Concepts for Designing Novel Materials Using Oxide Heterostructures

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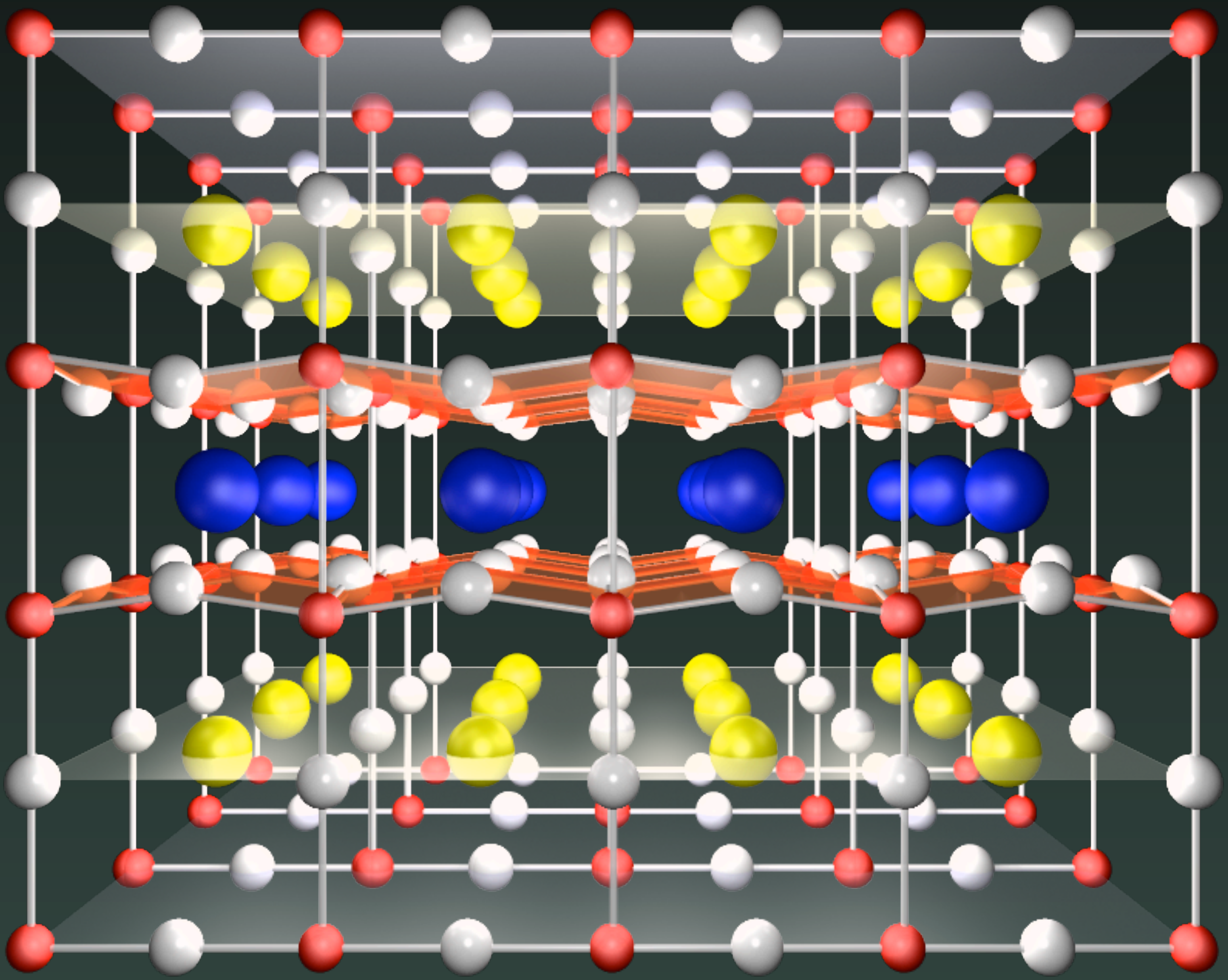
KITP - Materials by Design

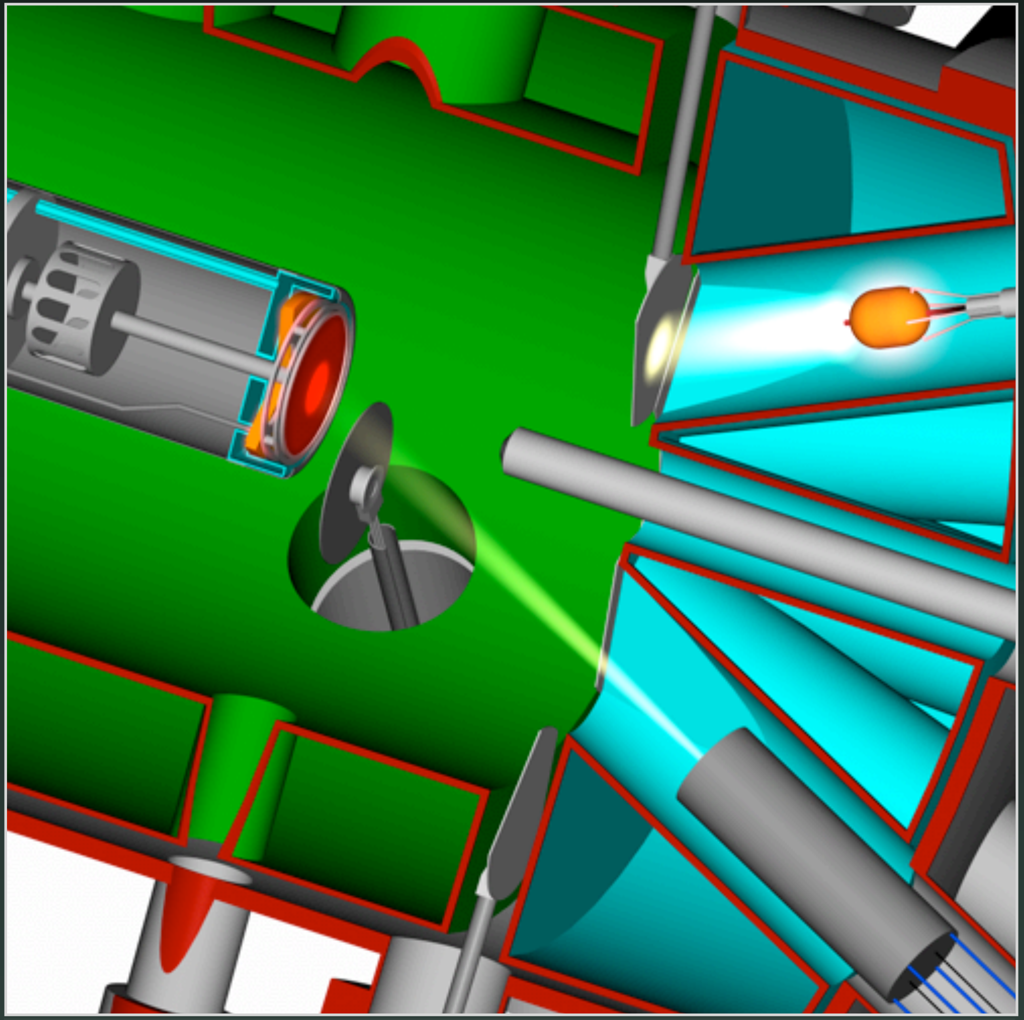


Y
Ba
Cu
O

Growth of Complex Oxide Multilayers

Growth by MBE:

- Bozovic, Eckstein, Schlom (Varian)
- Locquet (IBM Zurich)
- ...



A. Schmehl

Keys for progress

- Activated oxygen
- Accurate rate control
- RHEED
- Substrate termination
- ...

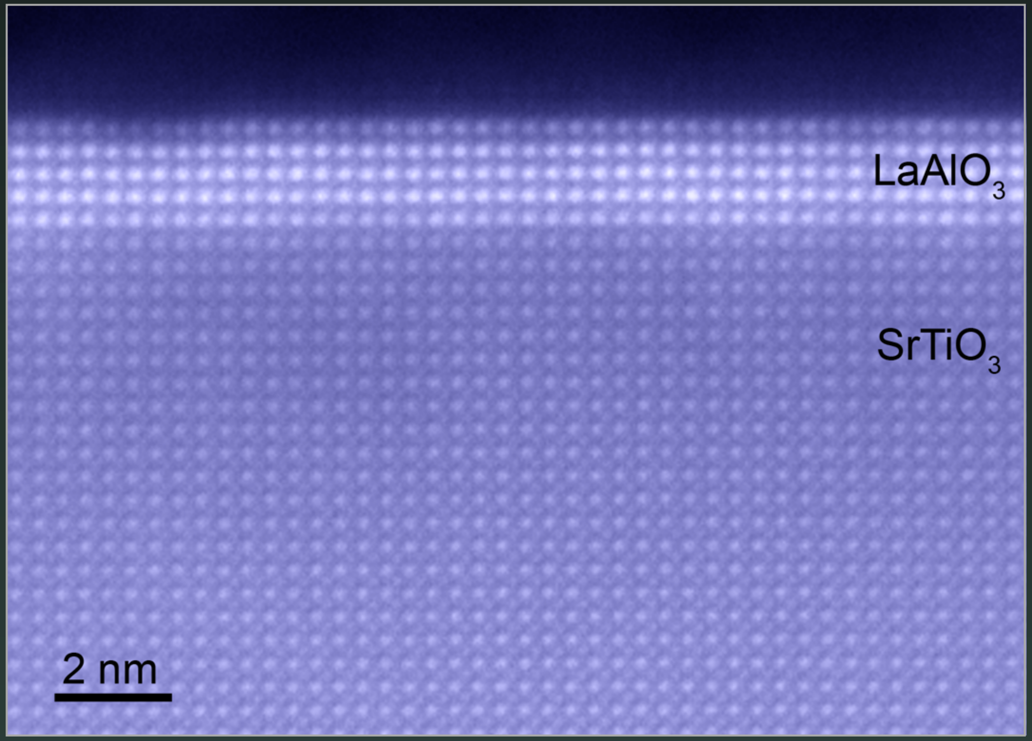
Now, growth of complex oxide multilayers also by pulsed laser deposition

