Effects of electron-electron interactions on a molecular ring junction out of equilibrium

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TUG

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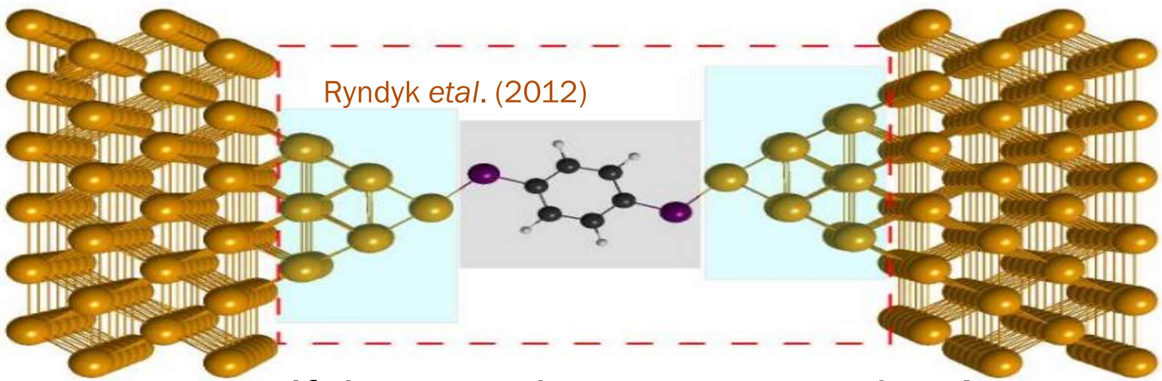
Agenda

I — **MOTIVATION:** transport at the nanoscale

II — **METHOD:** steady-state Cluster Perturbation Theory

III — **RESULTS:** current-voltage, charge distribution, ...



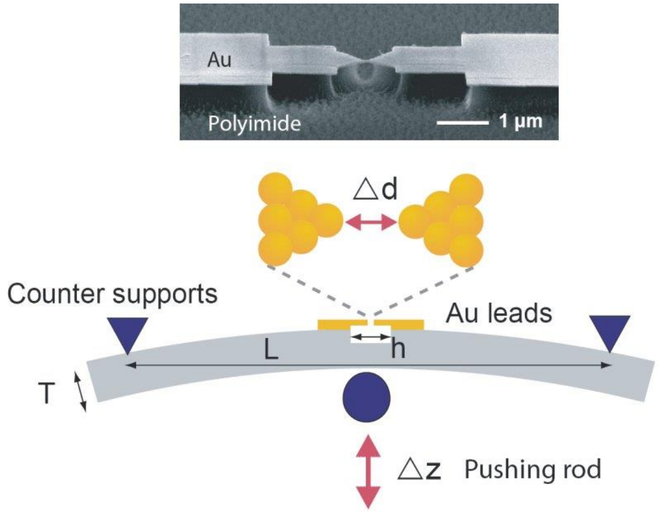
(If time permits or upon questions!

IV — **Theory + Results:** quantum dot

(see RG in S. **Andergassen**'s talk from Thursday)) alternative: STM (see R. **Berndt**'s talk from Wednesday morning)

Molecular junctions

1) Mechanically controlled break junction (MCBJ)



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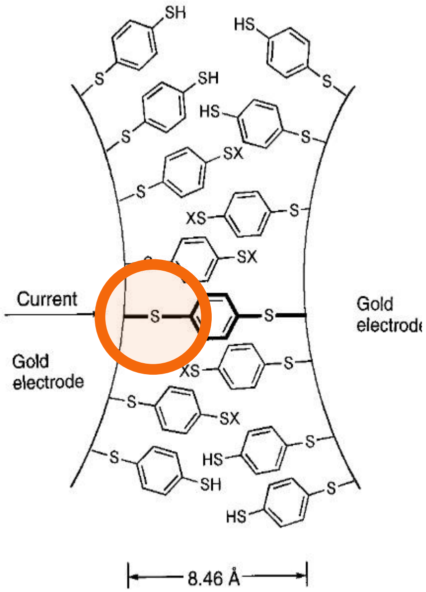
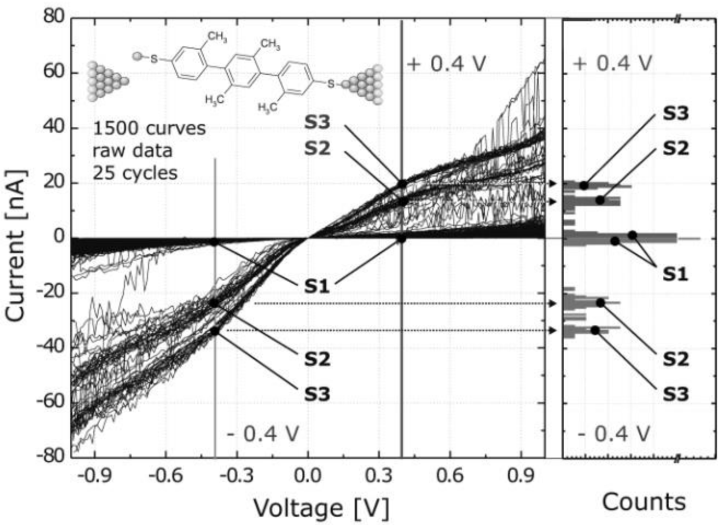
Agrait *etal*. Phys. Rep. **377** 81 (2003)

2) Anchor groups



Reed *etal*. Science **278**, 5336 (1997)

3) Statistical measurements



Lörtscher *etal*. PRL **98**, 176807 (2007)

4) Typical theoretical approach: Density Functional Theory + Non equilibrium Green's functions (DFT+NEGF)

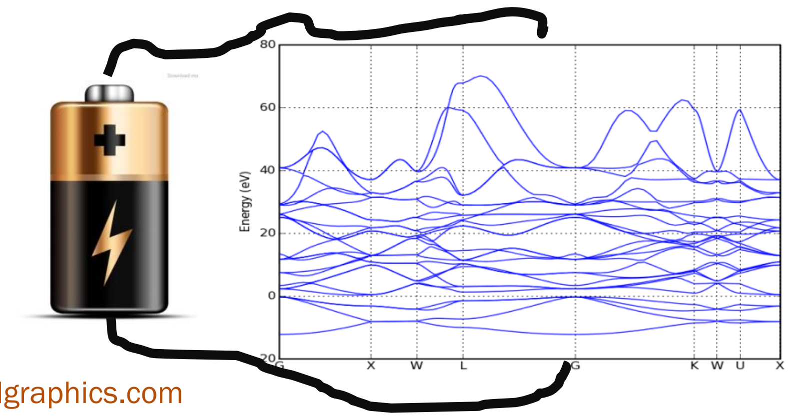
BUT: agreement between experiment and theory often poor – improve on electronic correlations – suitable methods ?

Dzhioev *etal*. J. Chem. Phys. **134** 044121 (2011);Toher *etal*. PRL **95**, 146402 (2005)

