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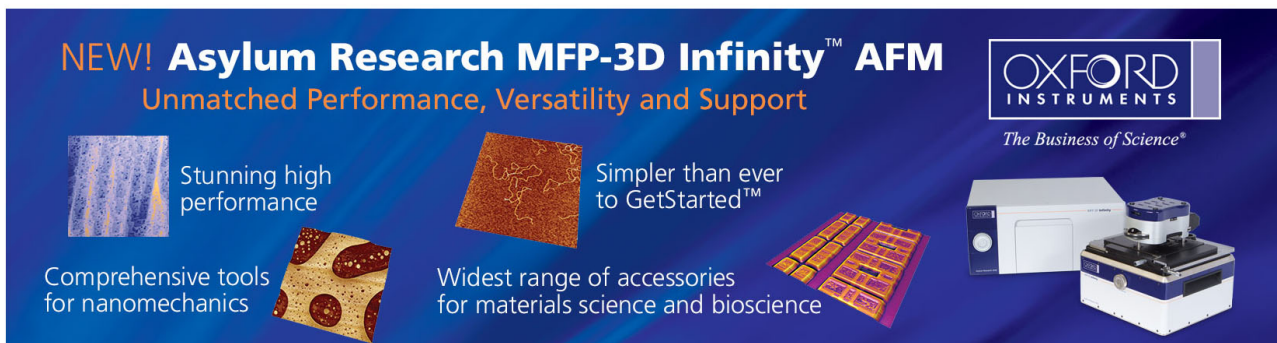
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## SiGe nanorings by ultrahigh vacuum chemical vapor deposition

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Formation of SiGe nanorings from Si capped Si<sub>0.1</sub>Ge<sub>0.9</sub> quantum dots (QDs) grown at 500 °C by ultrahigh vacuum chemical vapor deposition was investigated. SiGe nanorings have average diameter, width, and depth of 185, 30, and 9 nm, respectively. Based on both Raman and x-ray diffraction results, the formation of SiGe nanorings can be attributed to Ge outdiffusion from central SiGe QDs during *in situ* annealing. Moreover, the depth of SiGe nanorings can be controlled by Si cap thickness. The Si cap is essential for nanorings formation. © 2009 American Institute of Physics. [DOI: 10.1063/1.3116619]

Ringlike structures in III-V compound semiconductor system were widely reported recently.<sup>1–3</sup> Nanorings exhibit many interesting phenomena, such as unusual excitation properties and Aharonov–Bohm effect, due to the unique rotational symmetry<sup>4</sup> and the doubly connected topology of the ring,<sup>5</sup> respectively. SiGe nanorings grown by ultrahigh vacuum (UHV) chemical vapor deposition (CVD) have been reported previously. However, ring structures in the previous reports can only be observed in very limited process window due to specific growth mechanism.<sup>6</sup> In this work, SiGe nanorings with controllable depth and well-defined Ge content at edges are demonstrated. The ring structures were characterized by atomic force microscopy (AFM). Strain and Ge content in nanorings were analyzed through x-ray diffraction (XRD) using synchrotron radiation source and high resolution Raman spectroscopy. Ge outdiffusion mechanism is found to play a crucial role in the ring formation.

SiGe nanorings were grown by UHV/CVD system at 500 °C. The base pressure was  $\sim 10^{-9}$  torr. Silane (SiH<sub>4</sub>) and germane (GeH<sub>4</sub>) were used as reactant gases. The gas flows of GeH<sub>4</sub> and carrier gas (He) were fixed at 5 and 35 SCCM (SCCM denotes standard cubic centimeters per minute at STP), respectively. Ge layers transform into SiGe alloys due to Si/Ge interdiffusion.<sup>7</sup> After Stranski–Krastanov mode growth of quantum dots (QDs), Si cap with different thickness was deposited. The samples were *in situ* annealed for 1 h to form ring structures.

Surface morphology of as-grown SiGe QDs grown at 500 °C is shown in Fig. 1(a). Dot density is  $\sim 6 \times 10^8$  cm<sup>-2</sup>, and the average dot height and dot base are 15 and 115 nm, respectively. After Si cap layer deposition, small dots disappeared from AFM image [Fig. 1(b)] and only those with large dot size can be observed. Figure 1(c) shows the morphology of 4 nm Si capped QDs after 1 h of annealing. Ring structures are evident on the surface and shown by white rings. The density of nanorings from 4 nm Si capped

