In situ Three-Dimensional Imaging of Strain in Gold Nanocrystals During Catalytic Oxidation

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Supporting Information

Surface stress determination.

The surface stress σ_s can be estimated by the Young-Laplace equation:[1]

$$\sigma_s = -\frac{3K}{2}\frac{\Delta a}{a} \qquad (1)$$

where $K = 220\ GPa$ is the bulk modulus of gold, R the radius of the locally rounded region and $\frac{\Delta a}{a}$ the strain. The strain at the position indicated by the grey arrow of Fig. 2b is $-3.7 \cdot 10^{-4}$ for the nanocrystal under air and $2.6 \cdot 10^{-4}$ in CO/O_2. We can estimate the radius R of the locally rounded region is 20 nm. Equation (1) leads to a surface stress of 2.4 ± 0.3 $N \cdot m^{-1}$ for the nanocrystal under air, typical of tensile surface stress of metals in the range of 2 $N \cdot m^{-1}$.[1,2] On the other hand, the surface stress of the nanocrystal under CO/O_2 turns to be compressive and equals to -1.7 ± 0.1 $N \cdot m^{-1}$.

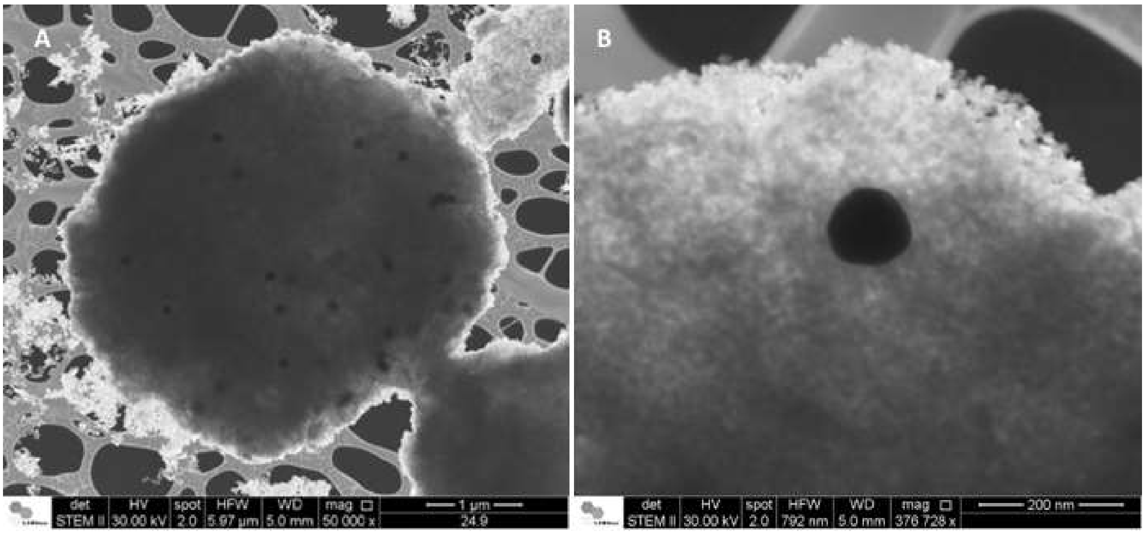

Fig. S1. Scanning transmission electron microscopy images of the Au/TiO_2 nanoparticles. (a) Low-magnification and (b) high-magnification.

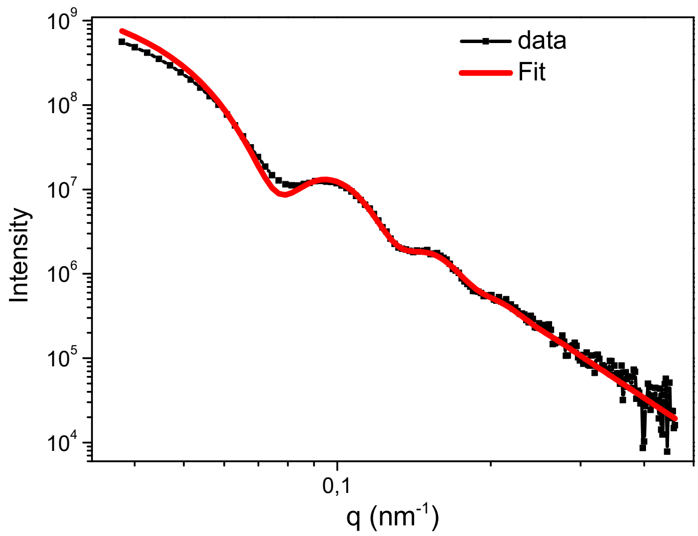

Fig. S2. SAXS pattern (black) of the synthetised gold nanoparticles and its fit (red) obtained with a sphere form factor and log-normal distribution.

