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Structure matters: asymmetric CO ignition at square and triangular Rh steps

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Tracking the active site of catalytic metallic nanoparticles is mandatory for tailoring new catalysts and promoting a clean and green environment. The activity of each of the facets of the nanoparticle will differ depending on the orientation and coordination of surface atoms [1], yet isolating their individual activity is typically challenging. Within this context, curved crystals possess several different planes, hence they are ideal for selectively probing different types of undercoordinated atoms [2,3]. Using this methodology, we have studied the CO oxidation on Rh stepped surfaces using a curved Rh(111) crystal. This peculiar sample features the flat (111) surface at the center of the crystal, and an increasing density of either A- (square) and B- (triangular) steps as one approaches each of the edges of the sample.

Preliminary Planar Laser-Induced Fluorescence (PLIF) experiments reveal that the B-side of the crystal ignites earlier during the CO oxidation, mimicking the results obtained using an identical curved Pd(111) crystal [2b], and in clear contradiction with the symmetric ignition observed for a curved Pt(111) sample [3]. Near-ambient pressure X-ray photoemission measurements conducted on the same curved Rh(111) crystal show that prior to the ignition of the whole sample, which is marked by an abrupt CO desorption and CO₂ production, the B-steps are partially CO-depleted and oxidized, while the A-steps feature a CO-saturated situation. Therefore, such large asymmetry in the chemical composition of A- and B-steps points to be the reason of the early ignition of the B-steps observed by PLIF.

References

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