Non equilibrium + many body interactions

non equilibrium

transient / dissipation / **steady state**



psdgraphics.com

+ many body interactions

c Goscinny & Uderzo Ehapa

methods

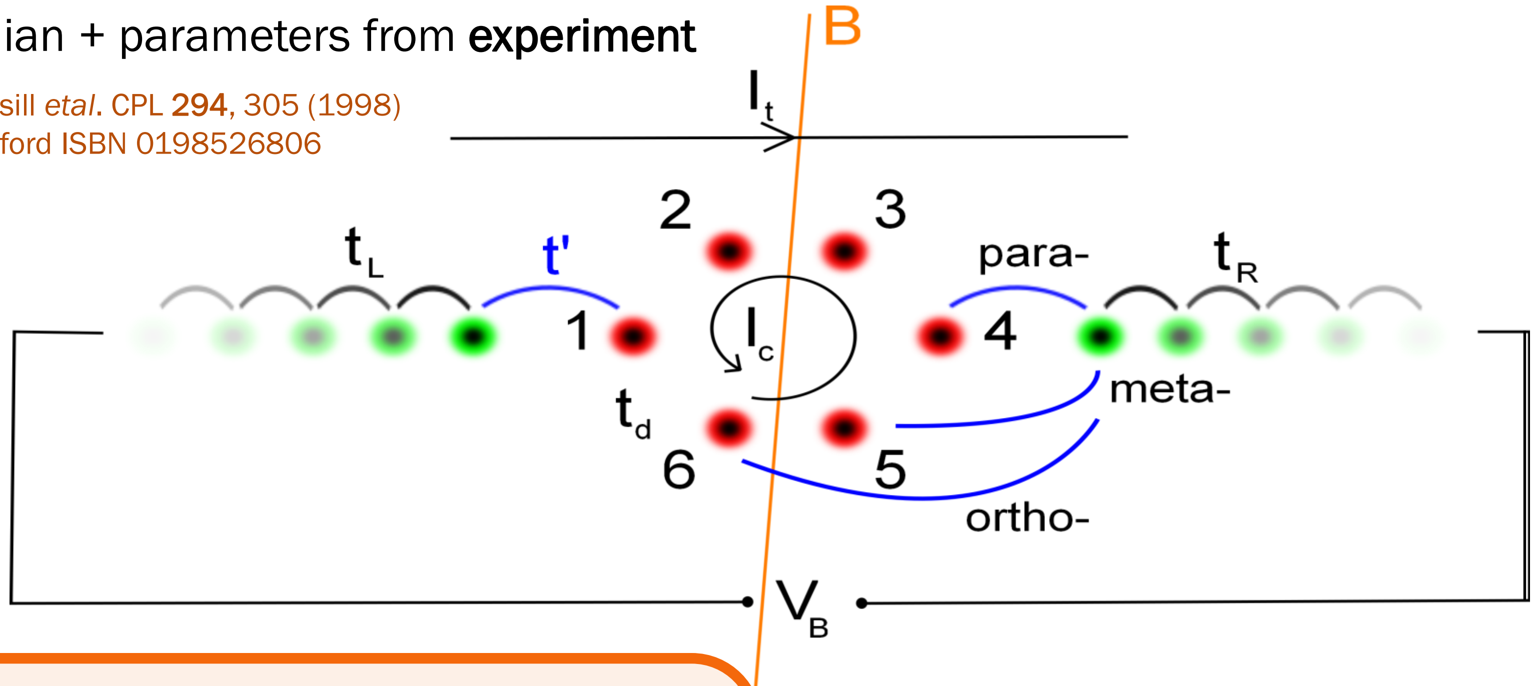
- non equilibrium **cluster perturbation theory**
- non equilibrium **variational cluster approach**
- non equilibrium dynamical mean field theory
- ...

(first two: this talk)

Metal - „Benzene“ - metal junction

model hamiltonian + parameters from **experiment**

Bursill *etal*. CPL **294**, 305 (1998)
Barford ISBN 0198526806



- 6 electronic orbitals + **e^- - e^- interaction**
- perpendicular **magnetic field B**, Peierls + Zeeman
- left + right **metallic leads + bias voltage V_B**
- **3 setups:** para, meta, ortho

- **transmission** current

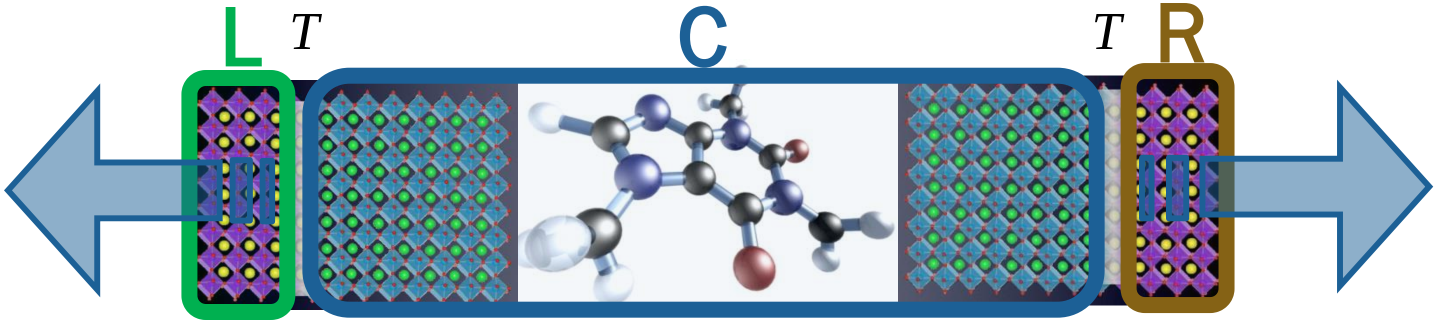
$$j_t = \bar{j}_2 - \bar{j}_1$$

- **circular** current

$$j_c = \frac{(\bar{j}_1 L_1 + \bar{j}_2 L_2)}{6}$$

Rai *etal*. PhysChemC **114**, 20583 (2010)

Steady-state Cluster Perturbation Theory



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- **Non-equilibrium Green's functions (Keldysh):**

Rammer *etal*. RMP **58**, 323 (1986)

$$\widetilde{G} = \begin{pmatrix} G^R & G^K \\ 0 & G^A \end{pmatrix}$$

- **Solve decoupled system exactly, T = 0:**
  - C = „molecule" + parts of the leads
  - L = remainder of left lead
  - R = remainder of right lead

$\widetilde{g}(\omega)$ = exact single particle Green's function of decoupled system

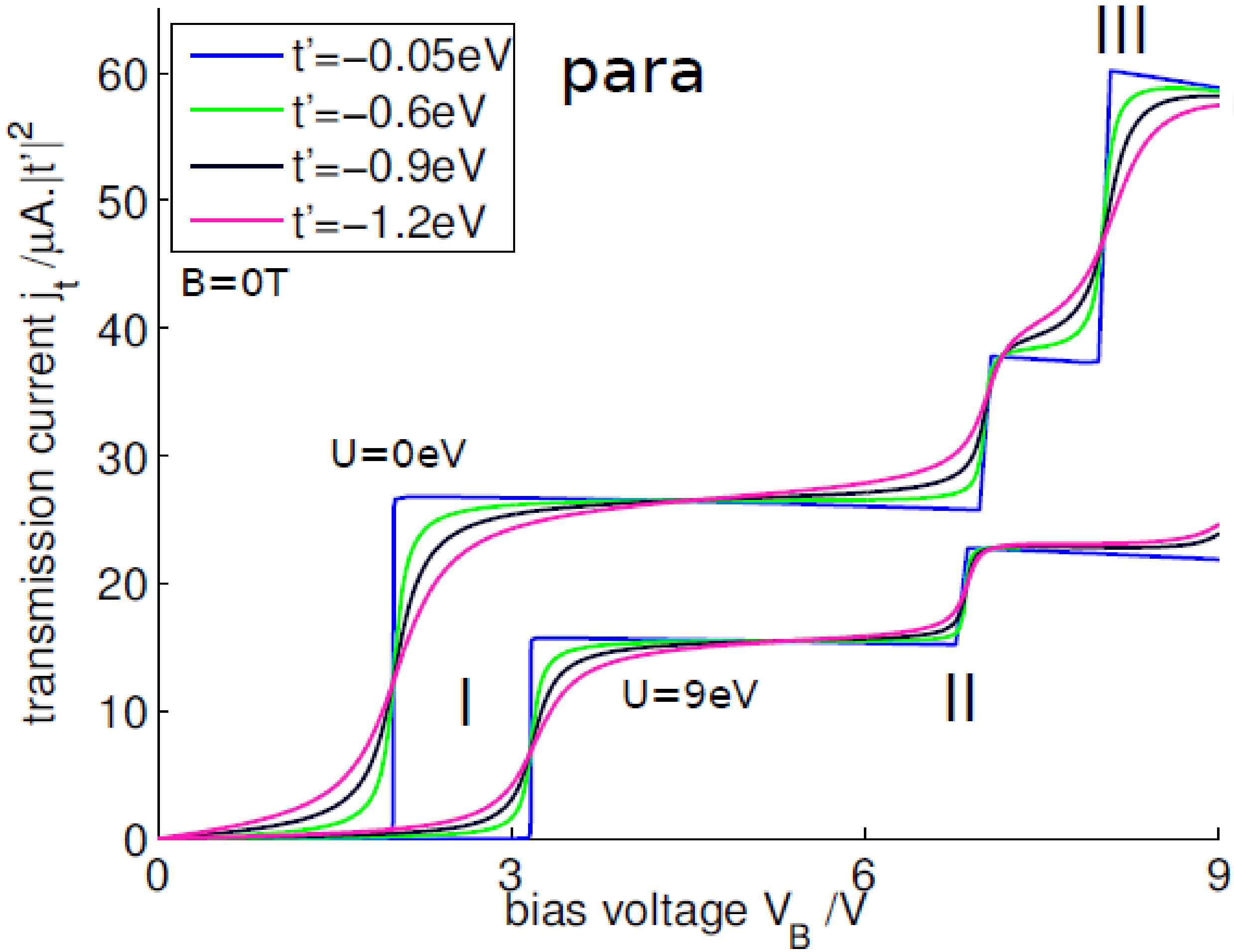
- **Steady-state CPT equation**

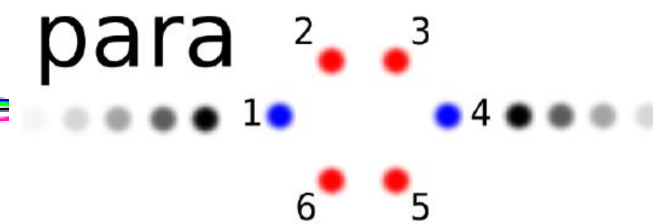
Gros *etal*. PRB **48**, 418 (1993)
Senechal *etal*. PRL **84**, 522 (2000)
Knap *etal*. PRB **84**, 115145 (2011)
Nuss *etal*. PRB **86**, 245119 (2012)