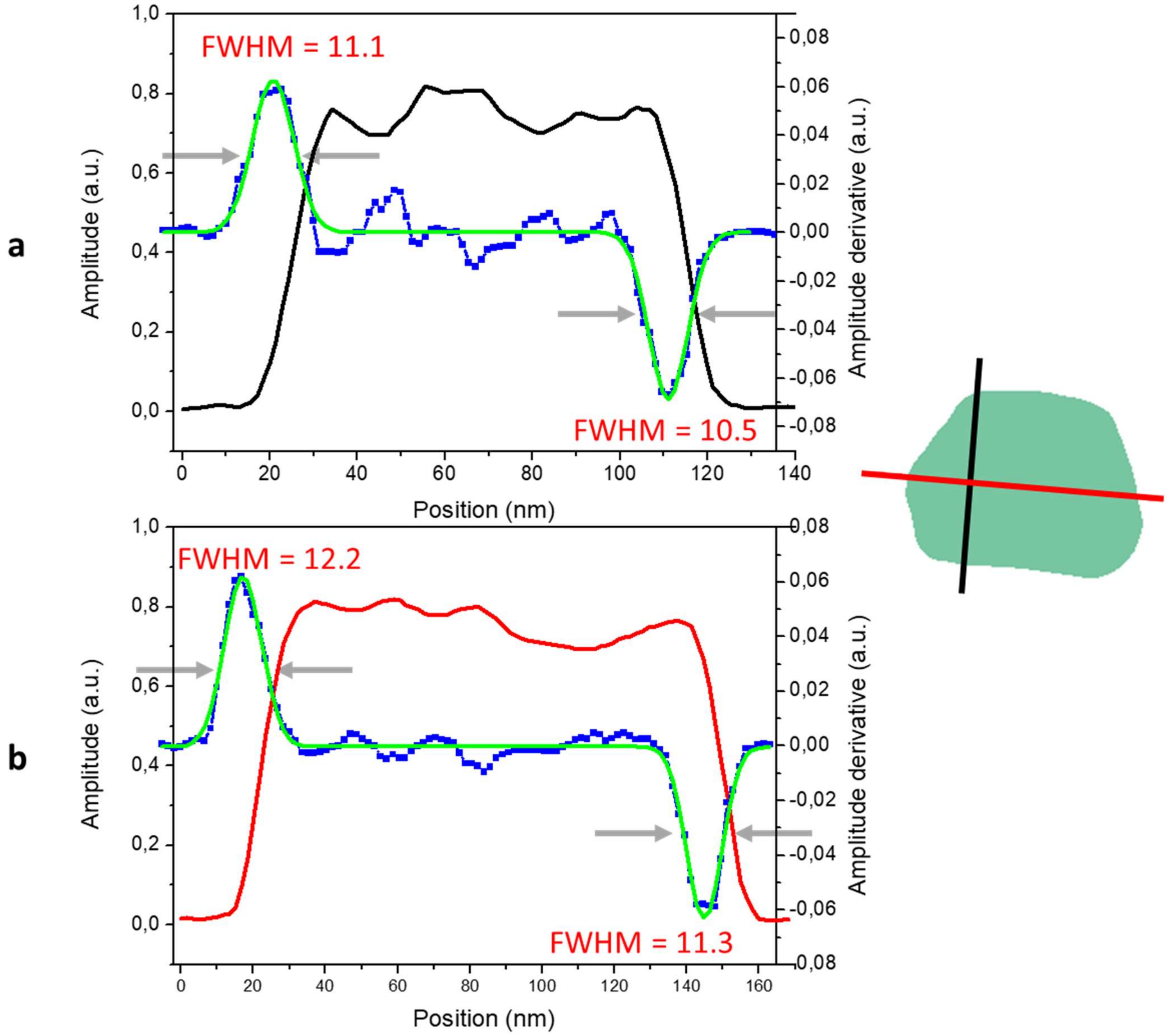

Fig. S3. Averaged line profiles. Lines (a, black) along the vertical and (b, red) horizontal directions of the particle cross-section corresponding to Figure 3 (shown here as green surface). The derivatives of the line scans are showing that the reconstruction resolution is 12 nm.