Many Good Reasons to Grow Materials on a Layer-by-Layer Basis

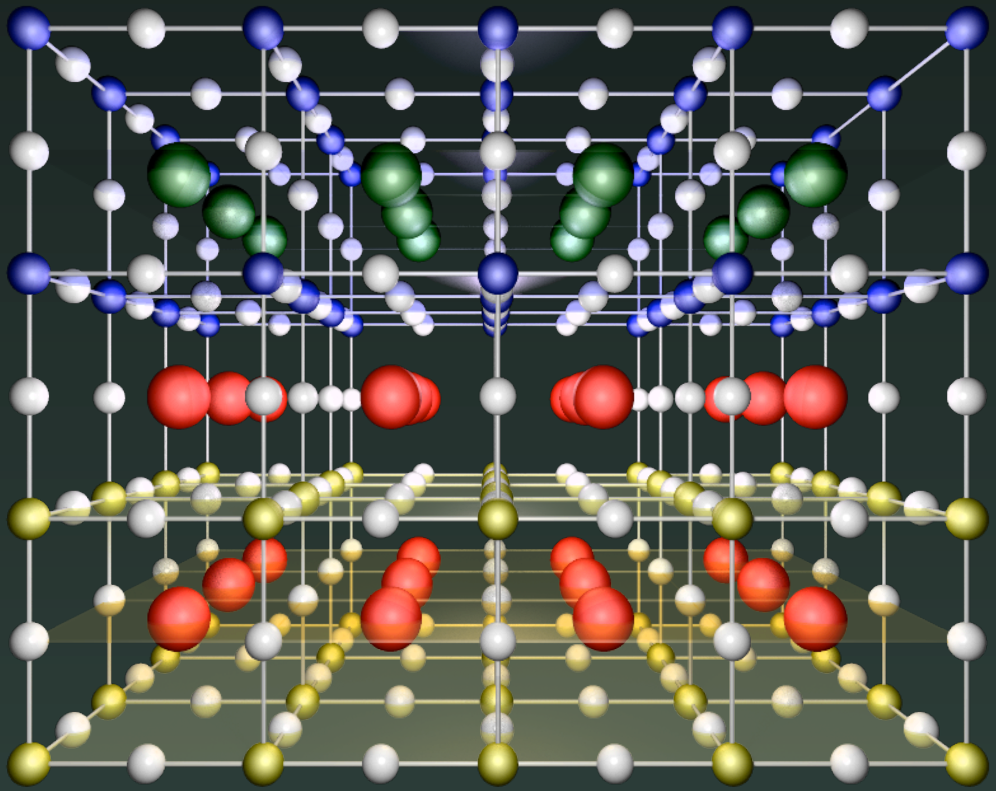
- Freedom to design, grow stacking sequences not realized by nature in the bulk (tricolor lattices)
- Grow device structures (tunnel junctions, FETs, ...)
- Create novel phases and metastable materials, large scale, single-crystal quality, easily measurable
- Utilize epitaxial stress and strain
- Add doping layers
- ...

Also Growth of Well Defined Interfaces Possible

seminal publications: Ohtoma & Hwang (2002, 2004)



L. Fitting Kourkoutis,
D. Muller (Cornell)



$LaAlO_3$ - $SrTiO_3$

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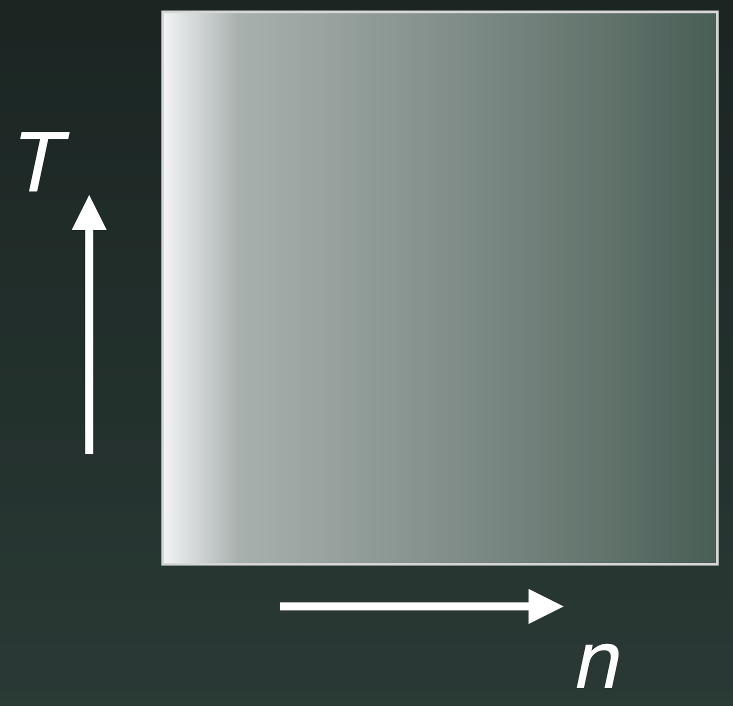
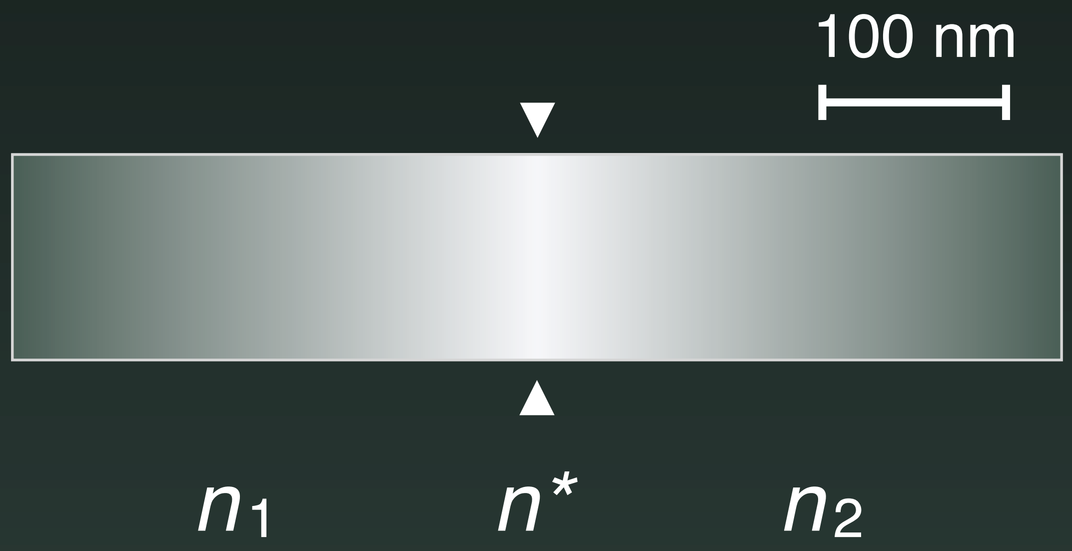
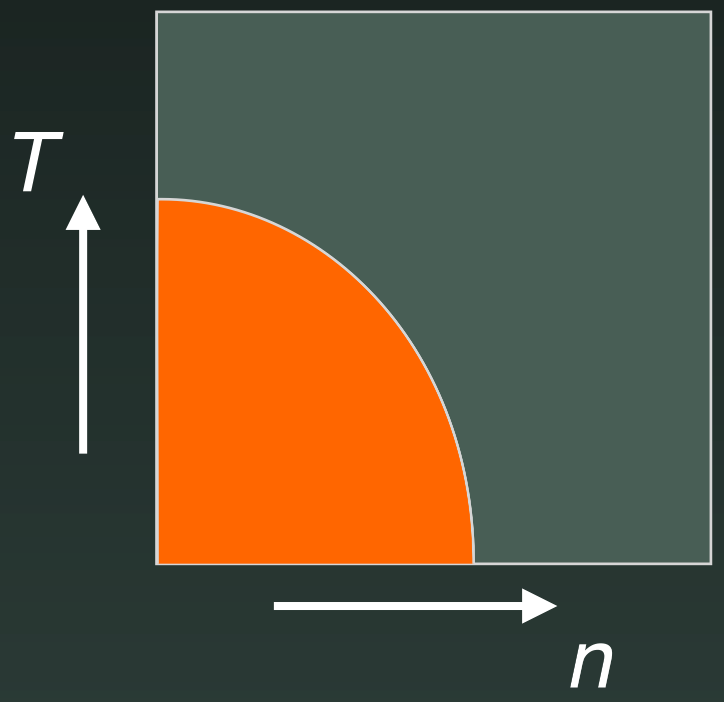
  large scale, single-crystal quality, easily measurable
- Utilize epitaxial stress and strain
- Add doping layers
- ...
- Create materials with novel electronic systems generated at interfaces in systems with strong electronic correlations

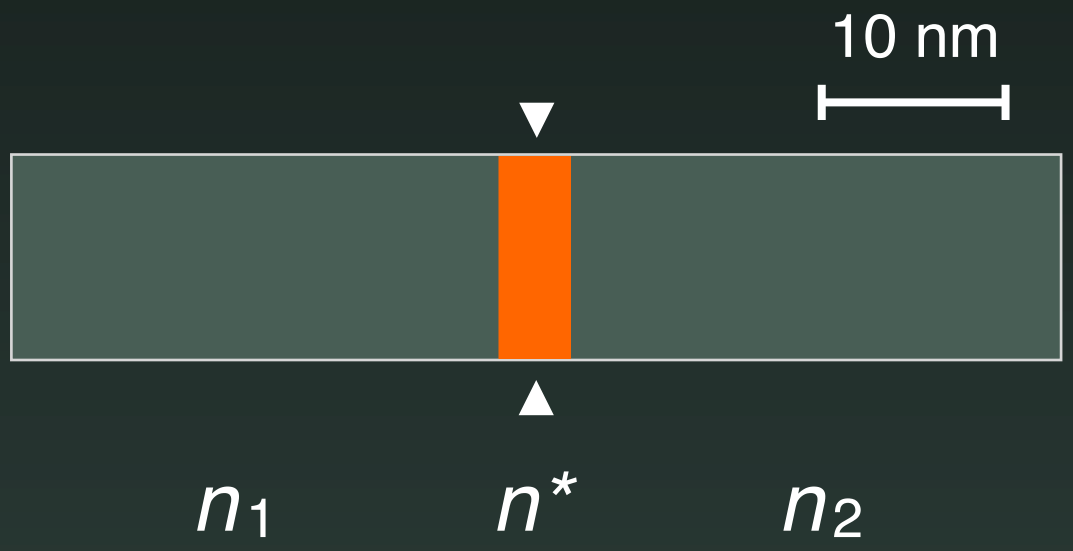
Standard Semiconductors - Band Bending at Interfaces