$$\widetilde{G}(\omega)^{-1} = \widetilde{g}(\omega)^{-1} - \widetilde{\mathbb{1}} \otimes T$$ = approximate Green's function of steady-state

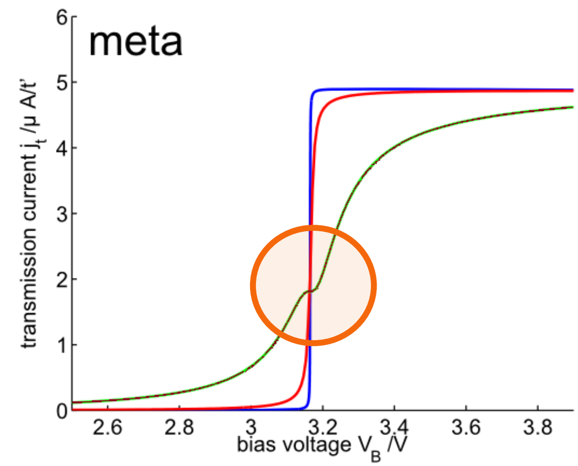
$\Sigma := \Sigma_C$ systematically improvable: increase size of C

I. Transmission current voltage characteristics



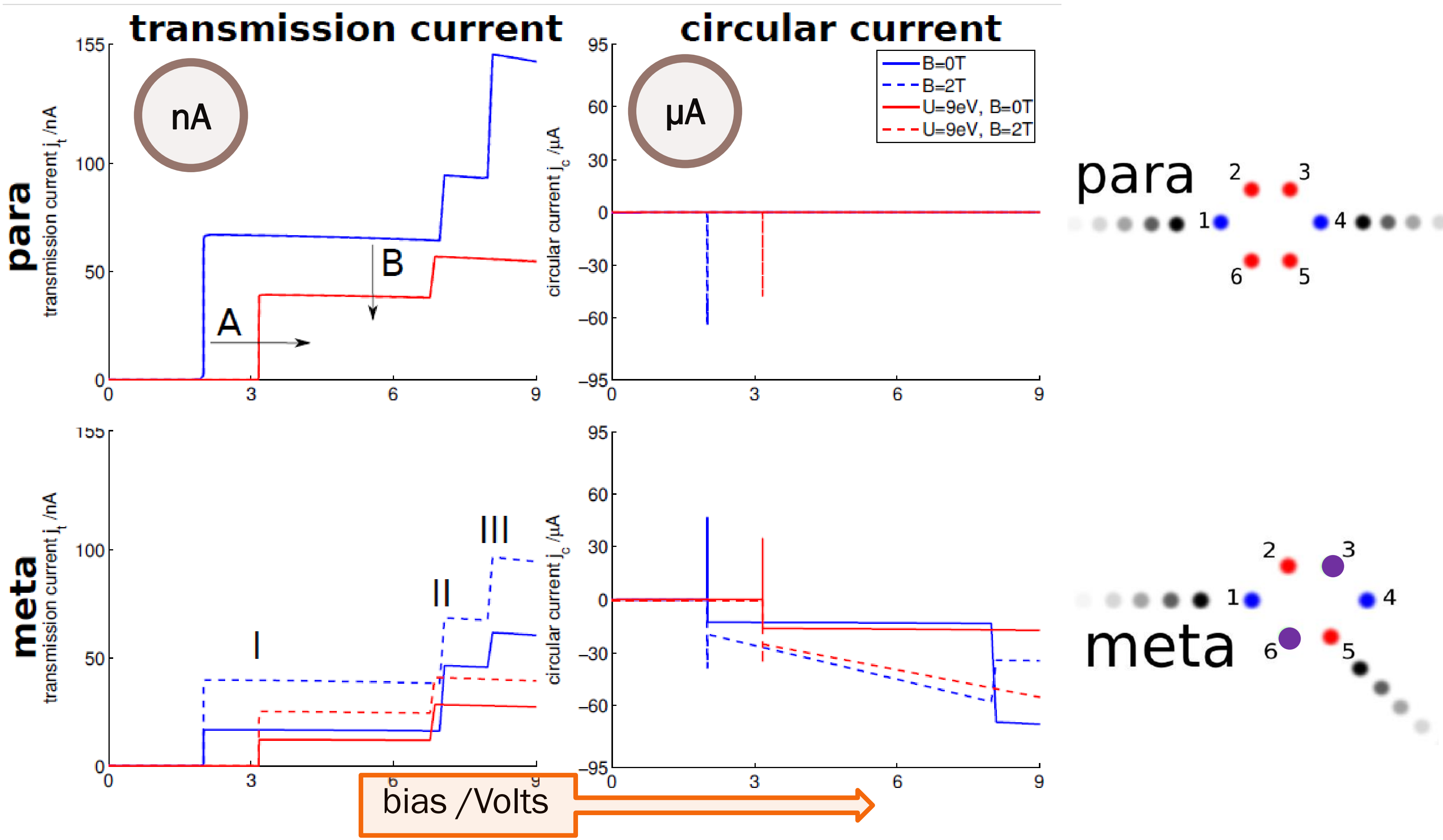


- interaction U
- broadening t'

