Trimetallic nanoclusters for the production of green H_2 by WGS (water-gas shift)

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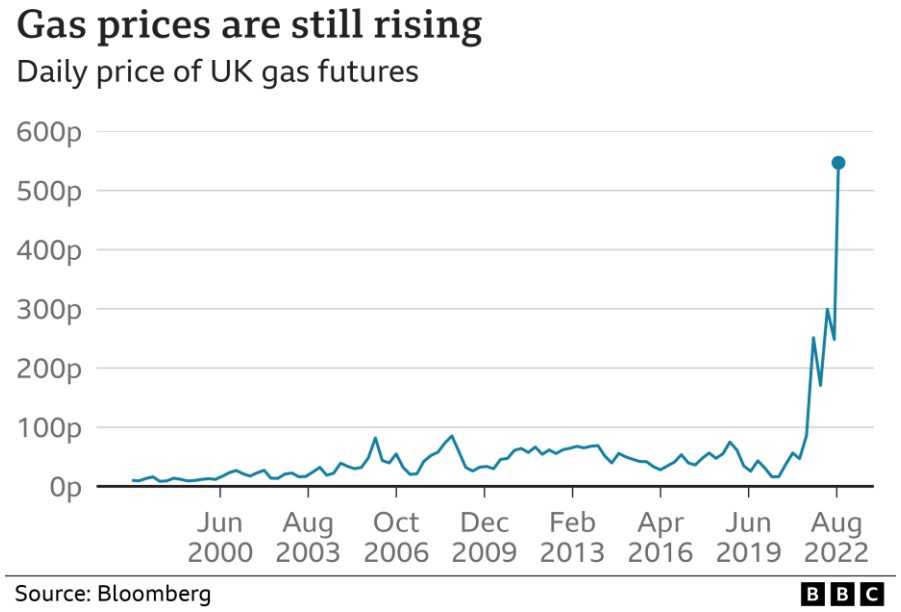
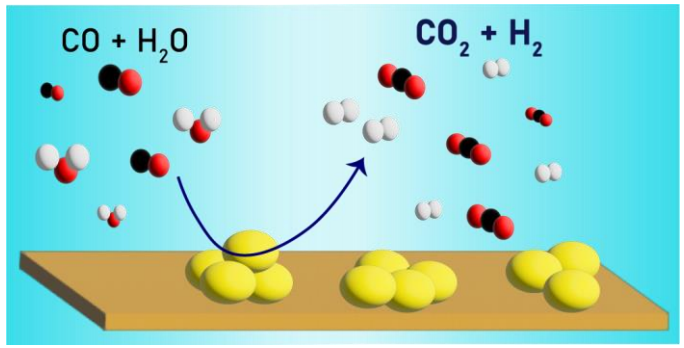
Water-gas Shift Reaction



Water-gas Shift Reaction



$$CO + H_2O \rightleftharpoons CO_2 + H_2$$

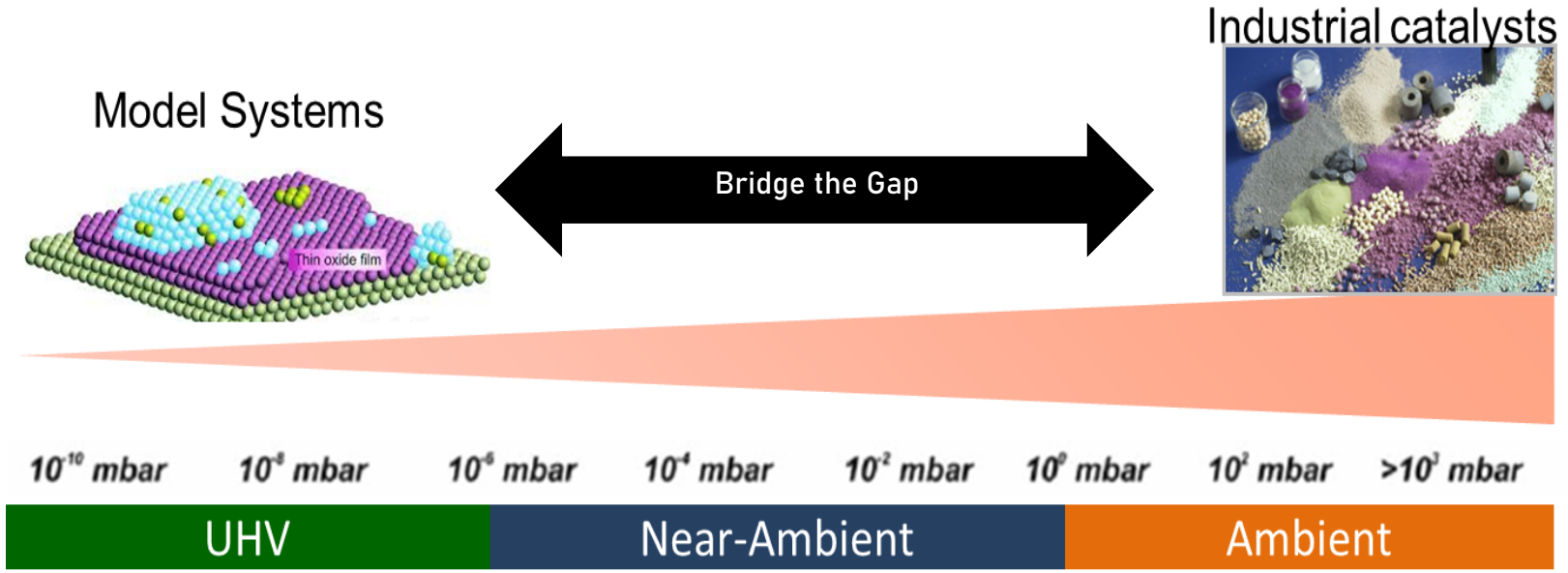
Catalyst

???

- Pure CO_2 is useful!!
  - ✓ Dry reforming of methane
    $CH_4 + CO_2 \rightarrow 2CO + 2H_2$
  - ✓ Food storage

