Supporting information

Stress-driven nucleation of three-dimensional crystal islands: from quantum dots to nanoneedles

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(1) GaAs nanoneedles

As discussed in the main text, GaAs NNs can be obtained by MOCVD on roughened GaAs and silicon[27] or on planar sapphire[28] substrates. Figure 1 presents typical SEM images of GaAs NNs grown on roughened GaAs(111)B and Si(111) substrates[27]. With increasing the growth time, the NN length is correspondingly increased without changing its shape, angle or tip dimension. These observations elucidate the growth of NNs being via continuous deposition on their initial 3D surface, favored along the [0001] WZ crystal orientation. Some NNs envelope each other during growth [Fig. 1 (a)]. The white hexagonal shapes in Fig. 1 (b) indicate well-aligned vertical, sharp NNs with a length of 2–3 μm. The NN sidewalls align to the <−1−12> ZB substrate directions. Figure 1 (c) shows a zoomed-in SEM image of a typical NN tip viewed nearly perpendicular to the growth axis. A linear array of NNs can be also attained as demonstrated in Fig. 1(d). Figure 1(e) shows 30° tilted and top-down views of a 4 μm long nanoneedle grown on a 4° off-cut Si (111) substrate. Typical taper angles of these NNs range between 6 and 9^0.

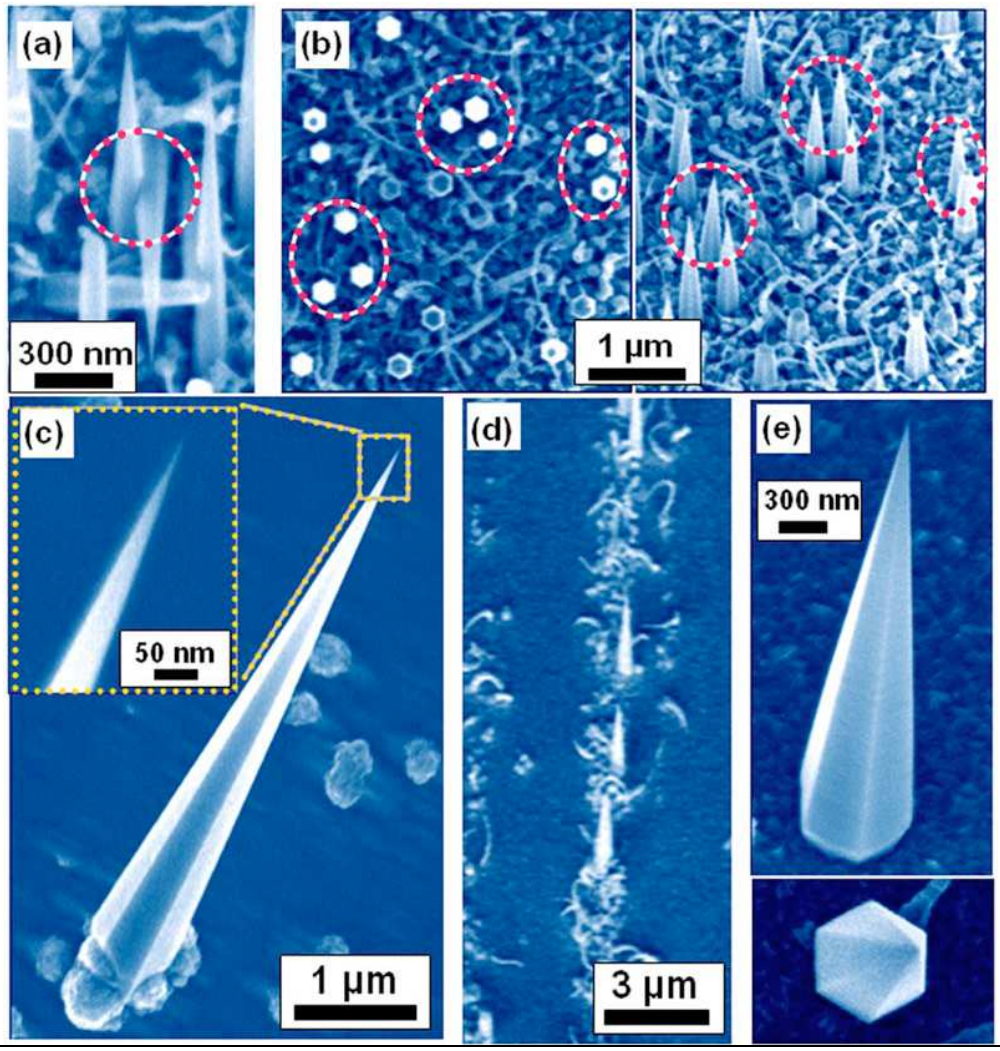

Fig.1. GaAs NNs grown by MOCVD on roughened GaAs(111)B (a-d) and Si (e) substrates.