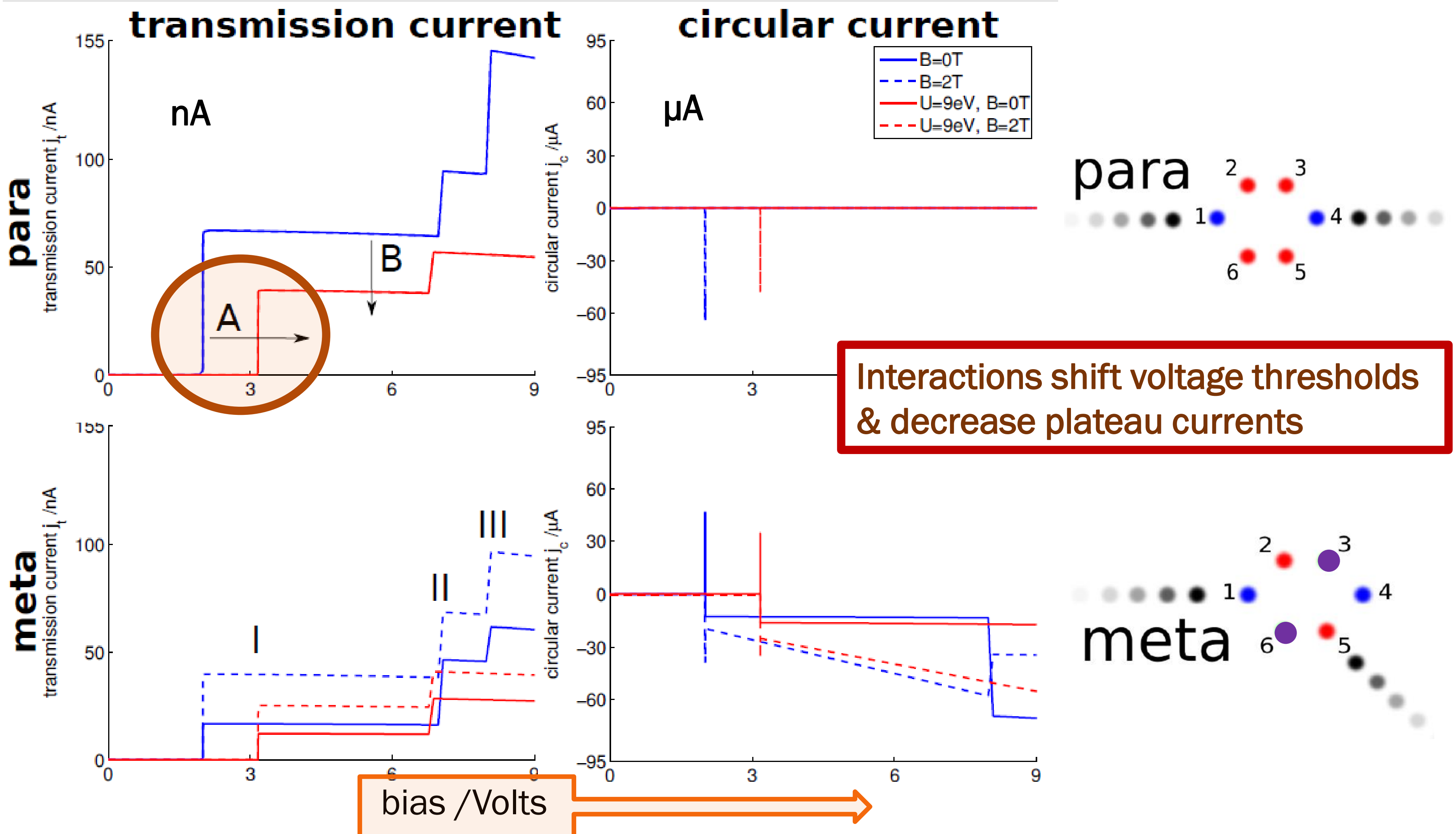


M. Nuss, W. von der Linden and E. Arrigoni (2013)

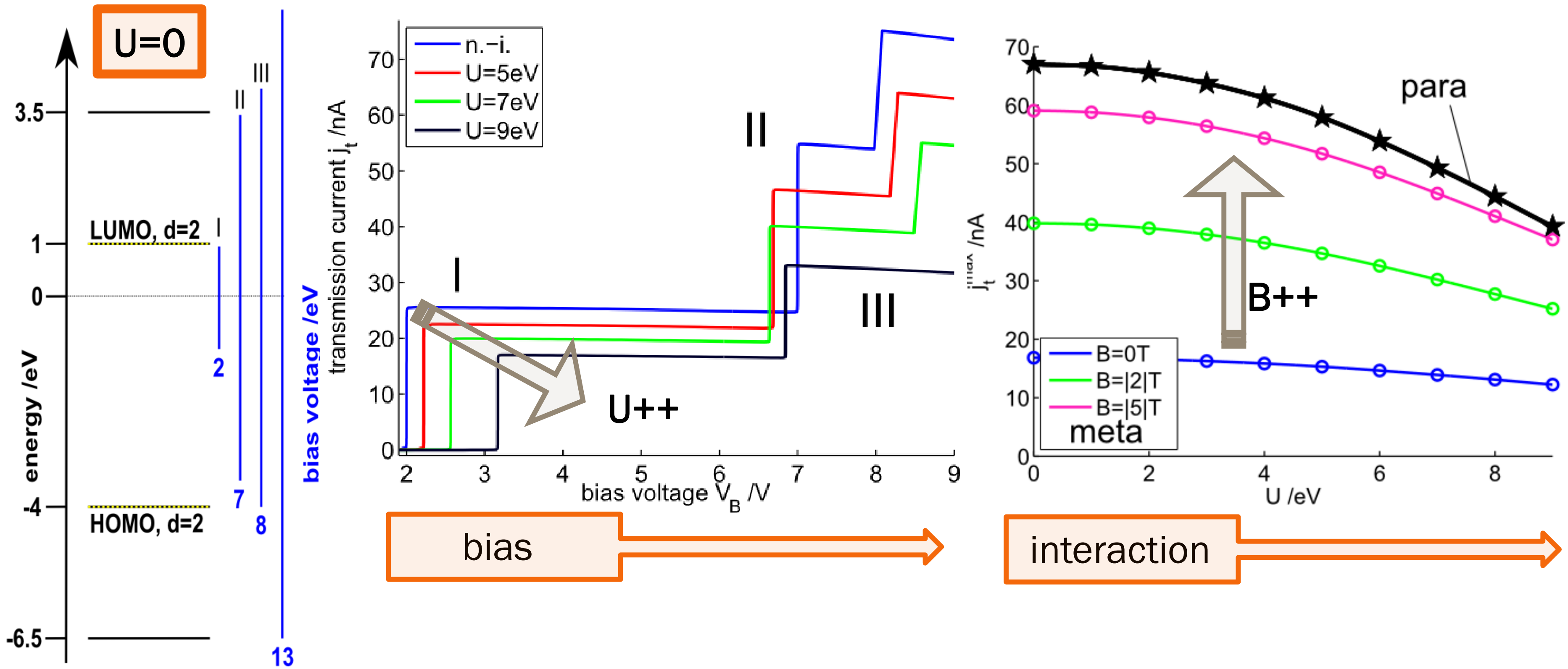
II. Current-voltage overview



II. Current-voltage overview

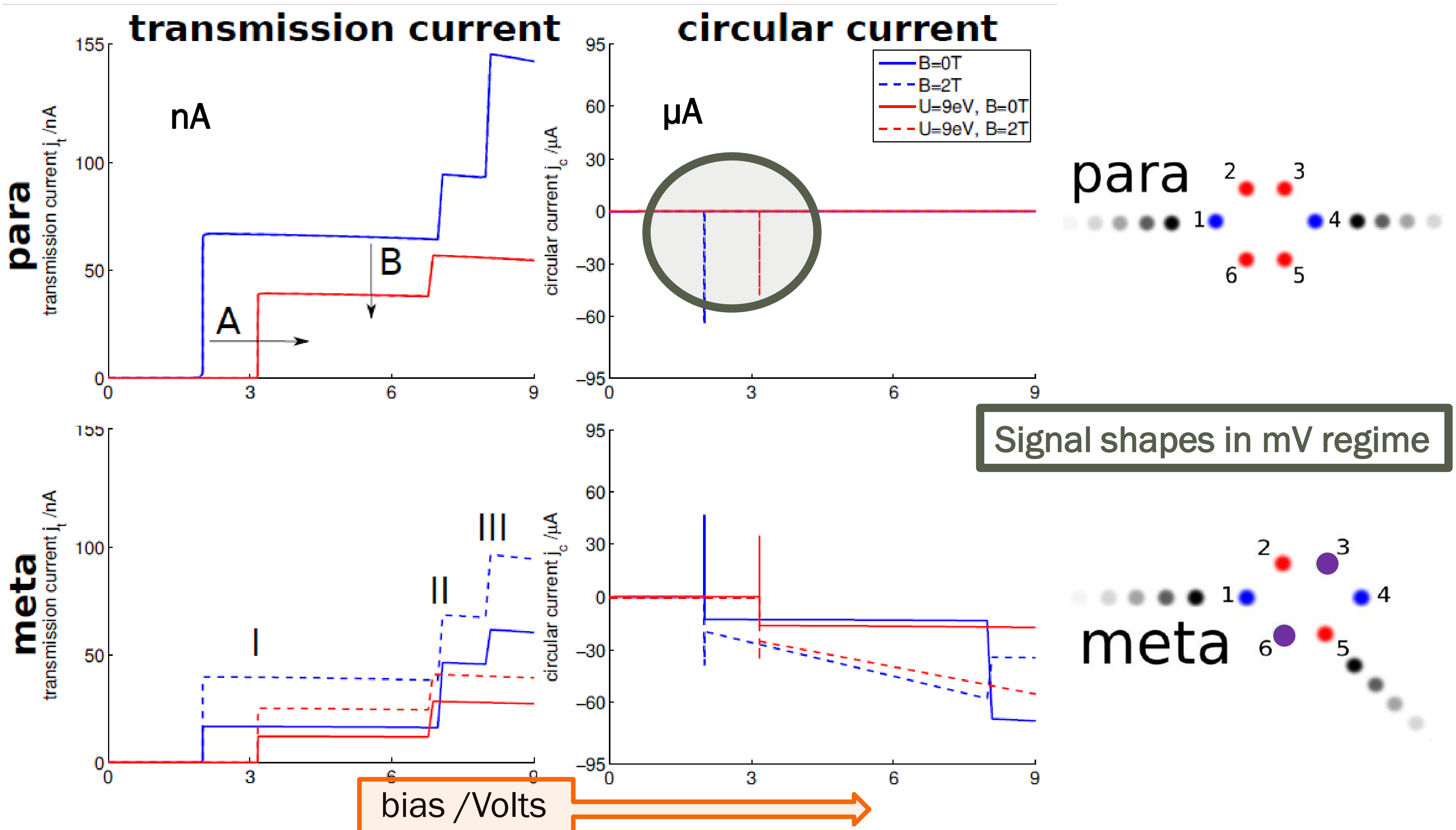


e⁻ - e⁻ interactions / transmission current

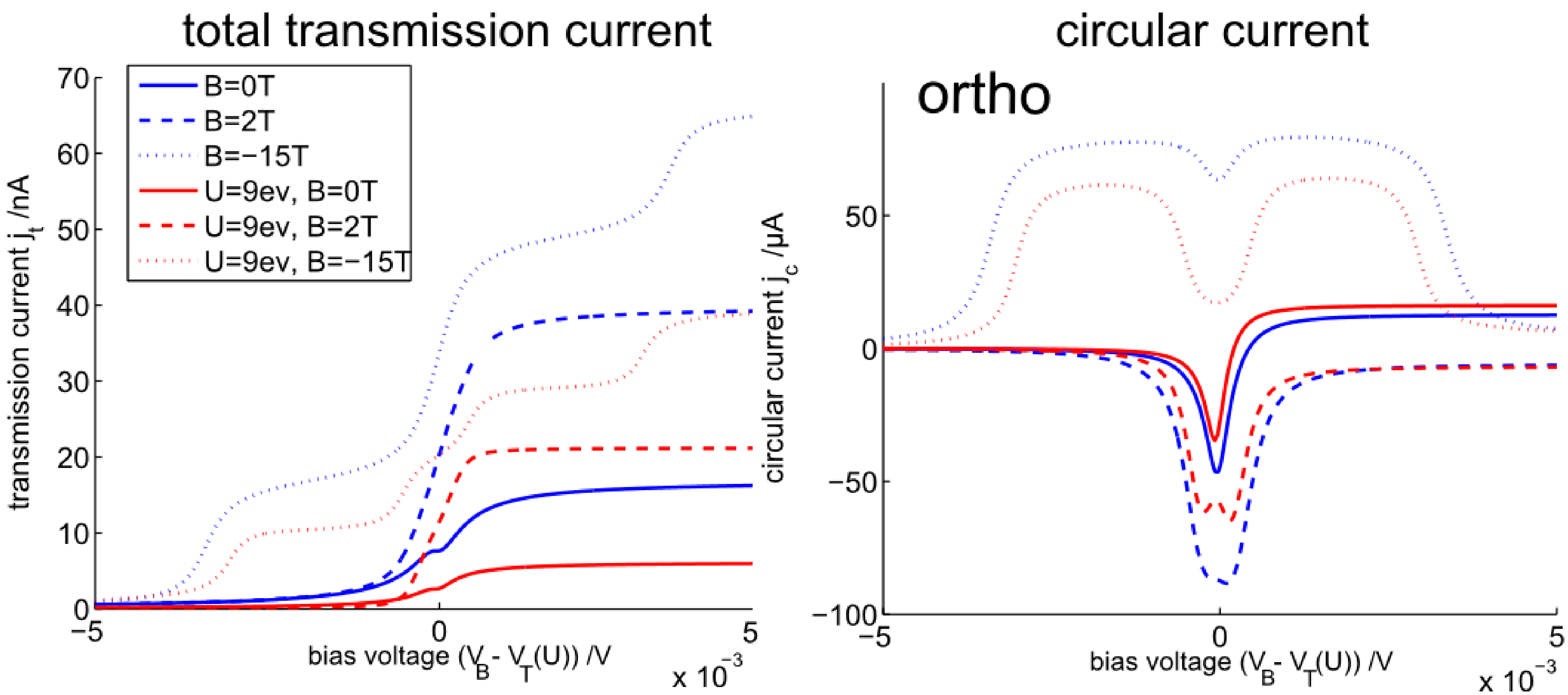


