Supporting information for:

Coherent Hole Transport in Selective Area Grown Ge Nanowire Networks

Santhanu Panikar Ramanandan¹,‡ Petar Tomić²,‡, Nicholas Paul Morgan¹, Andrea Giunto¹, Alok Rudra¹, Klaus Ensslin²,³, Thomas Ihn²,³, Anna Fontcuberta i Morral¹,⁴,⁵ *

¹Laboratory of Semiconductor Materials, Institute of Materials, École Polytechnique Fédérale de Lausanne EPFL, Lausanne 1015, Switzerland
²Solid State Laboratory, ETH Zurich, 8093 Zurich, Switzerland
³Quantum Center, ETH Zurich, CH-8093 Zurich, Switzerland
⁴Institute of Physics, Faculty of Basic Sciences, École Polytechnique Fédérale de Lausanne EPFL, Lausanne 1015, Switzerland
⁵Center for Quantum Science and Engineering, École Polytechnique Fédérale de Lausanne (EPFL), CH-1015 Lausanne, Switzerland

‡ Equal contribution

E-mail: anna.fontcuberta-morral@epfl.ch
1. Substrate preparation:

Germanium nanowires were grown on a quarter of 2-inch intrinsic Si (100) substrates with a resistivity of 2000 Ohm-cm. Samples were cleaned using the standard RCA procedure before the deposition of the dielectric mask layer. We used 175 nm thick thermally grown Si dry oxide as the dielectric material. Slits of nominal width varying from 50 nm to 200 nm and length varying from 2 µm to 20 µm were defined on the wafers by e-beam lithography with a 175 nm thick e-beam resist (50% ZEP). Reactive ion etching with a mixture of CH$_3$/SF$_6$ gases was used to transfer the patterns to the dielectric mask. After the RIE step, the substrates were sonicated in acetone (4 mins) and IPA (4 mins) to strip the e-beam resist. In order to remove any left-over organic residues from resist stripping, the samples were exposed to 600 W oxygen plasma for 10 mins. Finally, a short dip in diluted HF solution (1:10) for 10 secs was performed to remove the left out SiO$_2$ layer and native oxide of silicon before the NW growth. A schematic illustration of the substrate patterning process is shown in Figure SI 1.

![Figure SI 1: Schematic illustration of the substrate patterning process.](image-url)
Selective area growth of germanium nanowires was carried out using an AIX 200 horizontal metalorganic vapor phase epitaxy (MOVPE) system from Aixtron. Prior to the NW growth, the substrates were thermally cleaned in AsH$_3$ flow at 780 °C for 15 mins. For the thermal cleaning step, the AsH$_3$ flow rate was maintained at 60 sccm. The thermal cleaning step was found important to remove any native oxide of Si formed during the transfer process and for ensuing epitaxial growth. After the thermal cleaning step, AsH$_3$ flow was turned off and the substrates were cooled to the growth temperature in N$_2$ flow. SAG of Ge NWs was conducted at a reactor temperature of 700 °C using isobutyl germane as the precursor molecule and N$_2$ as the carrier gas. The precursor flow rate was kept at 1 sccm and the pressure was kept at 30 mbar. The NW growths were performed typically for 6 to 12 min depending on the width of the slits. The in-plane geometry of the NW is defined by the substrate patterning steps. In principle, in-plane geometries of any shape size, and orientation can be obtained by this approach. Figure SI 2 demonstrates the design flexibility of the SAG approach mentioned above.

Figure SI 2: Top view SEM images showing the design flexibility of SAG process. a) Top view SEM image of in-plane Ge NW for varying slit length. (b) Top view SEM image of Ge NW grown inside triangular shaped slits. (c) Top view SEM image of Ge NW grown inside circular shaped slits. Scale bar shown in b and c indicate 200 nm.
3. Orientation dependency of the Ge NW morphology

A strong dependency of the facet morphology of the NW on the in-plane orientation is observed. Nanowires with well-defined facets were obtained only along the two highly symmetric crystallographic directions i.e., $<110>$ and $<100>$. Figure SI 3 shows the magnified top-view SEM images of the Ge NWs as a function of misorientation with the $<110>$ crystallographic direction. Roughening of the side facets due to nano faceting can be observed for the NWs grown along the non-symmetric crystallographic orientations.

