Supporting Information

SI 1 Substrate design

The growth mask was obtaned using electron beam lithography by defining the arrays with different holes diameter and inter hole distances (pitch) as presented in Fig.1 below. The best and reproducable yield was obtained for 45 nm holes. The yiled or morphology of NWs were not affected by the pitch since we were focuing on the initial growth stages and using short growth times. At this stages NWs are to short to shadow each other or the observe As reemission effect[1]. To aquire enogh statistics (125 wires per sample were measured) we used 400 nm or 600 nm pitch. In Fig.2 we show SEMs of the same sample for these 2 pitches to demonstrate that growth outcome was the same.

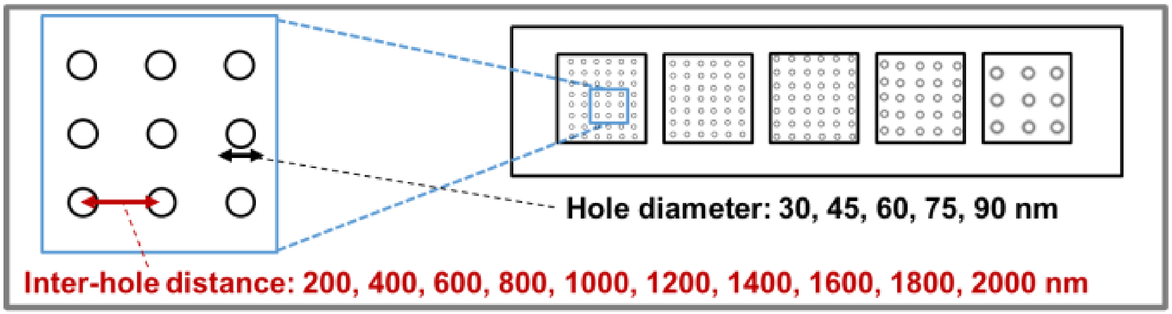

Figure 1 The schematics of pattern used to grow NW arrays.

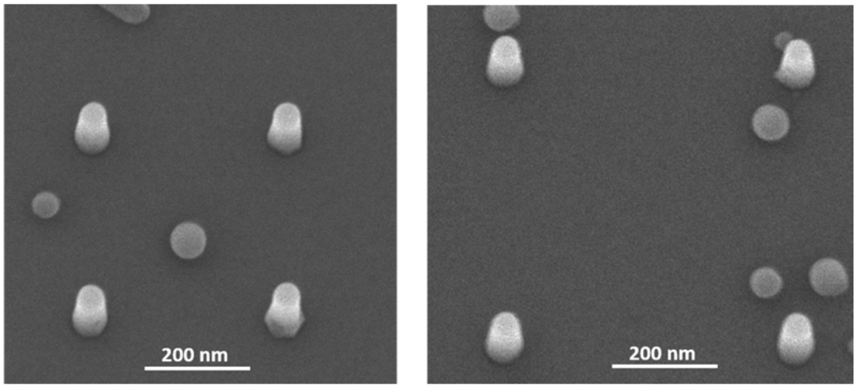

Figure 2. SEM images of the same sample for different inter - wire distances

SI 2 High yield arrays

In SEM below we present wide field SEM pictures of the arrays showing the consistent yield across a large area (about 100x100 μm^2) for two growth times.