- threshold renormalization beyond mean-field
- plateau current renormalized by U, B

II. Current-voltage overview



Current-voltage signals



- Lenz rule
- Zeemann splitting

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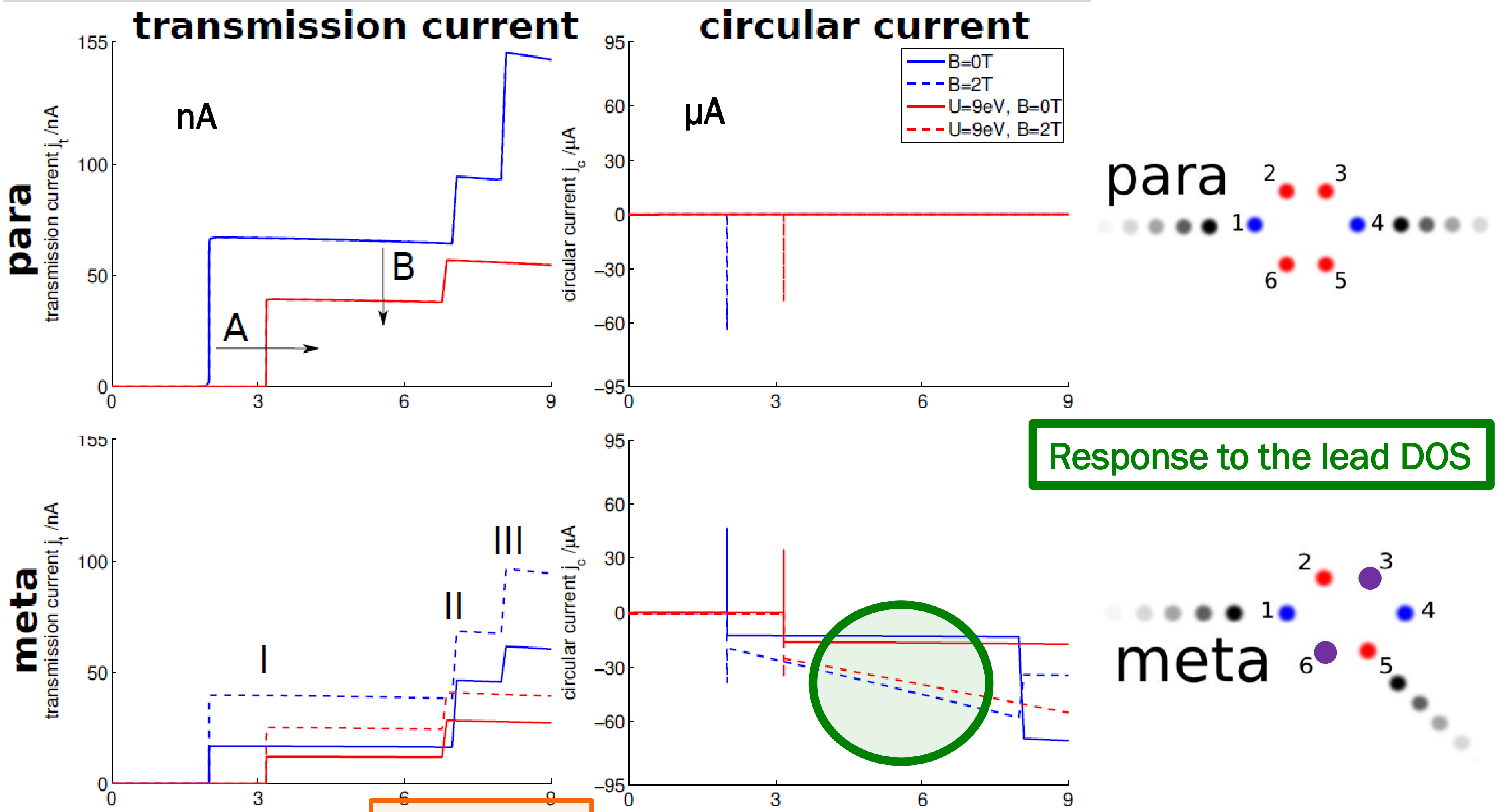
II. Current-voltage overview