Figure SI 3: Top view SEM images showing the orientation dependency of the Germanium nanowire morphology. The indicated angles represent the misorientation of the ridges with respect to [110] crystallographic direction. The scale bars indicate 400 nm.
4. Structural characterization of the in-plane Germanium nanowires.

To understand the effect of NW orientation on the defect density we performed transmission electron microscopy (TEM) studies. For this purpose, 100 nm thick TEM lamellas (longitudinal and perpendicular cross-sections) were prepared using Zeiss NVision 40 Focused ion beam (FIB). The in-plane Germanium nanowires were covered by a thin layer of protective carbon (SEM deposited) to prevent any surface damage during TEM lamella preparation.

The structural quality and the chemical composition of the prepared lamellas were investigated using FEI Talos transmission electron microscopy operating at 200 kV. A comparison between the perpendicular cross-sections of the NW grown along the two highly symmetric crystallographic orientations is shown in Figure SI 4. As explained in the main text, a clear dependency of the crystal quality on the NW orientation is observed. BF TEM images of the perpendicular cross-sections of the <100> oriented NW looks more defective than the <110> oriented NWs for similar growth condition and aspect ratio of the slits.

Figure SI 4: BF TEM of the perpendicular cross sections of NW grown along <110> (left panel) and <100> oriented nanowires. The white arrows show the few defects observed in the case of <110> oriented NWs. The scale bars shown (unless mentioned) indicates 50 nm.
Figure SI 5 shows the magnified image of the inclined defects found along the length of the NW. For the 100 oriented NWs, these defects make an angle of 54.7° with the (001) plane and extend till the top. We suspect these defects nucleate at the inhomogeneities created on the Si substrate surface after the RIE step as reported in the case of SAG of III-V on Si.

![Inclined defects image](image)

Figure SI 5: BF TEM image of the longitudinal cross section of the <110> oriented NW showing the presence of inclined defects along the NW length.

5. Device Fabrication

For the fabrication of hall bar devices, we patterned Si (100) substrate covered with SiO$_2$ with slits arranged in the form of hall bar crosses (Figure SI 6 a) using the method described in section 1. Figure SI 6 b illustrates the dimensions of the hall bar pattern used in this work. We varied the width (w) (from 30 nm to 120 nm) and orientation (<100> and <110>) of the pattern. The patterned substrates were loaded into MOVPE reactor and germanium NW hall bar structures were grown. The detailed description of the growth parameters is described in section 2 of this document. Figure SI 6 c shows the top view SEM image of the selective area grown <100> oriented Ge hall bar structures with a channel width of 70 nm after 12 min growth.

After the SAG growth of Ge hall bar structures, metallic contacts for the source, drain and hall probes were deposited using the standard metal lift off procedure. We used electron beam lithography to define the pattern of the metal contact on MMA/PMMA bi layer resist and room temperature dc sputtering to deposit the contact. A short dip (15 sec) in HF 1% was performed to remove native oxide layer before loading the sample to the load lock of the sputtering machine. Metal stack of Ti (10nm thickness)/ Al (30nm thickness) / Ti (20nm thickness)/ Au (100 nm thickness) was used as the
contact material. Finally, metal lift off was performed by sonicating the samples in acetone solution for 1 min. Figure SI 6 d shows the optical image of the final hall bar structure after depositing metal contact.

Figure SI 6: a) Schematic of the patterned Si (100) substrate used for the Ge hall bar device growth. (b) Dimensions of the hall bar used in this article, we varied the width from 70 nm to 120nm. (c) Top view SEM image of the SAG Ge hall bar structure with a channel width of 70 nm. (d) Optical micrograph of the NW device used for the hall measurement after the deposition of top contact.
6. NW cross section

The HAADF STEM images of the <110> oriented NW cross sections are shown in the Figure SI 7. The scans were taken from a different chip with the identical devices as the chip that was used in the transport measurements in the main text.

