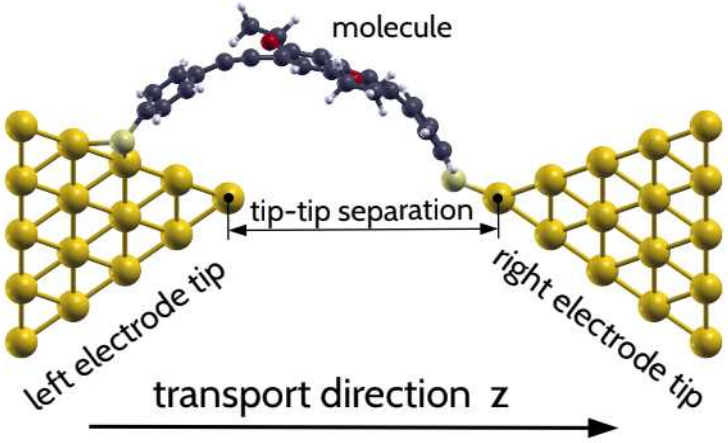

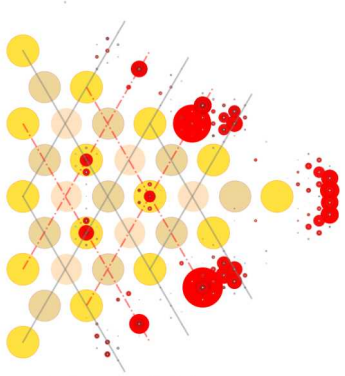

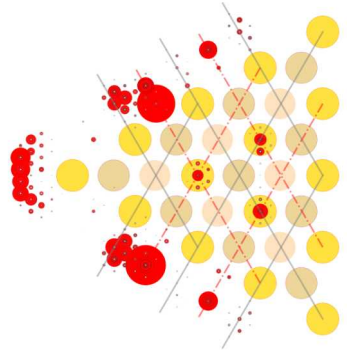

Evolution of single-level-model parameters in the mechanically controllable break junctions

Lokamani, F. Kilibarda, F. Günther, J. Kelling, A. Strobel, P. Zahn, G. Juckeland,

K. Gothelf, E. Sheer, S. Gemming and A. Erbe

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DRESDEN ROSSENDORF

Introduction

Mechanically controlled Break-Junctions

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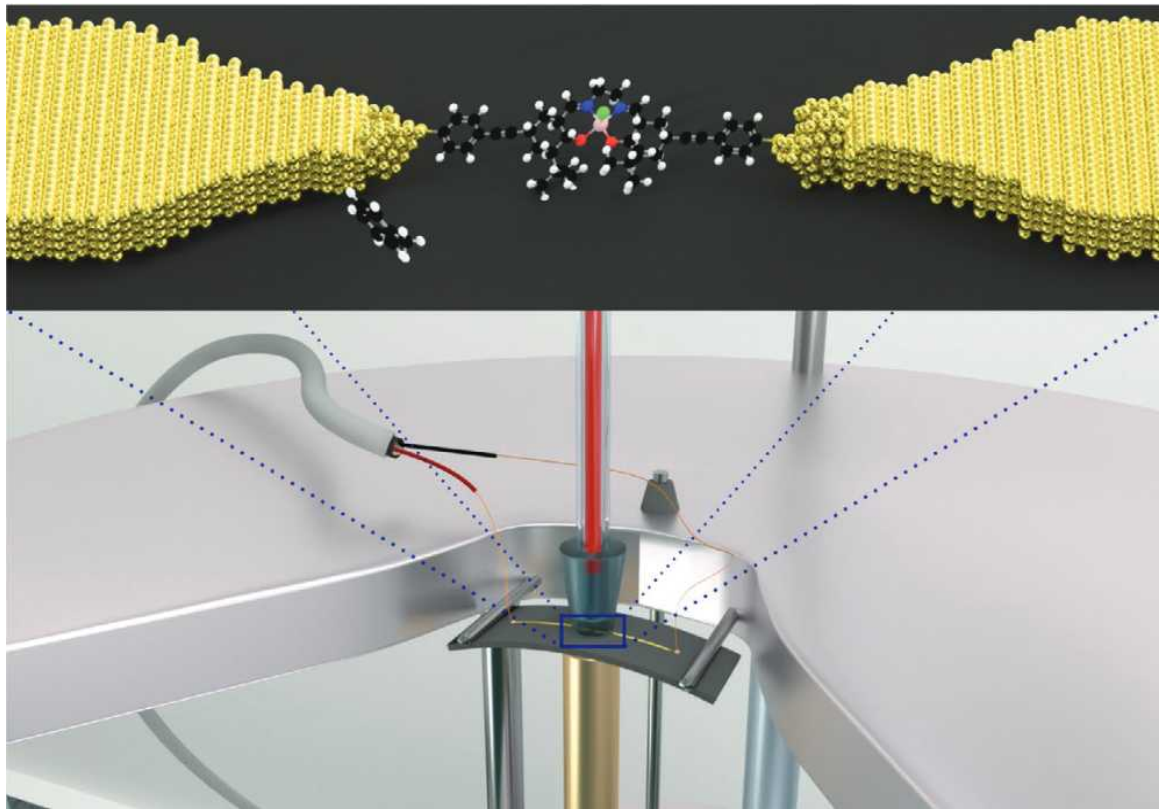
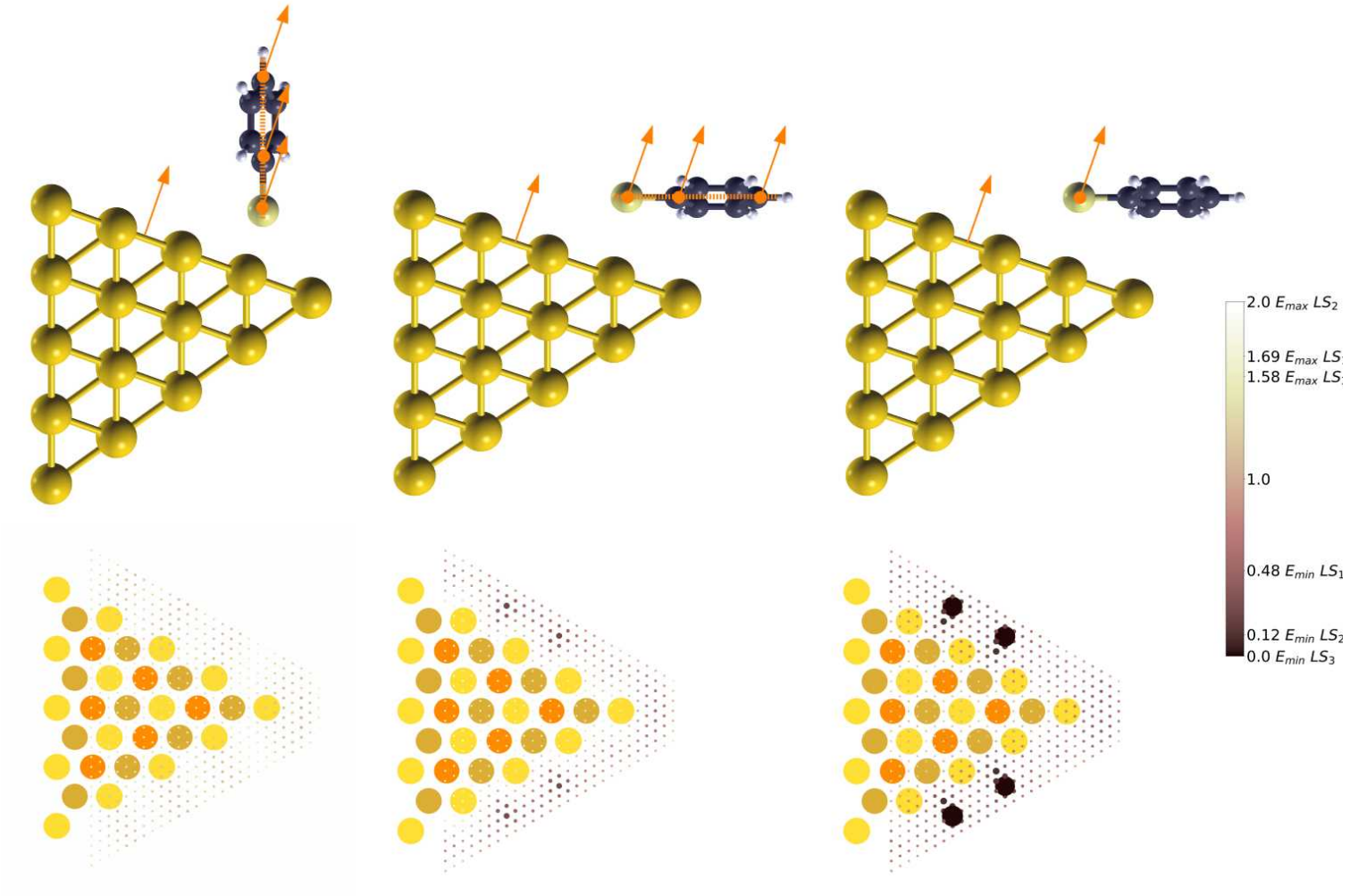
Mechanically controlled Break-Junctions

