SUPPORTING INFORMATION

High-Order Hilbert Curves: Fractal Structures with Isotropic, Tailorable Optical Properties.

Stefano De Zuani, ^{†,*} Thomas Reindl, [‡] Marcus Rommel, ^{‡,+} Bruno Gompf, [†] Audrey Berrier, [†] and Martin Dressel[†]

[†]1. Physikalisches Institut and Research Center SCoPE, Universität Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart, Germany

[‡]Max Planck Institute for Solid State Research, Heisenbergstrasse 1, 70569 Stuttgart, Germany

+Current address: Chalmers University of Technology, MC2 Kemivägen 9, 41285 Gothenburg, Sweden

*E-mail: stefano.de-zuani@pi1.physik.uni-stuttgart.de.

Hilbert Curve geometry:

The Hilbert curves for the first three iterations are plotted in Fig. S1:



Figure S1: Geometry of the Hilbert curves for the first three iterations (a) N = 1, (b) N = 2, (c) N = 3. The length *a* of the shortest line segment is indicated by the black arrow.

Design of the Hilbert curve pattern of order N = 9:

The Hilbert curves of order N = 1 is formed by three segments of length *a* which in our work is equal to 130 µm in an area of 130x130 µm² (writing field size for e-beam exposure). With increasing fractal order *N*, the length *a* of the shortest segment is scaled down according to the formula $a_{N=1}/(2^{N}-1)$ while the total length A of the curve increases following $A = (2^{N}+1)*a$.^{S1} In Table S1 the calculated lengths *a* and the Euclidian total length A of the Hilbert curves designed in an area of 130x130 µm² are shown for the fractal order N from 1 to 12. The values for N = 9 refers to the Hilbert curve presented in this work. The similarity dimensions *DS* of Hilbert curves of order $N \ge 2$ shown in Table S1 are calculated according to the formula:^{S2}

$$DS = \frac{\log[4^{N}-1)/(4^{N-1}-1)]}{\log[2^{N}-1)/(2^{N-1}-1)]}$$
(1)

Equation (1) vs *N* is also plotted in Fig. S2:



Figure S2: Similarity dimension *DS* of Hilbert curves of fractal orders *N* from 1 to 12 (red dots). The dashed black line shows the limit of $N \rightarrow \infty$.

The similarity dimension DS of the Hilbert curves increases rapidly towards DS = 2 which corresponds to a two dimensional structure (i.e., a closed thin film).

| Ν | a (µm) | Α (μm) | DS |
|----|--------|--------|--------|
| 1 | 127.8 | 383 | - |
| 2 | 42.6 | 639 | 1.465 |
| 3 | 18.3 | 1150 | 1.694 |
| 4 | 8.5 | 2173 | 1.834 |
| 5 | 4.1 | 4217 | 1.914 |
| 6 | 2.03 | 8307 | 1.956 |
| 7 | 1.01 | 16486 | 1.978 |
| 8 | 0.501 | 32845 | 1.989 |
| 9 | 0.250 | 65561 | 1.994 |
| 10 | 0.125 | 130995 | 1.997 |
| 11 | 0.062 | 261862 | 1.9986 |
| 12 | 0.031 | 523597 | 1.9993 |
| | | | |

Table S1: Shortest length a, total length A in an area of 130x130 μ m² and similarity dimensions *DS* for Hilbert curves of fractal order *N* from 1 to 12.

Fabrication of the Hilbert curve of order N = 9:

The sample was prepared using a Jeol JBX6300FS electron beam lithography (EBL) system. The Hilbert structure was fabricated on a Si substrate covered with 3 nm of native oxide. Two layers of PMMA with different sensitivity were spin coated on the substrate to obtain a resist mask with undercut, which improves the lift-off of the Au film. First PMMA 200k 3.5% was spin coated at 6000 rpm for 35 seconds and then prebaked on a hot plate for 4 minutes at 160°C. In the same way PMMA 950k 1.5% was spun on the sample and baked. The Hilbert curve pattern was then exposed into the resist over an area of 130x130 μ m². By repeating 20x10 times the writing field of area 130x130 μ m² with a gap of 250 nm in between fields, a total exposed area of 2.6 mm by 1.3 mm was obtained; a large patterned area is crucial to perform optical measurements with the ellipsometer, in particular at high angles of incidence where the incident light beam is about 2 mm wide. The exposure parameters were 100 kV of acceleration voltage, 1 nA of beam current, shot pitch 4 nm and a dose of 1100 μ m/cm². The sample was developed in a Methylisobutylketone (MIBK) and Isopropanol (IPA) mixture of 1:3 for 60s at 22°C. The development was stopped by putting the sample for 15 seconds in IPA and blow-dry with N₂ gas. Thermal evaporation was then used to deposit a 2-nm-thick adhesion layer of Cr with an evaporation rate of 1 Å/s and 50 nm of Au with an evaporation rate of 2 Å/s. Finally, a 2 hour-lasting N-Ethyl-2-pyrrolidone (NEP) bath at 75°C as well as followed by ultrasonic agitation applied at low power for few seconds at the end of the lift-off process. The sample was rinsed in Acetone and IPA and dried with N₂ gas.

