

Supporting information for:

Spatio-Temporal Imaging of the Acoustic Field Emitted by a Single Copper Nanowire

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Copper Nanowires Fabrication

To fabricate the nanowires, 30 μm thick polycarbonate foils (Makrofol N, Bayer AG) were irradiated at the UNILAC accelerator of GSI Helmholtz Centre for Heavy Ion Research (Darmstadt, Germany) with $\sim 2 \text{ GeV}$ Au ions at a typical fluence of $\sim 10^8$ ions/cm². On

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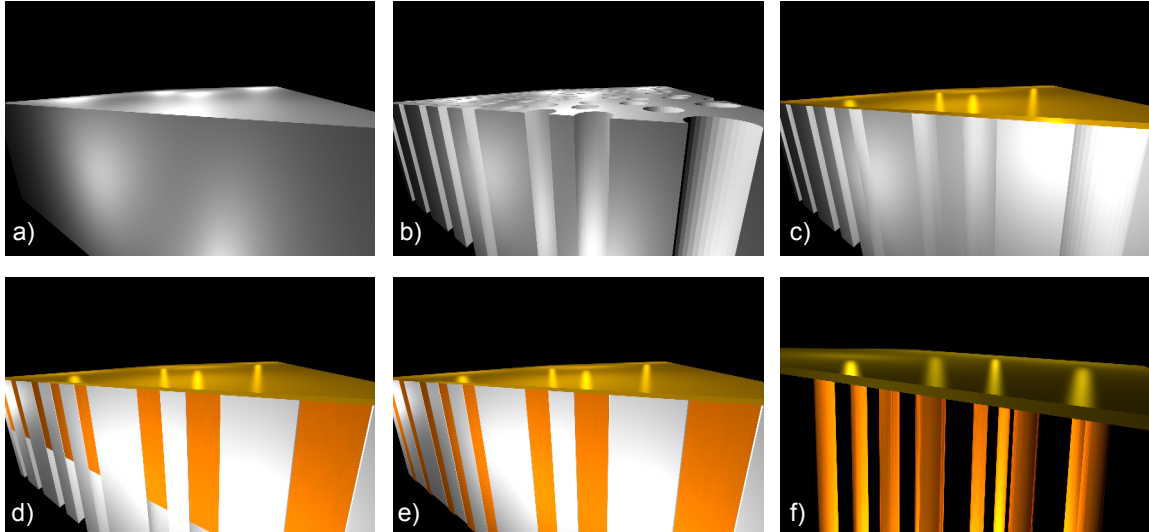


Figure S1: (a) 30 μm thick polycarbonate foils. (b) After ion irradiation, the tracks were selectively dissolved and enlarged resulting in cylindrical smooth nanochannels used as template to synthesize nanowires. (c) A cathode is prepared by sputtering a 100 nm layer of Au on one side of the membrane. (d)-(e) Cu was potentiostatically deposited in the channels in a two-electrode setup. (f) The polymer membrane was dissolved in dichloromethane.

its way through the foil, each ion creates a damaged cylindrical zone called latent track. After irradiation and exposition to UV light from both sides for 1 h (30 W, T-30M, Vilber Lourmat), these tracks were selectively dissolved and enlarged in a 6M NaOH solution at 50 $^{\circ}\text{C}$ for 15 min and 5 min resulting in cylindrical smooth nanochannels with diameter of 400 nm and 120 nm respectively, used as template to synthesize nanowires (Fig.S1(b)). The channel diameter increases nearly linearly with the etching time. A cathode is prepared by sputtering a 200 nm layer of Au on one side of the membrane (Fig.S1(c)). The layer was reinforced electrochemically with a 1 μm copper layer. Cu was electrochemically deposited in the channels in a two-electrode setup as constant applied cell voltage of $U = -0.03\text{ V}$ (Fig.S1(d)-(e)). The potentiostatic deposition of copper into the etched pores of the template was accomplished with an electrolyte solution of copper sulfate and sulfuric acid (238 g/L $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ and 21 g/L H_2SO_4). After nanowire growth, the polymer membrane was dissolved in dichloromethane (Fig.S1(f)), and the wires were detached

from the cathode by ultrasonification. In a final step, the nanowires were drop-casted onto the silicon membranes. If the dissolution step is well performed, the copper nanowires appear smooth and orange using optical microscopy (Fig.S2(f)).