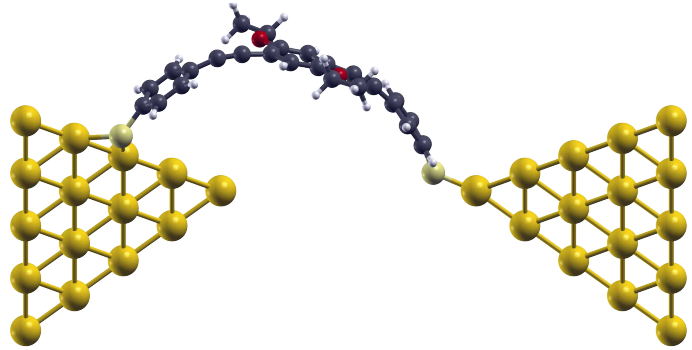


- Au-Au nanoconstriction fabricated using lithographic techniques
- Main principle: Controllably break a metallic wire to form atomistically shaped electrodes
- Probe molecules trapped btw. the sub-nanometer gap
- Attenuation factor?

Source: Phd-thesis, Filip Kilibarda, Uni Konstanz

Anchoring Position | Anchoring Angle | Curvature





- Thiophenyl
  – vertical, horizontal orientation
  – unrestricted
- Anchoring S-Atom moves along the facet normal

- Energy landscape
  – recurrent features and energy minimum at edges
  – tip region energectically unfavorable

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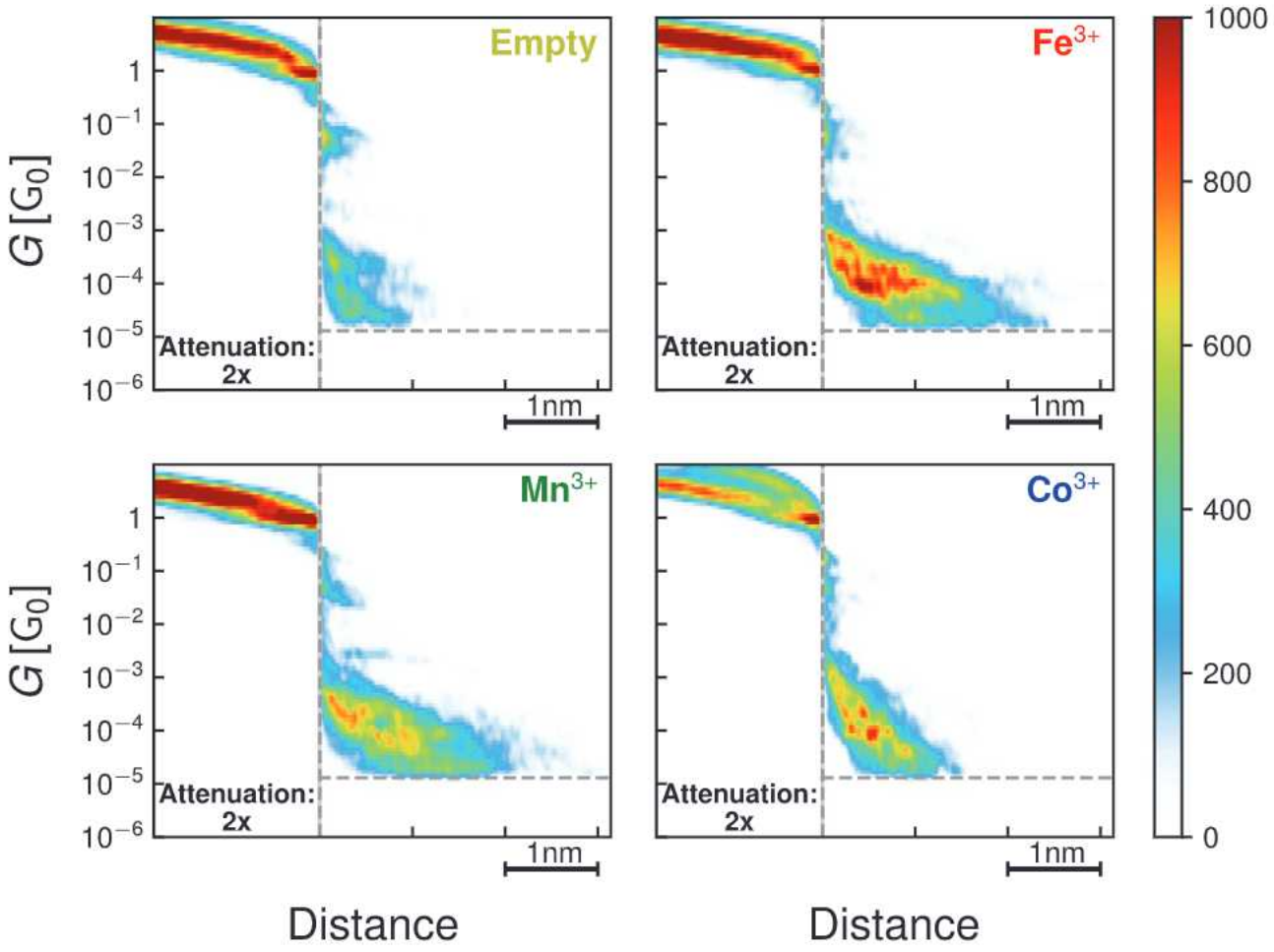
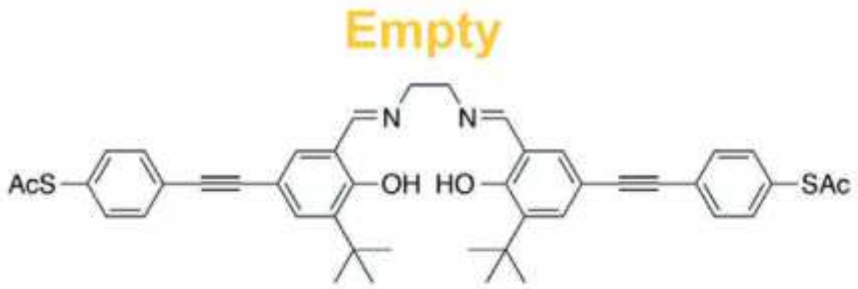
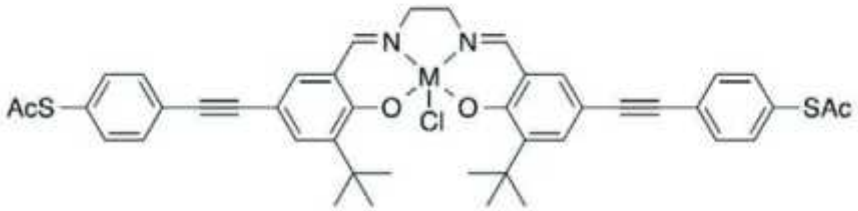
Statistical evaluation of SLM-parameters | Conductance histograms and plateaus