Figures 2 from Ref. [27] demonstrate pure WZ structure of GaAs NNs. Figure 2 (a) shows a HRTEM image on the [1−100] zone axis of a NN and its corresponding FFT. The tip in the image comes to an atomically sharp point of just 2–4 nm wide. The material remains single-crystal WZ all the way up until the tip. Figure 3(b) shows a FFT from another NN on the [1−210] zone axis, showing the distinct WZ pattern. The c and a axes for these NNs were determined to be 6.52 and 3.98 Å, respectively. This c/a ratio is 1.638, which is close to the ideal hexagonal c/a ratio of 1.633. More details regarding the NN growth and crystal structure can be found in Ref. [27].

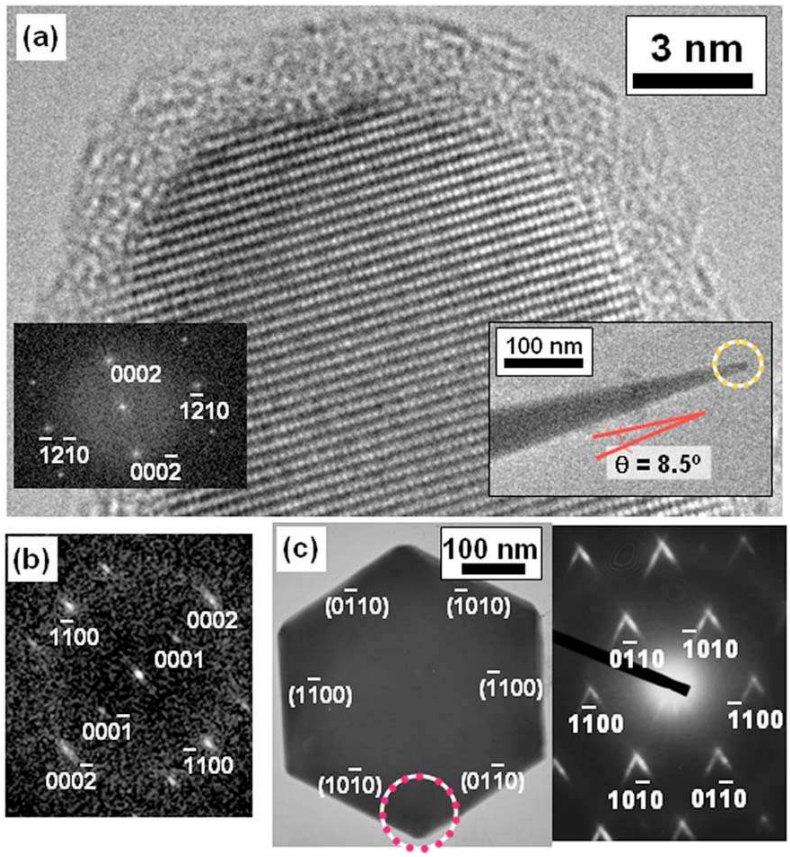

Fig. 2. HRTEM images of GaAs nanoneedles. (a) [1−100] zone axis HRTEM image of GaAs NN. The insets show the zoomed-out view, and also the image FFT. (b) FFT from another NN on its [1−210] zone axis with a distinct WZ pattern. (c) Top-down [0001] TEM image of a NN. The image to the right shows a selected area diffraction pattern from the circled area, with distinct wurtzite {1−100} spots matching the expected unique wurtzite 3.45 Å spacing.

(2) Details of calculations

To calculate the volume and sidewall surface area of pyramidal islands with tapered sidewalls, we consider the regular vertical facets of elementary length dl separated by the horizontal steps of width dr (see Fig. 1 (d) of the main text) so that

$$dl / dr = L / R = 2\beta = const \,. \tag{S1}$$

Here, L is the height, R is the base dimension and $\beta \equiv L / 2R$ is the aspect ratio. The cross-sectional area at distance l from the substrate surface equals $(C_1 / 2)r^2$ with $C_1 = 2\pi$ for a

cylinder; $C_1 = 3\sqrt{3}$ for a hexagonal prism and $C_1 = 8$ for a rectangular prism. In particular, $S_B = (C_1/2)R^2$ is the base surface area at $l = 0$. Obviously,

$$V = \frac{C_1}{2}\int_0^R r^2 dl = C_1\beta\int_0^R r^2 dr = \frac{C_1}{3}R^3\beta . \tag{S2}$$

This formula is equivalent to the well known expression for the volume of a regular prism. The surface area of vertical sidewalls of elementary height dl equals $(C_2/2)rdl$ with $C_2 = 4\pi$ for a cylinder, $C_2 = 12$ for a hexagonal prism and $C_2 = 16$ for a rectangular prism. Integration at constant β yields:

$$S_F = \frac{C_2}{2}\int_0^R rdl = C_2\beta\int_0^R rdr = \frac{C_2}{2}R^2\beta . \tag{S3}$$

Equations (S2) and (S3) give the formulas used in the main text with geometrical coefficients $k_1 = 1/3$ and $k_2 = 1/2$.