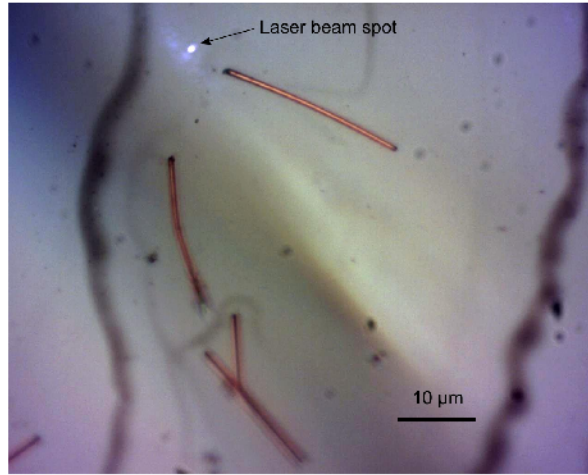


Figure S2: 400 nm diameter copper nanowires observed under white illumination with a 0.95 numerical aperture 100× objective. The probe beam laser is focused on the silicon membrane several microns away from a nanowire.

Silicon Membrane Fabrication

Anisotropic wet etching in KOH is undertaken on a 2 inches silicon on insulator (SOI) wafer consisting in a 300 μm thick Si(100) layer superimposed to a 1 μm thick SiO₂ layer and a last 10 μm thick Si(100) layer (Fig.S3(a)). A thermally grown 400 nm thick SiO₂ layer (using oxygen gas in a tube furnace) is used as a protective layer on the backside of the SOI wafer (Fig.S3(b)). A positive resist (Shipley S1813) was spin coated on the surface and exposed to UV light using a standard optical lithography system. After being developed, rinsed in water, blown dry, and postbaked, the sample was etched.

SiO₂ was locally removed in a buffered HF solution (BHF) and resist was dissolved in acetone (Fig.S3(c)). The substrate is then dipped in a hot 40% KOH solution until the buried silica layer of the SOI wafer is reached (Fig.S3(d)). The (100) plane is etched faster than the (111) plane due a high number of dangling bonds, resulting in the inverted structure.

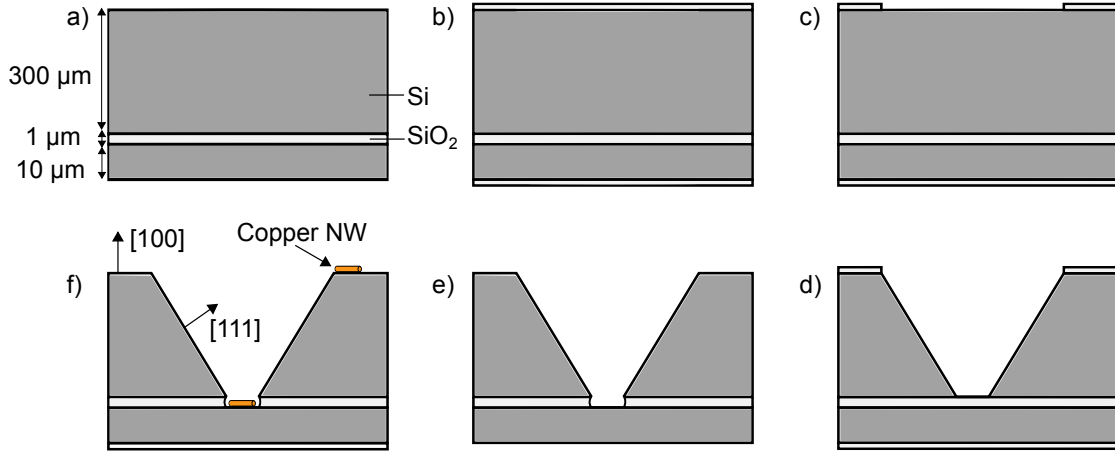


Figure S3: (a) 2 inches silicon on insulator (SOI) wafer : 300 μm – 1 μm – 10 μm thick Si(100)–SiO₂–Si(100) layers. (b) Thermally grown 400 nm thick SiO₂ layer. (c) SiO₂ locally removed using standard optical lithography and HF etching techniques. (d) Anisotropic Si etching using a 40% KOH solution. (e) The silica layers are then removed in HF. (f) The nanowires were drop-casted on the membrane.

The silica layers are then removed in HF (Fig.S3(e)). 10 μm thick silicon membranes at the bottom of inverted pyramids are thus obtained. For detection purpose, a 200 nm thick layer of aluminium is evaporated on opposite side of the inverted pyramids. In a final step, the nanowires were drop-casted at the bottom of the pyramids (Fig.S3(f)).