$M = Fe^{3+}$, $M = Mn^{3+}$, $M = Co^{3+}$



- Metallic bridges ▸ flat conductance plateaus | molecules ▸ reclining conductance plateaus
- Metallic bridges ▸ linear IV | molecules ▸ S-shaped IV ▸ *Fit single level model* [ε_0 | Γ]
- Typically multiple measurements are performed (statistical significance)

Source: Phd-thesis, Filip Kilibarda, Uni Konstanz

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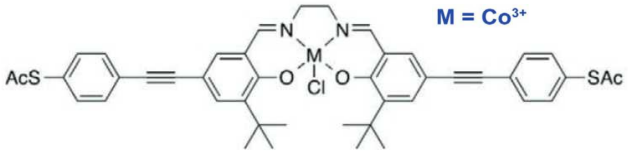
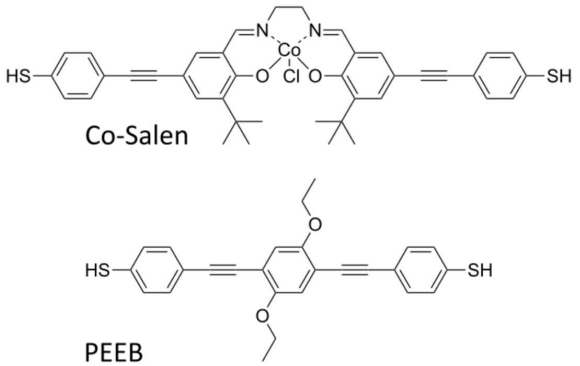
Stretch Evolution of SLM parameters in single opening curves



- Falling and rising trend with recurring maxima with tip-tip separation
- Time scales of measurement | mean of thermodynamically accessible junction geometries

Main Focus : Stretch Evolution of Γ



- Falling and rising trend with recurring maxima with tip-tip separation
- Time scales of measurement | mean of thermodynamically accessible junction geometries

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Setup - Dynamic Simulation Approach

Transport setup | Single Level Model | Energy Landscape | Descriptors

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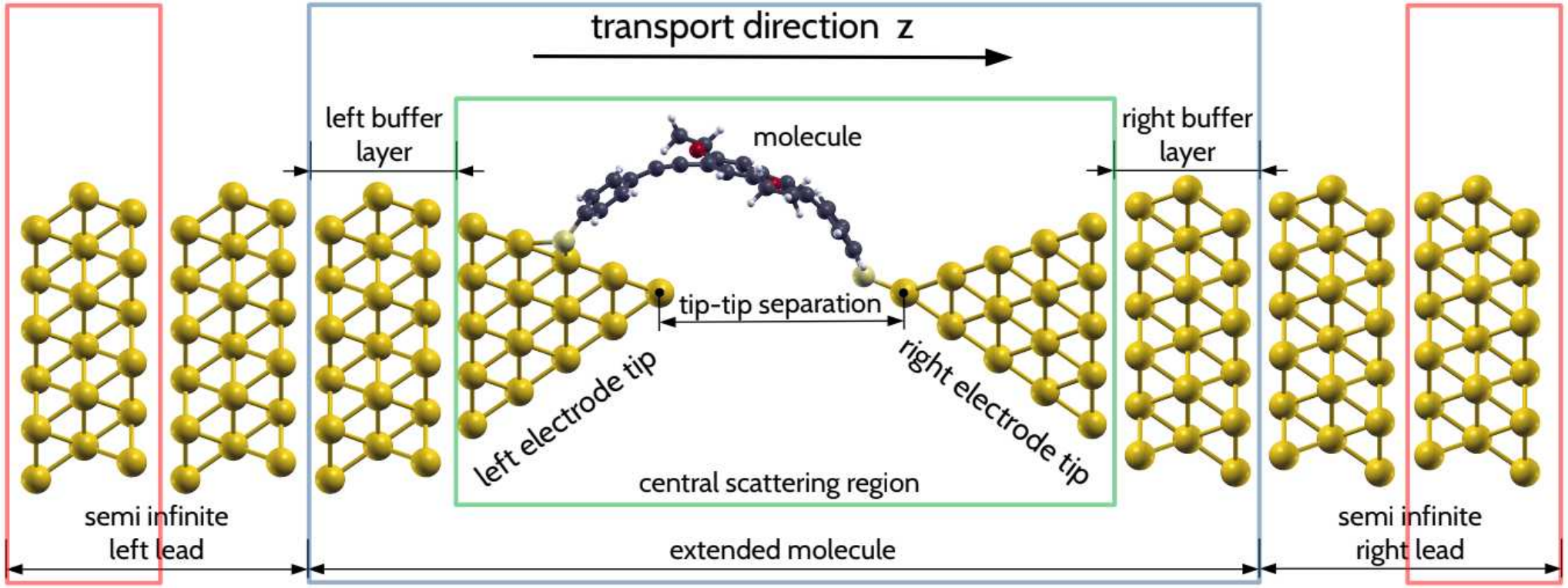
Transport Setup | evaluating zero-bias Transmission T(E) | Single Level Model