Response to the lead DOS

bias /Volts

Lead DOS effects

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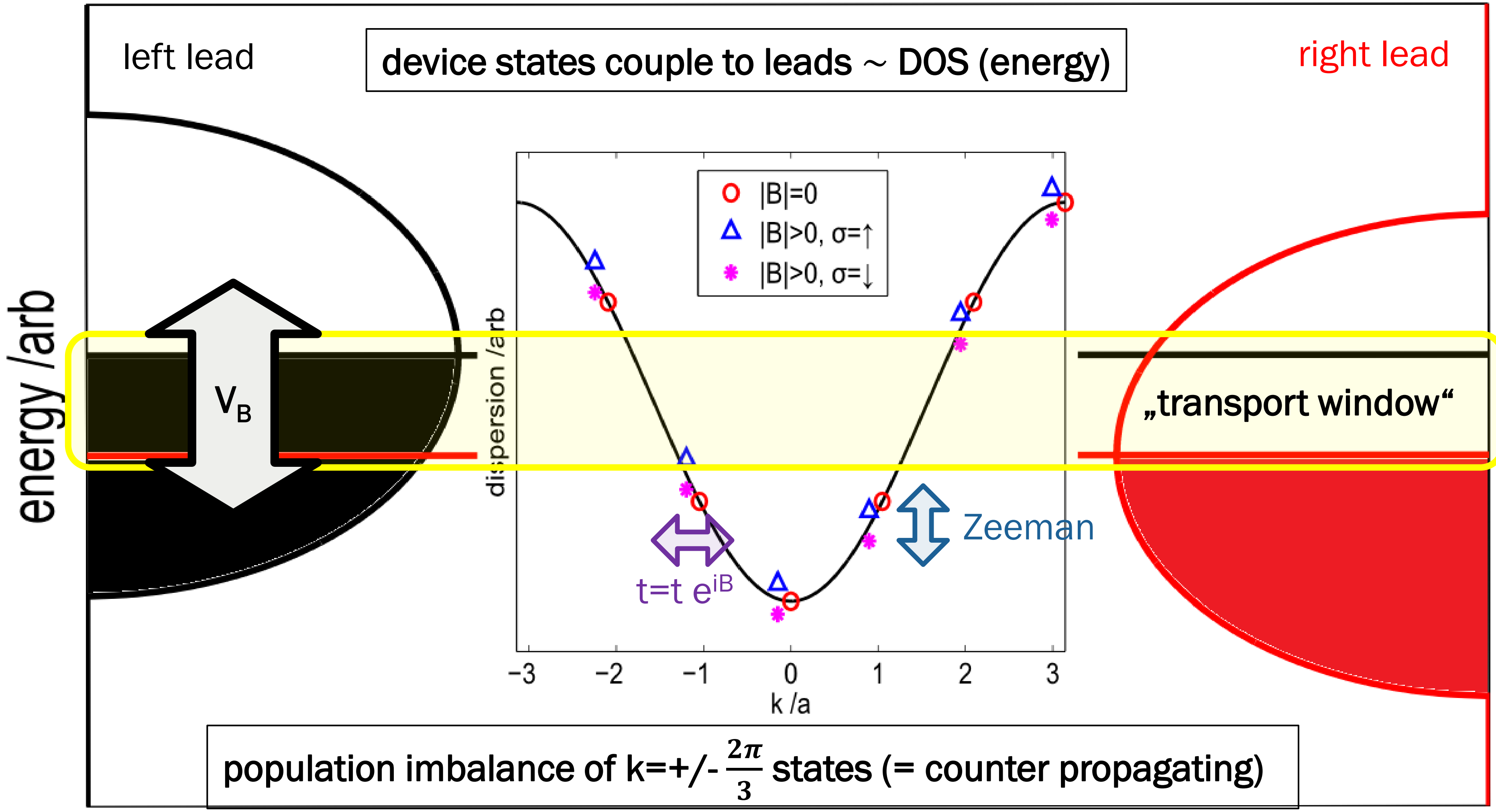
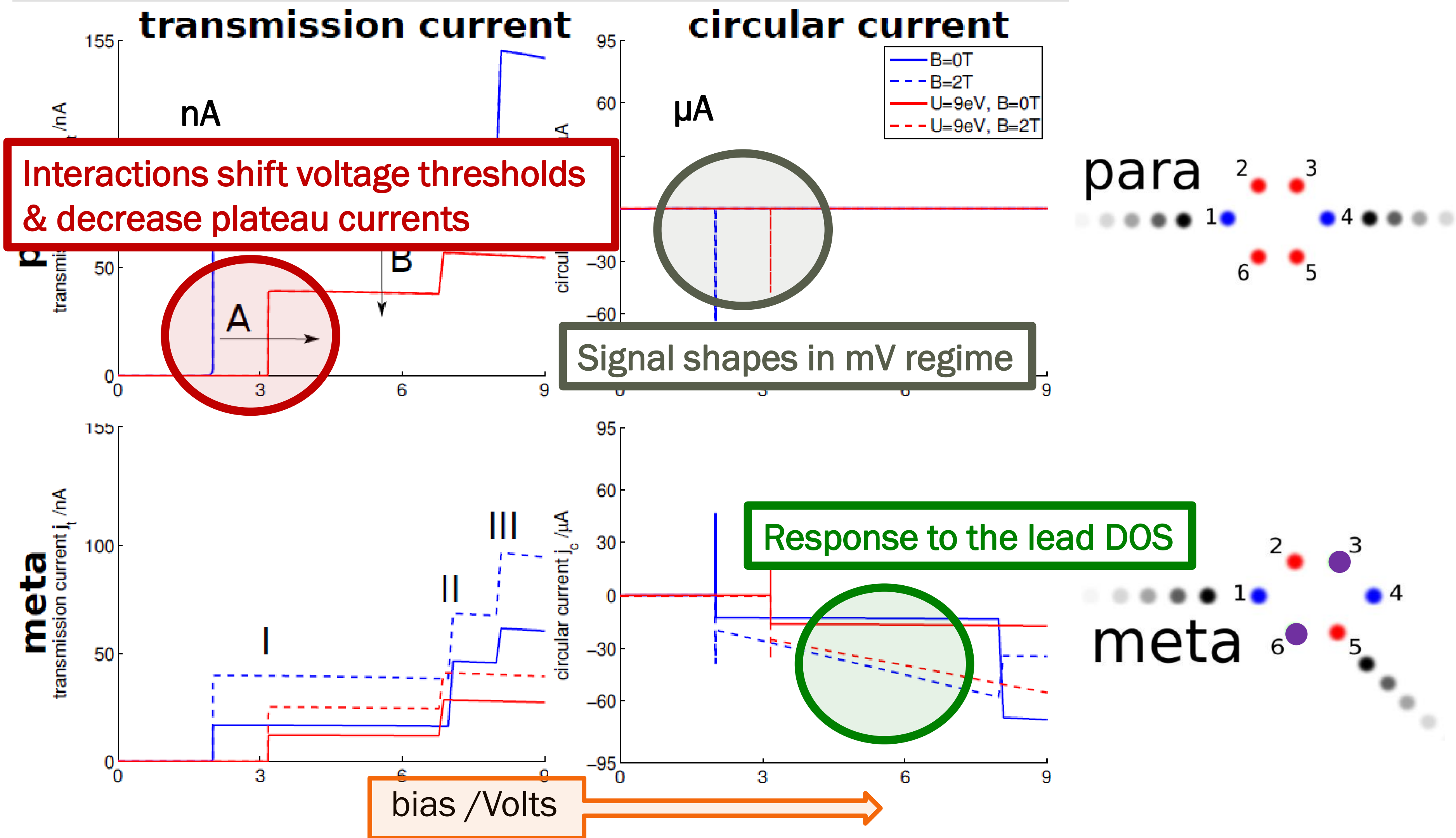
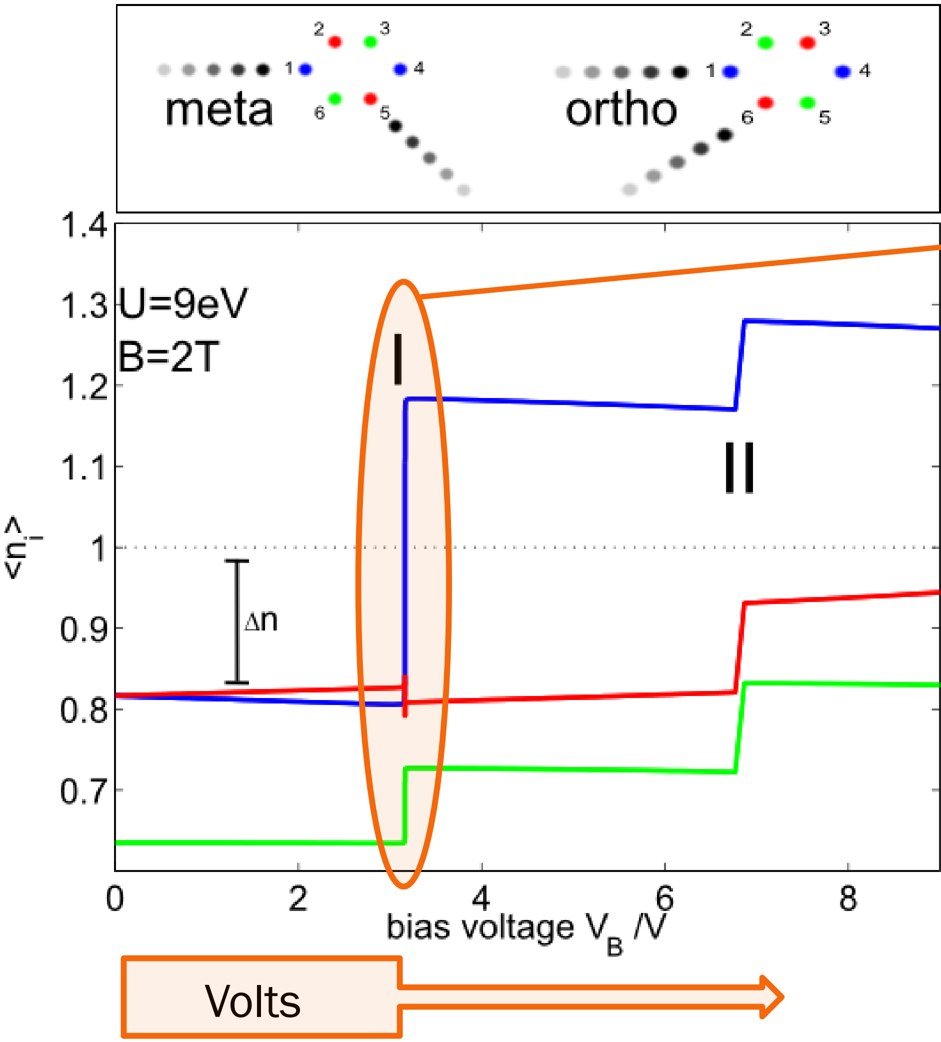


