Effects of electronic correlations and magnetic field on a molecular ring out of equilibrium

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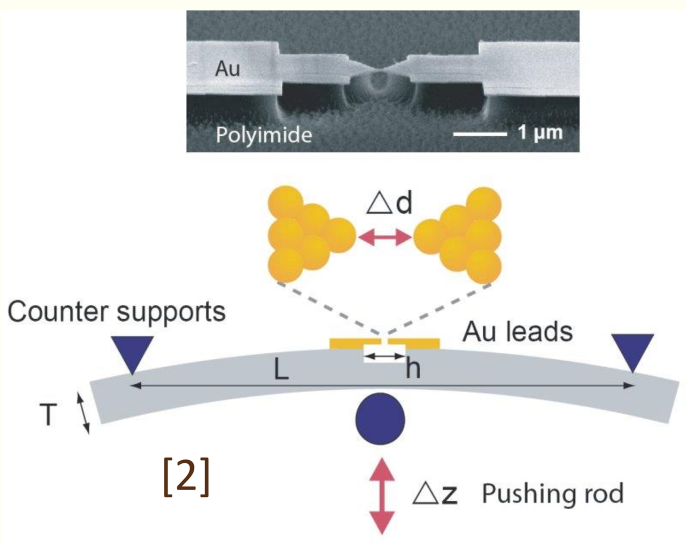
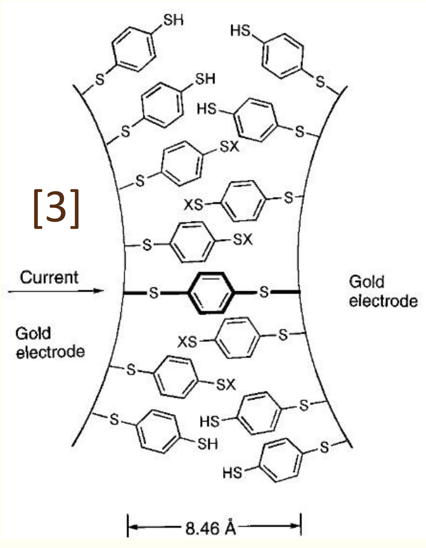
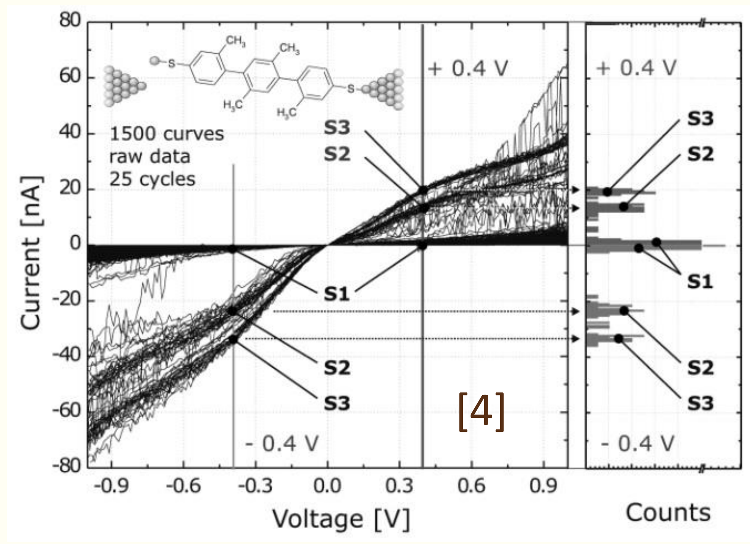
MOTIVATION



Experiment

- fabrication: **M**echanically **C**ontrolled **B**reak **J**unction (MCBJ) [2]
- anchor groups [3]
- statistical measurement [4]