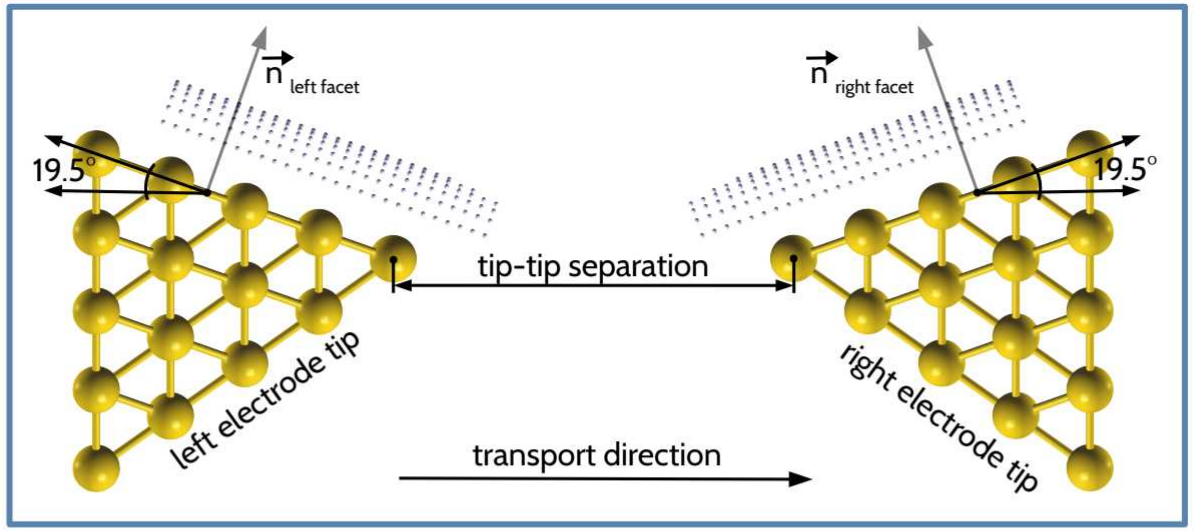


- electrode tips – equilateral triangular pyramids (baselength 4x Au-Au)
- buffer/semi-infinite layers (3 Au Layer 5x5)
- local-coordination-environment ▸ face-centered-cubic
- tip-tip separation [11.54Å, 27.24Å] in steps of 0.785Å
- Concurrent processes → Dynamic Simulation Approach

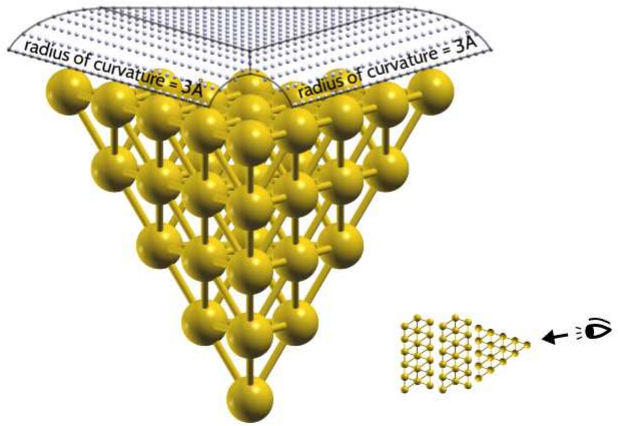
Single Level Model

$$T(E) = \frac{4\Gamma_{max}\Gamma_{min}}{(E-\epsilon_0)^2 + (\Gamma_{max}+\Gamma_{min})^2}$$

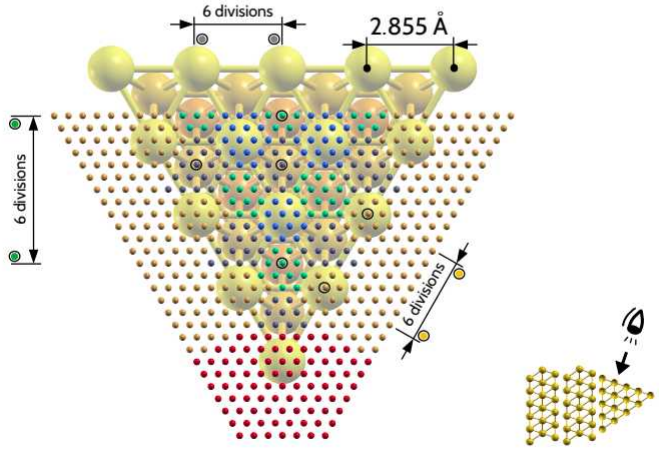
Anchoring positions (AP) | Diffusion Process