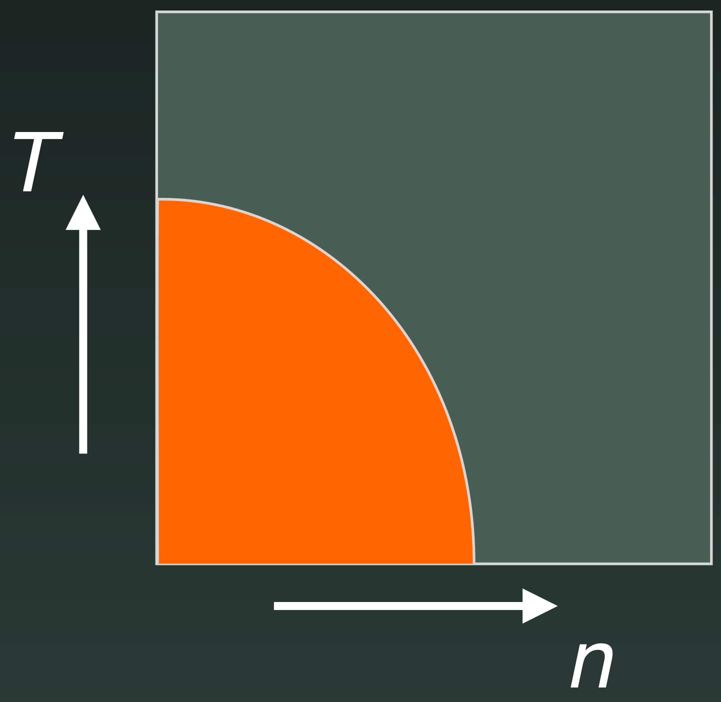


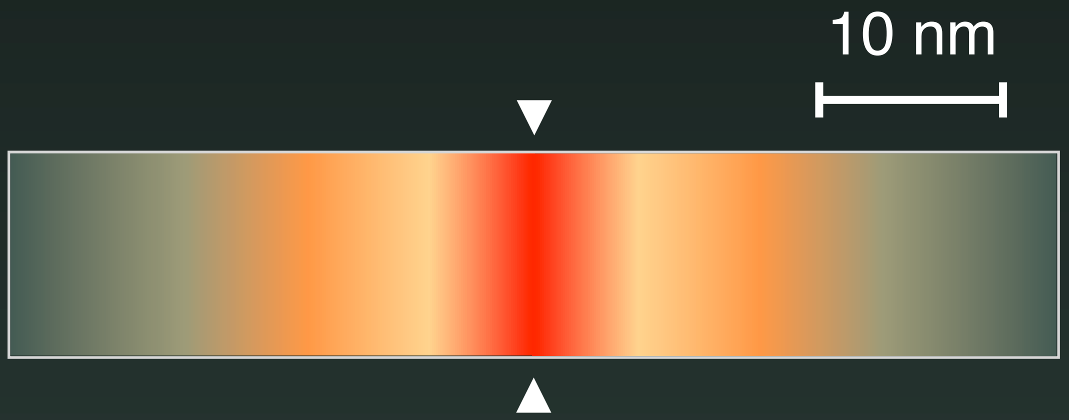
Electronic Reconstruction at Interfaces in Correlated Electronic Systems





Novel Electronic Phases at Interfaces in Correlated Electronic Systems





n_1 n^* n_2

J_1 J^* J_2

t_1 t^* t_2

U_1 U^* U_2

Plus:

- charge transfer, space charge effects, band bending ...
- possible misalignment
- orbital reconstruction
- altered:
  - Madelung energies
  - hybridization (also hybridization through interface)
  - screening, dielectric and magnetic environment
  - orbital order (including frustrated orbital order)
  - spin order (including frustrated spin order)

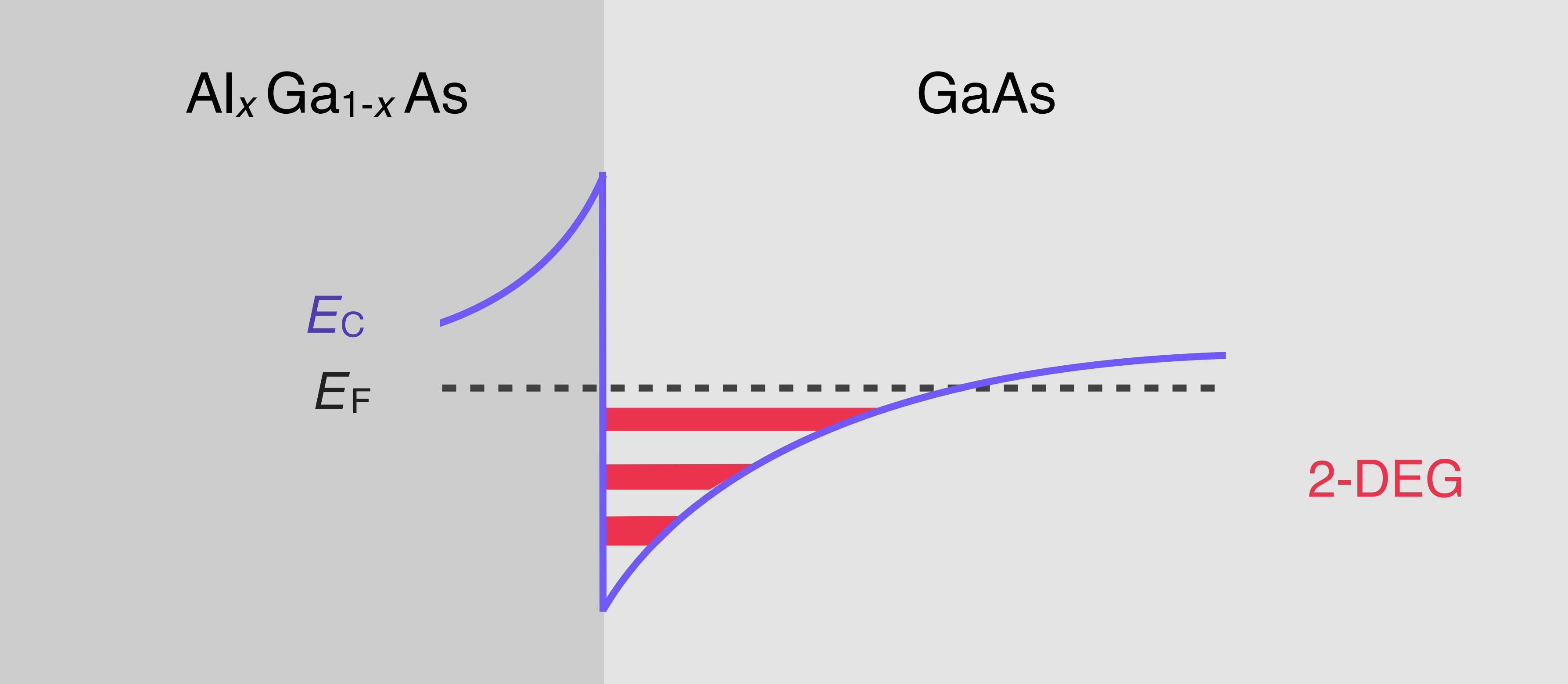
talks by Tchakhalian, Eckstein, Hwang, Abbamonte, Pentcheva, Okamoto, Bozovic, ...

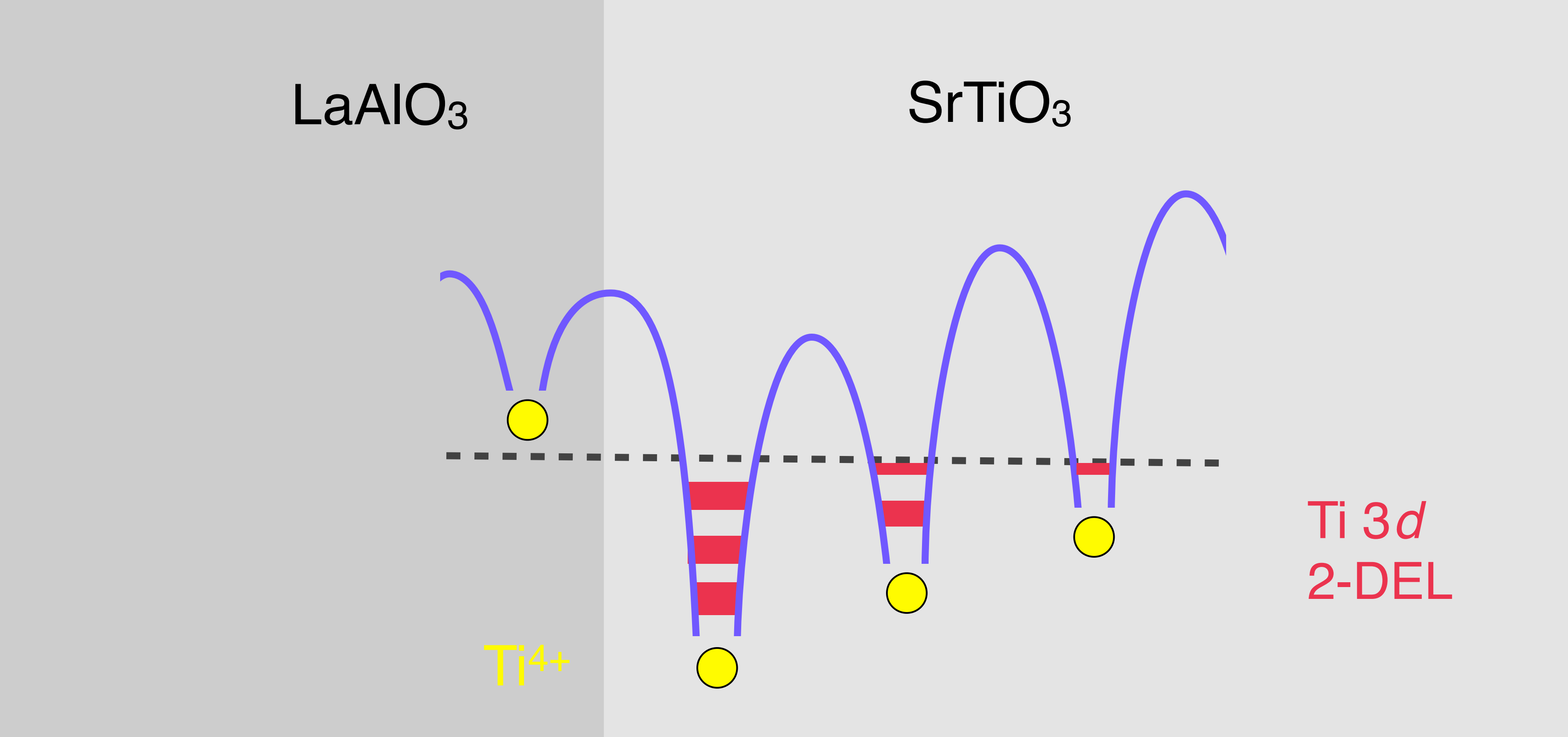
Plus:

- use gradients
- multiferroic coupling generated by interface
- use reduction of dimensionality
- decouple mobile carriers from excitations (superconducting pairing!)
- non-equilibrium effects, induced for example by current injection

talks by Tchakhalian, Eckstein, Hwang, Abbamonte, Pentcheva, Okamoto, Bozovic, ...