Theory

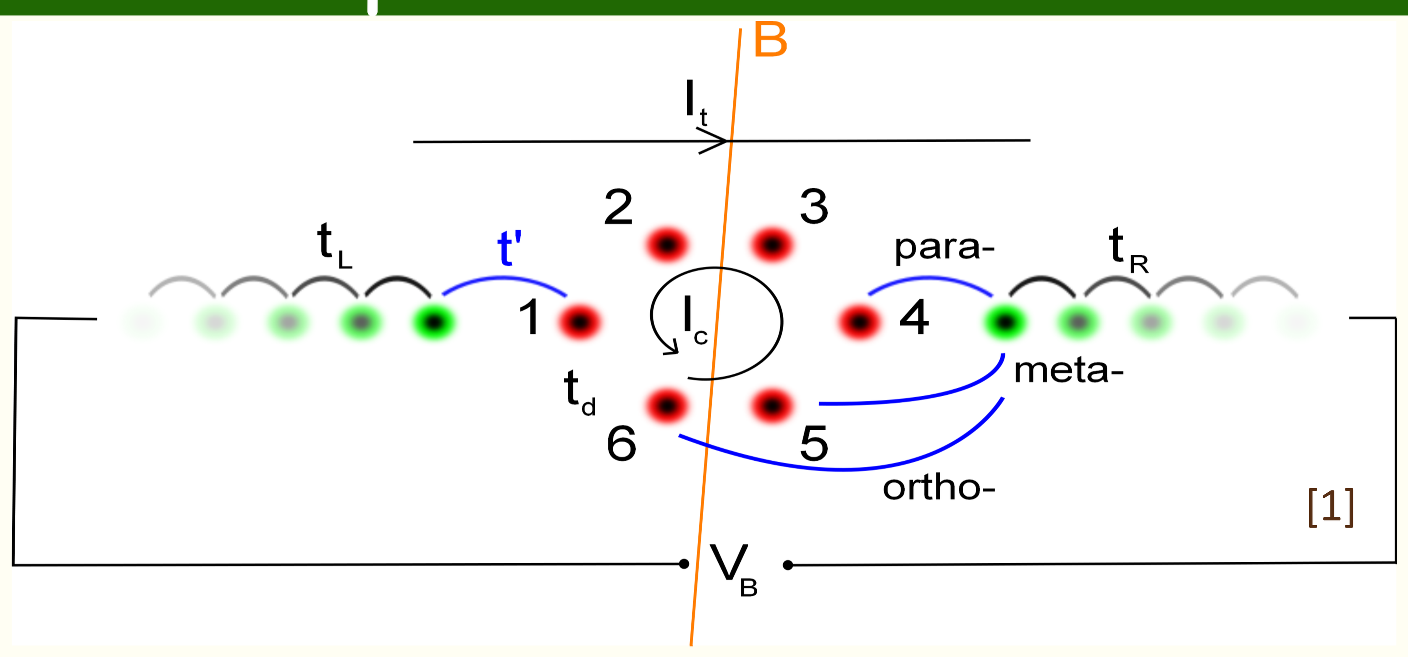
- **D**ensity **F**unctional **T**heory + **N**on-**E**quilibrium **G**reen's **F**unctions (DFT + NEGF)
- model calculations for strong correlations
- marriage of the above

agreement between experiment and theory often poor – improve on electronic correlations – suitable methods ?

I. Setup

metal - „Benzene" – metal junction

- 6 orbital ring + density interaction U_{loc}, $U_{n\text{-}n}$
- 2 metallic leads (semi.circ. DOS) + bias voltage V_B
- perp. magnetic field B (Peierls + Zeeman)
- 3 setups for connecting: (para, meta, ortho)



II. Keldysh formalism

- nonequilibrium single-particle Green's functions (see [5]) $\widetilde{G} = \begin{pmatrix} G^R & G^K \\ 0 & G^A \end{pmatrix}$
- **steady-state** density matrix $D^\sigma_{ij} = \frac{\delta_{ij}}{2} - \frac{i}{2}\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}G^{K\sigma}_{ij}(\omega)$
- current $j^\sigma_{ij} = \frac{ie}{\hbar}\left(h^\sigma_{ij}D^\sigma_{ij} - h^\sigma_{ji}D^\sigma_{ji}\right)$