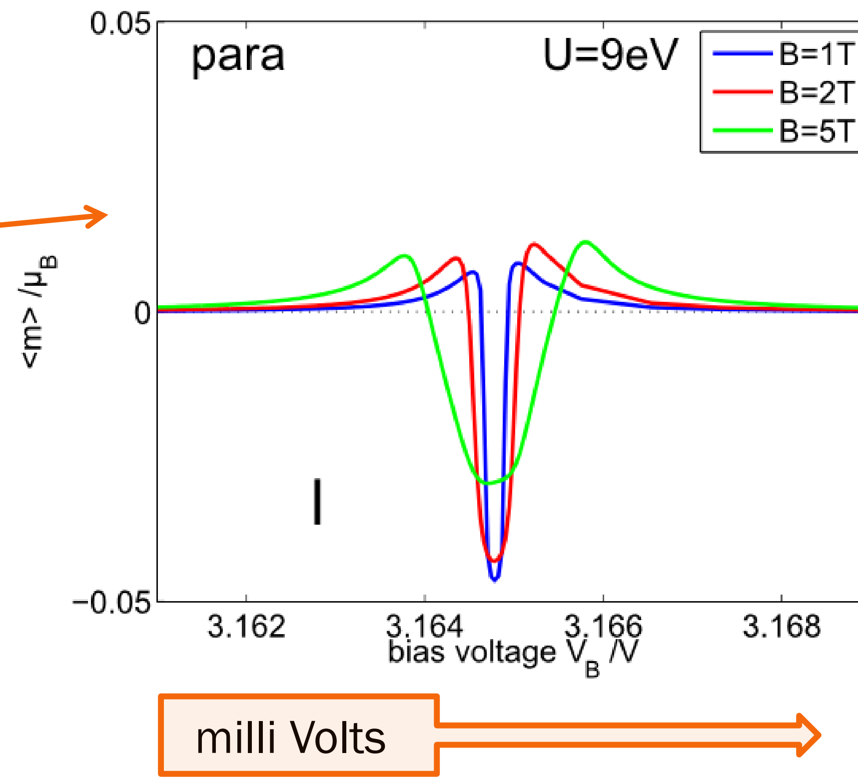
population imbalance of k=+/- $\frac{2\pi}{3}$ states (= counter propagating)

II. Current-voltage overview



III. Charge distribution + magnetization





- <n> substantially renormalized
- <m> signals due to Zeeman

Conclusions & Outlook

Jatakacs.edublogs.org

- Steady-state **Cluster Perturbation Theory**
- **U**: shifts voltage **thresholds** and current **plateaus**
- **B**: strong response of circular current on **lead DOS**
- **interactions** strongly renormalize **charge distribution**

wallpaperswide.com

- ***ab-initio*** (DFT) parameters (realistic leads) — Ryndyk *etal*. (2012)
- finite **T**emperature
- lattice **vibrations** — Knap *etal*. (2012), Sorantin *etal*. (2013)
- more **complicated** structures
- need for variational **feedback**? see quantum dot

Nuss *etal*. PRB **86**, 245119 (2012
Nuss *etal*. AIPcp. **1485** (2012)
Nuss *etal*. PRB **85**, 235107 (2012)
Nuss *etal*. PRB **88**, 045132 (2013)

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Thank you!

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arXiv:1307.7530

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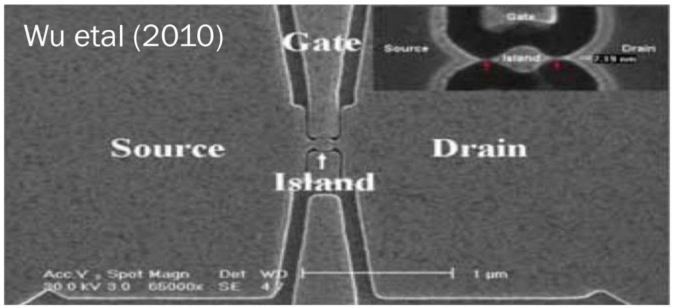
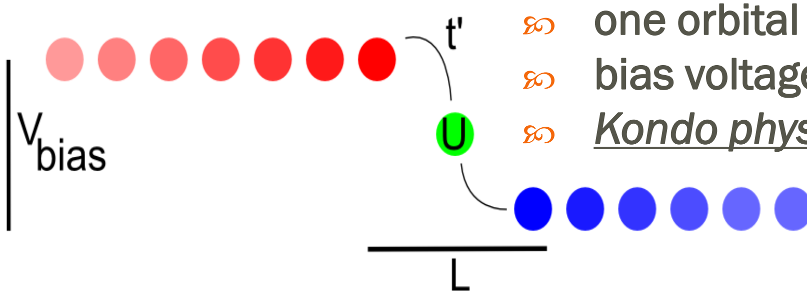
Quantum dot + non equilibrium Variational Cluster Approach