  - ✓ Plant growth

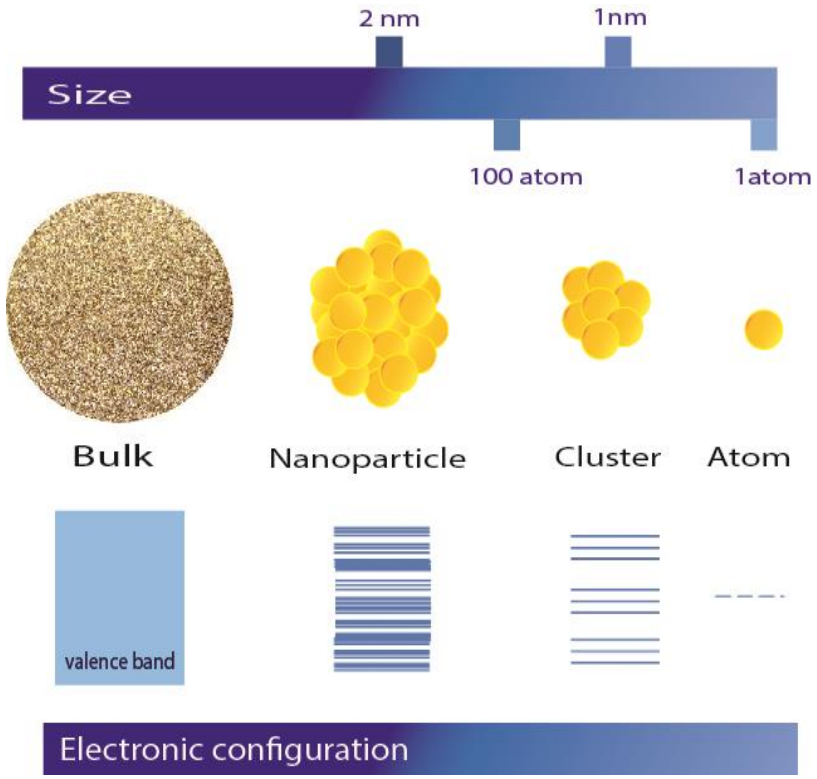
Industry and Research

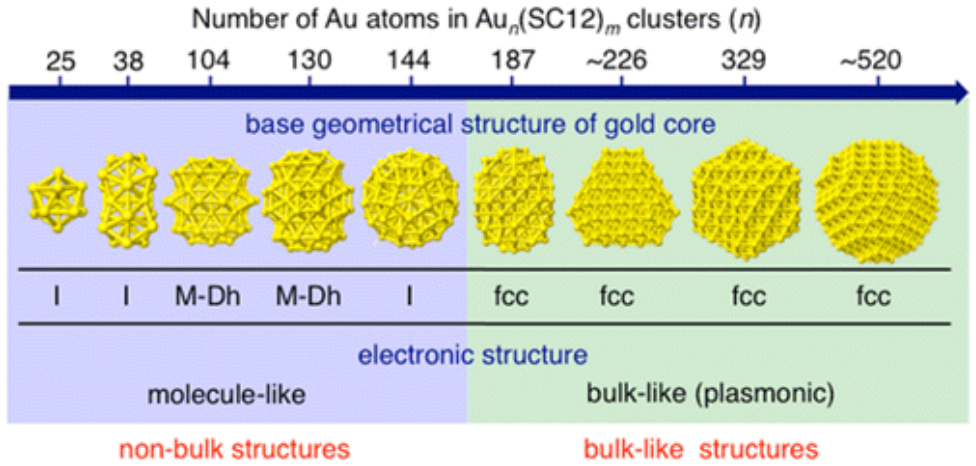


Needed:

- Well-defined material
- Working at ambient conditions

Nanoclusters



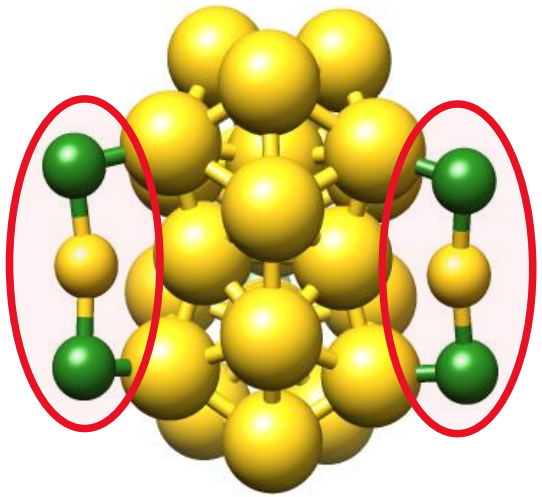


Y. Negishi *et al.* , *J. Am. Chem. Soc.*, **2015**, *137* (3), 1206

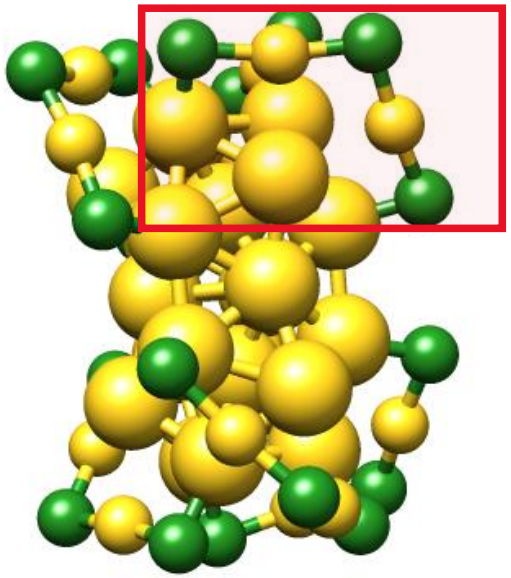
- ✓ Atomically defined
- ✓ Highly tuneable
- ✓ Unique structures

What are Nanoclusters?