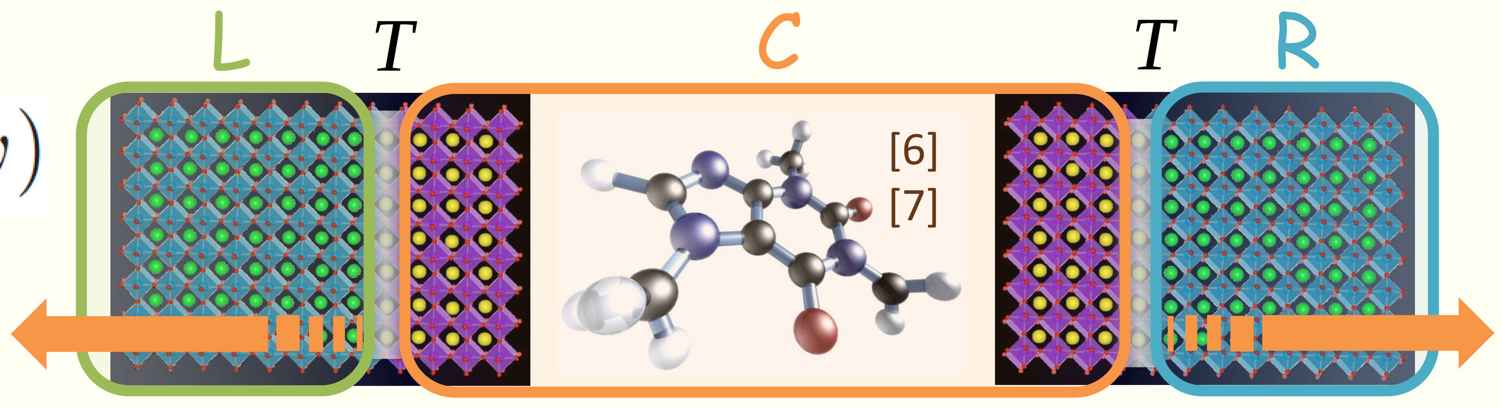
III. Steady-state Cluster Perturbation Theory (stsCPT)

1) $\tau < \tau_0$: solve disconnected parts exactly
C correlated molecule + part of leads
L rest of left lead
R remainder of right lead
→ $\widetilde{g}(\omega)$

2) couple systems using T

3) $\tau \to \infty$: stsCPT equation → $\widetilde{G}(\omega)$

„directly obtain steady-state"



„systematically improvable: increase size of C"

„here: rapid convergence in size of C, otherwise variational feedback" [9]

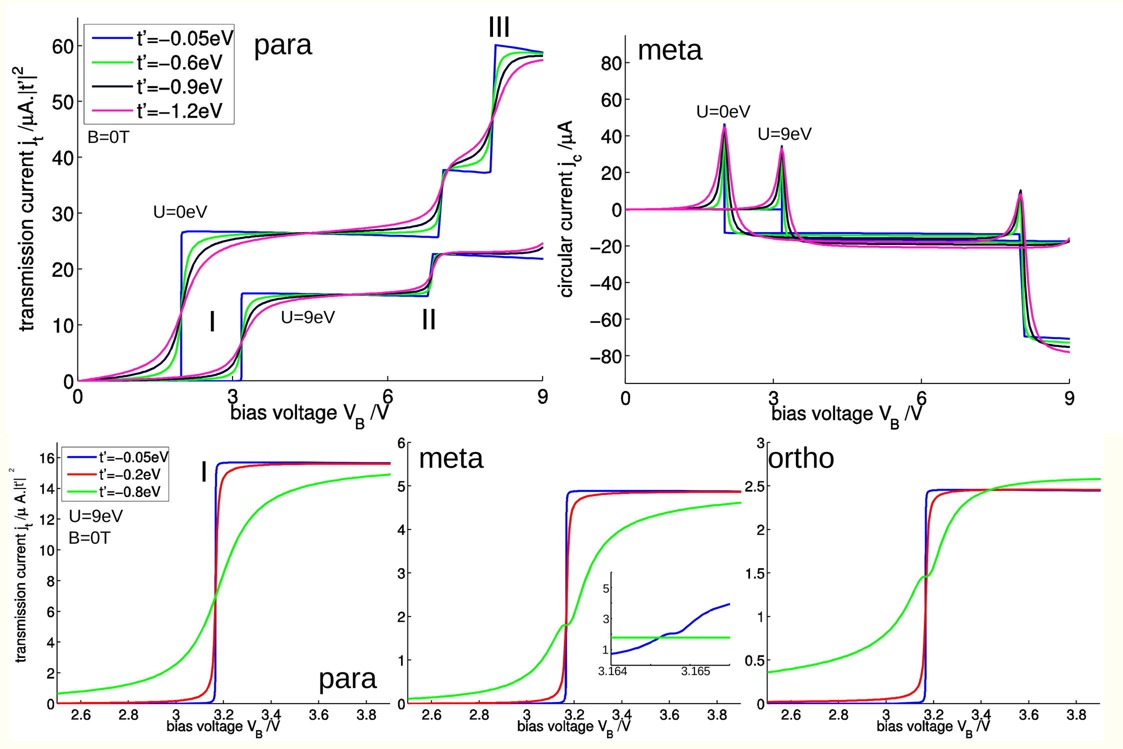
- approximation: take self-energy of the initially decoupled system
- stsCPT → single-particle Green's function