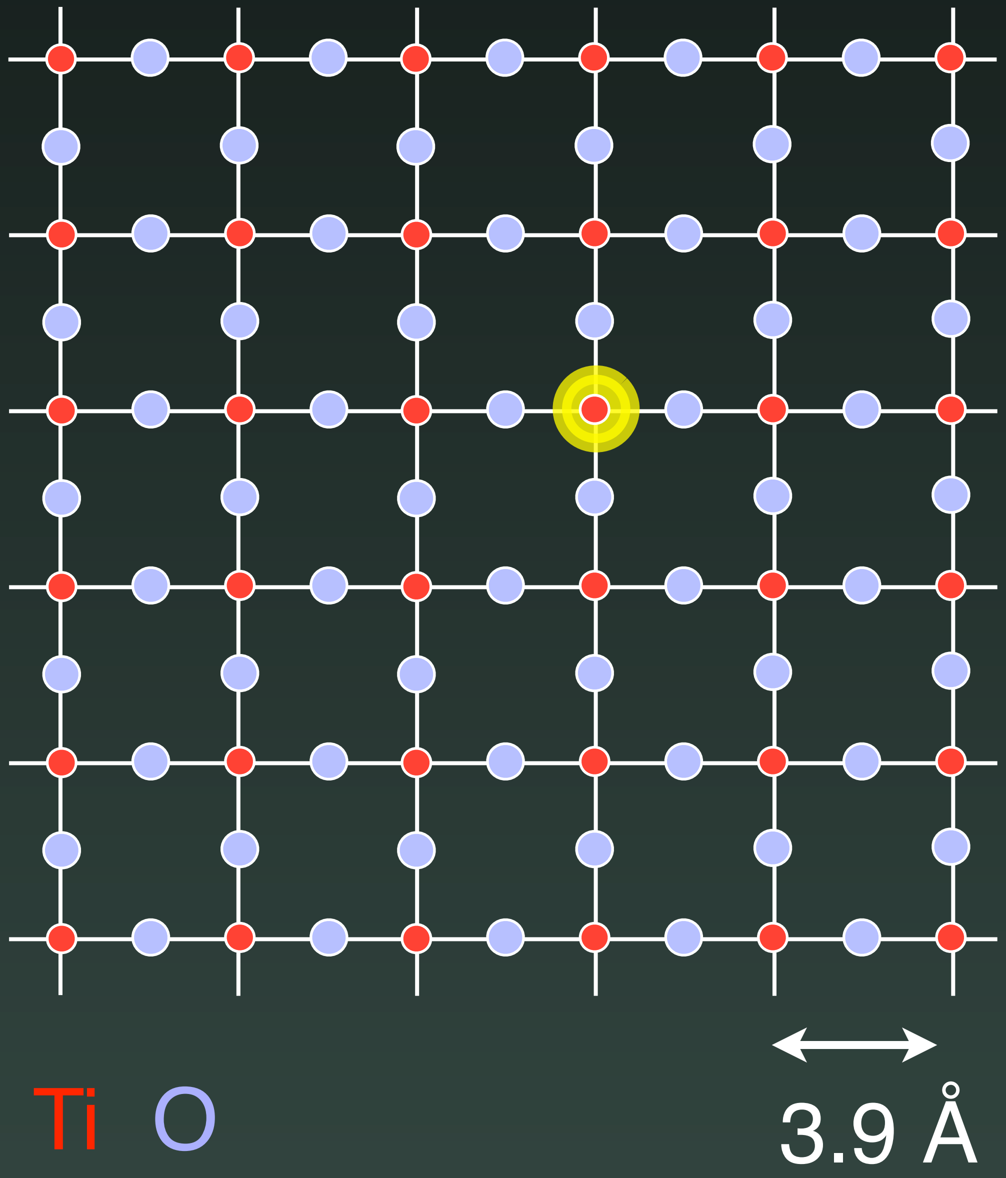
The LaAlO$_3$-SrTiO$_3$ Interface Electronic System





Martin Breitschaft *et al.*, arXiv:0907.1176

The $LaAlO_3$-$SrTiO_3$ Interface Electronic System



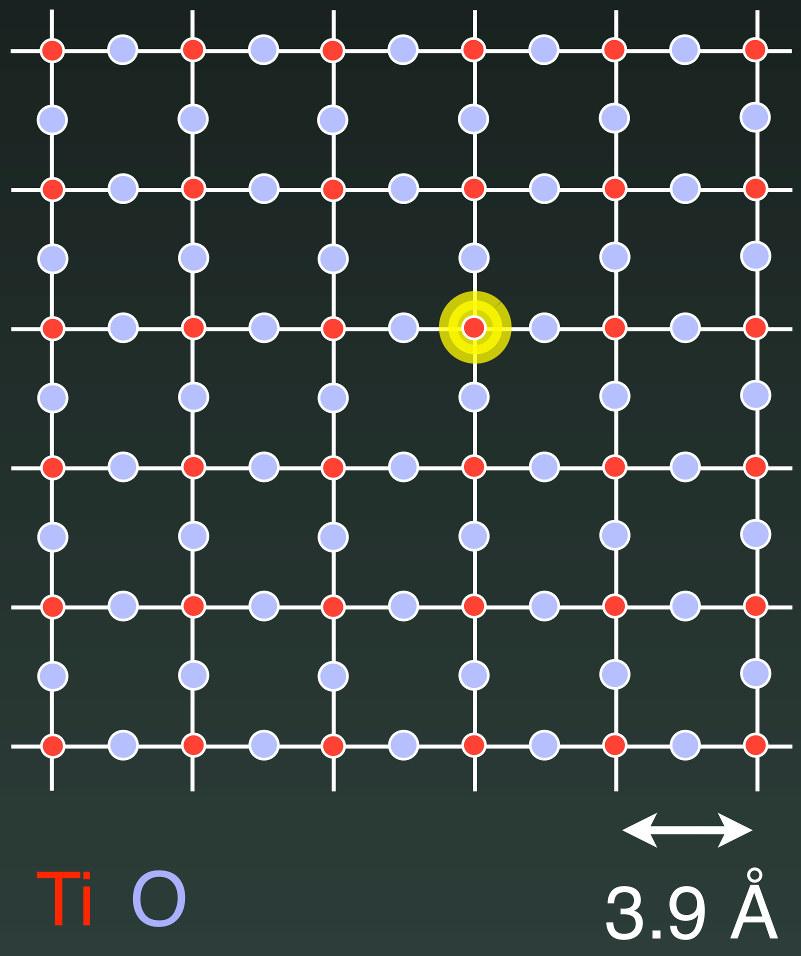
$n \sim 5 \times 10^{12} - 5 \times 10^{14}\, /\mathrm{cm}^2$

$r_s = (1/\pi a_B^2 n)^{1/2} \sim 5 - 50$

TiO_2-plane

The Energy of the Electron System

$$E = E_{\mathrm{H}} + E_{\mathrm{kin}} + E_{\mathrm{xc}} + E_{\mathrm{corr}} + E_{\mathrm{ext}}$$



The Energy of a Capacitor with LaAlO3-SrTiO3 Plates

$$E = E_{\mathrm{H}} + \sum_i E_{\mathrm{kin},i} + \sum_i E_{\mathrm{xc},i} + \sum_i E_{\mathrm{corr},i} + \sum_i E_{\mathrm{ext},i}$$

$$E = \frac{1}{2C} Q^2$$

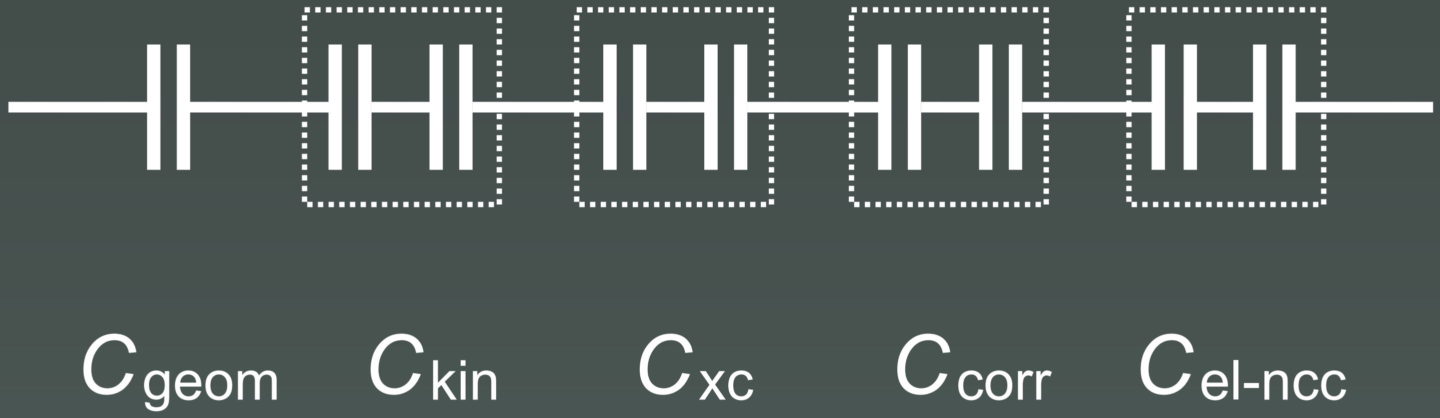
for homogenous electron systems:

$$E_{\mathrm{H}} + \sum_i E_{\mathrm{ext},i} = \frac{d}{\varepsilon_0 \varepsilon_r A} Q^2$$

neglecting tunneling and exchange contributions between the plates

The General Equation for the Capacitance of a Capacitor

$$1/C = 1/C_{\text{geom}} + \sum_i 1/C_{\text{kin},i} + \sum_i 1/C_{\text{xc},i} + \sum_i 1/C_{\text{corr},i} + \sum_i 1/C_{\text{el-ncc},i}$$



the electron systems of the electrodes have their own capacities

When is $C = C_{\text{geom}}$?

$$1/C = \underset{\dfrac{d}{\varepsilon_r}}{1/C_{\text{geom}}} + \sum_i \underset{a_{\text{B}}}{1/C_{\text{kin},i}} + \sum_i \underset{r_s a_{\text{B}}}{1/C_{\text{xc},i}} + \sum_i \underset{r_s a_{\text{B}}}{1/C_{\text{corr},i}} + \sum_i \underset{0}{1/C_{\text{el-ncc},i}}$$

Transition to Quantum Electronics

$$C = \varepsilon_0 \varepsilon_r \frac{A}{d}$$ is only valid in the limit of large d/ε_{r}

The General Equation for the Capacitance of a Capacitor

$$1/C = 1/C_{\mathrm{geom}} + \sum_i 1/C_{\mathrm{kin},i} + \sum_i 1/C_{\mathrm{xc},i} + \sum_i 1/C_{\mathrm{corr},i} + \sum_i 1/C_{\mathrm{el\text{-}ncc},i}$$