QDs is  $\sim 1.3 \times 10^8$  cm<sup>-2</sup>, which is similar to the dot density ( $\sim 1.2 \times 10^8$  cm<sup>-2</sup>) of the sample before annealing within 10% error bar of measurement. Surface morphology of nanorings from 2 nm Si capped QDs is also shown in Fig. 1(d), and the density and diameter of nanorings are similar to those from 4 nm Si capped QDs. Note that QDs without Si cap cannot be transformed into rings after annealing. After Si capping, large QDs (>150 nm) are still observable in AFM image, but the contrast of small dots (<90 nm) becomes much weaker [Fig. 1(b)]. Surprisingly, the large QDs have no Si capping, while small dots were buried in Si cap as shown by cross-sectional transmission electron microscopy (TEM) image in Fig. 2. An additional {111} facet appeared at the edge of QDs in Fig. 2(a), probably due to Ge surface diffusion from small dots in neighborhood during long Si cap growth time.<sup>8</sup> Note that the growth rate of Si cap is extremely small (0.1 nm/min) at 500 °C and growth time is 40

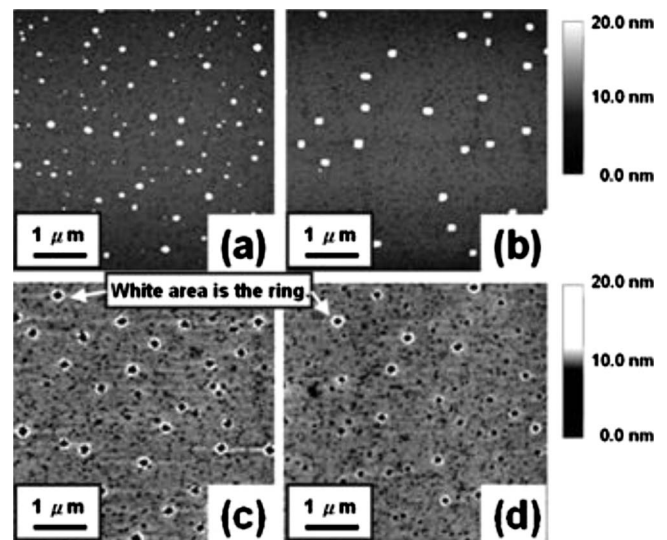


FIG. 1. AFM images ( $5 \times 5 \mu\text{m}^2$ ) of (a) uncapped, (b) 4 nm Si capped QDs, (c) nanorings from 4 nm Si capped QDs, and (d) nanorings from 2 nm Si capped QDs.

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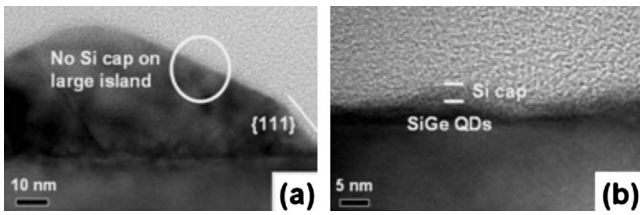


FIG. 2. TEM images of 4 nm Si capped QDs: (a) large QDs and (b) small QDs.

min. SiGe QDs were *in situ* annealed during Si cap growth. Both Ge content and strain relaxation in QDs may increase. Moreover, dot height of large QDs at 500 °C is much larger than Si cap thickness. Si atoms adsorbed directly on QDs may suffer tensile strain due to the larger lattice constants of QDs and tend to diffuse to relatively strain-free region around dots.<sup>6</sup> SiGe dots shown on the surface are therefore not covered by Si cap.

Outdiffusion mechanism of InGaAs nanorings has been extensively discussed. The relatively large diffusivity of In is responsible for the formation of nanorings.<sup>2,9</sup> Surface diffusion of Ge is generally larger than that of Si.<sup>10</sup> Figure 3 illustrates the outdiffusion model of SiGe nanorings. Si cap layer only covers the edge of SiGe QDs and can block Ge surface diffusion out of the region covered underneath. Ge surface diffusion is faster than Ge interdiffusion into Si cap,<sup>11,12</sup> and Ge atoms inside SiGe dots can diffuse outward to the top of Si cap over the edge of QDs [Figs. 3(b) and 3(c)]. The Ge outdiffusion continued with annealing and left a hole near the center of former SiGe dots. Ring depth (the distance from the bottom of ring to the ridge at edge) can be controlled by Si cap thickness. Thicker Si cap thickness leads to a larger ring depth of nanorings. Note that the Ge surface diffusion of SiGe QDs was reportedly suppressed by the Si cap layer deposition.<sup>13</sup> Moreover, the preserving shape and composition of SiGe QDs during Si capping at deposition temperature  $T_{\text{dep}}$  lower than the QDs growth temperature of 600 °C was reported.<sup>14</sup> In principle, the outdiffusion can be slow and the ring shape should be preserved after Si capping if the growth temperature of Si cap is lower than the nanorings formation temperature of 500 °C.