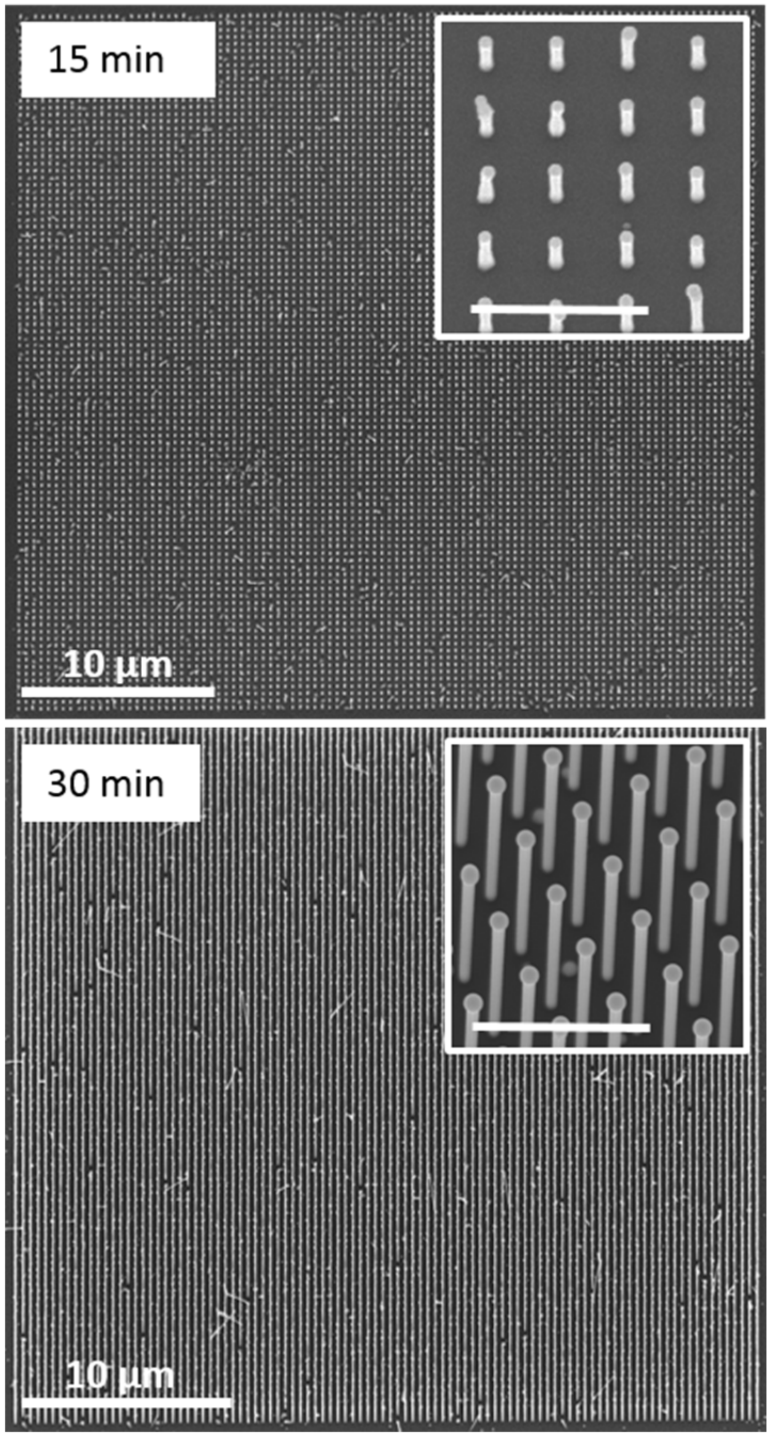

Figure 3. SEM images of samples grown for 2 different times demonstrating high yield in large area with small insets showing closer view. The scale bar on the insets is 1 µm and the tilt angle 20˚.

SI3 Ga - contact angle estimation

In the geometrical description of our system, used for modeling of the growth, one of the parameters required was a droplet contact angle. The exact contact angle of the droplet within the opening is hard to access, but we combined two techniques, SEM and AFM, to try to estimate its value. In the Fig.3 below we present the 3D image of the droplet acquired by AFM and also SEM images of the droplets taken in cross section under the 50˚ tilt. We see that the value of the contact angle is around 90˚. Additional fact that tells us that the contact angle should be around this value is the fact that we obtained high yield of vertical wires. The direct impact of the contact angle of Ga and NW orientation was previously demonstrated by Matteini et al [2]. They found that only droplets with the contact angle around 90˚ will lead to the vertical NW growth.