2D Homogenous Electron Systems:

$$\frac{C_{\mathrm{geom}}}{A} = \frac{\varepsilon_0 \varepsilon_{\mathrm{r}}}{d}$$

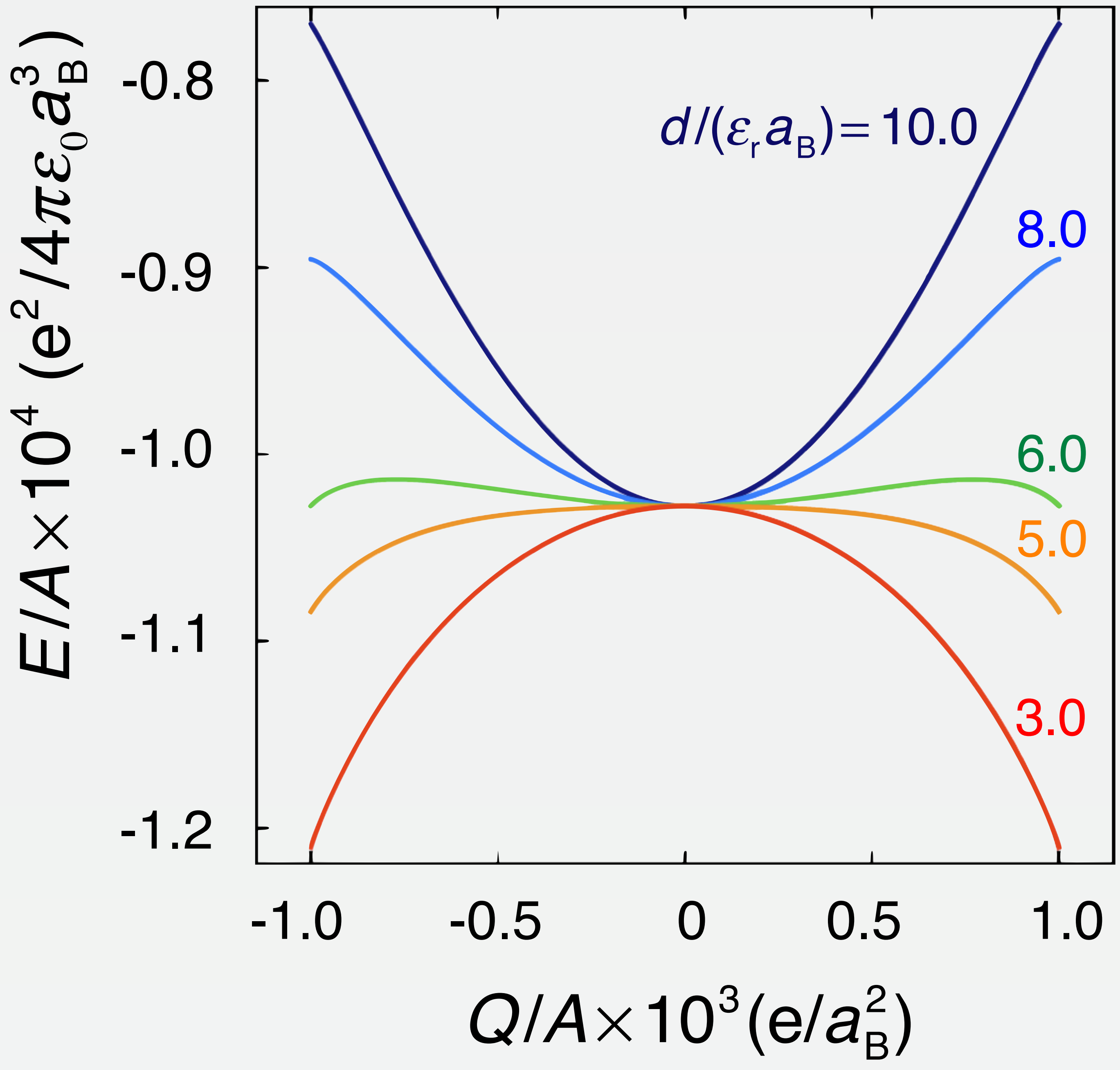
$$\frac{C_{\mathrm{kin},i}}{A} = 4\varepsilon_0 \frac{m_i^*/m}{a_{\mathrm{B}}} = \mathrm{e}^2 \rho_i^{(2\mathrm{D})}\left(E_{\mathrm{F}}\right)$$

$$C_{\mathrm{el\text{-}ncc},i} = 0$$

$$\frac{C_{\mathrm{xc},i}}{A} = -2^{3/2} \frac{\varepsilon_0 \varepsilon_{\mathrm{eff},i}}{a_{\mathrm{B}} r_{\mathrm{s}}}$$

$$\frac{C_{\mathrm{corr},i}}{A} = -\frac{\varepsilon_0 \varepsilon_{\mathrm{eff},i}}{a_{\mathrm{B}}} f(r_{\mathrm{s}})$$

Energy of Capacitors with 2D Electron Gases in the Plates



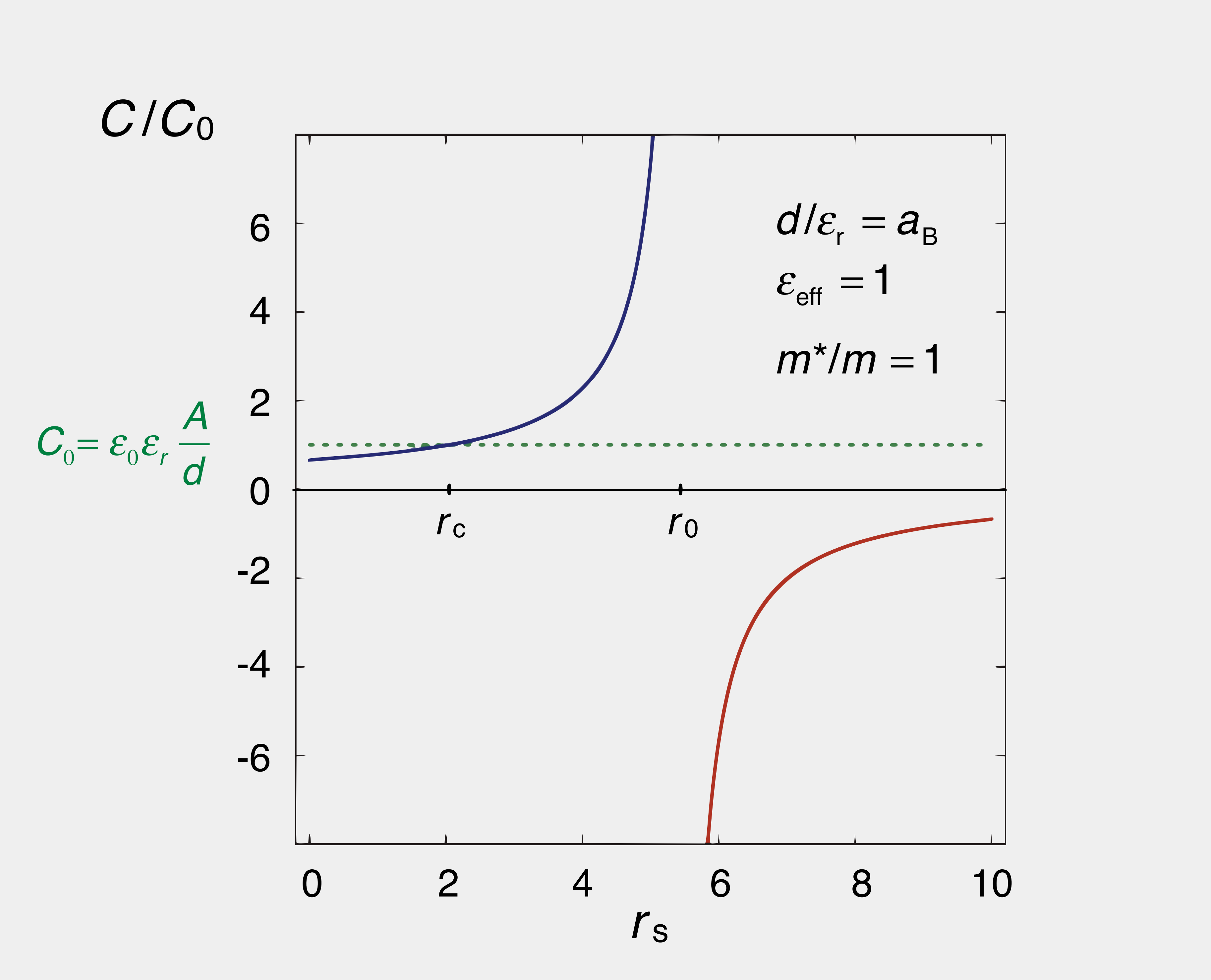
$n = 3.6 \times 10^{13}\ \mathrm{cm}^{-2}$