$$\widetilde{G}(\omega)^{-1} = \widetilde{g}(\omega)^{-1} - \widetilde{\mathbb{1}} \otimes T$$

„exact in noninteracting system" [8, 9]

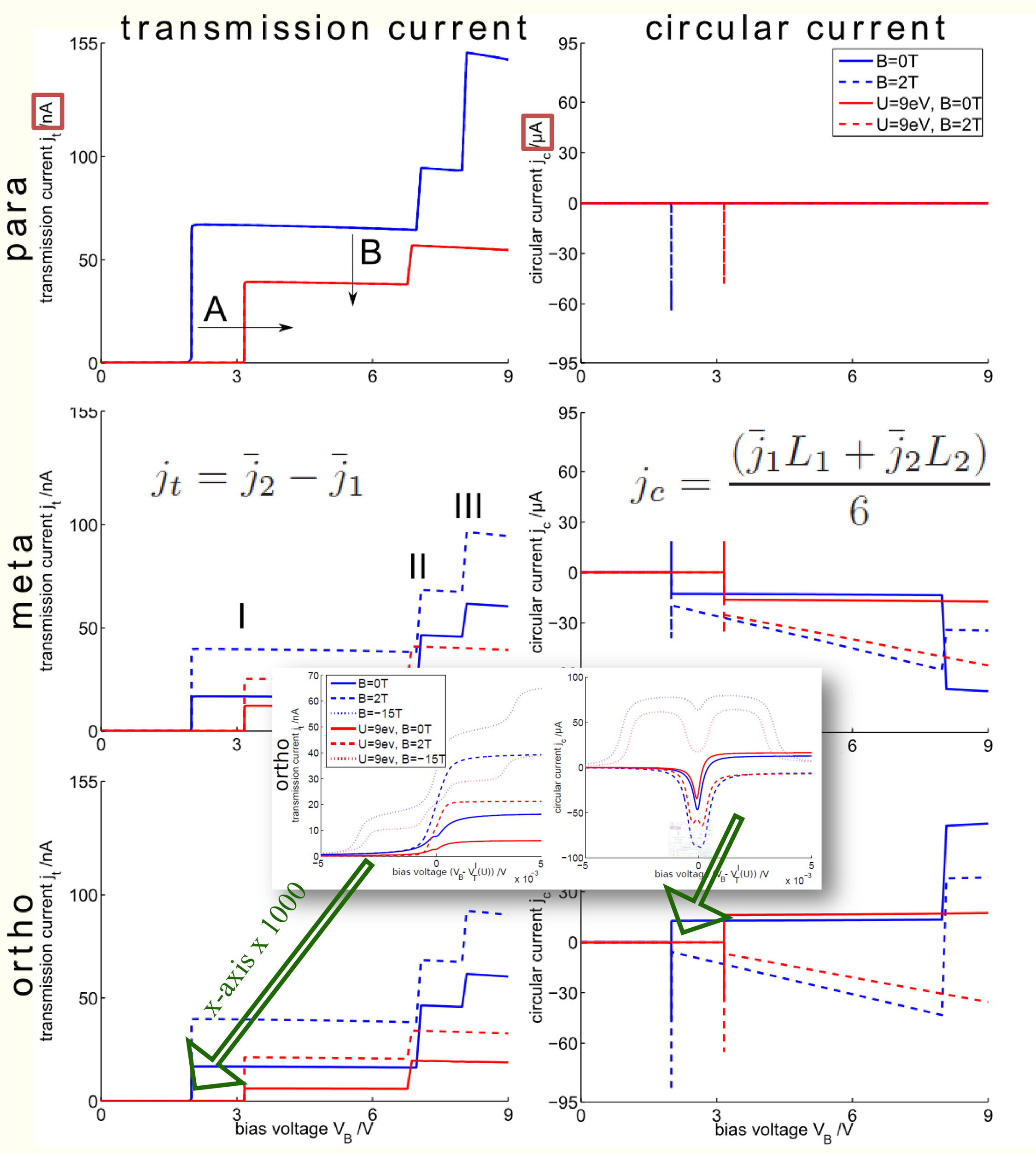
IV. Steady-state results

a) Lead induced broadening

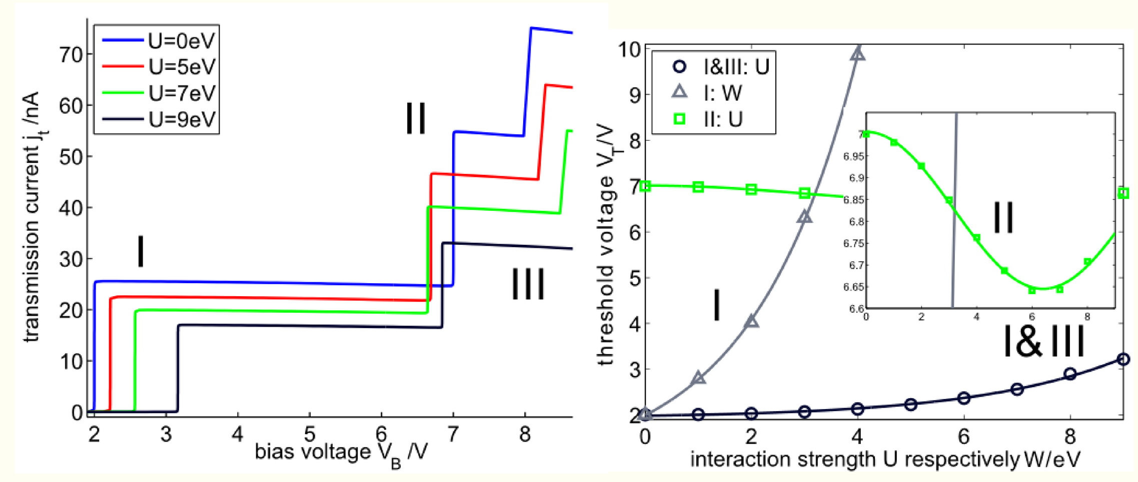


- **nA** currents in transmission, **µA** circular currents
- **broadening** effects described accurately
- e-e **interactions** beyond mean-field
  - reduce current plateau magnitues
  - modify threshold voltages
- j_c strongly **setup** dependent