- one orbital + on-site interaction
- bias voltage
- *Kondo physics*

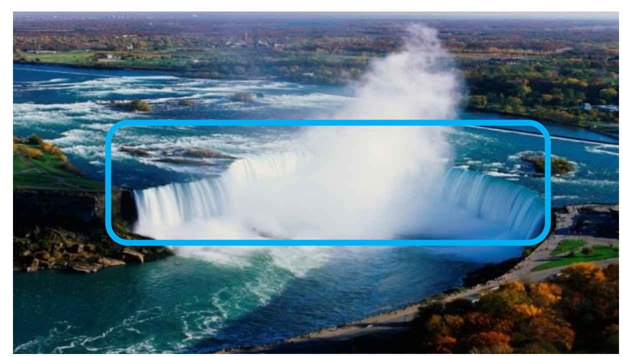
initial reference system *as similar as possible to* **the steady-state system**

$$\tau < \tau_0 : \hat{h} \mapsto \hat{h} + \sum_i x_i \hat{\Delta}_i$$

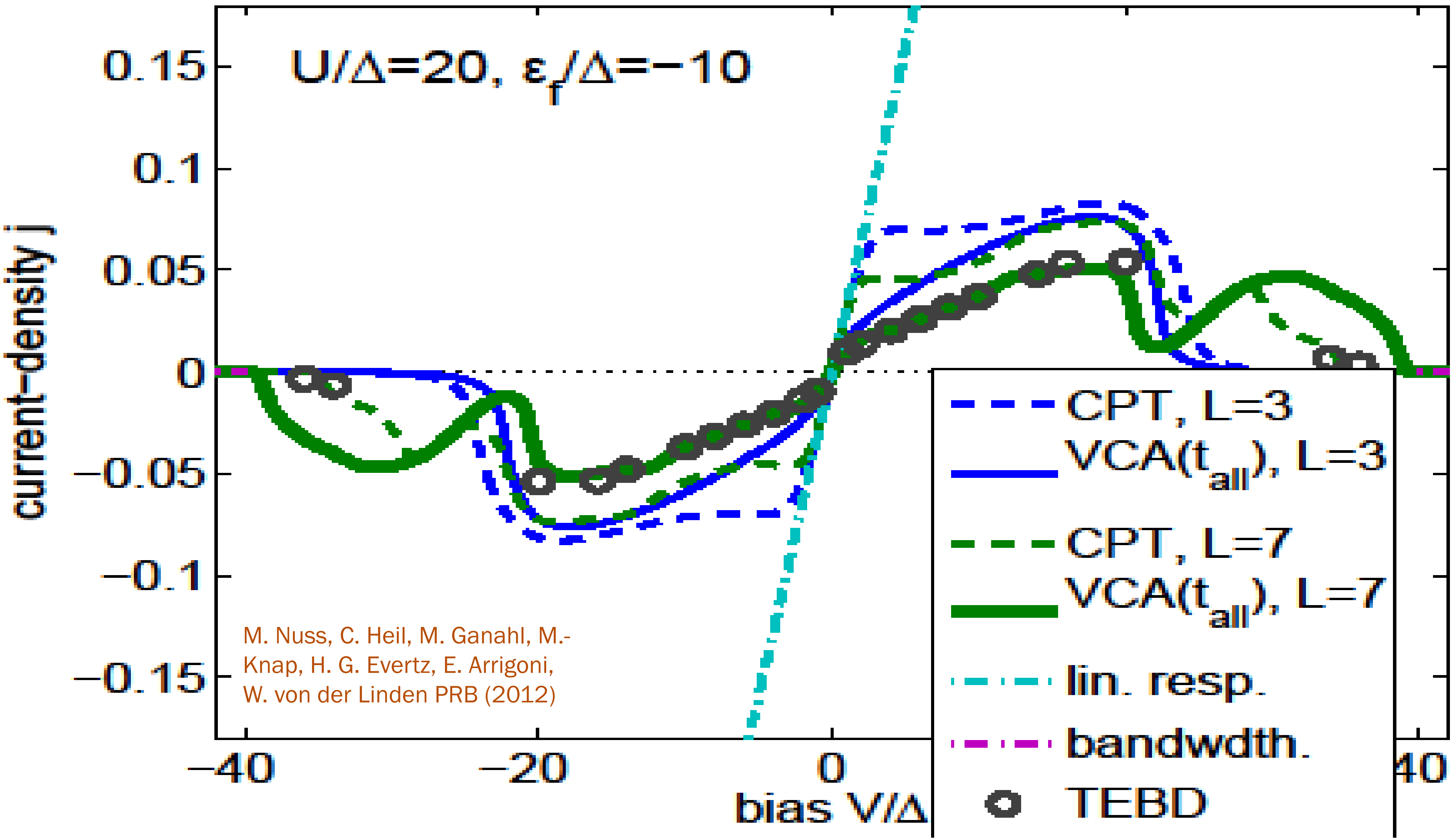
$$\tau > \tau_0 : T \mapsto T - \sum_i x_i \hat{\Delta}_i$$

$$\langle \hat{\Delta}_i \rangle_{\text{initial-state}} \overset{!}{=} \langle \hat{\Delta}_i \rangle_{\text{steady-state}}$$ = self-consistent feedback

$\cong$

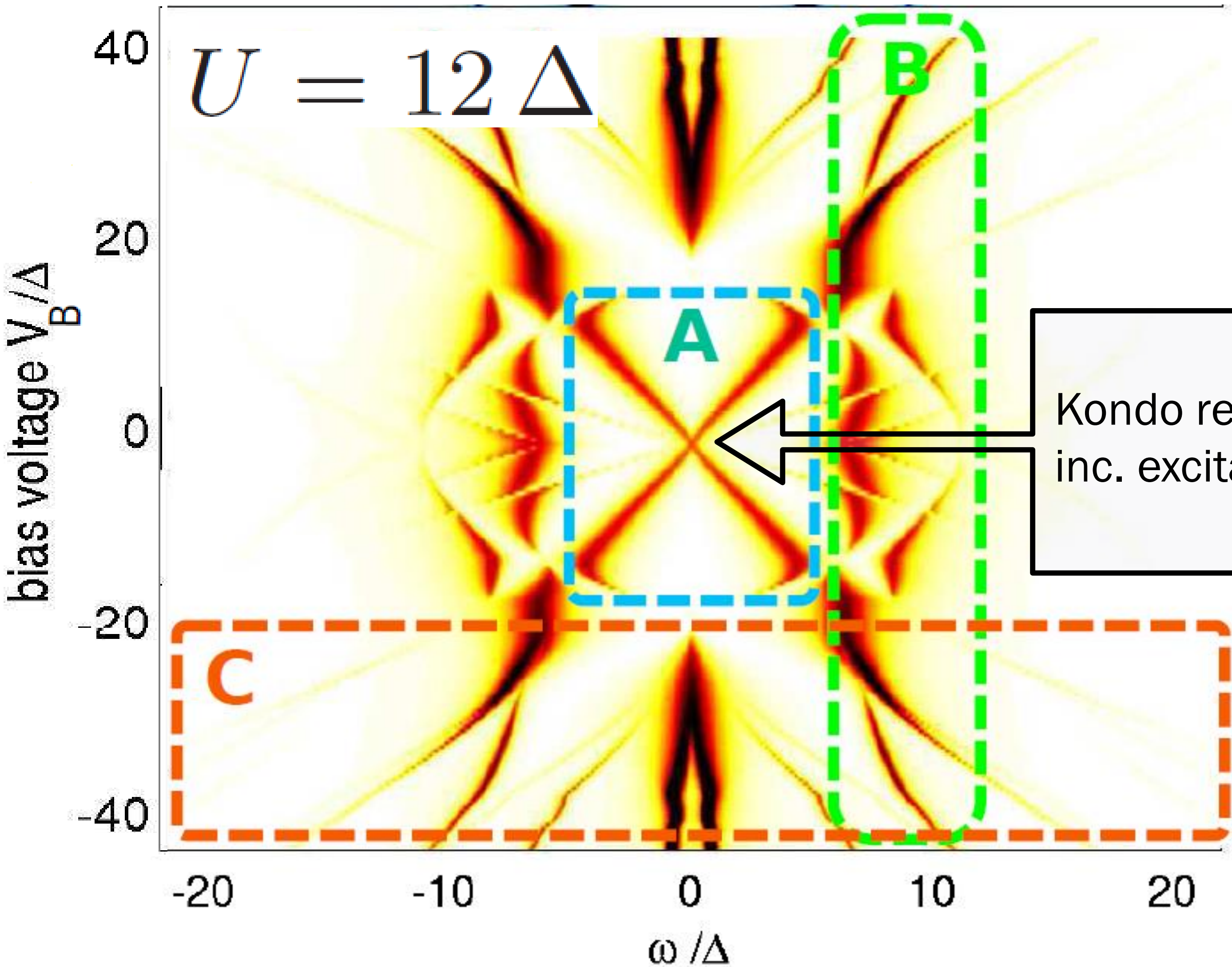


I. current voltage characteristics



M. Nuss, C. Heil, M. Ganahl, M.-Knap, H. G. Evertz, E. Arrigoni, W. von der Linden PRB (2012)

II. Non equilibrium local density of states



A: Kondo

Equilibrium

Kondo resonance @ $\omega = 0$

inc. excitations @ $\omega \cong \pm\frac{U}{2}$

B: Hubbard band

C: lead-band

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III. Conductivity + gate voltage