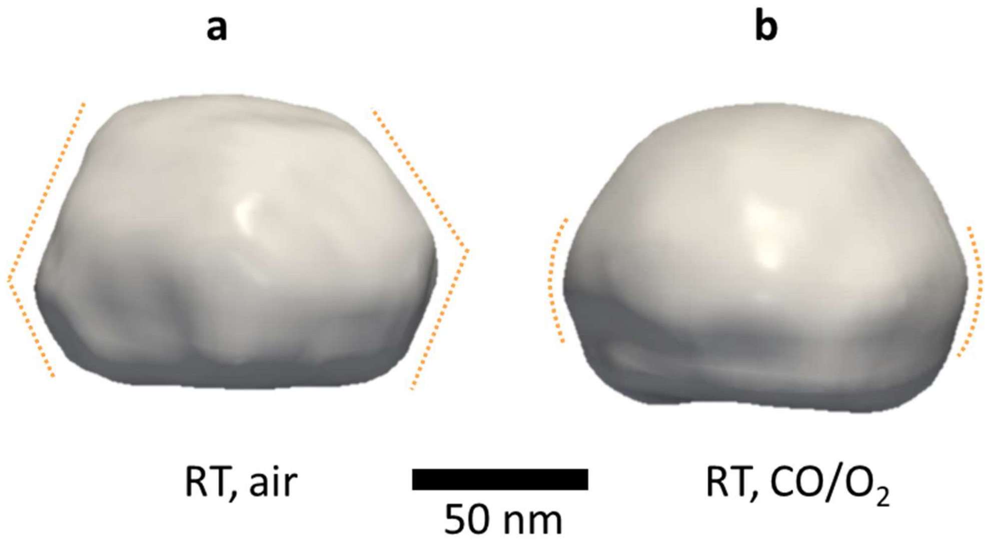

Fig. S4. *In situ* Bragg coherent X-ray diffraction imaging. A grey isosurface (30 %) representing the particle shape of the same Au/TiO_2 nanoparticle in side view at RT under air (a) and under CO/O_2 (b). The dash lines are showing the facetted and rounder shapes of the nanoparticle.

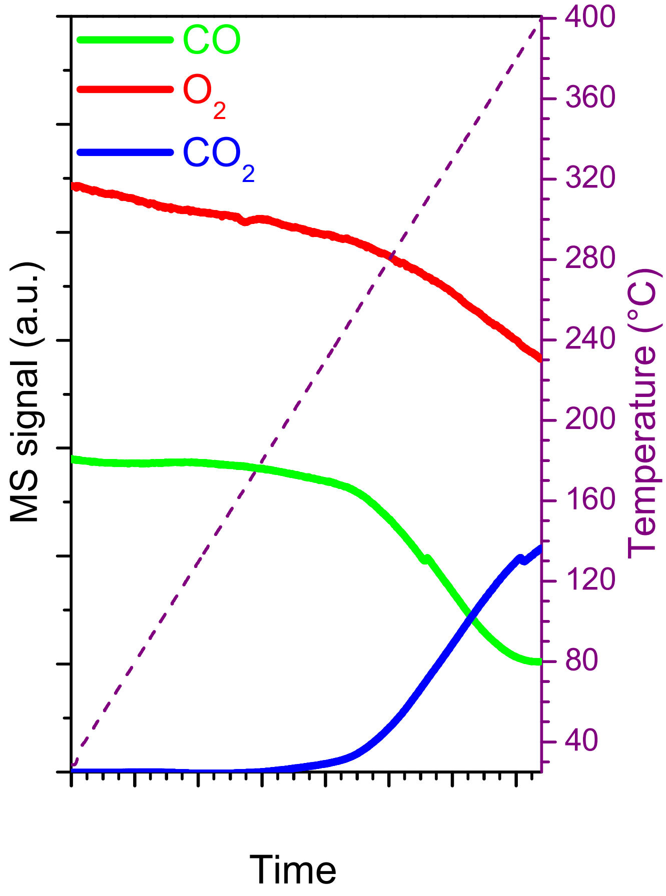

Fig. S5. Mass spectrometry signal of O_2, CO and CO_2 during a separate experiment, heating the gold catalyst inside the CDI cell.[3]

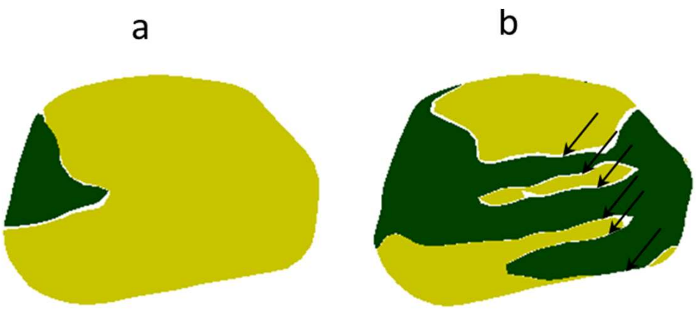

Fig. S6. Displacement map from a cross section of the Au nanocrystal at 200 and 400 °C under CO/O_2. Cross section of the distribution of the low (yellow) and high (green) phase shift of the same Au/TiO_2 nanoparticle under CO/O_2 at 200 °C (a) and 400 °C (b). The black arrows indicate the position of the nanotwin network.