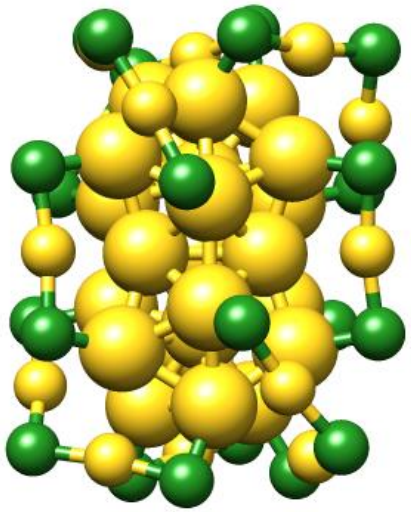
Short Staples



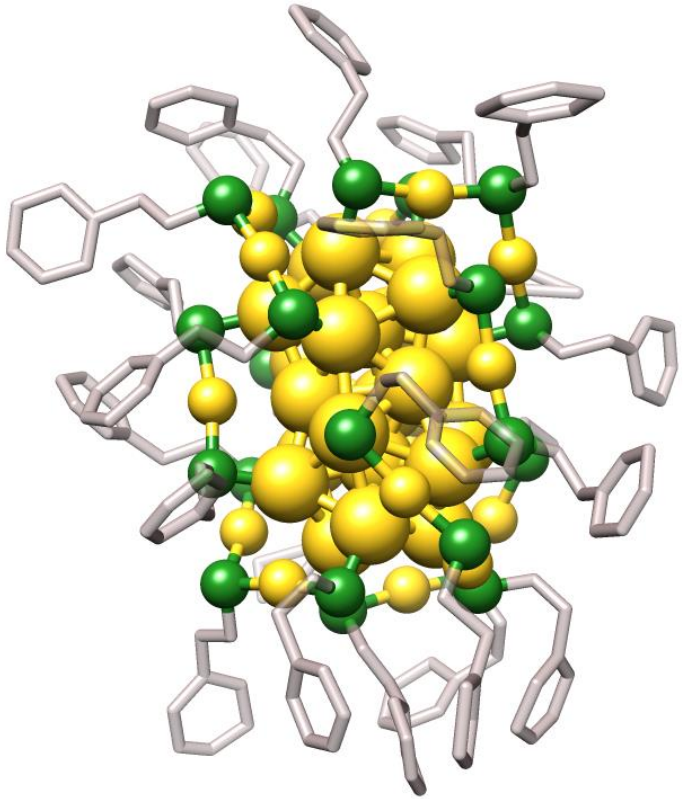
Long Staples



Full Ligand Shell



Full Structure



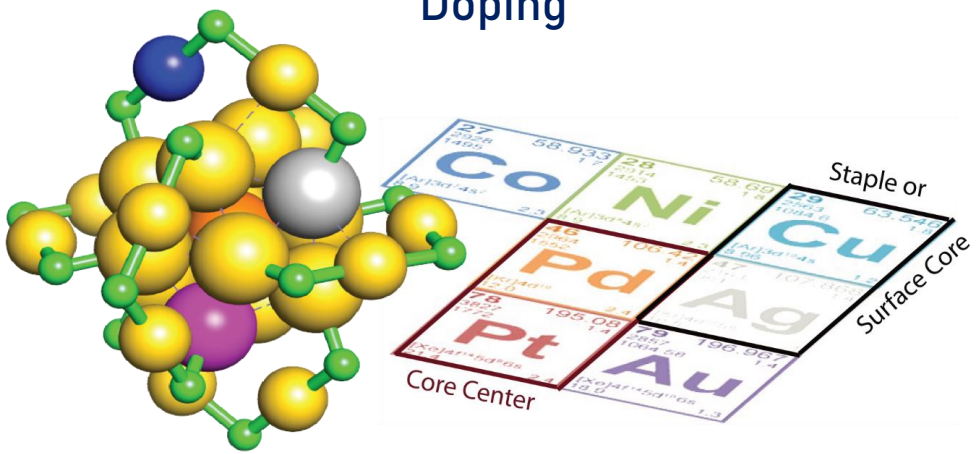
Au

S

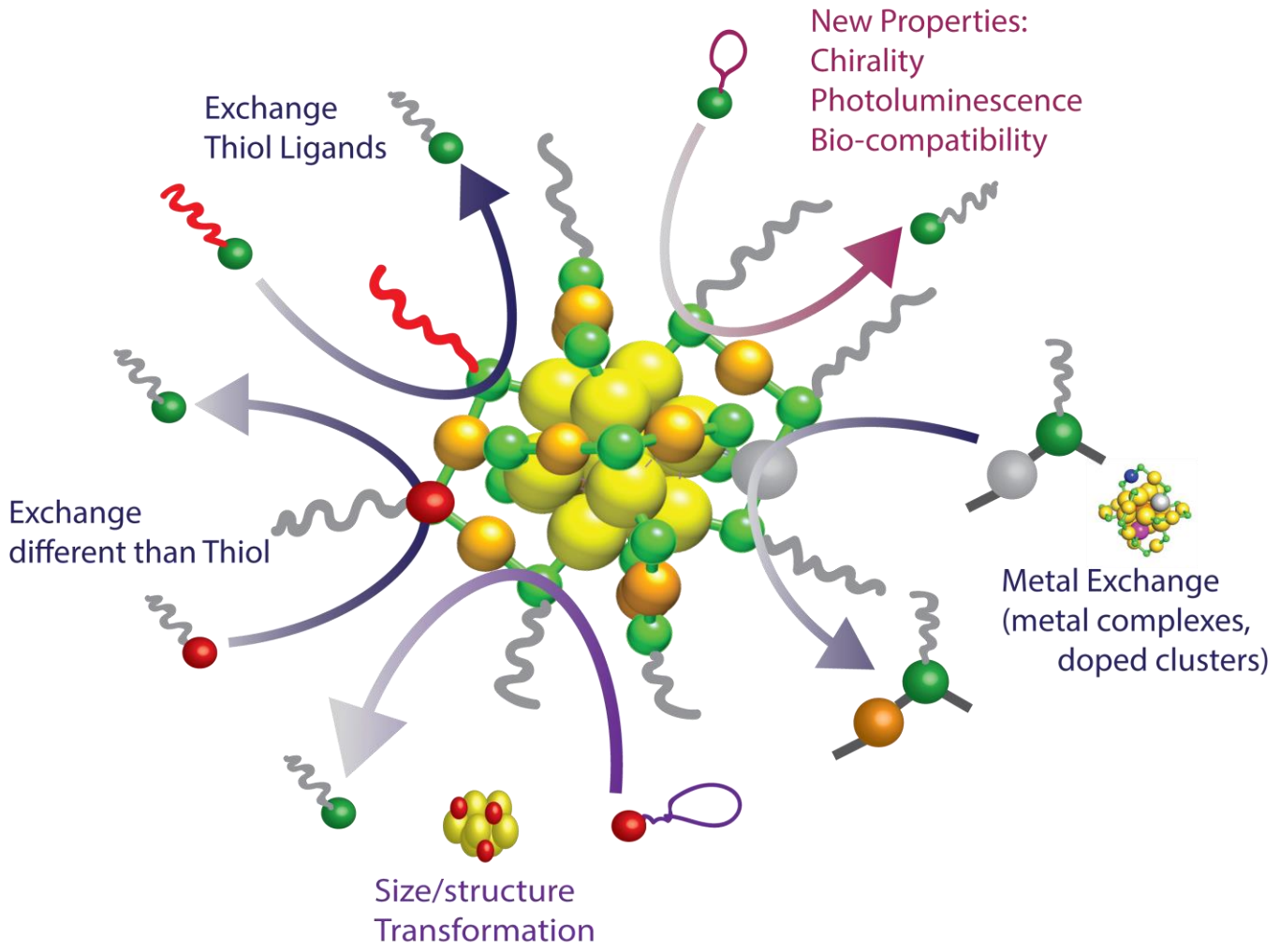
C

Tuneable Properties