Optical Setup

The optical setup is a pump-probe ultrafast laser setup. The laser source is a mode locked Ti:Sapphire (MAI-TAI Spectra) operating in the range of 720 nm up to 910 nm, with a repetition rate around 79.3 MHz. The pulse time width is less than 100 fs at the laser output. The laser beam is split in two parts with a polarization beam splitter (PBS1). A half-wave plate is used to tune the amount of laser power sent to the pump or to the probe. The pump beam is modulated at 1.8 MHz using an acousto-optic modulator. The pump beam is focused on the silicon membrane by a 0.95 numerical aperture 100 \times objective or

by a 0.50 numerical aperture 50 \times objective. Because the scattered light coming from the pump beam could strongly affect the lock-in detection, we realize a two-color experiment by doubling the pump frequency with a non-linear crystal (BBO). The beam diameter at $1/e^2$ is thus estimated to be respectively $< 1 \mu\text{m}$ and $< 2 \mu\text{m}$ in the focal plane, for the $\times 100$ and $50\times$ objectives.

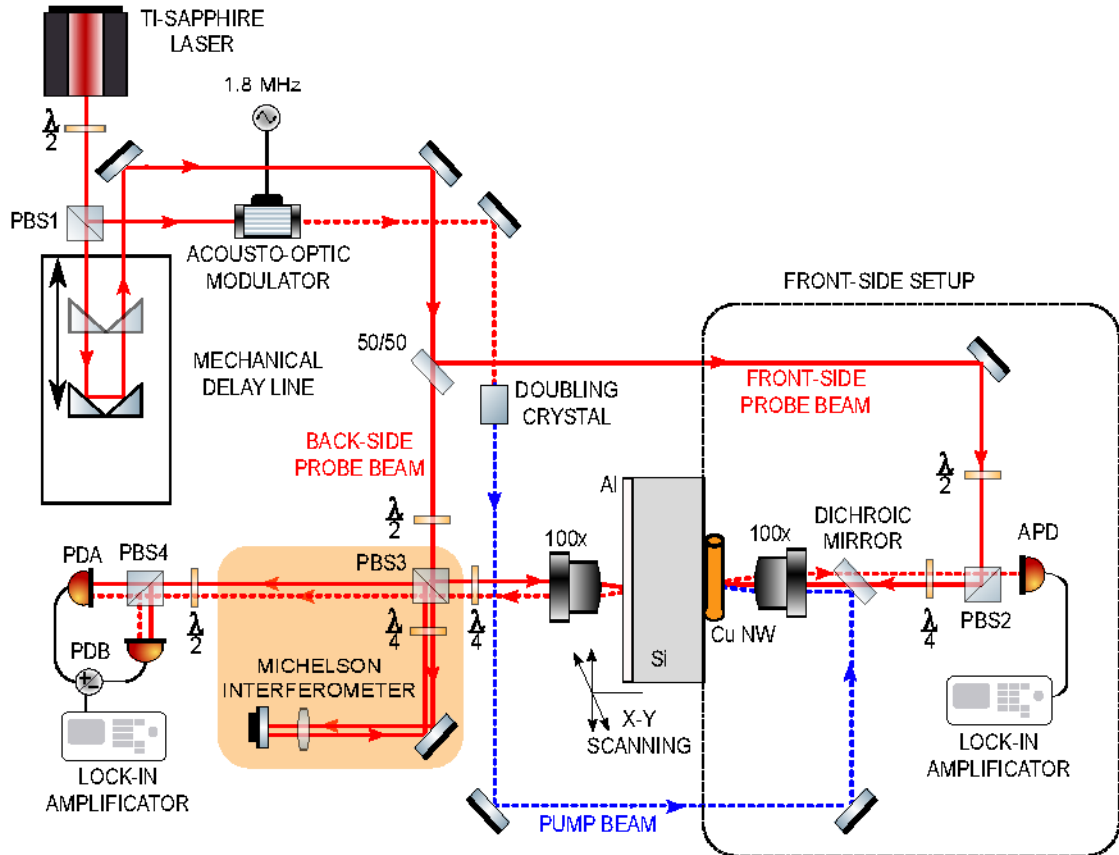


Figure S4: Simplified scheme of the set-up. The two probe beams can be simultaneously focused on both sides of the silicon substrate. On the nanowire side, the reflectometry signal is acquired thanks to an avalanche photodiode (APD). On the other side, the reflected red probe is sent to a Michelson interferometer.

