for θ_C=0.75 ML. This indicates that the Si surface for θ_C=0.25 ML was exposed to the air, that is the formation of VW-mode Ge dots without the wetting layer. Here, in-situ RHEED patterns during QD formation were also monitored, but a significant difference was not observed between VW-mode and SK-mode QDs. VW-mode QDs are preferred for strong carrier confinement, but SK-mode QDs can also induce sufficient carrier confinement effect if the volume of the wetting layer is much smaller than that of the dots. Since infrared photoluminescence was reported in SK-mode Ge dots with the wetting layer of 2–3 MLs (0.28–0.42 nm) [17,18], the carrier confinement in SK-mode QDs obtained in this study could be expected (thickness of wetting layer in SK-mode QDs was about 0.2 nm which is shown later in Fig. 7.) These results indicate it is possible to form small and dense SK-mode Ge QDs at low temperature by C-mediated SPE for θ_C=0.75 ML.

C binding states in Ge QDs were investigated by the XPS in order to analyze the effect of C mediation on Ge QD formation. The C 1s XPS spectra of Ge QDs for θ_C=0.25 ML at T_S=650 °C, and for θ_C=0.25 and 0.75 ML at T_S=450 °C are shown in Fig. 4. The C 1s spectrum was deconvoluted into five components of C–O–O (~289 eV), C–O (~286.8 eV), C–C (~285.2 eV), C–Ge (~284.5 eV), and C–Si