Identification of the correct model:

Highly anisotropic spectroscopic ellipsometry was performed to look for the possible presence of non-zero offdiagonal elements of the Jones matrix (Ψ_{PS} , Ψ_{SP} , Δ_{PS} , Δ_{SP}) which would indicate the presence of anisotropy in the structure.



Figure S4: Spectroscopic ellipsometry angles $\Psi_{PS}(^{\circ})$, $\Psi_{SP}(^{\circ})$ measured at angles of incidence from 45° to 75° every 10° in the frequency range between 400 and 1500 nm.

As we can see from Fig. S4 the off-diagonal elements $\Psi_{PS}(^{\circ})$, $\Psi_{SP}(^{\circ})$ are below 1 for angles of incidence up to 75° hence it is safe to say that mixing of polarization can be neglected. In the case of our structure, although an isotropic model can describe quite well the ellipsometry data, it fails to capture the difference in the absorption between p-polarized light and s-polarized light around 530 nm caused by the non-zero thickness of the two-dimensional Hilbert structure. The presence of this out-of-plane resonance forces us to adopt a uniaxial model with in-plane isotropy to better describe the optical properties of the Hilbert structure. These results confirm the uniaxial model as the right model for the Hilbert structure and it allows us to conclude that further measurements at other angles of incidence will not change the behavior of the reflected intensity predicted by the model. In particular the isotropic azimuthal behavior shown in Fig. 2(d) is confirmed.

Model 1: Uniaxial General Oscillator Layer (GenOsc) Model

The scheme of the uniaxial general oscillator layer model is shown in Fig. S4.



Figure S4: Scheme of the uniaxial GenOsc layer model.

Model 1 is made of a uniaxial general oscillator layer formed by two different GenOsc layers to model the effective permittivity of the Hilbert nanostructures along the x and y axis (in-plane direction) and z axis (out-of-plane direction). The permittivity tensors along the x and y axis are equal. The in-plane GenOsc layer is built with four Lorentz oscillators and a Drude component while the GenOsc layer in the out-of-plane direction is formed by two other Lorentz oscillators.

Model 2: Uniaxial General Oscillator Layer (GenOsc) Model with in-plane Bruggeman Effective Medium Approximation (BEMA) Model

The scheme of the uniaxial Bruggeman effective medium approximation (BEMA) model is shown in Fig. S₅.



Figure S5: Scheme of the uniaxial BEMA model.

In model 2 the GenOsc layer in the in-plane direction has been substituted by a BEMA layer composed by the dielectric constants of a 50 nm closed gold film with a filling factor f = 20% and 80% voids. The GenOsc layer in the out-ofplane direction is exactly the same as in model 1 to take into account the three dimensionality of the Hilbert structure. The Au filling factor f is equal to 20% and the depolarization factor L is equal to 0.18.

Reflectance at normal incidence over a broad frequency range:

Figure S6 shows two reflectance measurements in the frequency range between 700 and 1230 nm (red) and between 1250 and 12500 nm (black) measured by a Bruker IFS 66/s Fourier-transform infrared spectrometer at normal incidence using un-polarized light.



Figure S6: Reflectance measured at normal incident between 700 and 1230 nm (red) and 1250 and 12500 nm (black). The inset is a zoom-in of Fig. S6 between 700 and 2000 nm. The noise seen around 1200 nm in the red curve is due to the decrease in detectivity of the Silicon detector in this frequency region.

Figure S6 confirms the quasi-flat reflectance of the Hilbert structure even at higher wavelengths up to 12.5μ m. It also shows the appearance of three more broad features at about 3.4, 5.5 and 8.2 μ m together with the peak at about 1.8 μ m already seen in Fig. 2(c) of the article, which are probably related to further resonance modes of the Hilbert curve in this frequency range. However one can see that in the whole range from 700 nm to 12.5 μ m the reflectance is constant within 12%.

As addition check of the validity of the uniaxial model the reflectance measured at normal incidence with the FTIR spectrometer is compared with the one extracted at normal incidence with model 1 and model 2:



Figure S7: Reflectance measured at normal incident between 700 and 2200 nm (black) compared to the reflectance at normal incidence extracted from (a) model 1 (blue) and (b) model 2 (red).

Figure S7 confirmed the good agreement of both models with the measured data.

References:

(s1) Schroeder, M. R. Fractals, Chaos, Power Laws. In *Introduction*; W. H. Freeman and Company: New York, o 1991; pp 9-13.
(s2) Sanchez-Hernandez, D. A. Multiband Integrated Antennas for 4G Terminals. In *Printed Multiband Fractal Antennas*; Artech House Inc: Boston, London, 2008; pp 101-125.