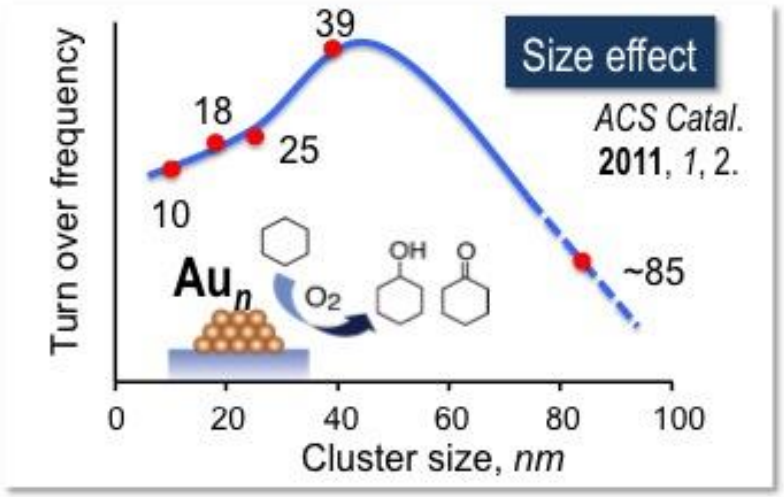
Doping

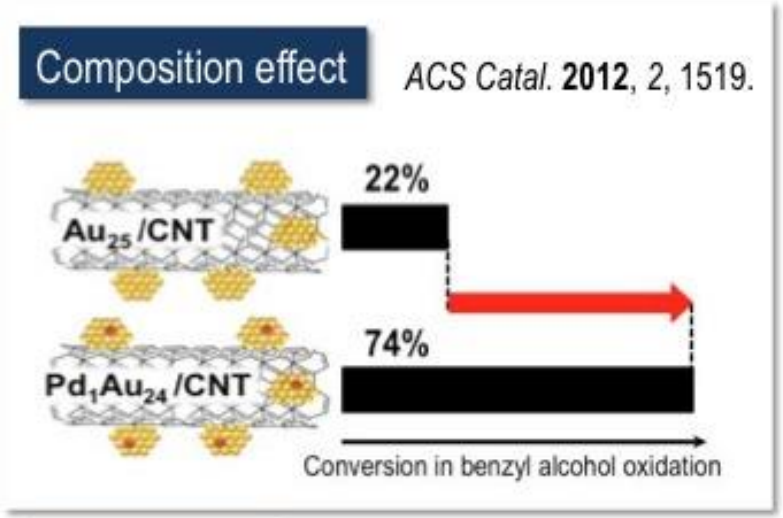


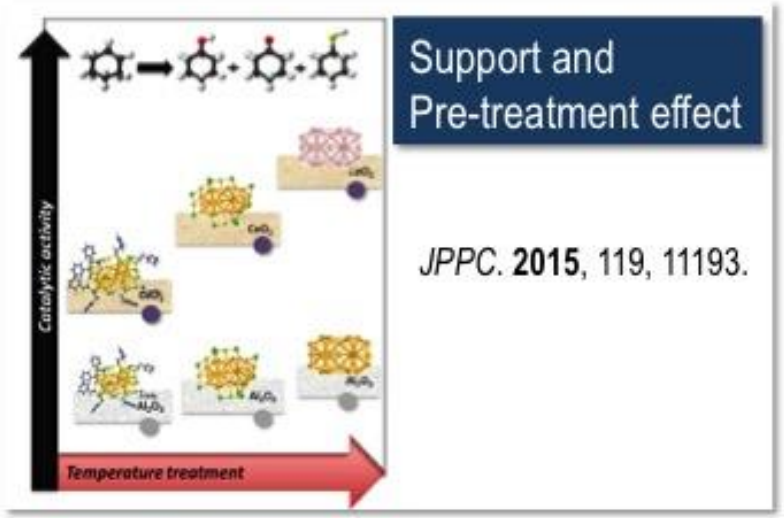
Ligand Exchange

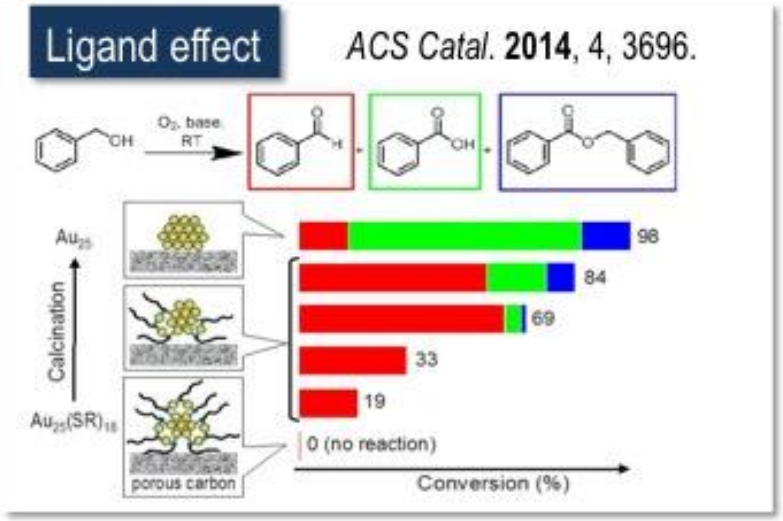


Influencing Catalytic Activity

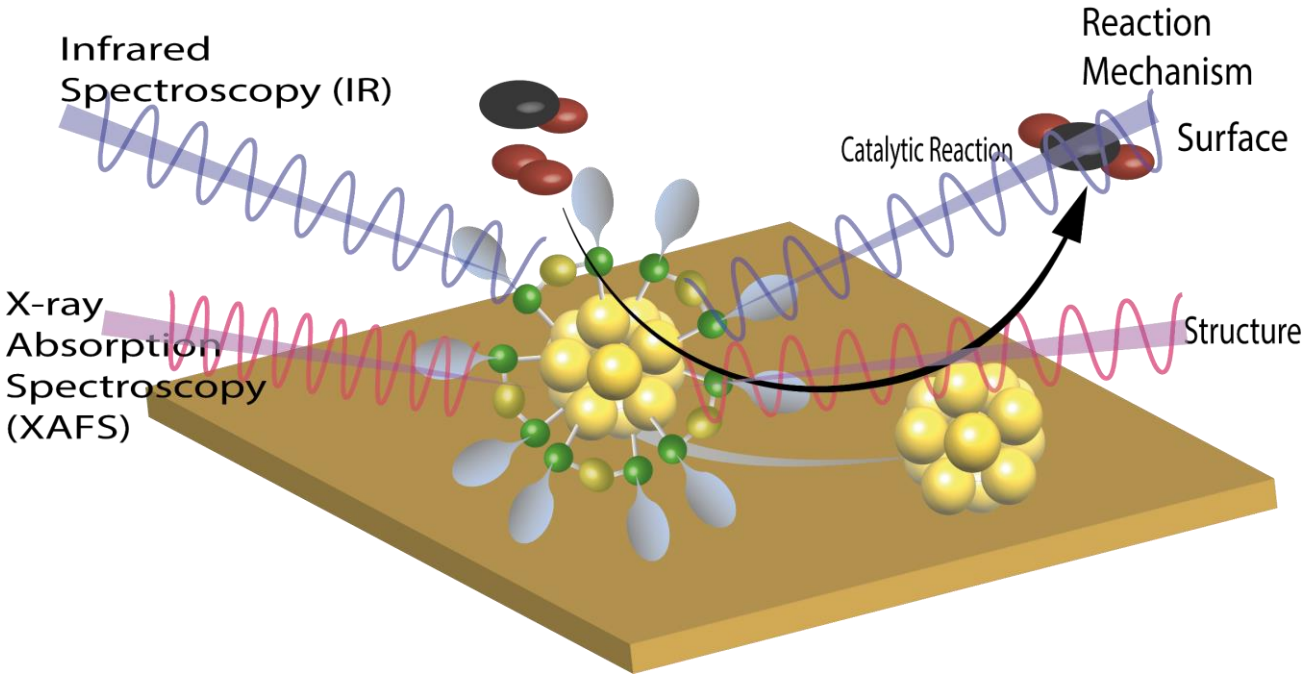


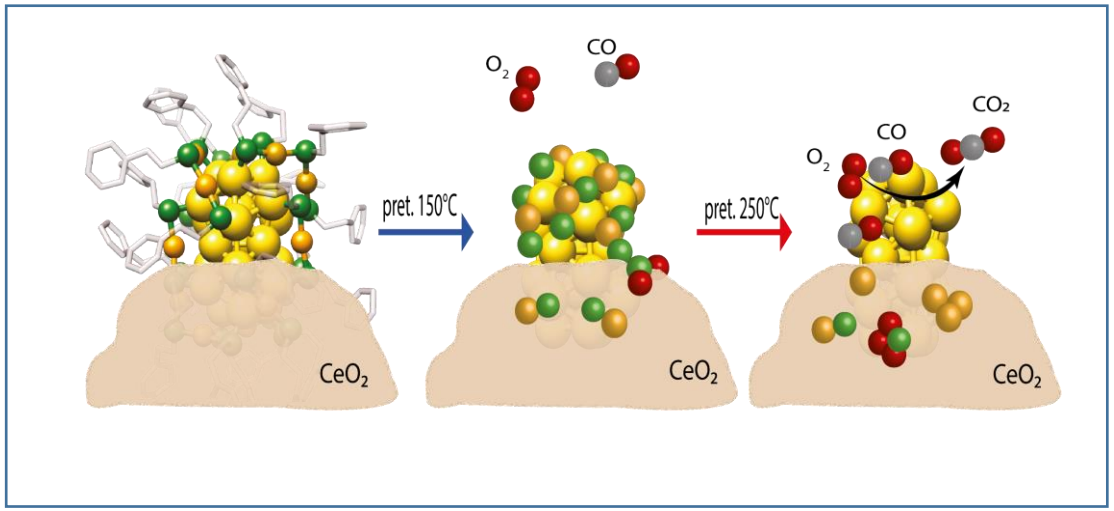