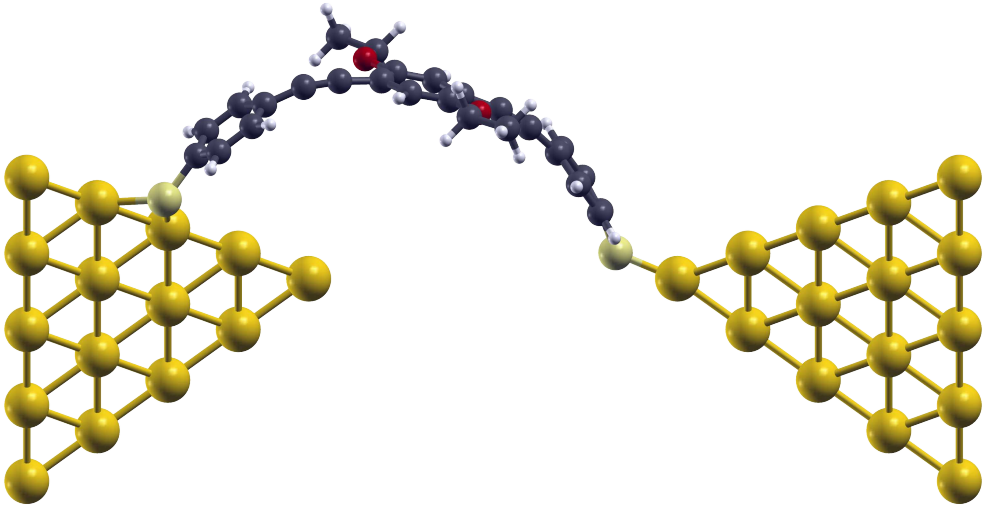
docking grids on left-/right facets



curvature of docking grid about the facet edges



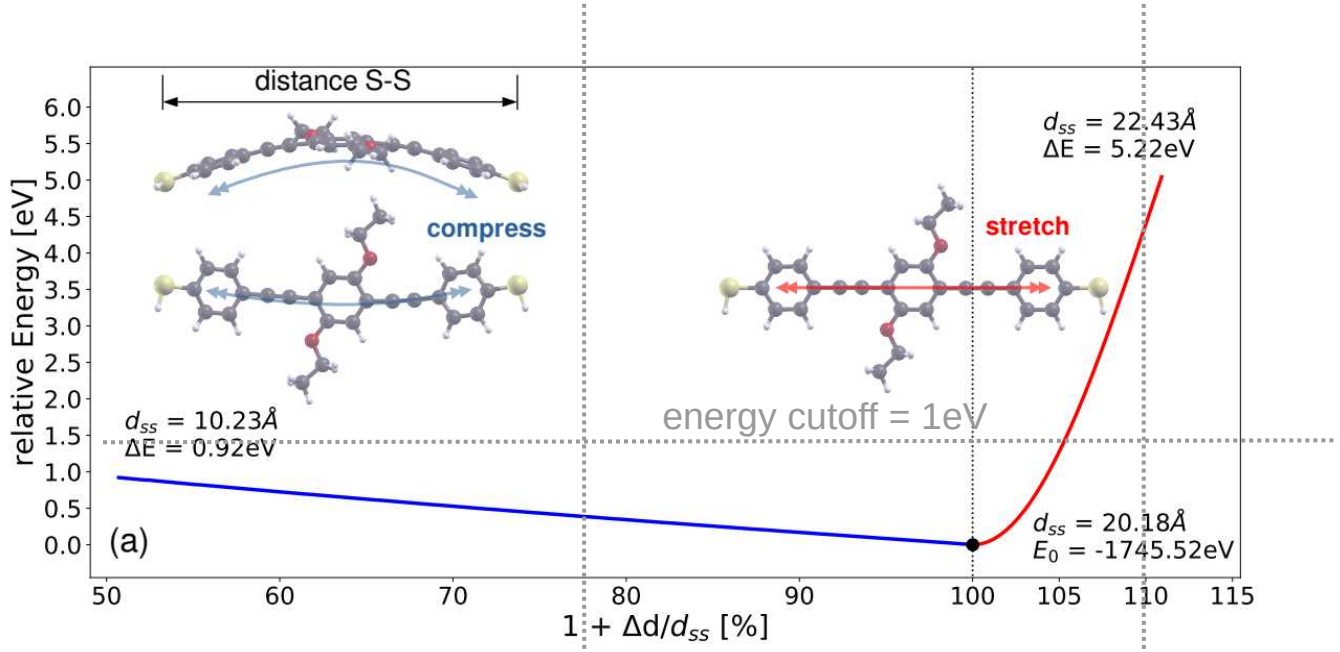
grid division of docking positions on the facet edges

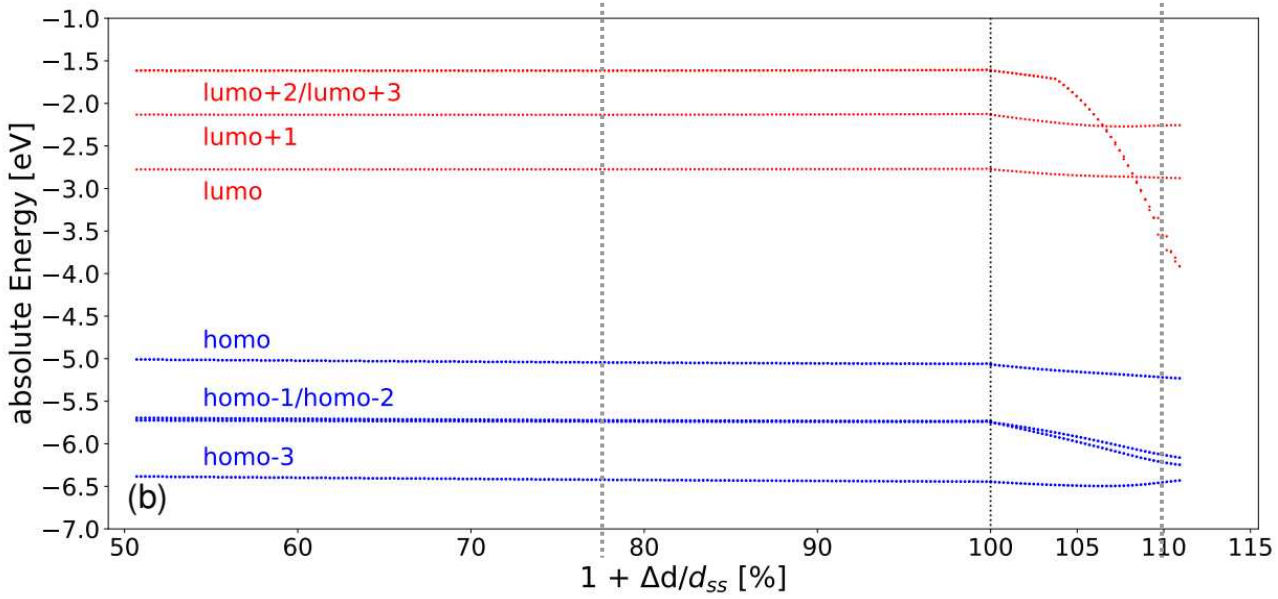


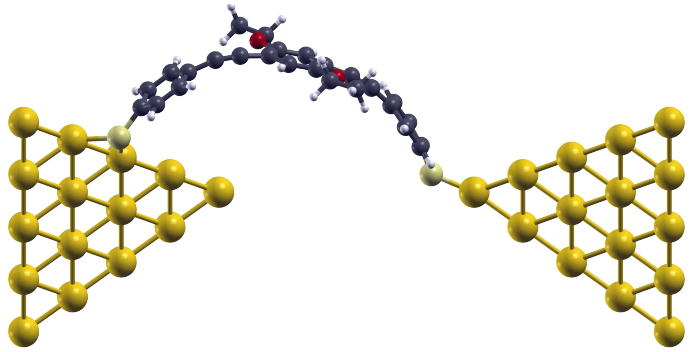
- Is the molecule with thiol anchoring groups at equlibrium for all tip-tip separations?
- Can the anchoring S-atoms slide freely on the gold facets?
- How much can the molecule be bend or streched? Alteration of electronic states?

