The Germanium "Halo": Visualizing invisible Ge interstitials

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An elaborate theory has been constructed[1] to explain the unique interaction of Ge, O, and Si interstitials at high temperature (900^{o}C) under oxidizing conditions. These interstitial interactions include: (a) the coalescence by Ostwald Ripenin of Ge nanocrystallites (Fig. 1), (b) migration and burrowing[2] of Ge Quantum Dots (QDs) (Fig. 2), Ge doping of Si substrates by proximal Ge QDs to form thin SiGe "shells", and, ultimately, (d) the "explosion" of large Ge QDs (Fig.3). Despite the overwhelming preponderance of experimental evidence that points to the presence and interactions Ge, O, and Si interstitials as responsible for facilitating these novel phenomena, no actual experimental evidence for the presence of these interstitials could be produced until now. Interstitial detection in bulk materials is generally an indirect and tedious process. Electron Paramagnetic Resonance (EPR) and Extended X-ray Absorption Fine Structure (EXAFS) measurements are notable for detecting the presence of interstitials in atomic lattices. However, the extremely small geometries of the nanopatterned pillar structures used for our experimental demonstrations precludes the use of either EPR or EXAFS. Instead, we have discovered that Z-contrast, even in conventional Transmission Electron Microscopy (TEM), coupled with Energy Dispersive X-ray fluorescence (EDX) mapping are powerful tools to directly visualize the distribution of Ge interstitials within the amorphous SiO_2 layers surrounding Ge nanocrystallites (Fig. 4) and QDs (Fig. 5) within the nanopatterned pillar structures. This is because contrast in TEM images is either due to inelastic scattering (Z-contrast) or due to elastic scattering (Bragg diffraction). The SiO_2 matrix being amorphous has a uniform elastic scattering contrast over which the additional Z-contrast of the Ge interstitials is superimposed. Using this novel approach of visualizing Ge interstitial concentrations within nanopillar structures, we present and discuss the results for Ge interstitial distributions observed for the various phenomena described above.

Keywords: Ge interstitials, Z-contrast, Visualization, TEM, EDX Mapping

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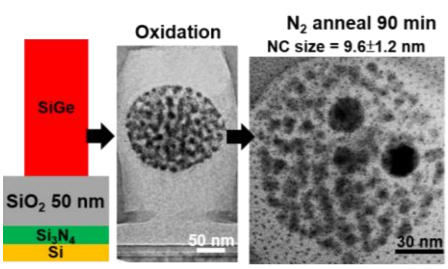

Fig. 1 CTEM micrographs showing coarsening of Ge nanocrystallites subjected to further post-oxidation anneal in a N_2 ambient at 900ºC The underlying Si_3N_4 buffer layer provides the Si interstitials that facilitate Ge Ostwald Ripening.

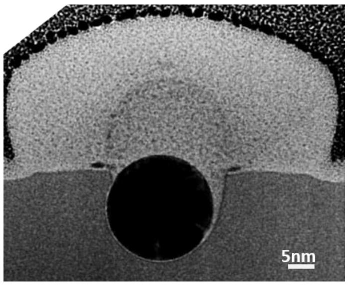

Fig. 2 CTEM micrograph of a migrating Ge QD showing the depth and trajectory of penetration from its as-formed SiO_2 matrix into a 150nm-thick Si_3N_4 buffer layer following a 900ºC, 60in oxidation.

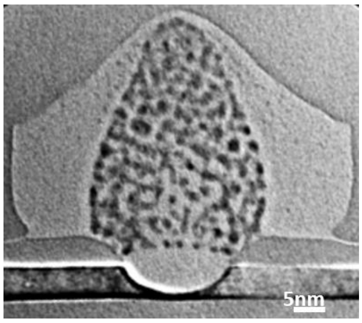

Fig. 3 CTEM micrograph of the “explosive” breakup of a Ge QD attempting to penetrate deeper into the Si substrate. Note the SiGe “shell” created by Ge interstitial migration from the QD to the Si substrate surface.

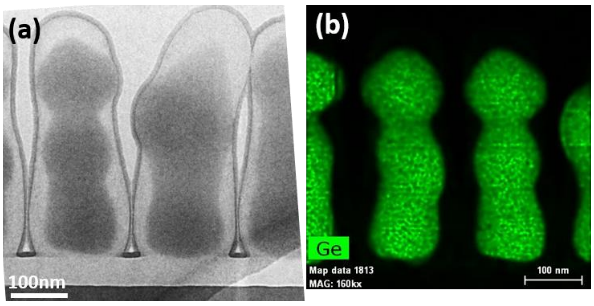

Fig. 4 (a) CTEM micrograph of showing Ge interstitial Z-contrast within an SiO_2 pillar and (b) EDX map of the Ge interstitial distribution showing good agreement with the Ge Z-contrast..

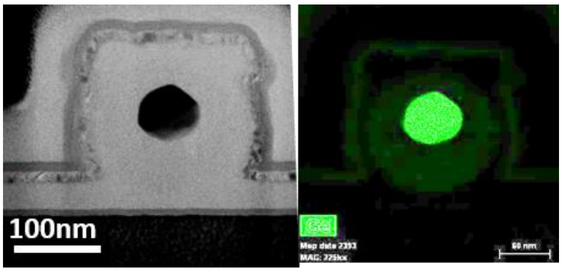

Fig. 5 (a) CTEM micrograph of a Ge QD formed within a SiO_2 pillar. Note the depleted Ge Z-contrast within the pillar, which is borne out by (b) the EDX map of a lower concentration of Ge interstitials surrounding the Ge QD.