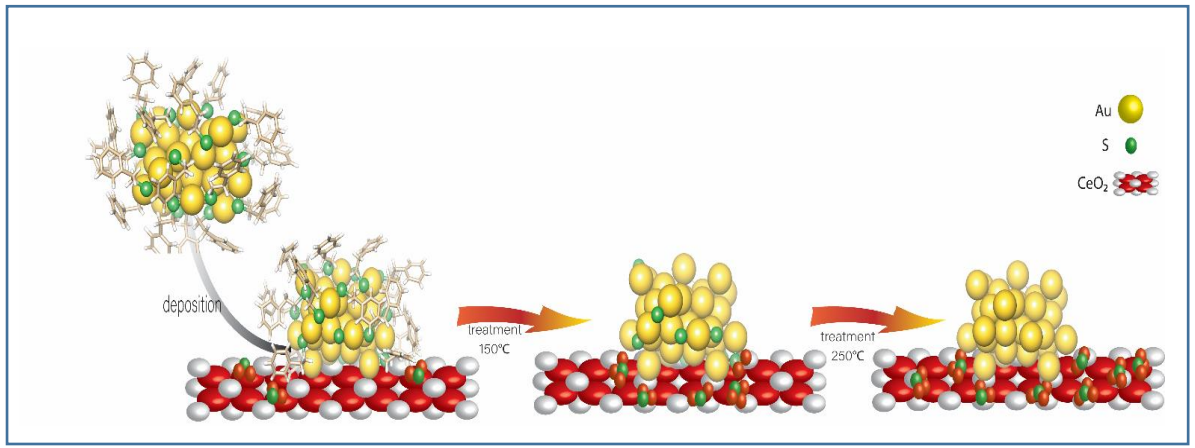


Nanoclusters in Heterogeneous Catalysis

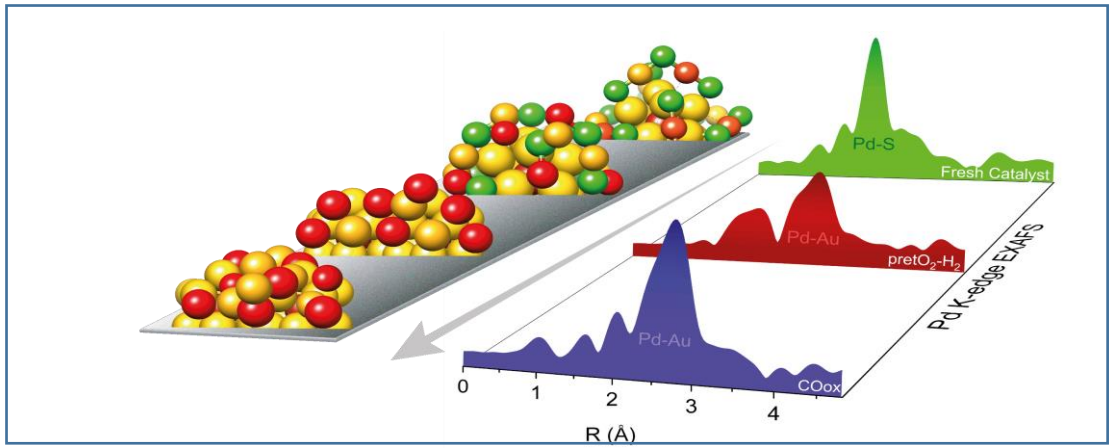




Pollitt et al. ACS Catalysis 2020



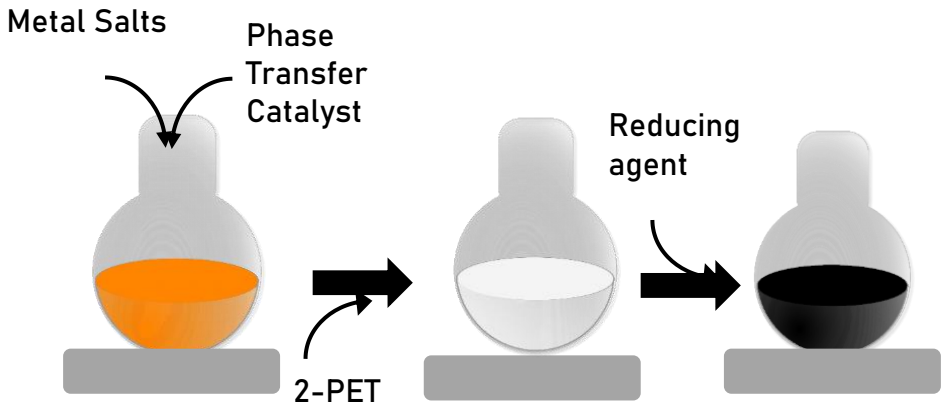
B.Zhang et al. ChemCatChem 2018, V.Truttmann et al. ChemCatChem 2022