$m^*/m = 1$

$\varepsilon_{\mathrm{eff}} = 1$

T. Kopp and J. Mannhart, J. Appl. Phys. **106**, 064504 (2009)

Capacitance of a Capacitor with 2D Electron Gases in the Plates



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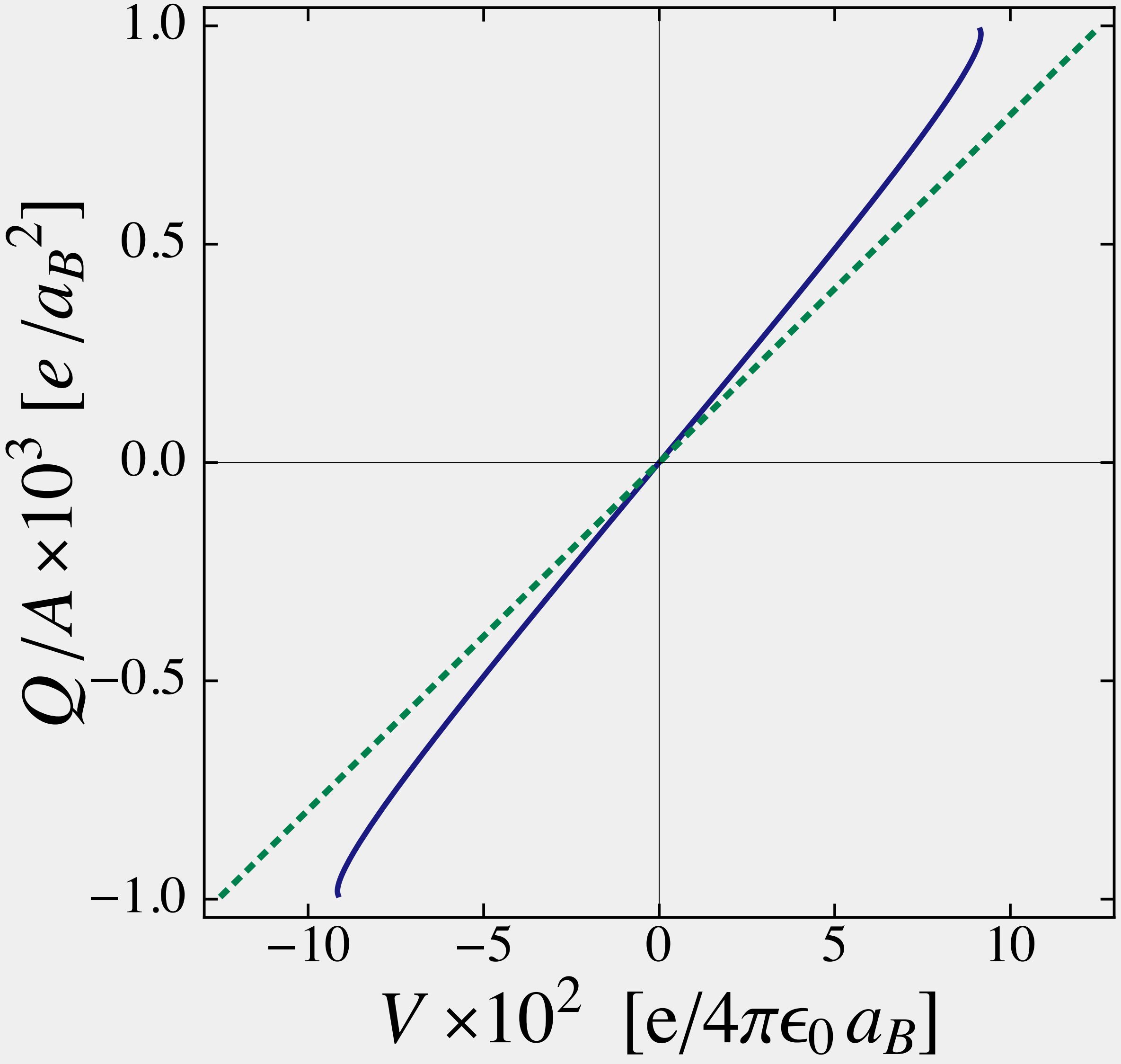
$C(V)$-Characteristic of a Capacitor with 2D Electron Gases in the Plates