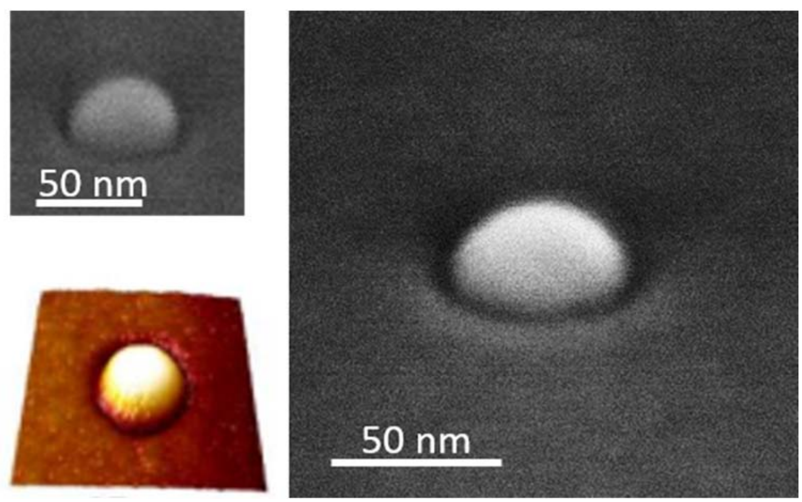

Figure 4. SEM and AFM images of the GA droplet after 10 minutes of GA deposition with the Ga rate of 1.1 Å/s within the opening in the silicon oxide mask. The contact angle is estimated to be around 90˚.

SI 4. Estimation of the effect of kinetic fluctuations to the size distributions broadening

Starting from the NW length distribution (without droplets), we present the nucleation distribution as

$$f(L,L_*) = A\exp\left[\frac{L_*-L}{\Delta L} - \exp\left(\frac{L-L_*}{\Delta L}\right)\right], \tag{1}$$

with L_* as the most representative length and $\Delta L = \chi_5 v_5 \Delta t$ as the distribution width. The latter corresponds to standard deviation in the Gaussian approximation of the double exponential distribution. In the growth stage, the nucleation distribution is broadened due to kinetic fluctuations. This process is described by the Poissonian Green's function [3–5]

$$G(s,\langle s\rangle) = \frac{1}{\sqrt{2\pi\langle s\rangle}}\exp\left[-\frac{(s-\langle s\rangle)^2}{2\langle s\rangle}\right]. \tag{2}$$

Same expression applies also for the i variable in the Eq. (3) in the main text. . The variance equals the mean size $\langle s\rangle = [(b\chi_5 v_5)/h](t-t_c)$ acquired at the growth stage. Using the normalization condition [3] $g(L)dL = G(s)ds$, we obtain Green's function in terms of the NW length

$$g(L,\langle L\rangle) = \frac{1}{\sqrt{2\pi h\langle L\rangle}}\exp\left[-\frac{(L-\langle L\rangle)^2}{2h\langle L\rangle}\right]. \tag{3}$$

The variance added by kinetic fluctuations $\sigma_{growth} = hb\chi_5 v_5 (t-t_c)$ approximately equals 45 nm^2 after 10 min, which is much smaller than the measured variance of 260 nm^2. Therefore, we can neglect the effect of kinetic fluctuations in the initial growth stage and use the nucleation distribution given by Eq. (1) to fit the statistical data for the NW lengths.

Repeating similar consideration for the NW diameter, the variance of Green's function $8R_{Ga}^3/(9\langle R\rangle) << 1$ is extremely small so that it looks as delta-function. The resulting radius distribution is then entirely determined by the nucleation stage. The double exponential solution for the i variable is re-scaled in terms of the radius as

$$f(R,R_*) = \frac{3R^2}{R_{Ga}^3} B\exp\left(x - e^x\right), \quad x = \frac{R_*^3 - R^3}{R_{Ga}^3 \Delta i}. \tag{4}$$

For $|R_* - R|/R << 1$, we can use the approximation $x \cong 3R_*^2(R_* - R)/(R_{Ga}^3 \Delta i)$, yielding the double exponential radius distribution

$$f(R,R_*) = CR^2 \exp\left[\frac{R_* - R}{\Delta R} - \exp\left(\frac{R - R_*}{\Delta R}\right)\right], \tag{5}$$

with the width $\Delta R = \frac{R_{Ga}^3(\beta_D)\Delta t}{3R_*^2 \tau}$.

