Supporting Information

Visualising Discrete Structural Transformations in Germanium Nanowires during Ion Beam Irradiation and Subsequent Annealing

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Figure S1. Schematics of the experimental set-up used for concurrent TEM imaging/Ga-ion FIB implantations.

For the concurrent imaging/implantation experiments, Si chips with silicon nitride membranes were used (Ted Pella), which were patterned using the FIB to number and open slits in the membrane (Fig. S1). The patterning is of benefit for two reasons; the slits allow high resolution imaging of sections of the nanowire as well as facile navigation to locate the same nanowire. The parameters for patterning were: 7 lines 10 μ m apart, 70 μ m long and 0.3 μ m wide at 30 kV, 3 nA for a nominal thickness of 300 nm. Nanowires that were fully crossing the 0.3 μ m wide slits were used for implantations. The mounting of the membrane chips to the TEM holder was done under optical microscope using a dedicated positioning system taking care that the grids were placed with the same rotation.



Figure S2. Lattice resolution TEM image of 64 nm diameter Ge nanowire irradiated with a fluency of 7.6 $\times 10^{13}$ ion cm⁻², demonstrating amorphous pockets at the nanowire surface, accompanied by regions of accumulated point defects (marked with circles).



Figure S3. Consecutive 30 kV Ga ion irradiations of a 50 nm diameter Ge nanowire with the beam tilted to about 6 degrees off the [110] zone direction at a fluency of 1.9×10^{13} ion cm⁻² (a) weak beam dark field imaging taken under g, 2g with g = 220 conditions. (b) Corresponding lattice resolution image taken close to the [110] zone direction at the nanowire side surface. (c) Weak beam dark field imaging taken under g, 2g with g = 220 conditions after irradiation with a1.1 ×10¹⁴ ion cm⁻² , (d) lattice resolution image taken at the same nanowire side surface as in (b).



Figure S4. Various cross-sectional TEM images of nanowires subjected to Ga-ion implantations at 30 kV ($\mathbf{a} - \mathbf{e}$) and at 5 kV (\mathbf{f}) with the irradiation direction from the top. Excluding the nanowire shown in (\mathbf{a}) all other nanowires were defect free and grown in the [111] direction. (\mathbf{a}) 64 nm Ge

nanowire grown along the [211] direction irradiated with a fluency of 3.8×10^{13} ion cm⁻², featuring intrinsic stacking fault defects along the (111) set of planes. (b) 29 nm Ge nanowire irradiated with a fluency of 3.8×10^{13} ion cm⁻², featuring a fully amorphous structure. (c) 60 nm nanowire irradiated with a fluency of 7.6×10^{13} ion cm⁻² featuring extended but not full amorphisation leaving crystalline domains towards the middle bottom of the nanowire cross-section. (d) Lattice resolution image of the area marked in **e**, demonstrating uneven amorphous/crystalline interface. (e) 74 nm diameter Ge nanowire irradiated with a fluency of 1.9×10^{13} ion cm⁻², featuring amorphous pockets within mostly crystalline structure. (f) 39 nm diameter Ge nanowire irradiated with a fluency of 3×10^{13} ion cm⁻² at 5 kV, insets show fast Fourier transforms (FFTs) of the areas from the image marked with arrows confirming amorphous and crystalline regions of the cross-section.



Figure S5. Iradina maps of **(a)** cascade recoils, **(b)** ion paths, **(c)** implanted ions and **(d)** atom displacements based on a cylindrical Ge nanowire with a diameter of 45 nm being irradiated by a 30 kV Ga ion beam orthogonal to the nanowire. Images **(e-f)** are based on a 25 nm nanowire irradiated by a 5 kV Ga ion beam.

Iradina is a static Monte Carlo simulation similar to SRIM, except the target shape can be defined within the nanoscale simulation volume. The material of the wire was defined as Ge and the surrounding material is set as a vacuum. For this experimental procedure the nanowire was defined as a cylinder with a varied diameter, 65 nm, 45 nm or 25nm, and a varied accelerating voltage, 5 kV or 30 kV. A section of the nanowire was defined by the periodic boundary conditions (PBC), essentially defining a 2 dimensional cross section of the nanowire, which in turn was divided into a number of cells (defined by the user). The ion beam direction was kept orthogonal to the direction of the nanowire as the crystallinity of the material was disregarded due to random phase approximation (RPA) of the target atoms.



Figure S6. (a) and **(b)** Cross-sectional TEM images at different tilt angles of almost fully amorphised 25 nm nanowire that was subjected to 30 kV Ga-ion implantation at a fluency of 1.9×10^{13} ions cm⁻². Initially the nanowire was imaged along the growth direction (image on the left) which was determined by using the electron diffraction of the Si carrier wafer. Image **(b)** is after 28 degrees tilt in one-direction. Selected area electron diffraction patterns for both nanowires showed attenuated amorphous rings. In contrast, panel **(c)** shows a high resolution TEM image of the same 22 nm Ge nanowire , confirming retained single crystal structure viewed along the [111] zone after irradiation at 5 kV and fluency of 1.9×10^{13} ions cm⁻².



Figure S7. 30 kV, Ga ion irradiation of a 25 nm diameter Ge nanowire with a beam tilted to about 3 degrees away from the [210] zone direction. (a) Before irradiation and (b) after irradiation at 1.9 $\times 10^{13}$ ion cm⁻². Lattice resolution images are taken close to the [210] zone direction. 5 kV, Ga-ion irradiation of a 20 nm diameter Ge nanowire with the beam tilted to about 5 degrees away from the [110] zone direction; (c) before irradiation and (d) after irradiation at 7.6 $\times 10^{13}$ ion cm⁻². Lattice resolution images are taken close to the [110] zone direction; (c) before irradiation and (d) after irradiation at 7.6 $\times 10^{13}$ ion cm⁻². Lattice resolution images are taken close to the [110] zone direction.



Figure S8. Cross-sectional TEM images of Ge (001) wafers subjected to Ga-ion implantations at 7 degrees off the normal direction at increasing fluencies for 30 kV ($\mathbf{a} - \mathbf{c}$) and 5 kV ($\mathbf{d} - \mathbf{f}$) implantations. Measured average thicknesses of amorphous layers are given in the insets.