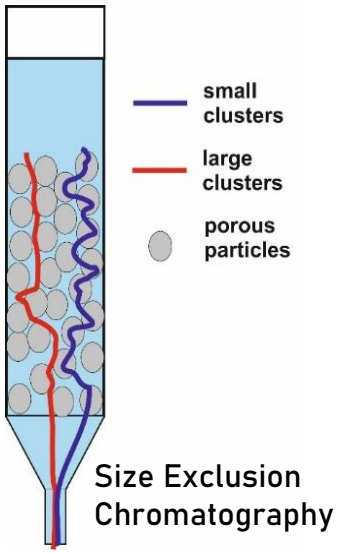


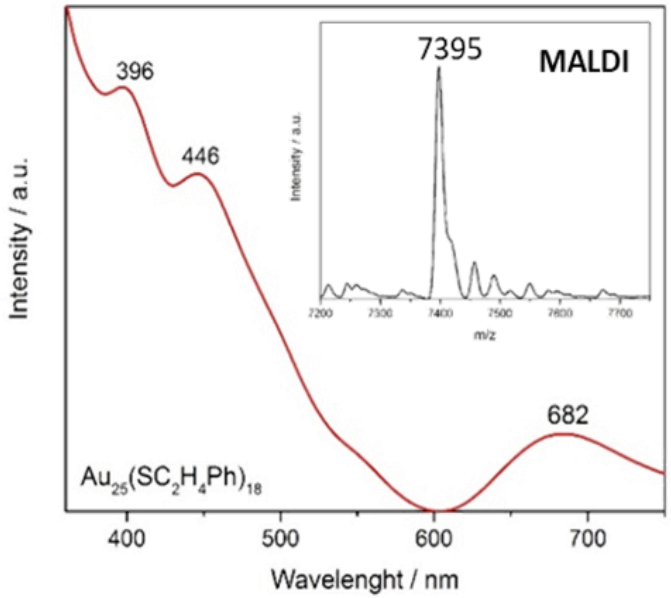
Garcia et al. JPCC 2020

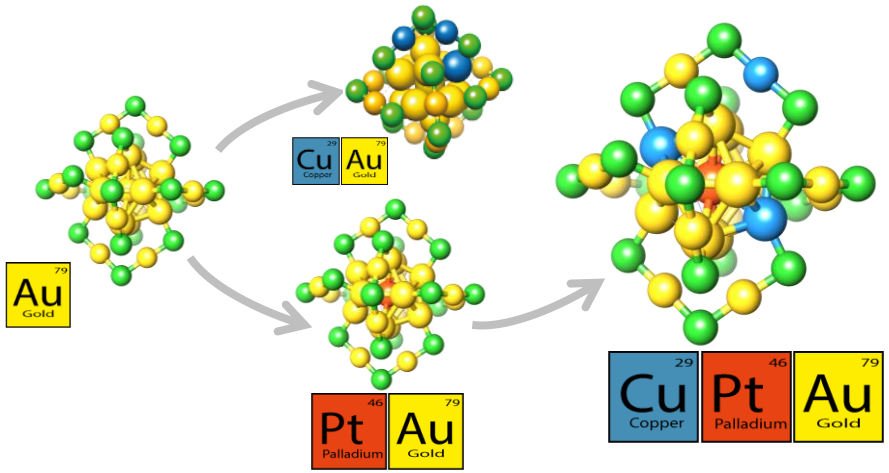
Nanocluster preparation

$Au_{25}(2\text{-PET})_{18}$ Synthesis





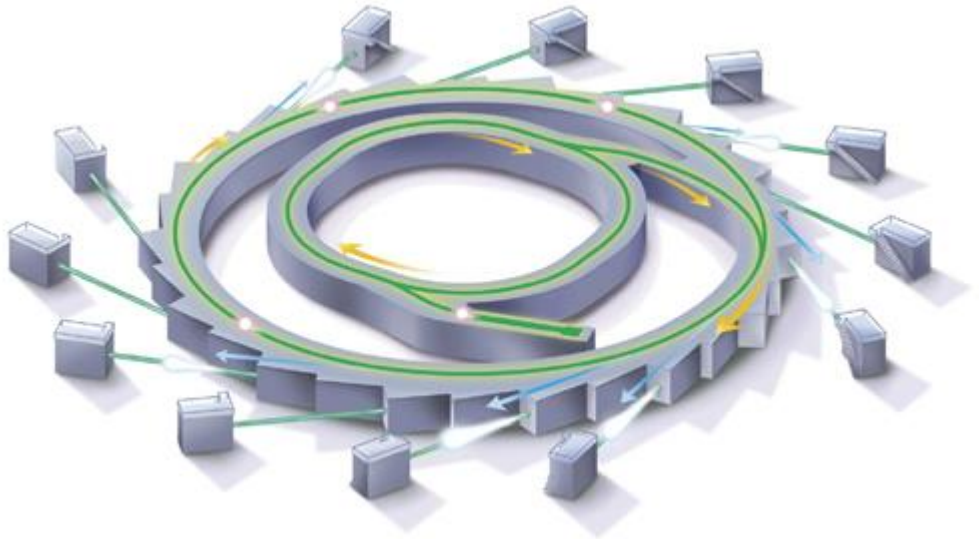




Mix with CeO_2

Supported Catalyst

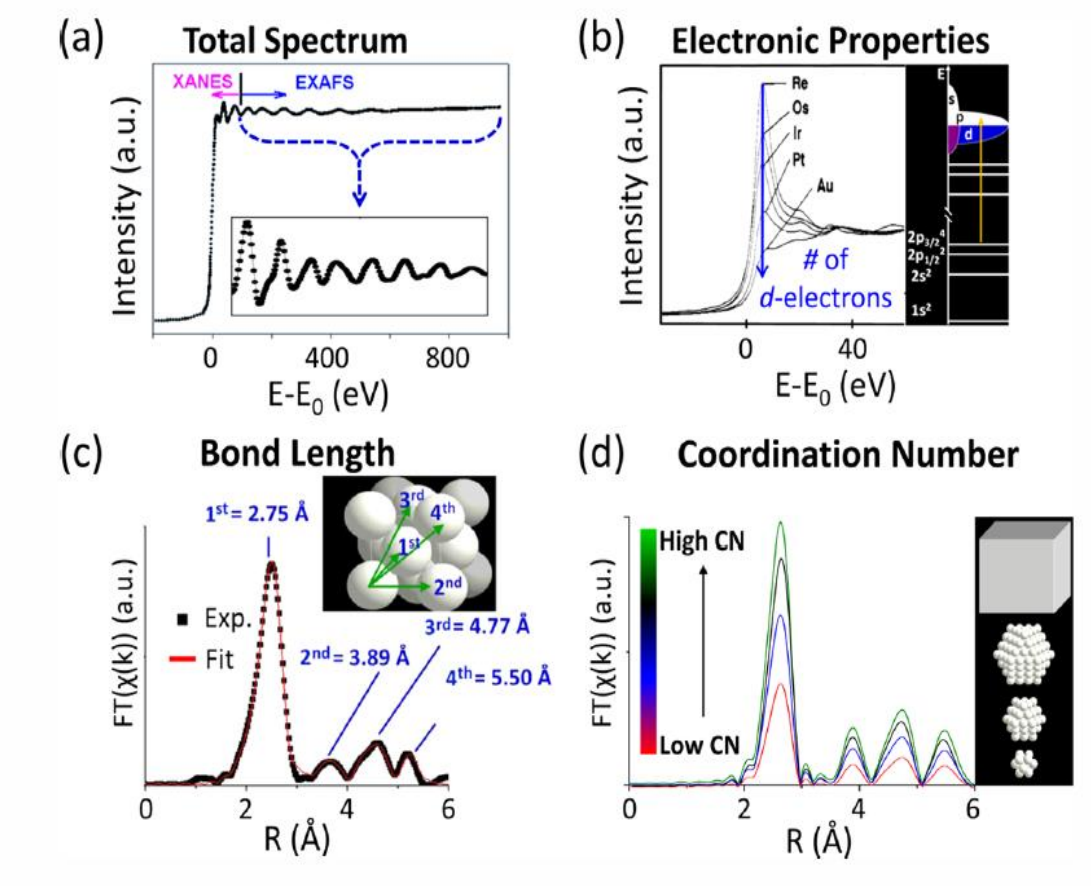
Synchrotron

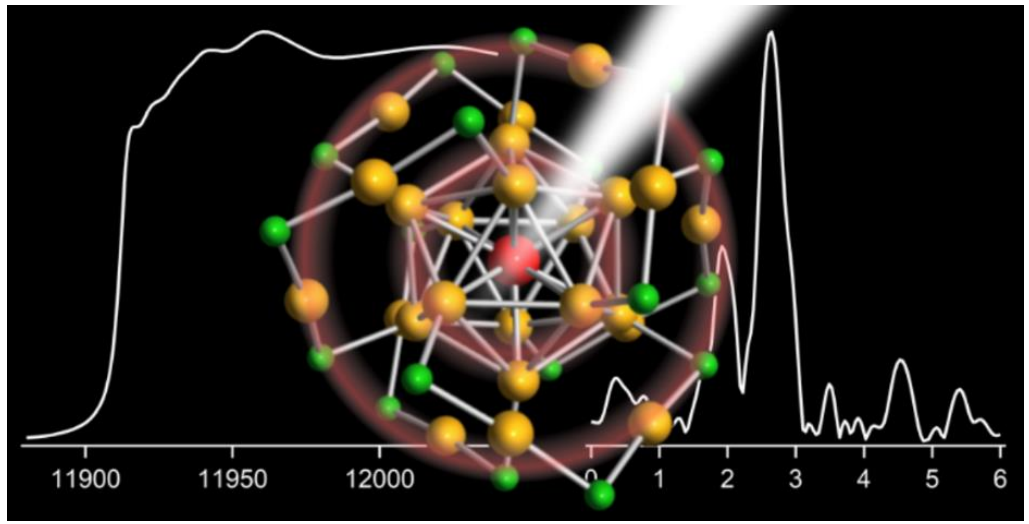


XAFS

X-Ray **A**bsorption **F**ine **S**tructure Spectroscopy