- Grid spacing accomodates midpoints between high symmetry sites
- Additional grid points chosen protruding over the facet-edges

Molecular Deformation



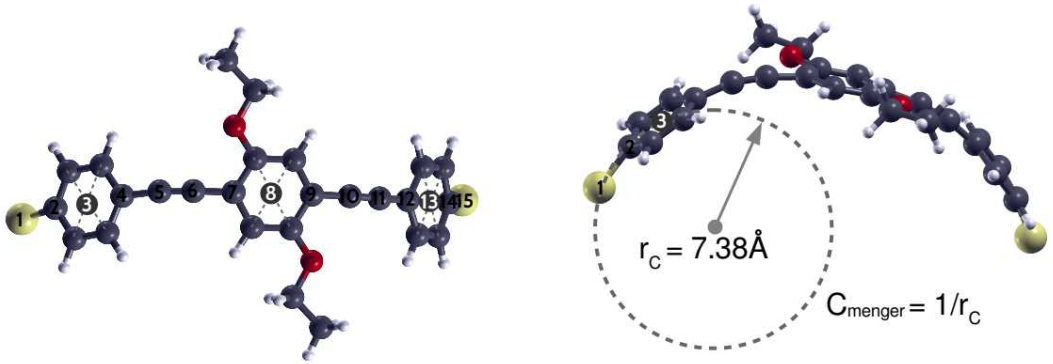


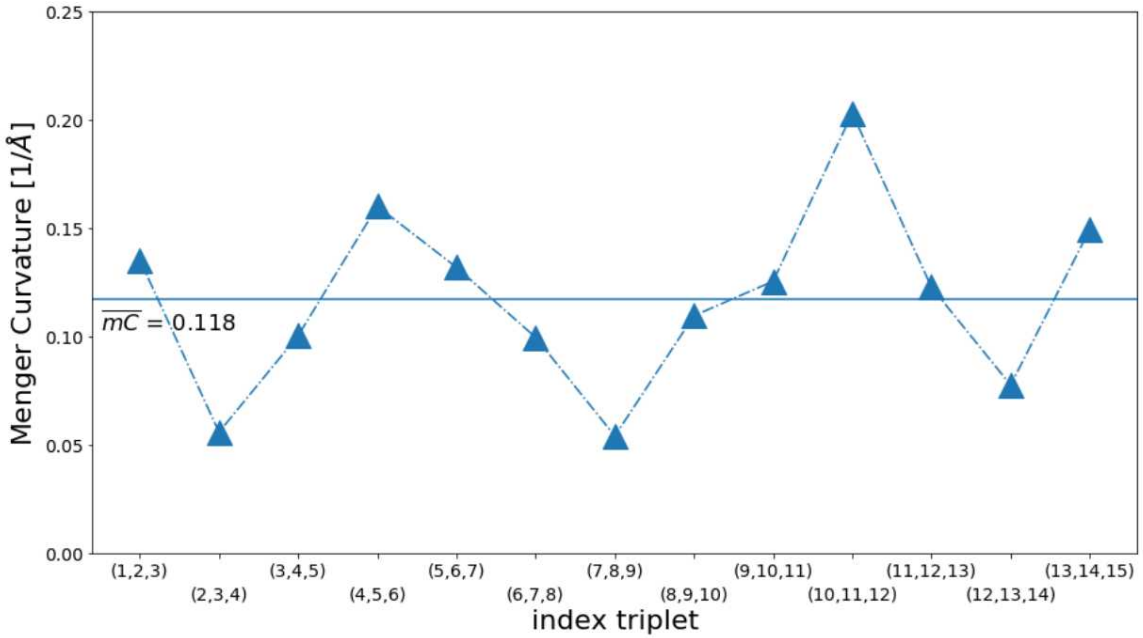


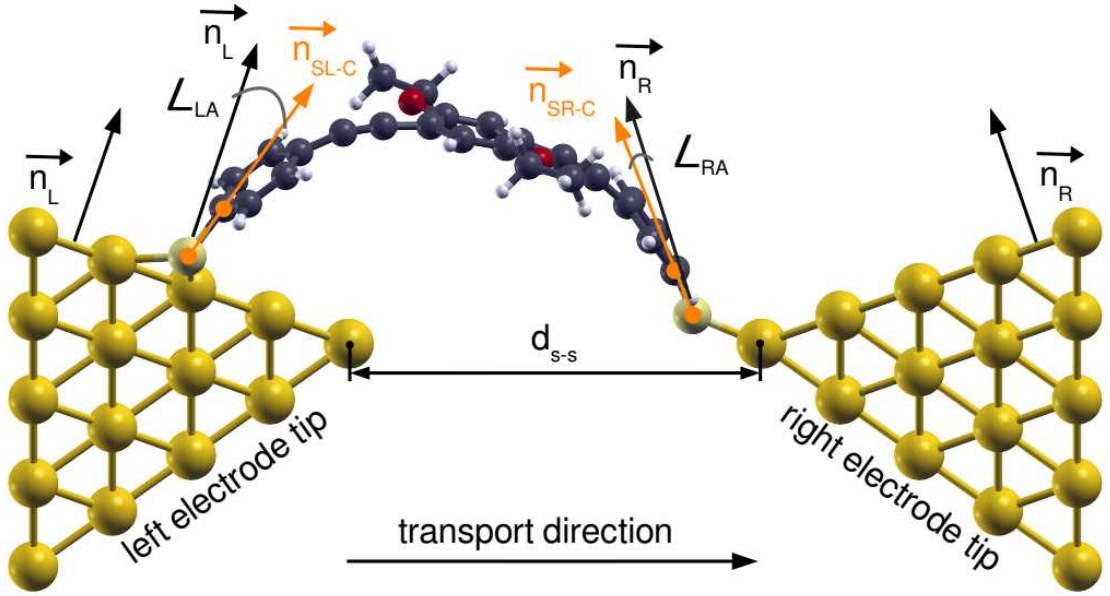
- Stretching follows Hooke‘s Law (parabola)
- Compression linear ▸ in-plane and out-of-plane buldging
- Initial energy cutoff 1eV
- Distortions included [0.75, 1.05] * d_{ss}

- Energy crossover of unoccupied levels [1.05, 1.10] * d_{ss}
- Splitting on HOMO-1/-2 neglible

Curvature (mC) | Anchoring Angle (AA)