$d/(\varepsilon_r a_B) = 10.0$

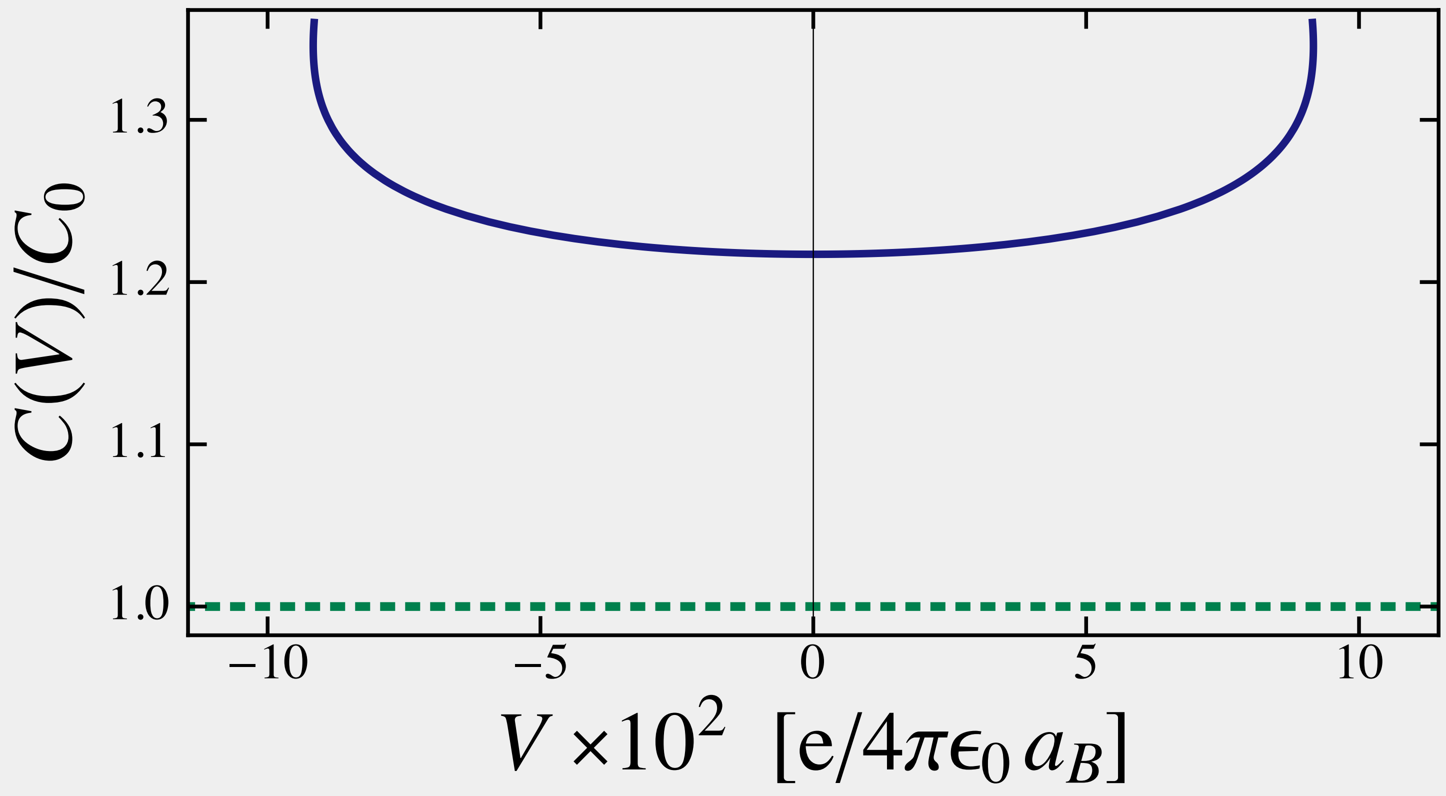
$n = 3.6 \times 10^{13}$ cm^{-2}

$m^*/m = 1$

$\varepsilon_{\mathrm{eff}} = 2.5$

T. Kopp and J. Mannhart, J. Appl. Phys. **106**, 064504 (2009)

$C(V)$-Characteristic of a Capacitor with 2D Electron Gases in the Plates



$d/(\varepsilon_r a_B) = 10.0$

$n = 3.6 \times 10^{13}$ cm^{-2}

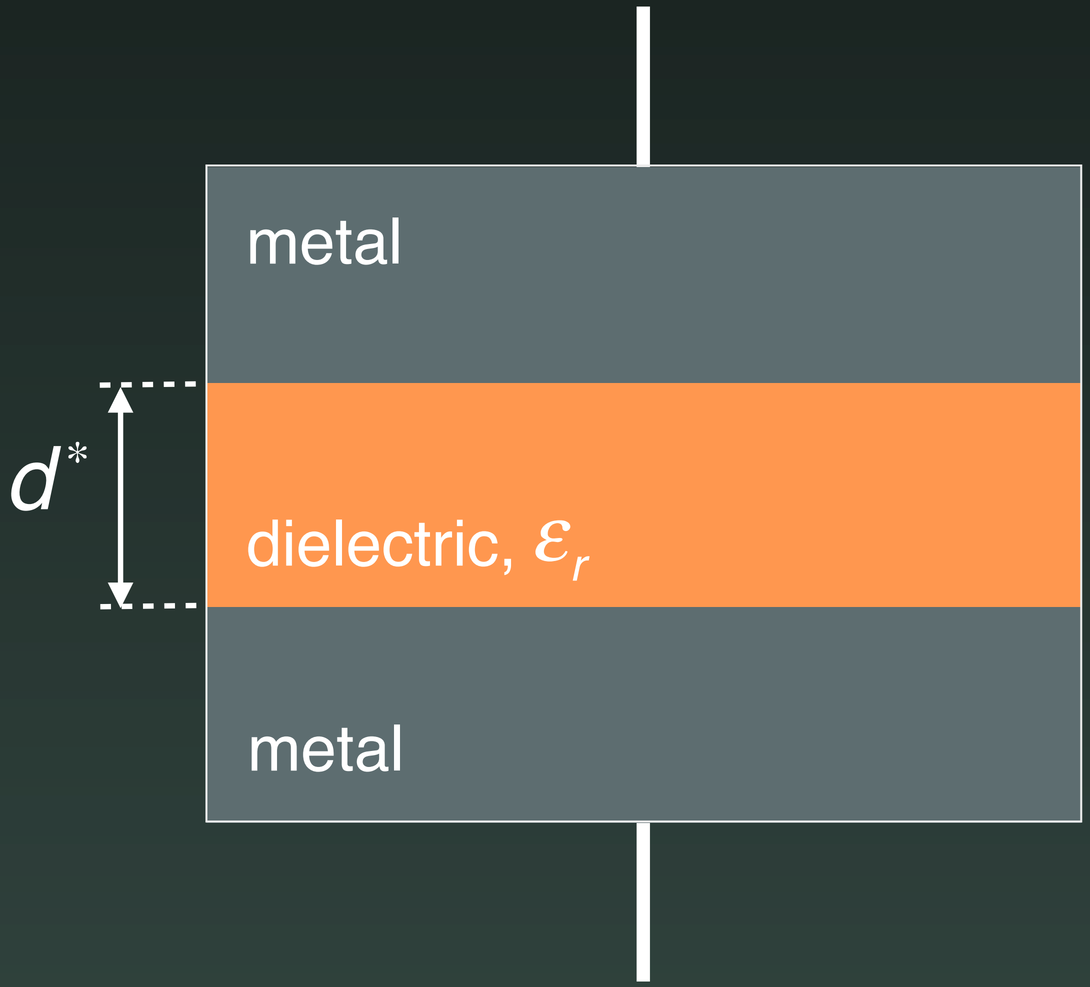
$m^*/m = 1$

$\varepsilon_{\mathrm{eff}} = 2.5$

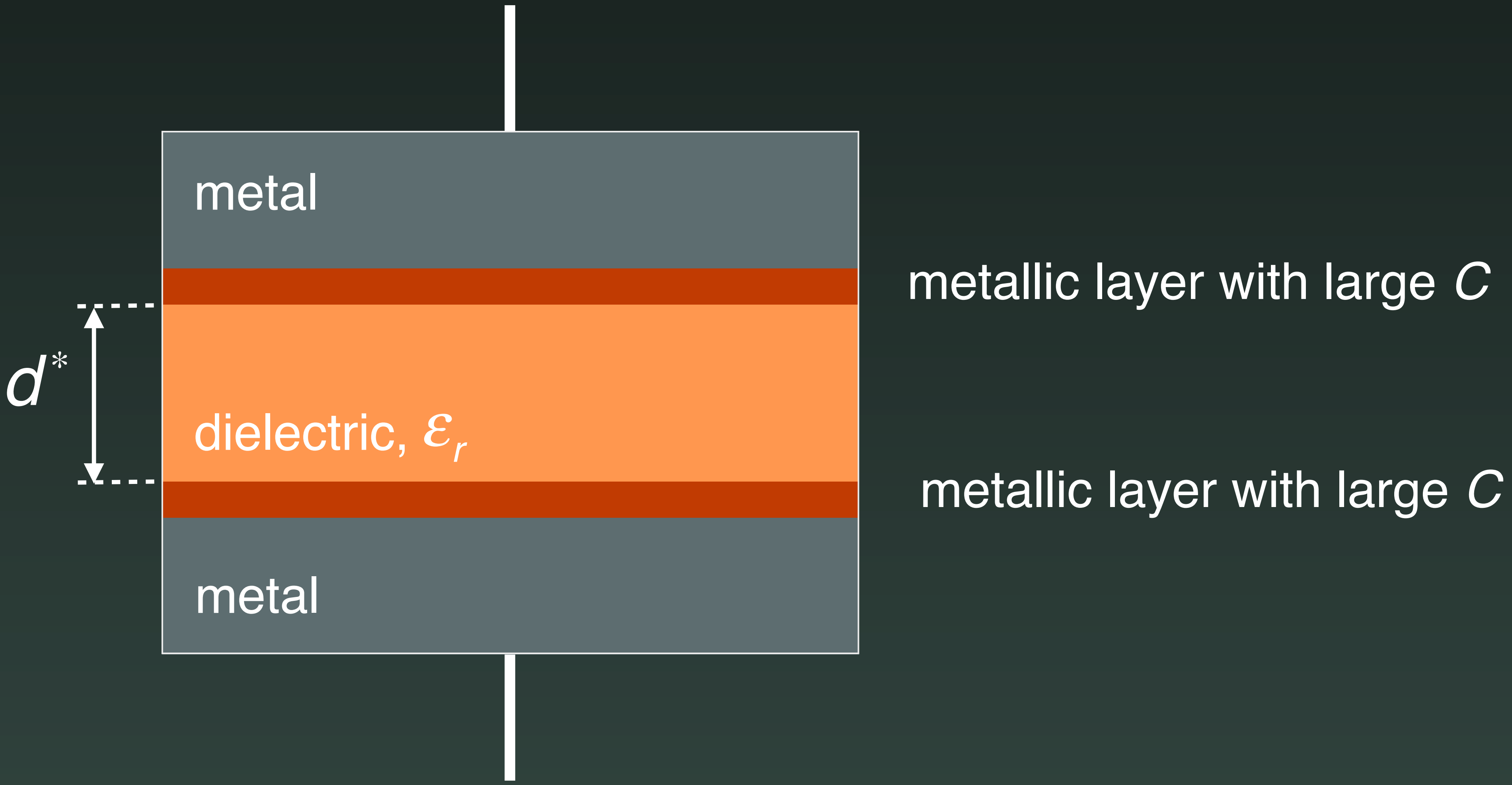
$$C = \varepsilon_0 \varepsilon_r \frac{A}{d}$$

T. Kopp and J. Mannhart, J. Appl. Phys. **106**, 064504 (2009)

Capacitor with $C > \varepsilon_0 \varepsilon_r \frac{A}{d^*}$



Capacitor with $C > \varepsilon_0 \varepsilon_r \dfrac{A}{d^*}$

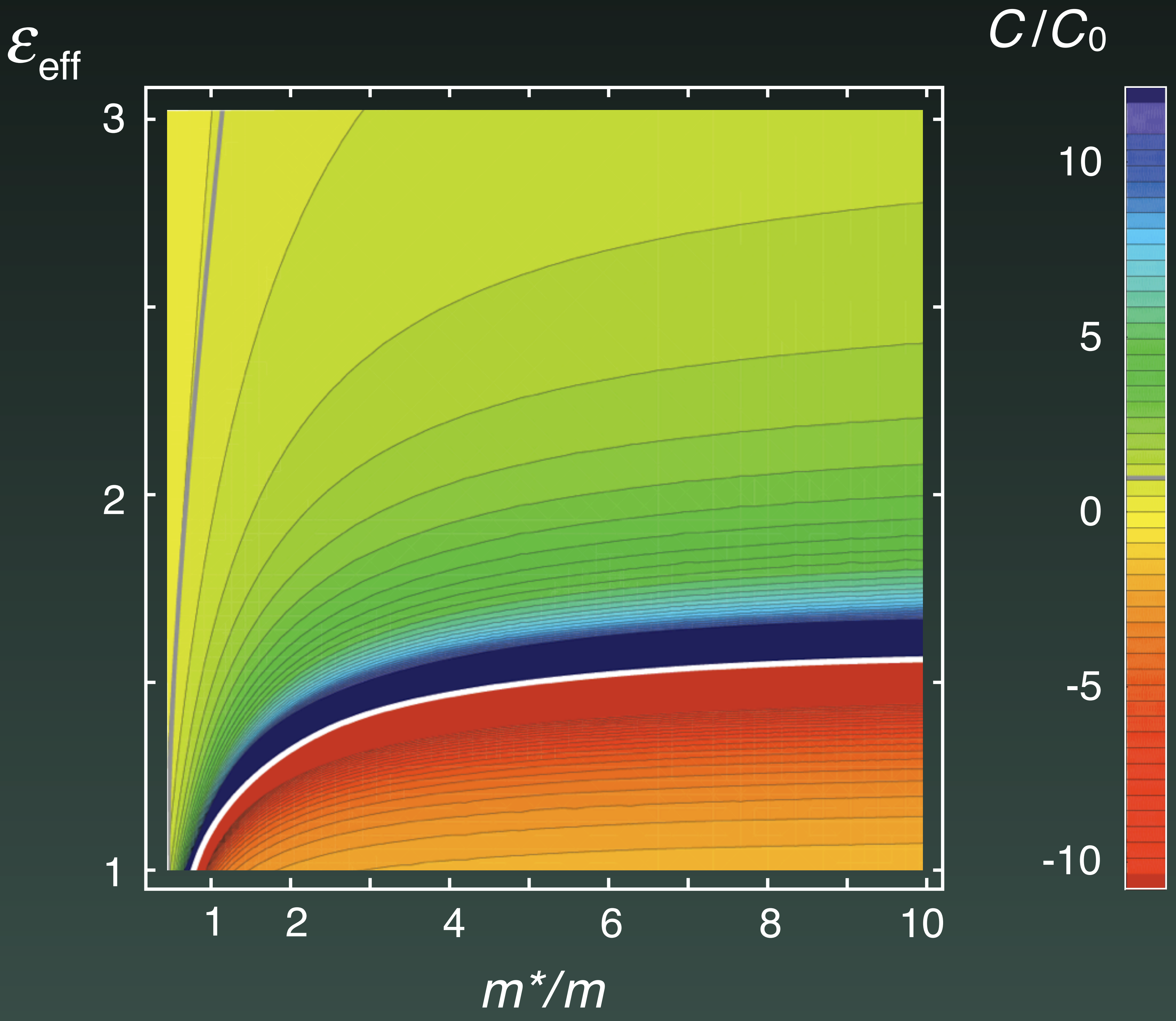


Materials of Interest

- interface electron systems, *e.g.*, $LaAlO_3$-$SrTiO_3$, $LaVO_3$-$SrTiO_3$
- standard metals with low carrier densities
- strongly correlated systems, transition-metal oxides
- graphene, nanotubes (Latessa *et al.*, PRB (2005): Ilani *et al.*, Nature Physics (2006))
- van-Hove systems (for small-*C* devices)

...

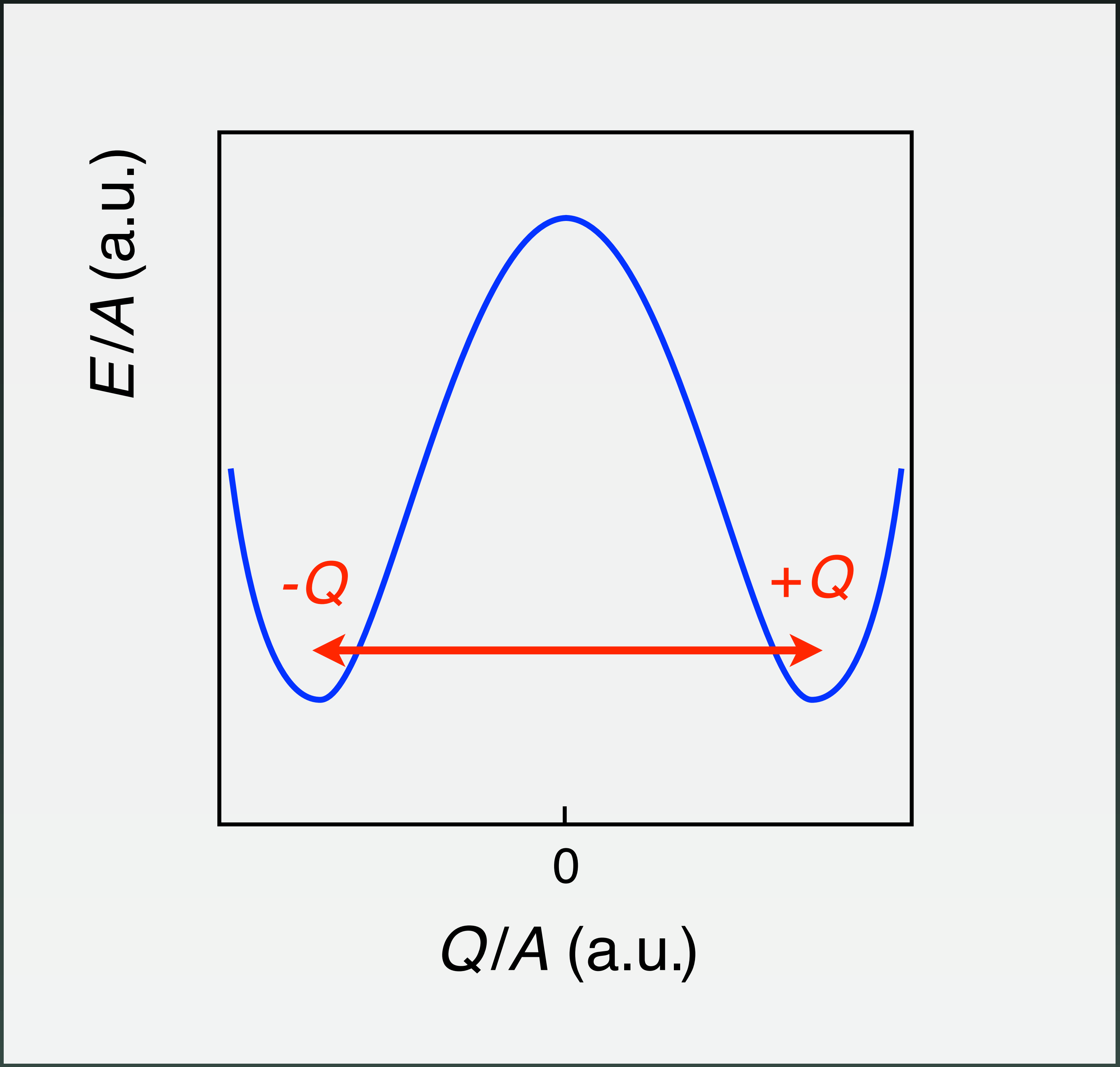
Capacitance of a Capacitor with 2D Electron Gases in the Plates



$r_s = 7$

$d/\varepsilon_r = 1.23\, a_B$

States of Capacitors with Negative *C*



switching between the two states with bias pulses

- memory devices?
- coherent states?