Figure SI 7: HAADF STEM images of the NWs’ cross sections of the four devices characterized by the transport measurements. The contrast in figure (b) is different because a hole was unintentionally made in the NW during the FIB lamella preparation process. Scale bar indicates 50 nm.

7. Estimation of the depletion width:

We have used two different approaches for the estimate of the depletion width at the surface of the NW. First, an empirical model where we extrapolate to the average width of the NW corresponding to zero conductance by linearly fitting the data. We take this value as an estimate of twice the width of the depletion region. This model yields depletion widths of 12.7 nm, 10.5 nm and 11.0 nm for the data measured at 1.8 K, 20 K and room temperature respectively. In the second model, we estimated the depletion width by modelling the transport cross section area as \( A_{\text{trans}} = A_{\text{c.s.}} - C_{\text{c.s.}}d \) where \( A_{\text{c.s.}} \) is the NW cross-section area, \( C_{\text{c.s.}} \) is the circumference of the NW cross section and \( d \) is the depletion width. Both \( A_{\text{c.s.}} \) and \( C_{\text{c.s.}} \) are known from the TEM scans of the cross sections (see Figure SI 7). Then we can fit the data with the equation

\[
G_{xx}L_x = \sigma_{xx}(A_{\text{c.s.}} - C_{\text{c.s.}}d)
\]

where \( \sigma_{xx} \) and \( d \) are the two fitting parameters. The result of this model are depletion widths of 10.9 ± 3.5 nm, 10.2 ± 4.6 nm, and 12.2 ± 3.5 nm for 1.8 K, 20 K and room
temperature respectively which are denoted by black error bar in Figure SI 8. These results coincide well with the results from the first model. Since both estimates are based on crude approximations they should be taken only as rough estimates.

![Figure SI 8: Normalized conductance $G_{xx}L_x$ as a function of the average NW width measured at $T = 1.8$ K (a), $20$ K (b) and room temperature (c). Dashed lines are a linear fit to the data and the result from the transport cross section model, twice the depletion width, described in the main text is indicated with the black error bar.](image)

8. Mobility estimate

From the measurements of the longitudinal $R_{xx}(B,V_{TG})$ and Hall resistance $R_{xy}(B,V_{TG})$ as a function of the magnetic field ($\Delta B = 0.5$ T) and the top gate voltage ($\Delta V_G = 0.1$ V) we were able to estimate the Drude mobility $\mu(V_G) = \left[ G_{xx}(B = 0) \frac{dR_{xy}}{dB} \right] L_x/W_{eff}$ where $L_x$ is the length between the $V_{xy}$ voltage contacts, $W_{eff}$ is the effective nanowire width, and $G_{xx}(B = 0)$ is the longitudinal conductance at zero magnetic field. To suppress the UCF, the data was averaged over the top gate voltage interval of $1.2$ V. Furthermore, to remove the WAL peak from $G_{xx}(B = 0)$ we fitted $G_{xx}(B)$ with the Drude model expression: $G_{xx}(B) = G_0/(1 + \mu^2 B^2)$ where $G_0$, zero field conductance, and $\mu$ are the two fitting parameters (see Figure SI 9(a)). The fit was performed at $|B| > 2T$ to exclude the WAL peak from the Drude fit. The result of the fit, $G_0$, is plotted in Figure SI 9 b as a function of the top gate voltage. Using the corrected values for $G_{xx}(B = 0)$ we estimated the mobility using the following expression:

$$\mu(V_G) = \left[ G_0 \frac{dR_{xy}}{dB} \right] L_x/W_{eff}$$ (2)
The second fitting parameter, $\mu$, is within 30% of the mobility estimated with Equation (2). Since the fit was performed over a small part of the full Lorentzian described by the Drude mobility model we argue that the mobility extracted from the Drude fit is a less reliable estimate compared to the estimate of Equation (2) relying on the geometrical assumption. Still, the estimate of the mobility using Equation (2) should be taken as a crude approximation.

