Supporting Information

Anomalous Oriented Attachment Growth Behavior on SnO_2 Nanocrystals

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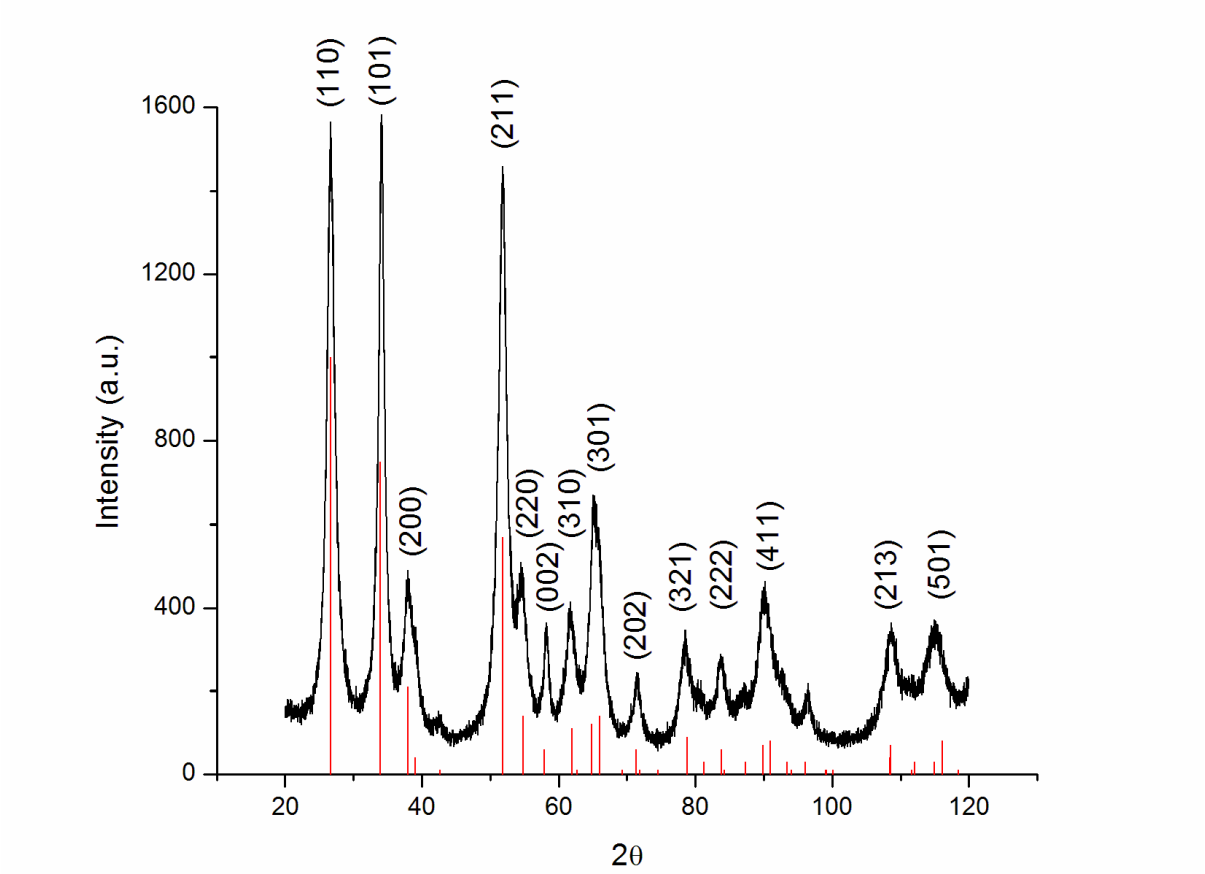


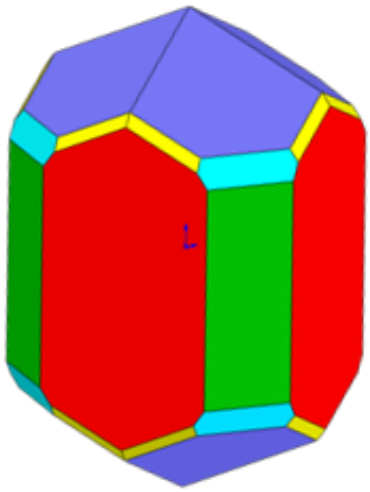
S1 - XRD Characterization

Figure S1. Indexed X-ray diffraction pattern of dry SnO_2 powder sample obtained with a rotary anode Rigaku D/MAX 2500 PC diffractometer using Cu-Kα radiation. Red lines denote the reference spectrum[1] accordingly to JCPDS 41-1445. The XRD peaks indicate small crystalline domains with dimensions of approximately 12 and 16 nm for (110) and (101) planes, respectively, as evaluated by Scherrer's equation.

[1] - McCarthy, G. J. & Welton, J. M. X-Ray Diffraction Data for SnO_2. An Illustration of the New Powder Data Evaluation Methods. *Powder Diffr.* **4**(3), 156-160 (1989).