The probe beam passes through a mechanical delay line. Two retro-reflectors are mounted on a 60cm length translating stage, in front of the static one. The three round trips of the light in this setup gives us a maximum length of 3.6m resulting in a 12 ns maximum delay between the two laser pulses (almost reaching the repetition rate of the laser which is

12.6 ns). The probe beam has been split in two arms using a 50R/50T plate beamsplitter, the first one is focused by the same optical system applied to focus the pump beam on the sample surface; the second arm is focused by a long working distance 0.50 numerical aperture 100× objective (100× Mitutoyo Plan Apo SL Infinity Corrected Objective) on the aluminium side (back-side) of the sample (see Fig.S1(f)). The pump beam and the first probe beam constitute the front-side setup, which is a very standard two-color pump-probe reflectivity experiment. A half-wave plate and a PBS (PBS2) are positioned just before the objectives entrances in order to collect the reflected probe beams. The $\lambda/4$ retardation plates convert the linear polarized light of the laser in a circularly polarized light. On the back-side setup, the probe beam has been split in two arms using the PBS3. The first one is the “sample” beam, the second one is reflected by a reference mirror mounted on a piezoelectric device. In this Michelson configuration, the sample and the reference beam which exhibit perpendicular polarization are collected. In order to mix these two arms, a $\lambda/2$ plate and a polarizing beam splitter (PBS4) are used to obtain interference on two photodiodes (PDA and PDB). In the past,^{S1} it has been demonstrated that controlling the reference mirror position with a feedback loop, the A–B locking signal represent the imaginary part of $(\Delta R/R)(t)$ in contrast with reflectivity measurement which gives the real part of the same quantity. Usual reflectometry measurements may be performed by occulting the reference arm.

Isotropic Model

As a first and rough approximation before exploiting the Finite Difference Time Domain (FDTD) simulations, the surface displacement mapping is investigate through the isotropic approximation to grasp the general tendancies. In this model, the nanowire generates longitudinal spherical waves with a temporal periodicity $T_0 = 125$ ps (Fig.S5). The surface displacement mapping undergone in the article is expected to display concentric rings according to this isotropic approximation.

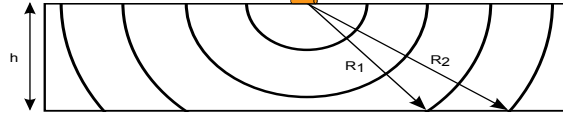


Figure S5: Schematic side view of the silicon membrane of thickness h at the time delay t_d . The nanowire generates concentric spherical waves with a temporal periodicity $T_0 = 125$ ps.

Let us consider the surface displacement mapping done at a time delay $t_d > t_{L1}$, where t_{L1} is the time of flight of a longitudinal wave across the silicon membrane of thickness h : $v_L^{\text{SI}} t_{L1} = h$. N concentric rings are observable. The radius R_1 of the inner ring is $R_1 = \sqrt{(v_L^{\text{SI}}(t_d - (N - 1)T_0))^2 - h^2}$, the radius R_2 of the following ring is $R_2 = \sqrt{(v_L^{\text{SI}}(t_d - (N - 2)T_0))^2 - h^2}$ and the last ring has a radius $R_N = \sqrt{(v_L^{\text{SI}} t_d)^2 - h^2}$. Generally speaking, the n^{th} ring has a radius $R_n = \sqrt{(v_L^{\text{SI}}(t_d - (N - n)T_0))^2 - h^2}$. Let us consider 5 concentric rings at a time delay $t_d = 1.8$ ns. The figure S6a is the radius R_n and the figure S6b is the separation d between two successive rings $d = R_{n+1} - R_n$.

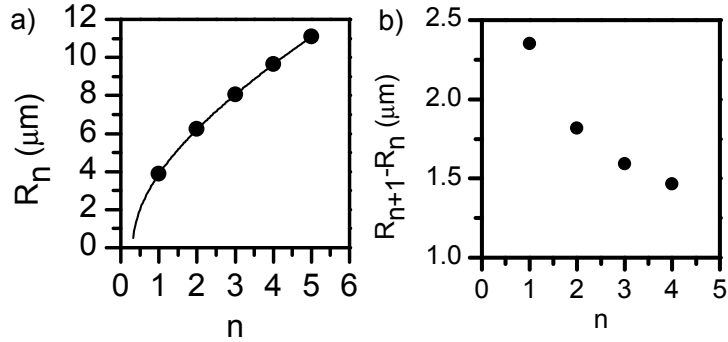


Figure S6: a) Radius R_n of the concentric rings. b) Separation d between two successive rings $d = R_{n+1} - R_n$.

Supporting Movies

Movie S1 : It shows the evolution in time of the acoustic pattern of the Figure 3(d).

Movie S2 : It shows the evolution in time of the acoustic pattern of the Figure 3(a).

References

- (S1) Perrin, B.; Rossignol, C.; Bonello, B.; Jeannet, J. C. *Physica B*. **1999**, *263-264*, 571–573.