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Dynamic Simulation Approach

Modeling Surface Dynamics | Statistical Evaluation of electronic coupling Γ

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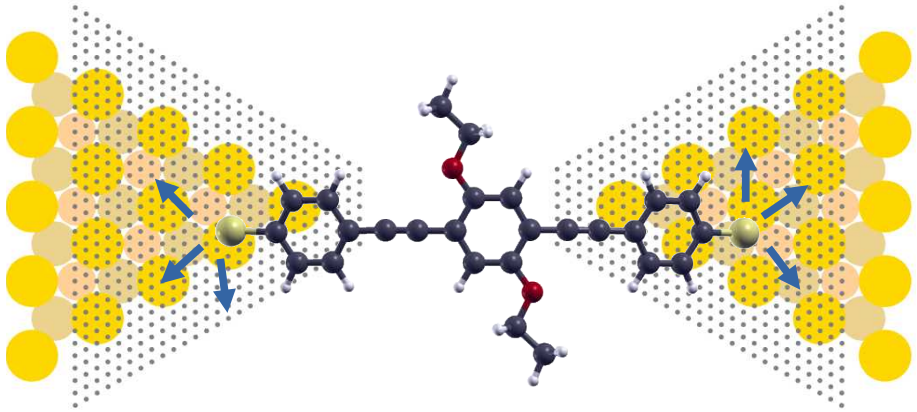
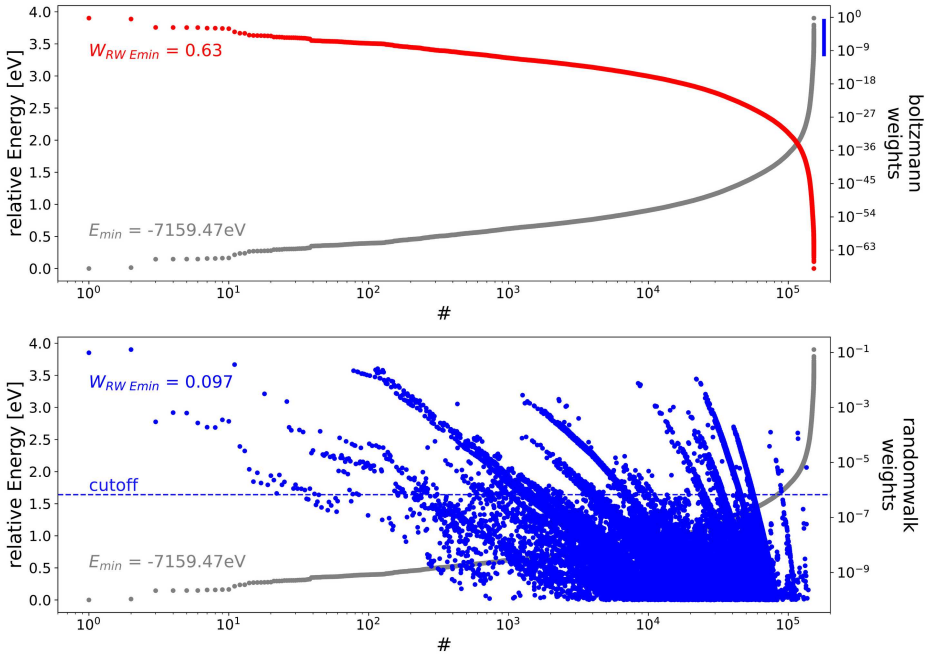
Modeling Surface Dynamics





- Deep local minima enclosed by high-energy configurations
- Contribution of geometrically restricted configurations
- Thermal sampling – random walk 4D configuration space
- Master equation – Transition rates – metropolis probabilities for nearest neighbours
- Statistical averaging using non-equilibrium weights

Stretch Evolution of Γ

Stretch Evolution of Γ

- Region I – falling trend, Region IV – rising trend
- Region II, III – intermediate region with recurring peaks
- Focus on measurement from a single opening curve | not multiple
- No „*a priori*“ assumptions

Comparison with Geometrical Descriptors

- Strong Γ
  - edge-edge & tip-tip anchoring positions (AP)
  - planar configurations
- Influence of curvature stronger than anchoring angle
- Intermediate regions II,III – mixture of concurrent processes

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Realspace Projection - Evolution of Γ

Microstates of junction geometries

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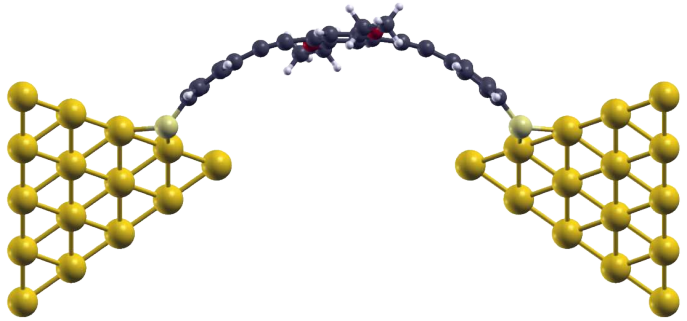
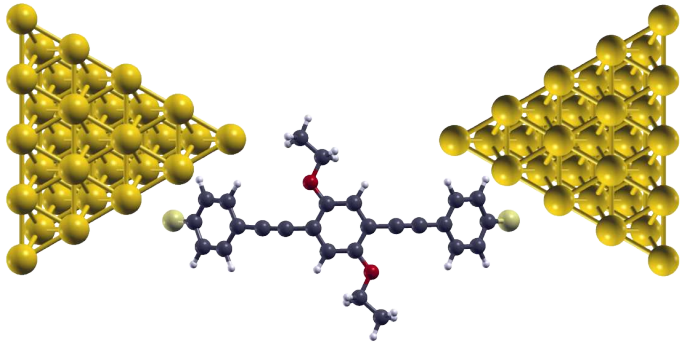
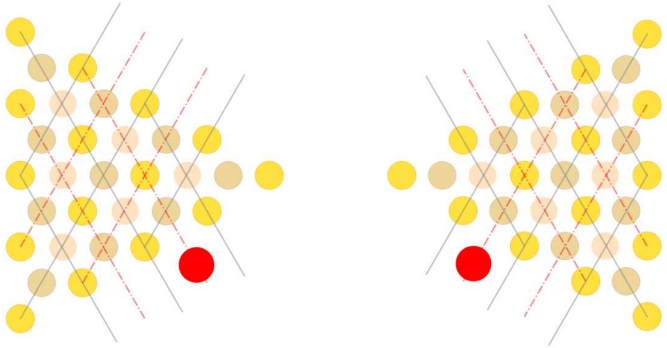
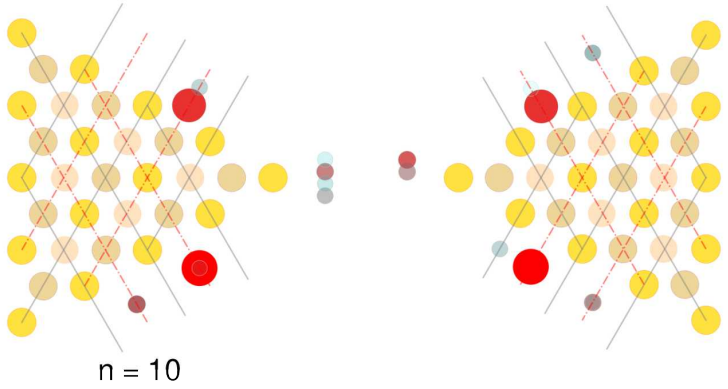
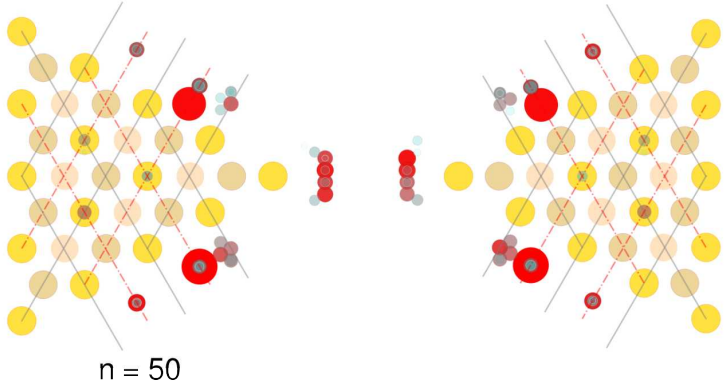
Realspace Projection of Γ | energectically favorable configurations