Figure SI 9: (a) Longitudinal conductance plotted as a function of the magnetic field for three different top gate voltages (dots). The data included in the Drude fit is highlighted with gray dots, while the resulting fit is denoted with red dashed lines. (b) Zero conductance fitting parameter plotted as a function of the top gate voltage.

9. Coherence length from the autocorrelation field

The coherence length $l_\phi$ can be extracted from the autocorrelation field of the conductance fluctuation amplitude as a function of the magnetic field shift $\Delta B$. The autocorrelation function is calculated according to

$$F(\Delta B) = \langle [\Delta G(B + \Delta B) - \langle \Delta G(B + \Delta B) \rangle][\Delta G(B) - \langle \Delta G(B) \rangle] \rangle$$

where $\langle \ldots \rangle$ is the ensemble average and $\Delta G(B)$ is the conductance with the background subtracted, i.e., $\Delta G(B) = G(B) - G_{bg}$. Then, the correlation field $\Delta B_c$ is determined from $F(\Delta B_c) = F(0)/2$, which, for the 1D case $W < l_\phi$, is related to the coherence length through the relation1,2
\[ \Delta B_c = C \frac{h/e}{W l_\phi} \]  

(4)

where \( W \) is the NW width, \( h \) is Planck’s constant, \( e \) is the elementary charge, and \( C \) is a constant within the interval \([0.42, 0.95]\). Its value depends on the relation between the coherence and thermal length \( l_T \), where the case of \( l_\phi \ll l_T \) corresponds to the lower and the case of \( l_\phi \gg l_T \) to the upper bound. The thermal length is \( l_T = (hD/k_B T)^{0.5} \) where \( D \) is the diffusion constant, \( k_B \) is the Boltzmann constant, and \( T \) is the temperature. It is a characteristic length scale over which the coherence is lost due to energy-averaging over the \( 4k_B T \) interval around the Fermi energy.

Figure SI 10: Longitudinal conductance \( G(B) \) as a function of the perpendicular magnetic field measured at \( V_G = 0V \) (blue), background signal \( G_{bg} \) (orange) and the conductance amplitude \( \Delta G(B) = G(B) - G_{bg} \) (red). (b) Normalized autocorrelation function of the red trace in (a). We analyze the magnetoconductance data measured at \( V_G = 0 V \) and temperature \( T = 1.8 K \) (see Figure SI 10 a). The background conductance \( G_{bg} \) is determined using the Savitzky-Golay filter\(^3\), and we only used the conductance amplitude data for magnetic fields \( B > 1 T \) to exclude the WAL peak (as depicted in Figure SI 10a). The correlation field extracted from the autocorrelation function in Figure SI 10 b is \( 260 \) m\( T \). Depending on the value of the \( C \) factor in Equation (4), this yields a correlation length in the interval between 110 nm and 250 nm for the estimated NW width of 50 nm. Since we cannot accurately estimate the effective mass in the nanowire, we can only find the limiting bounds from the bulk heavy and light hole masses\(^4\) leading to \( l_T \approx 100 \) nm – 290 nm which leaves the \( C \) factor in Equation (4) undetermined.
Furthermore, the autocorrelation function does not properly decay to zero, and therefore the correlation field has a large uncertainty. Therefore, we argue, this analysis only estimates the order of magnitude of the coherence length, $l_\phi \sim 100 \text{ nm}$.