SI 5. Selection of correct features to include in the statistical analysis

In case of the growths under higher As pressure we observed additional broadening of the length distributions towards lower values (Fig.5 in the main text). By carful and detailed observation of the sample under SEM we found that many wires stop to grow since their droplet was consumed. Therefore, they should not be taken into account when studying the statistical distributions. In the Fig.2 , left, we present one 3D image of AFM scan of the array and corresponding profile underneath. . The features marked by arrows are obviously much shorter them majority, but relying only on the AFM it is not possible to exclude them from the analysis. Combining AFM and SEM it becomes clearer that short features should not be included in the population of NWs that which length distribution we want to study and understand. These structures are marked with red frames in SEM image in Fig.2 right.

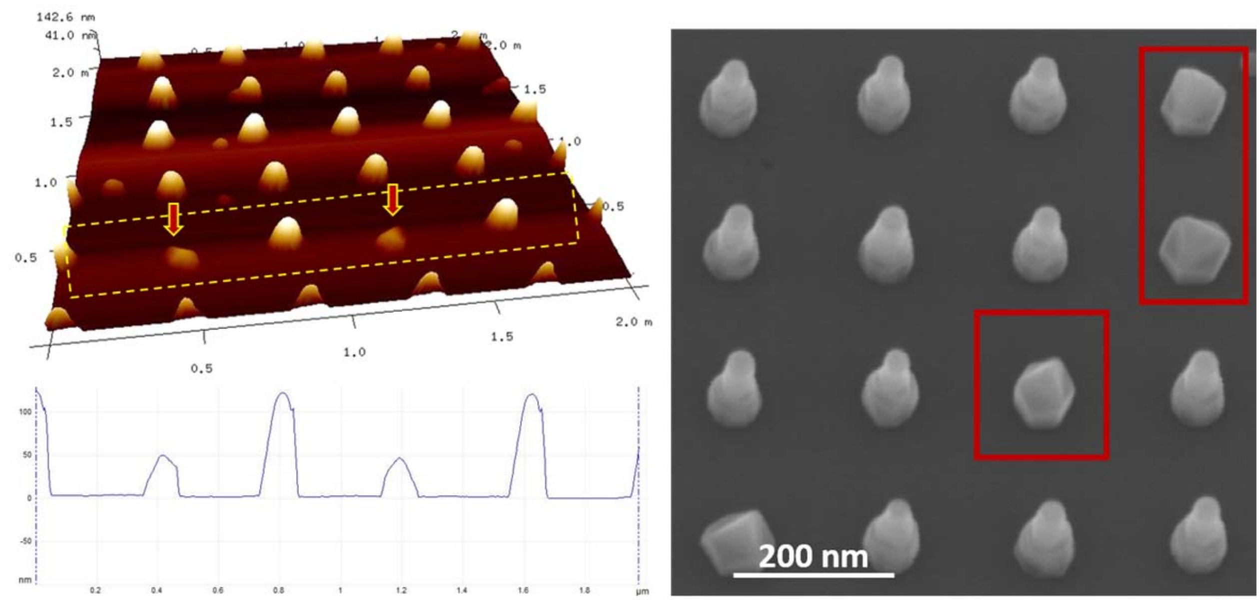

Figure 5. Left – AFM 3D image of the array and corresponding profile underneath. Arrows are pointing at the shorter features. Right – SEM image of the arrays. In the red squares on ecan see the structures without the droplet that correspond to the shorter features in the AFM scan.

References :

[1] Ramdani M R, Harmand J C, Glas F, Patriarche G and Travers L 2013 Arsenic Pathways in Self-Catalyzed Growth of GaAs Nanowires

[2] Matteini F, Potts H and Jabeen F 2015 Wetting of Ga on SiO 1–5

[3] Dubrovskii V G and Nazarenko M V 2010 Nucleation theory beyond the deterministic limit . I . The nucleation stage **114507**

[4] Dubrovskii V G 2016 Kinetic narrowing of size distribution *Phys. Rev. B - Condens. Matter Mater. Phys.* **93** 1–6

[5] Dubrovskii V G 2013 Self-regulated pulsed nucleation in catalyzed nanowire growth *Phys. Rev. B - Condens. Matter Mater. Phys.* **87** 1–7