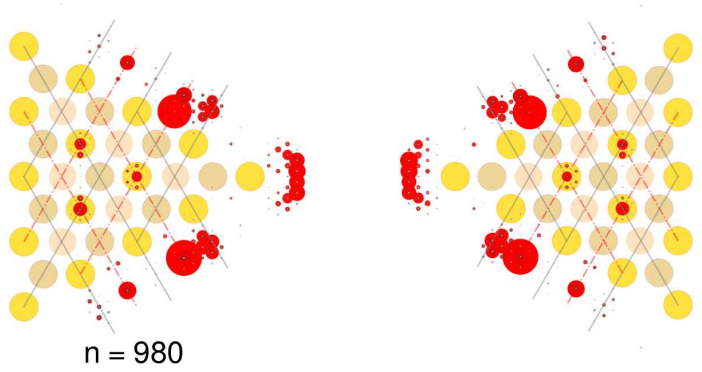












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Conclusion | Outlook | Acknowledgements

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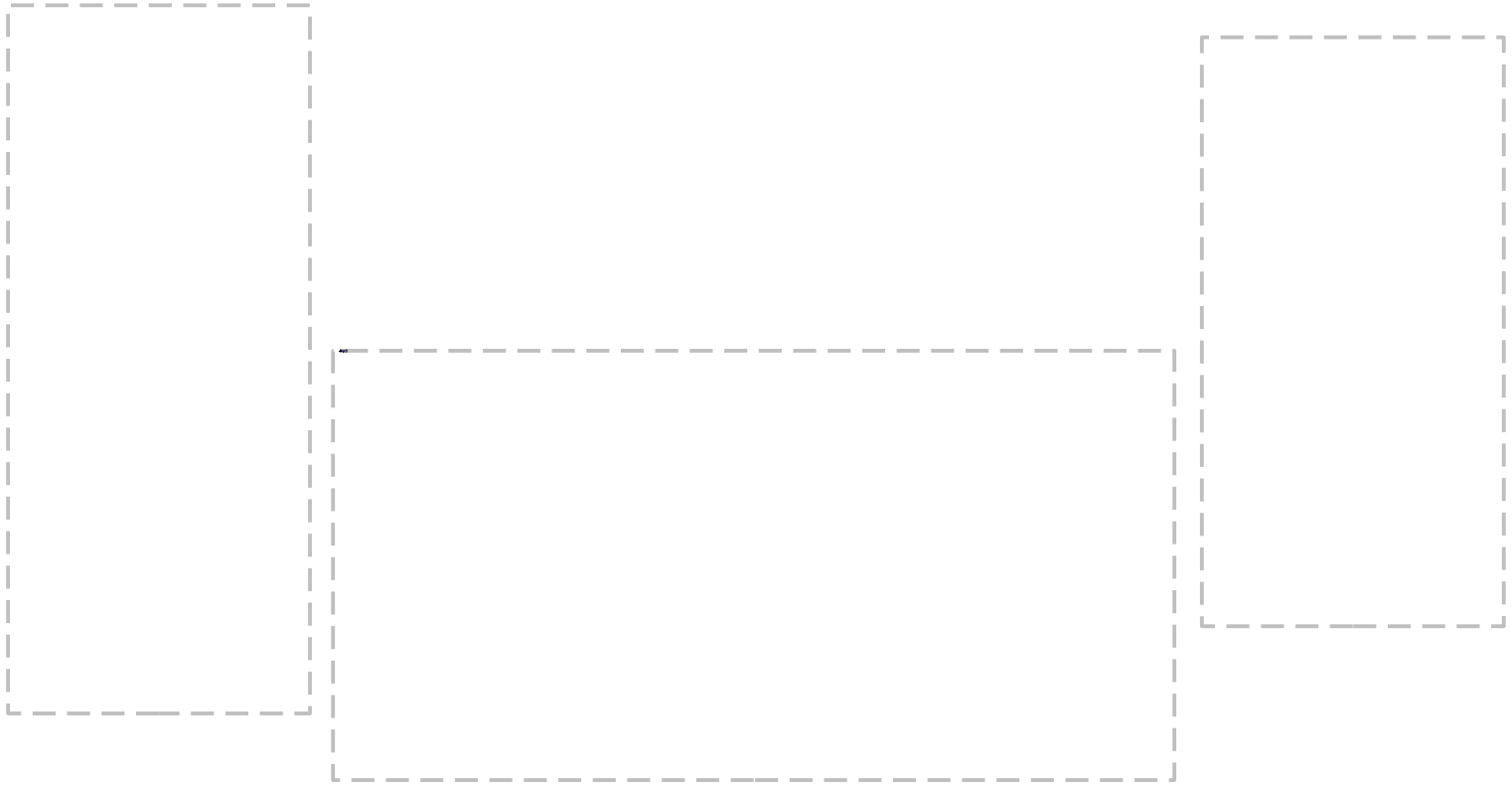
Conclusion

- Investigate single opening measurement
- Dynamic simulation approach
  - Falling/Rising trend in the stretch evolution of Γ
  - Recurrent maxima

- Geometrical descriptors
  - edge-edge & tip-tip Anchoring Positions (AP)
  - Planar configurations

- Influence of curvature stronger than anchoring angle
- Intermediate regions II,III – mixture of concurrent processes

Conclusion



- Complex interplay of anchoring atoms sliding between bridge and top sites along Au(111)
- Strong Γ – symmetric anchoring in region I, optimal geometry/angle in region IV
- Intermediate region II, III mixed tip-edge configuration and symmetric/asymmetric anchoring
  - Local maxima/minima alternate in this region – recurrent peaks
  - link between the evolution of Γ and microstates of junction geometries

Outlook and Acknowledgement

- Evolution of energy level of the dominant transport channel
- Extending single level model to high-bias measurements

- Computational resources – ZIH TU Dresden and HZDR
- SCADS