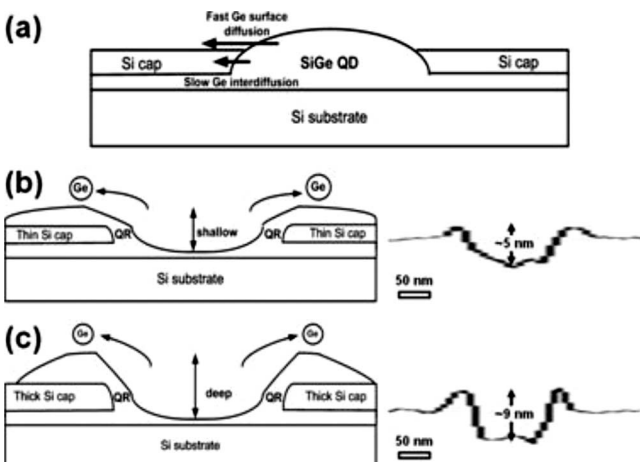


FIG. 3. Outdiffusion mechanism of SiGe nanorings: (a) SiGe QDs with Si cap, (b) Ge outdiffuse from 2 nm Si capped QDs, and (c) Ge outdiffuse from 4 nm Si capped QDs. AFM profiles show 4 nm Si cap sample has a larger depth (9 nm) than the 2 nm Si cap sample (5 nm).

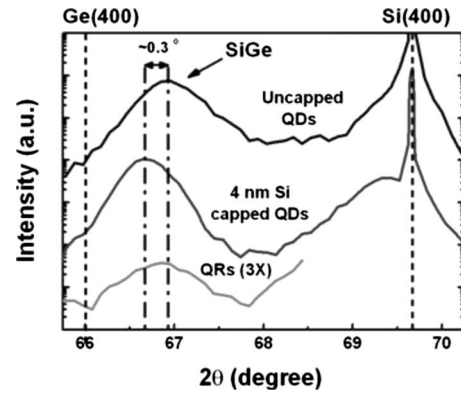


FIG. 4. (400) XRD in-plane radial scan of uncapped QDs, 4 nm Si capped QDs, and nanorings. X-ray wavelength used is 1.5406 Å.

Strain variation between nanorings and QDs can be measured by XRD and Raman measurements. X-ray scattering radial scans along in-plane and surface normal directions were performed to determine the lateral and vertical lattice constants, respectively. Taking both the Poisson ratio and the unstrained SiGe lattice constant<sup>15</sup> into account, the ~90% Ge content in SiGe QDs can be derived by the XRD results in this work. Figure 4 shows XRD lateral radial scans across Si (400) and Ge (400) reflections of the uncapped, 4 nm Si capped QDs, and nanorings. The peak centered at ~67° and is contributed from SiGe islands. An increase of lateral lattice parameter, as revealed by observed shift of ~-0.3° in Fig. 4, upon Si capping is attributed to the increase of Ge content and/or strain relaxation in QDs. Ge surface diffusion from small dots to large dots may be responsible for the Ge content increase and strain relaxation. When QDs transformed into nanorings by annealing, SiGe peak shifts back to the higher angle and the intensity decreases. The increase of compressive strain or/and the decrease of Ge concentration are responsible for the peak shift. Nevertheless, these two effects cannot be distinguished by XRD results since no non-surface reflection can be detected to provide vertical lattice constant. Raman spectroscopy with 488 nm laser excitation on uncapped, Si capped QDs, and nanorings are shown in Fig. 5 with the resolution of 0.2 cm<sup>-1</sup>. Ge-Ge peak of Si capped QDs sample is ~0.5 cm<sup>-1</sup> higher than the uncapped QDs sample due to the increase of Ge content in large islands, which is similar to XRD results. However, the peak of nanorings sample shifts more to higher wave number (~0.5 cm<sup>-1</sup>) as compared with Si capped QDs sample. The

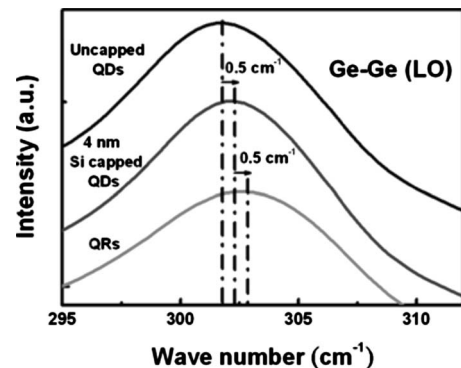


FIG. 5. Raman spectroscopy of uncapped QDs, 4 nm Si capped QDs, and nanorings.

increase of Ge content or/and compressive strain is responsible for the blueshift. However, combined with XRD results, the increase of compressive strain is the only possible origin. Ge diffuses gradually from the top of QDs and the left nanoring has more compressive strain. Note that the bottom of SiGe dots is more compressively strained than the top of QDs according to the simulation work.<sup>16</sup>

In summary, SiGe nanorings with controllable depth have been investigated. The Ge outdiffusion mechanism is proposed to nanorings formation at 500 °C. Note that the true quantum ring nature has been demonstrated by photoluminescence and spectral response measurement in the III-V system.<sup>2,3</sup> However, in our case, the interband detector based on the quantum behavior of SiGe nanorings is still under investigation.

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