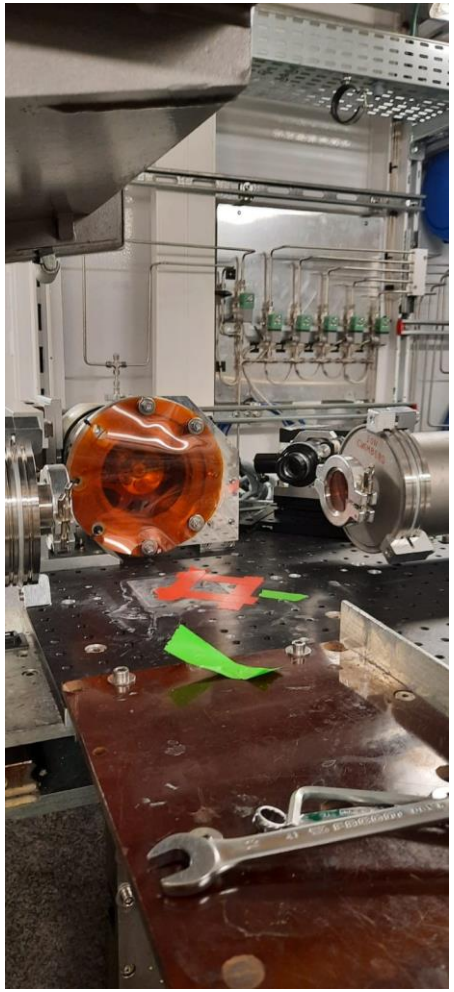
EXAFS & XANES





P. Zhang; *JPCC.,* (2014) ; S.Yamazoe & T.Tsukuda *Bull. Chem. Soc. Jpn.*, (2019)

Measurement Set-Up

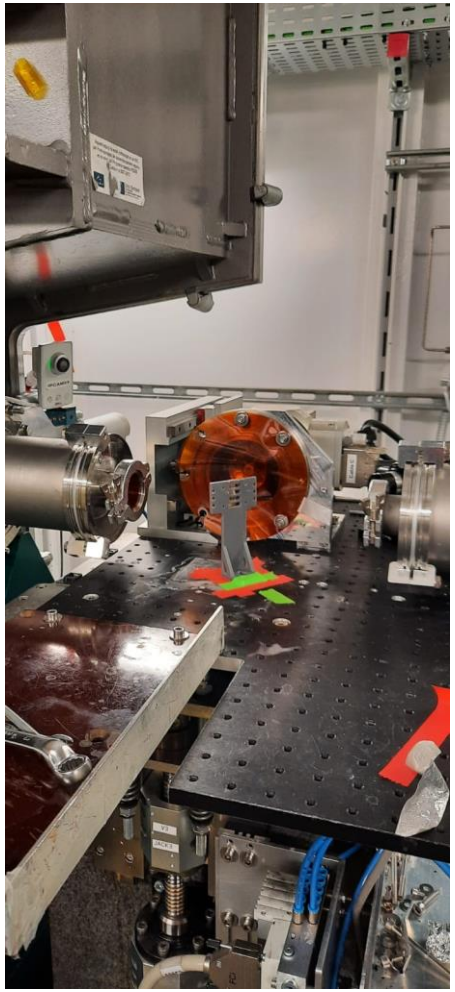


2 Types of Detection

- Transmission
- **Fluorescence**

2 Different Measurements

In-Situ

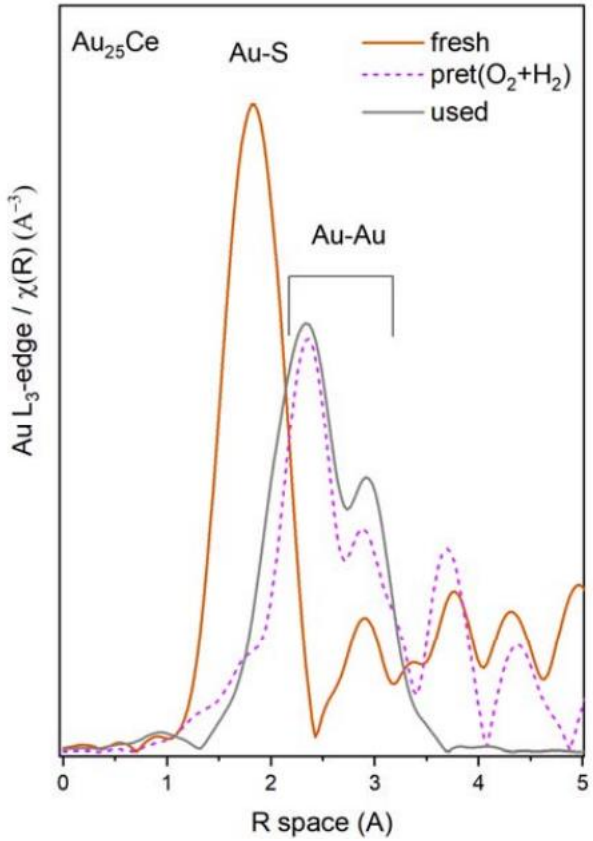


Static

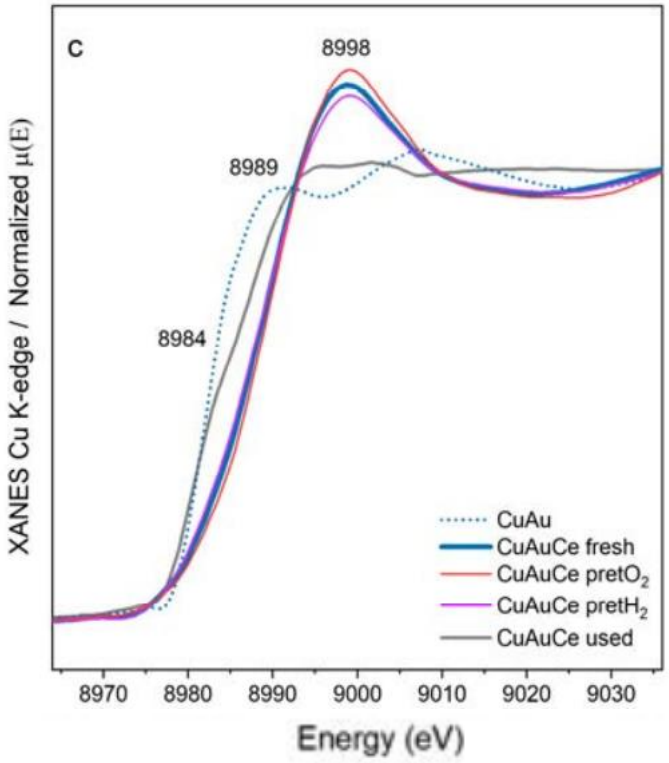
Structure Dynamics: *operando XAFS spectroscopy*