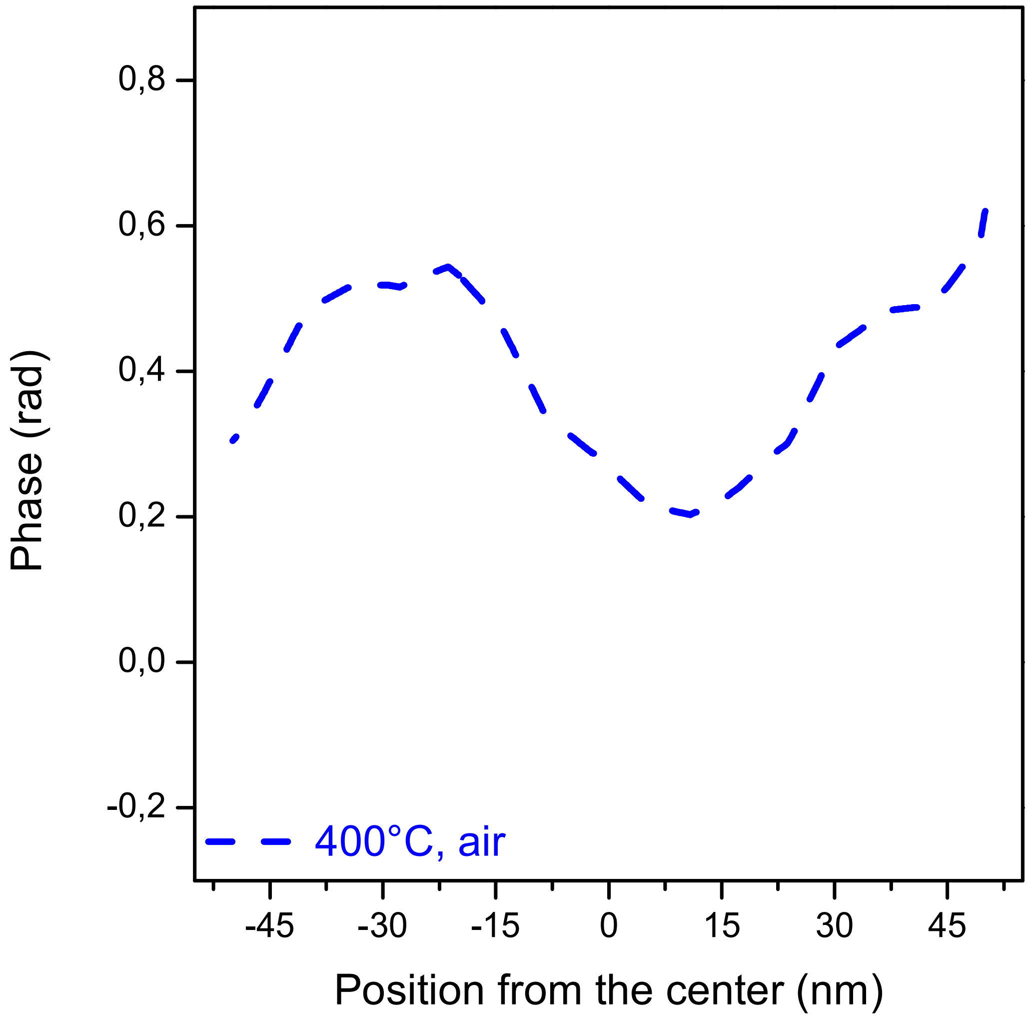

Fig. S7. Line scan of the phase at 400 °C under air. The line scan is corresponding to the values of the position of the dashed line shown (for 400 °C under CO/O_2) in Fig. 2a.

References:

1 I. Robinson, *J. Phys. Soc. Japan*, 2013, **82**, 1.

2 W. Haiss, *Reports Prog. Phys.*, 2001, **64**, 591.

3 A. Rochet, A. F. Suzana, A. R. Passos, T. Kalile, F. Berenguer, C. V. Santilli, S. H. Pulcinelli and F. Meneau, *Catal. Today*, 2018, **in press**, 10.1016/j.cattod.2018.12.020.