10. Weak antilocalization fitting:

To fit the WAL peak in the magneto-conductance $G(B)$, first, the background must be subtracted. The background is removed by fitting the Drude mobility model in the same manner as described in the Section 3 of the Supplementary. In Figure SI 11 a we show measured $G(B)$ (red trace) averaged over the top gate voltage interval from $-1.1 \text{ V}$ to $0.4 \text{ V}$ ($\Delta V_G = 100 \text{ mV}$) where the data used for the Drude fit is highlighted in blue. The result of the fit is plotted with a lime dashed line. The mobility extracted from the Drude fit is $\sim 350 \text{ cm}^2/\text{Vs}$. Inserting the extracted mobility into Equation (2) we obtain an estimate on the effective width of the NW, $W_{\text{eff}} \sim 50 \text{ nm}$.

Figure SI 11: (a) Longitudinal conductance measured as a function of the perpendicular magnetic field averaged of the top gate voltage interval [-1.1V, 0.4V] (red), measured data used for the Drude model fit (blue) and the Drude model fit (dashed lime). (b) Conductance amplitude obtained from the data in (a) (blue). Dashed lines are WAL fits for 40, 50 and 60nm of effective NW width denoted by gold, orange and red dashed traces.

By subtracting the fit, $G_{bg}$, from the measured data, $G(B)$, we obtain the conductance amplitude $\Delta G(B)$ that we fit with the WAL correction given in the main text (blue data
points in Figure SI 11 b). The WAL fit is performed in the dirty metal regime, with the elastic mean free path $l_e$ set to 10 nm, and the effective NW width set in the range $W_{\text{eff}} = 50 \pm 10 \, \text{nm}$, while the spin-orbit length $l_{SO}$ and the coherence length $l_{\Phi}$ are the only fitting parameters. For the effective NW widths of 40 nm, 50 nm, and 60 nm, we extract a spin-orbit length of 12 nm, 8 nm and 5 nm, and a coherence length of 105 nm, 98 nm and 94 nm respectively.

11. Transport measurements along <110> and <100> oriented nanowires

As discussed in the main text of our manuscript, to determine density and mobility of the NWs, assumptions on the transport cross section must be made which introduced large uncertainty in these parameters. As <110> and <100> nanowires have different geometry of the cross-sections, making direct comparisons in terms of density and mobility is difficult. For example, <110> NWs grow taller than <100> NWs. Furthermore, an additional difference is the width of the NWs caused by the etching of the SiO$_2$ mask from the side of the trenches before the growth process (before loading the sample into the growth chamber, we do an HF dip to remove the native oxide on Si which also etches sides of the SiO$_2$ mask). At this point, we can only claim that all measured NWs have a density of the order of $n \sim 5 \cdot 10^{18} \, \text{cm}^{-3}$ and mobility $\mu \sim 300 \, \text{cm}^2/\text{V}s$.

Moreover, during the fabrication process we have only managed to fabricate a top gate on one of the devices (the one discussed in the main text), hence we have not managed to average out the influence of UCF in magneto transport measurements, shown

Figure SI 12, when comparing <100> and <110> NWs. This makes the extraction, and hence the comparison between <100> and <110> direction in terms of spin orbit strength unreliable.
Figure SI 12: $\Delta G_{xx}$ defined as $G_{xx}(B) - G_{xx}(0)$ (a,b) and $R_{xy}(B)$ (c,d) as a function of perpendicular magnetic field obtained from magneto transport measurements performed on ungated NW Hall bars with nominal widths of 50 and 120 nm.

References

1. Lee, P. A., Stone, A. D. & Fukuyama, H. Universal conductance fluctuations in metals: Effects of finite temperature, interactions, and magnetic field. *Phys. Rev. B* **35**, 1039–1070 (1987).

2. Beenakker, C. W. J. & van Houten, H. Quantum Transport in Semiconductor Nanostructures. *Solid State Phys. - Adv. Res. Appl.* **44**, 1–228 (2004).

3. Savitzky, A.; Golay, M. J. E. Smoothing and Differentiation. *Anal. Chem* **36**, ...
1627–1639 (1964).

4. Lawaetz, P. Valence-Band Parameters in Cubic Semiconductors. Phys. Rev. B 4, 3460–3467 (1971).