S2 – SnO_2 Surface Energy *ab initio* Calculation

Surface energy calculations[2] for SnO_2 nanocrystals were performed with the CRYSTAL06 program package. Oxygen atoms were described by the standard 6-31G* basis sets and the Tin centers in the PS-21G* scheme, where PS stands for Durand-Barthelat's non-relativistic large effective core potential. Becke's three-parameter hybrid nonlocal exchange functional combined with the Lee-Yang-Parr gradient-corrected correlation functional (B3LYP) was used. Full optimization of the cell parameters (a and c) and internal coordinate (u) for the bulk SnO_2 was carried out. The low index (110), (101), (100) and (001) surfaces were modeled by unreconstructed (truncated bulk) slab models using the calculated equilibrium geometry. Because these surfaces have a different number of atoms in each layer, the low-index surfaces were modeled with different thicknesses in the z-direction but periodic in x- and y-directions. After the corresponding convergence test, slab models containing nine SnO_2 layers for the (110), (100), and (101) surfaces and eleven layers for the (001) one were selected. A complete relaxation of all the atoms in each model was performed.



Tetragonal SnO_2 - $P4_2/mnm$		
(hkl)	Surface Energy (J/m^2)	Relative Distance
100	1.74	1.000
001	2.71	1.557
110	1.56	0.897
101	2.11	1.213
111	4.07	2.339
211	2.18	1.253
301	2.08	1.195

Figure S2. Wulff geometry[3] for SnO_2 nanocrystal after the *ab initio* surface energy calculations.

[2] - Beltrán, A.; Andrés, J.; Longo, E.; Leite, E. R.; *Appl. Phys. Lett.* **2003**, 83 (4), 635-637.

[3] - Herring, C.; *Physical Reviews* **1951**, 82 (1), 87-93.

S3 – Monte Carlo Model

A custom Monte Carlo based algorithm was implemented using Microsoft Visual Basic programming language for the evaluation of aspect ratio distribution for a system with 2D isotropic attachment behavior. Firstly, a 20x20 matrix M_{ij} with a seed in its center was built, according to Figure S3a. Then, two random integer numbers ranging from 1 to 20 were drawn denoting the *i* and *j* matrix coordinates. The applied selection rule was determined by the neighborhood: for the *ij* drawn coordinates just next to the seed, it would be integrated to the seed; otherwise, other random *ij* coordinates would be drawn.

This process was repeated iteratively until the seed reached 20 blocks, as depicted in Figure S3b. This process was repeated 50000 times in order to assure a statistical representativeness for L and W dimensions measurements. The aspect ratio was considered as the ratio between lengthen and shorten directions on *i* and *j* axes.

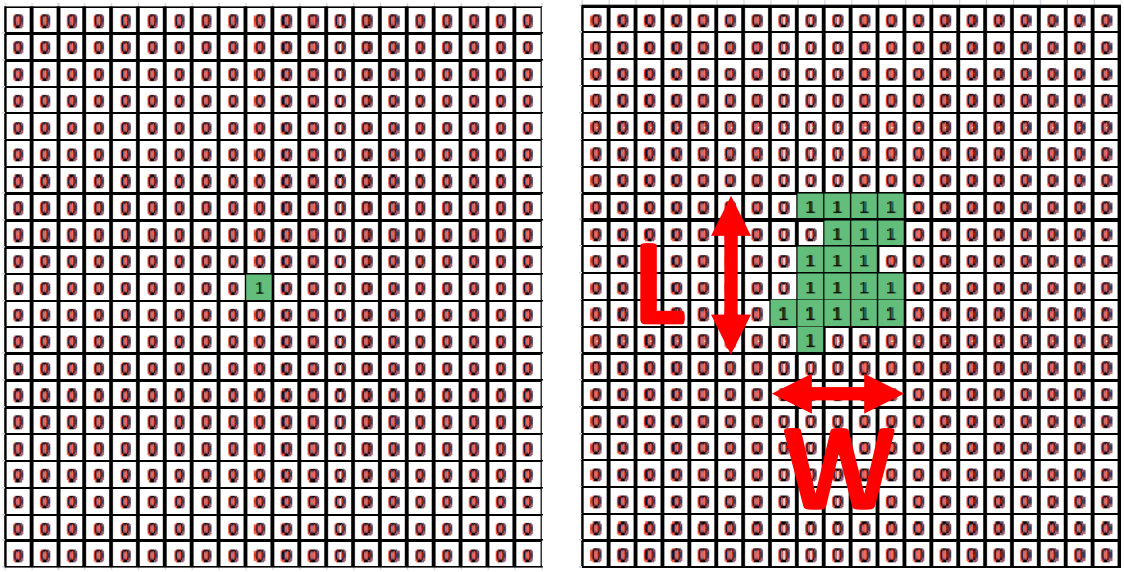


Figure S3. (a) Initial M_{ij} matrix with a seed and (b) result after the iterative calculation until 20 neighbor blocks. The L and W refer to the maximum dimensions along *i* and *j* directions.

The results indicated 1.308 and 0.081 as the mean aspect ratio value and variance, respectively.