b) Current



c) Threshold voltage

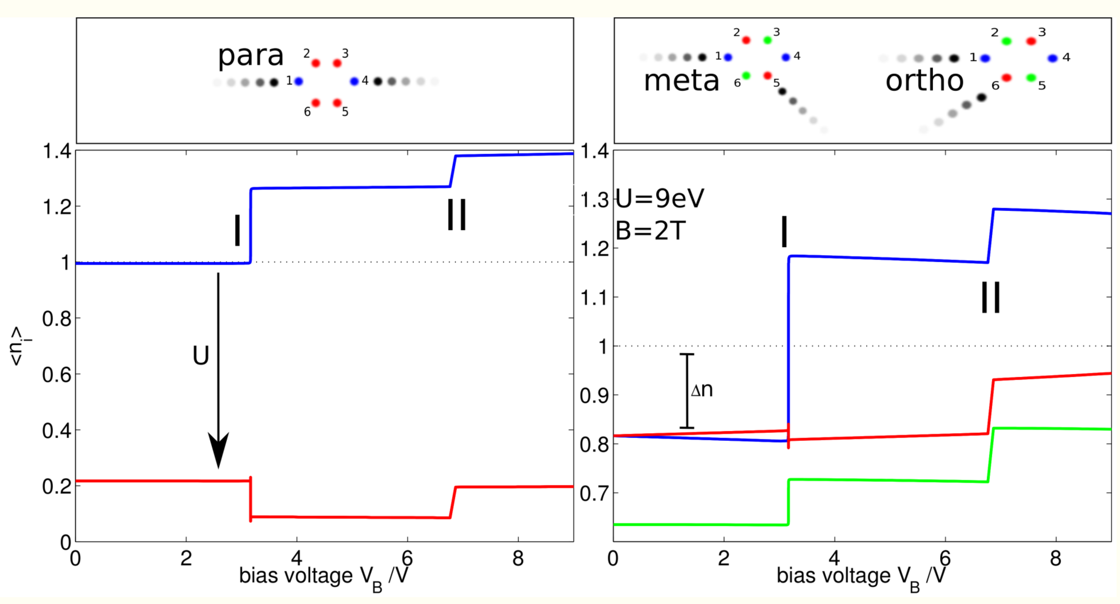


- basic structure understandable by noninteracting electronic levels of molecule
- thresholds renormalized by e-e interactions beyond mean-field

V. Literature

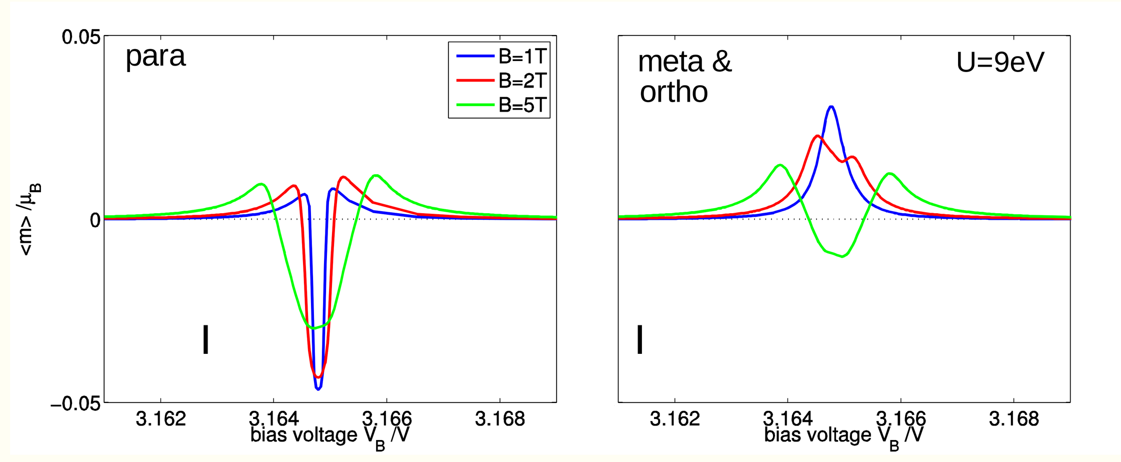
1. Nuss, von der Linden and Arrigoni, arXiv 1307.**7530** (2013)
2. © Dep. Physics, Nanoelectronics , Univ. Basel, 2008-2013
3. Reed *etal.* Science **278**, 5336 252-254 (1997)
4. Lörtscher *etal.* PRL **98**, 176807 (2007)
5. Rammer *etal.* RMP **58**, 323 (1986)
6. © xl8r, http://xl8r.deviantart.com/ (2013)
7. Shen, NGSCES (2013)
8. Gros etal. PRB **48**, 418 (1993), Senechal etal. PRL **84**, 522 (2000)
9. Knap etal. PRB **84**, 115145 (2011), Nuss etal. PRB **86**, 245119 (2012)

d) Charge distribution



- charge density strongly renormalized by e-e interactions beyond mean-field

e) Magnetization



- Zeemann term leads to magnetic response at voltage thresholds

VI. Conclusions

- stsCPT = systematically improvable method to study steady-state including *ab-initio* and finite T
- lead induced broadening
- e-e interactions beyond mean-field:
  - renormalize current thresholds + plateau magnitudes
  - considerably renormalize charge distribution
- j_c shows strong response on lead DOS

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