Au L_3-edge

EXAFS



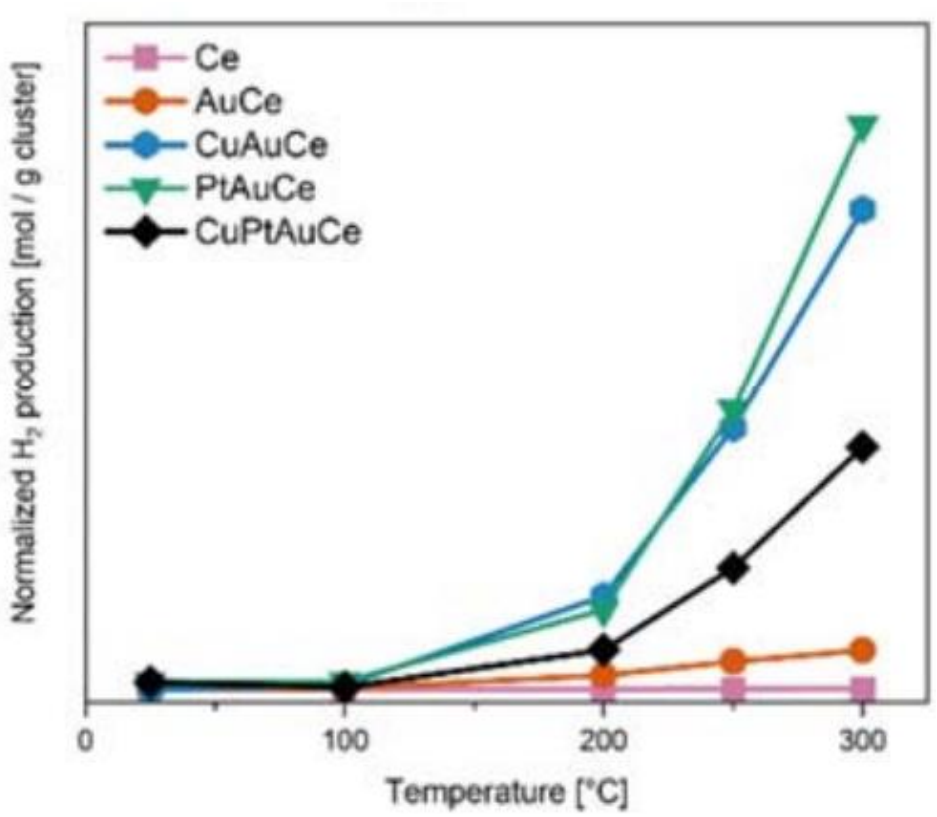
XANES



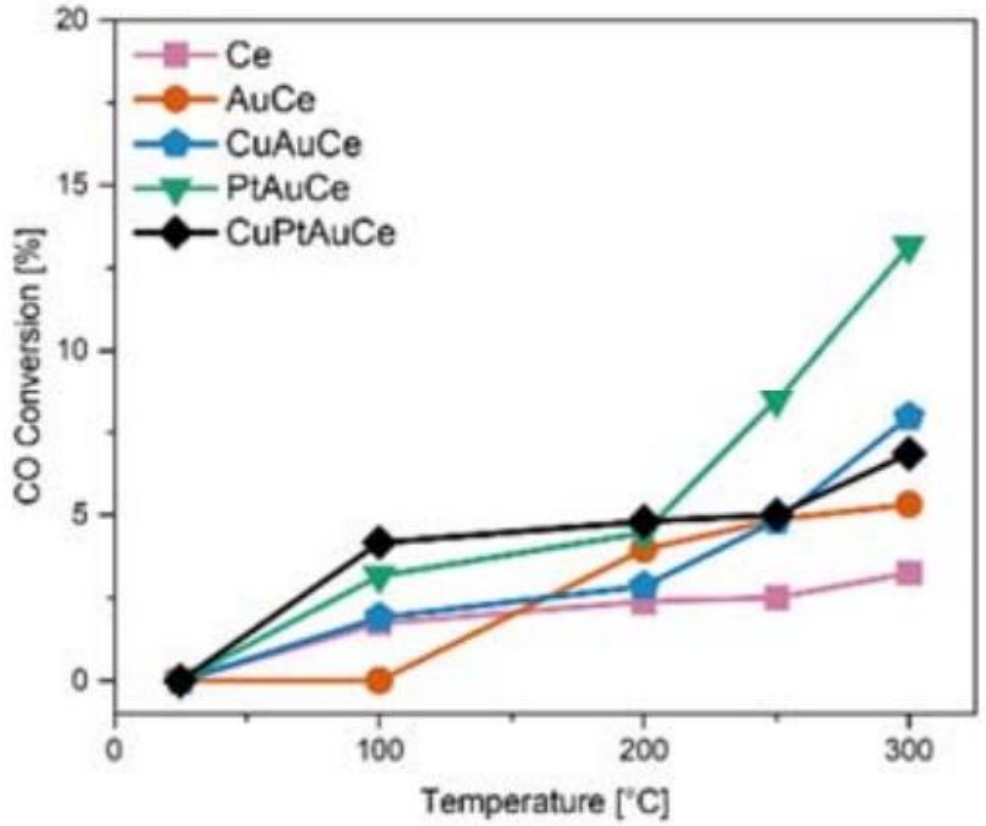
- Staples are dismounted after pre-treatment
- Au oxidation state changes

Catalytic Activity Studies

Formation of H_2



Conversion of CO



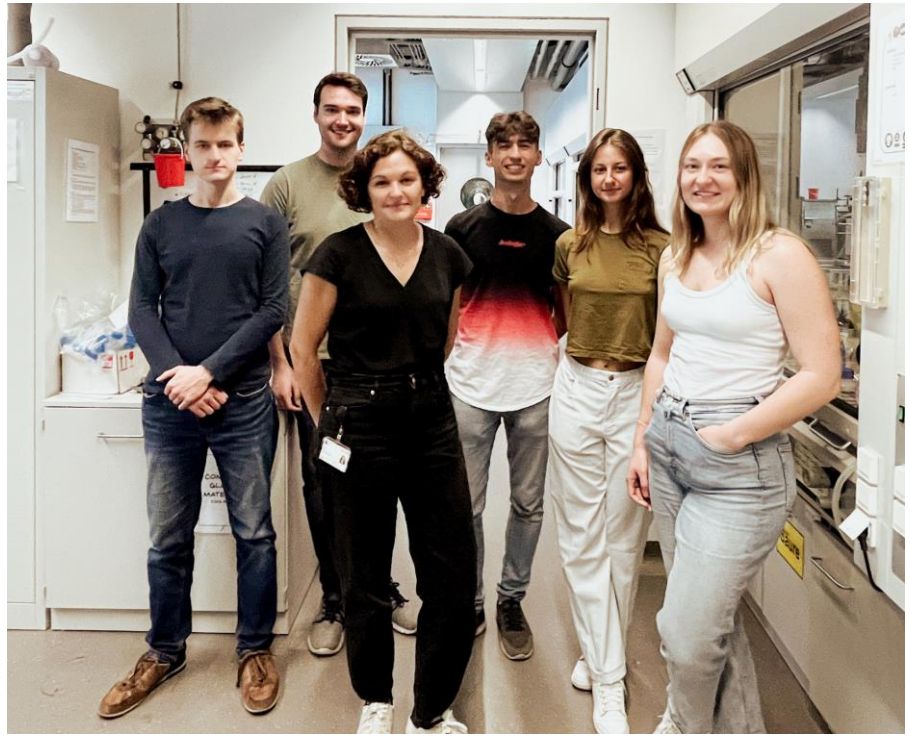
- Doping increases catalytic activity -> synergistic effect

Conclusions & Outlook

- ✓ Atomically defined systems allow to understand reaction at atomic level
- ✓ Doping increases activity
- ✓ Bimetallic clusters more stable
- ✓ Migration of dopants
- ✓ Dynamic structure of the clusters

- ➢ Check for effect of support
- ➢ Try other cluster sizes
- ➢ Check effect of the ligand

Acknowledgements



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