TEM Image Simulations of Structural Dynamics on CeO_2-supported Pt Catalysts

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Structural Dynamics & TEM Data

- **Heterogeneous catalysts** are critical in chemical conversion processes that are important in many societal areas
  - **Pt NP on CeO_2 system**, often used in clean energy materials and catalytic converters in cars
- Transmission electron microscopy (TEM) movies of **Pt NPs on CeO_2 in CO & O_2 gas** were studied to understand the catalytic structure and structural dynamics
- **Average structure**/ average atomic locations determined by combining frames from TEM movies to create a single ***summed image***

Can we determine what is happening to Pt in CO & O_2 and apply this method to other situations?

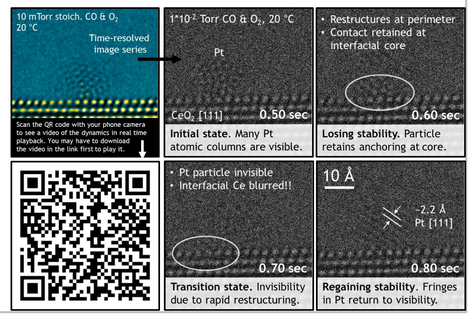
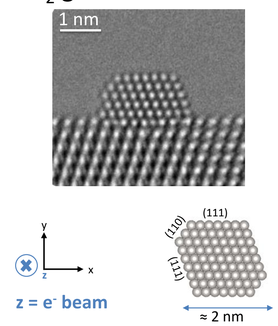


Figure credit to Joshua Vincent

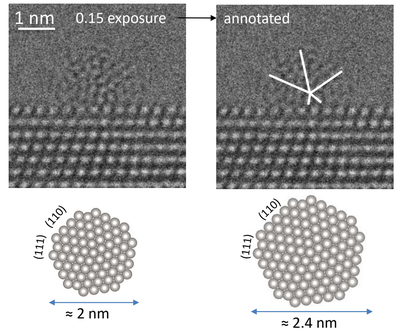
Single Crystal v Twinned Structures

N_2 gas environment



Pt is believed to have a single crystal structure in the experimental TEM images because of the visible repeated planes.

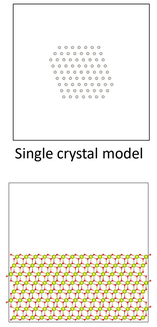
CO & O_2 gas environment



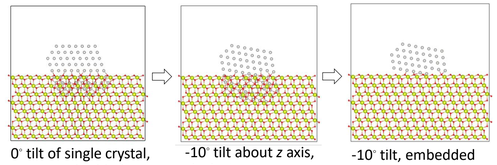
Some frames of Pt in CO & O_2 show **twin planes (annotated). Twinning** occurs when the planes on each side of the grain boundary mirror each other.

Tilt Manipulation of the Pt

A Python script rotated all Pt atoms, then deleted the embedded Pt atoms in the ceria. Embedded atoms removed after rotations to prevent an empty wedge at interface.

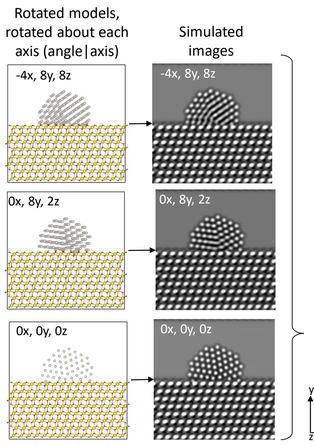


Pt NPs rotated about each principal axis **(x, y, z)** from **-10° to 10° in 2° increments**

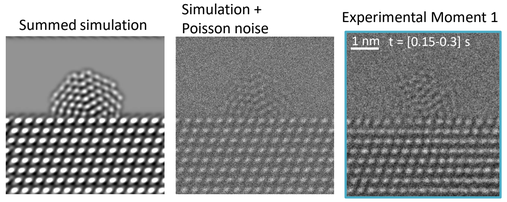


11^3 rotations * 3 models **≈ 4,000 possibilities**

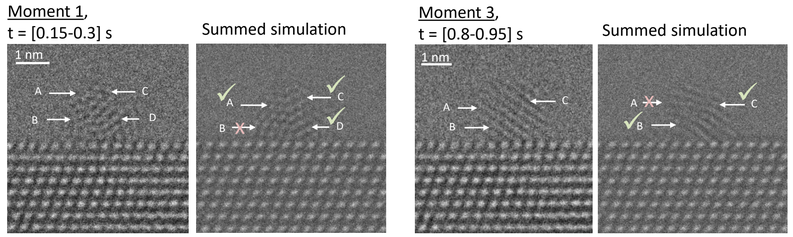
Creating Simulated Images from Experimental Data



- **Three moments** in experimental data chosen to simulate.
- The chosen rotations (far left) were used because each had one feature seen in the experimental data.
- **Twinned models created best match.**



Comparing Simulated Images to Experimental Data



- Moment 1: Best simulation, **dynamics occurring at the Pt and CeO_2 interface** resulting in different Point B.
- Moment 3: Worst simulation, **likely needs a different structure!**

Conclusions

Static models and TEM image simulations were **summed** to replicate contrast features in **experimental CO and O_2 gas TEM time series**

1. Directional contrast streaks from specific rotations provide **insight into structures**
2. **Twinned NP structures** provided the most accurate models and simulations
3. Unlikely to be rotating **more than 8 degrees**

Future work:

- Investigate quantifiable methods to determine quality of simulated match
- Use DFT/MD to use physics-informed atomistic dynamic models

Acknowledgements

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