Ferroelectric-type Behavior without Ferroelectric Compound

Reminiscent of ferroelectrics, but

- no ferroelectric compounds
- no discharging, no depolarization
- electronic effect, fast

Capacitors with Electron Systems with Negative Compressibilities

1. negative compressibilities of the electron system cause instabilities:
   charge density wave, stripes, other inhomogeneous phases

2. coherent states of both plates? (Moon *et al.* (1995), Zheng *et al.* (1997))

3. charge imbalance between plates, negative C

controlling parameters:

- material properties (strongly correlated systems?)
- device geometry
- experimental conditions (T, V, history)

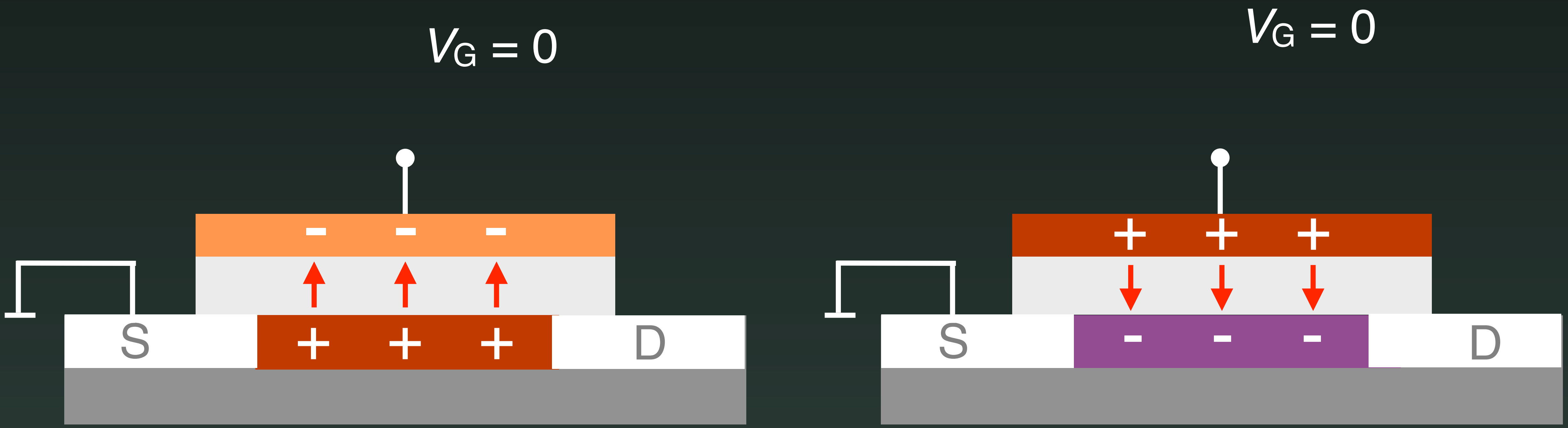
Other Work on Total Negative Capacitances

for example:

- generated by using amplifier circuits (Beavis, 1954)
- found for electron injection through interfaces (Omura, 2000)
- proposed for structures comprising ferroelectrics (Salahuddin and Datta, 2007)

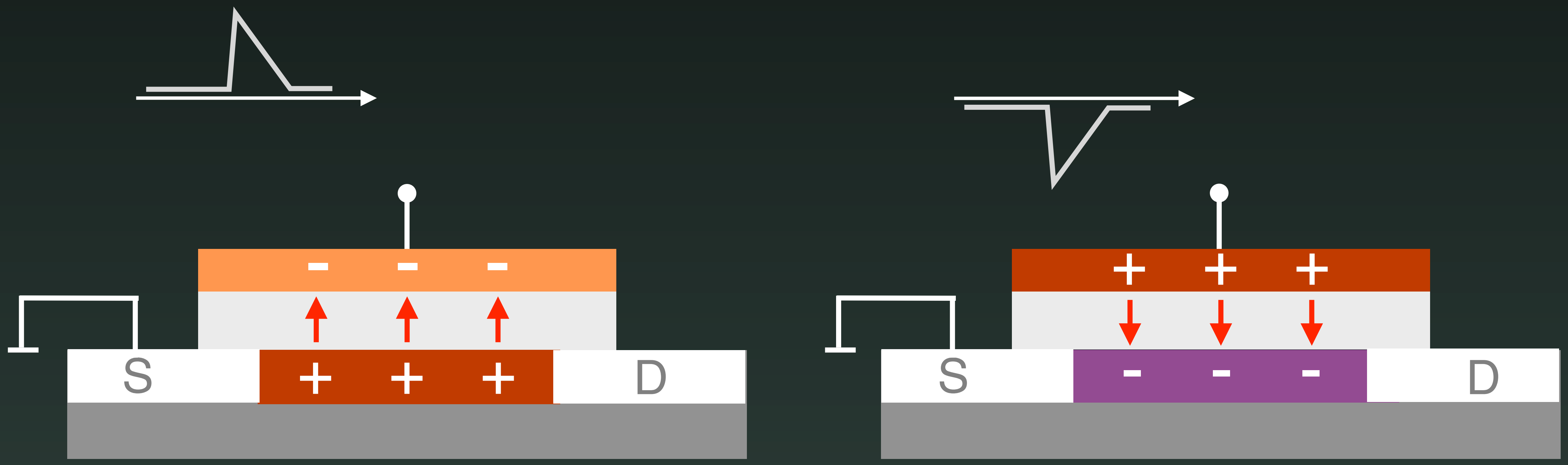
plus considerable experimental and theoretical work on negative contributions to a total positive capacitance

MOSFETs with Negative-*C* Gate Stacks



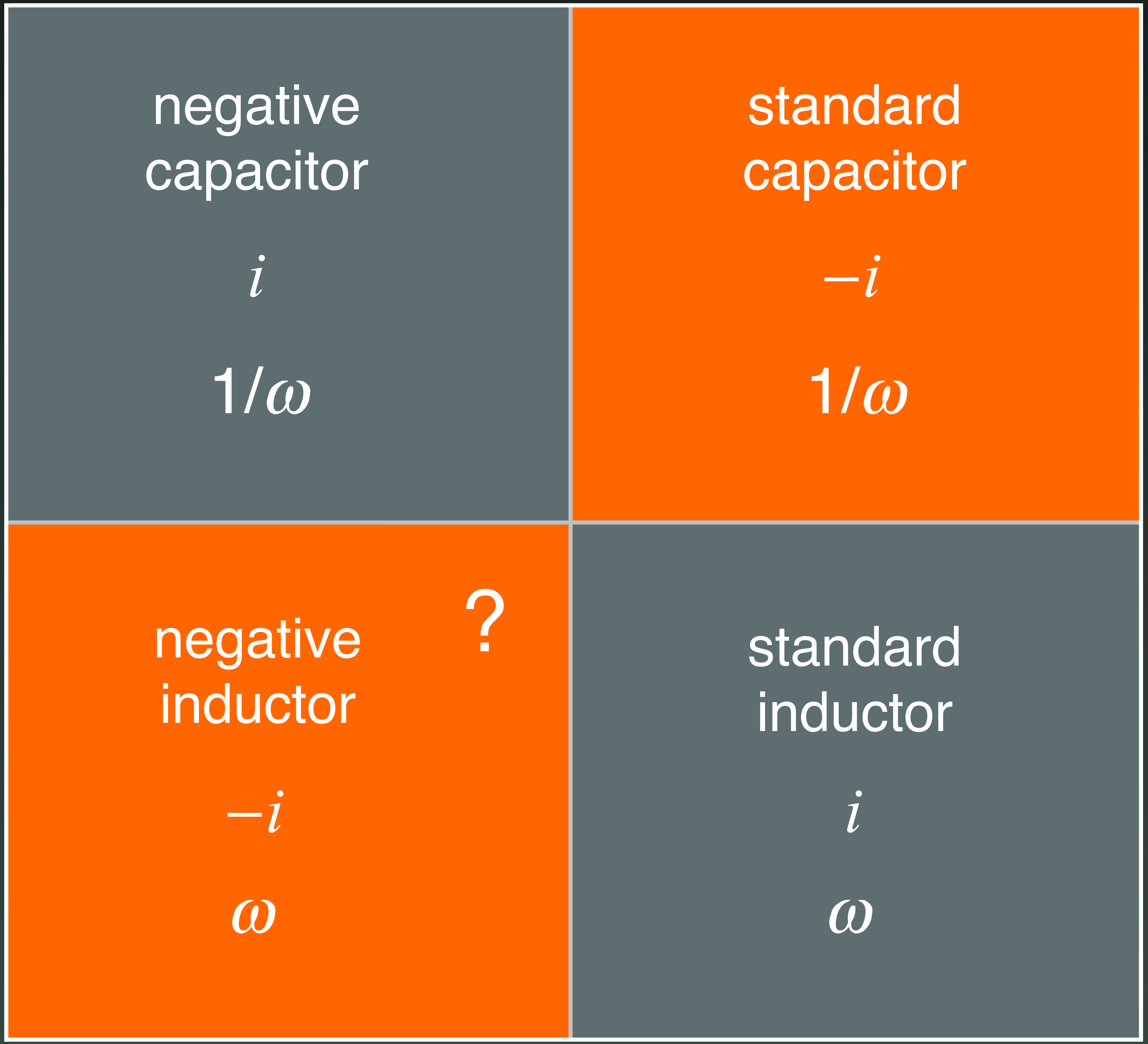
Information carriers: electrons with neutralized global Coulomb energies

MOSFETs with Negative-*C* Gate Stacks



Information carriers: electrons with neutralized global Coulomb energies

Overview on Impedances



negative capacitor i $1/\omega$	standard capacitor $-i$ $1/\omega$
negative inductor ? $-i$ ω	standard inductor i ω

$$Z = -i\,\frac{1}{\omega C}$$

phase shift

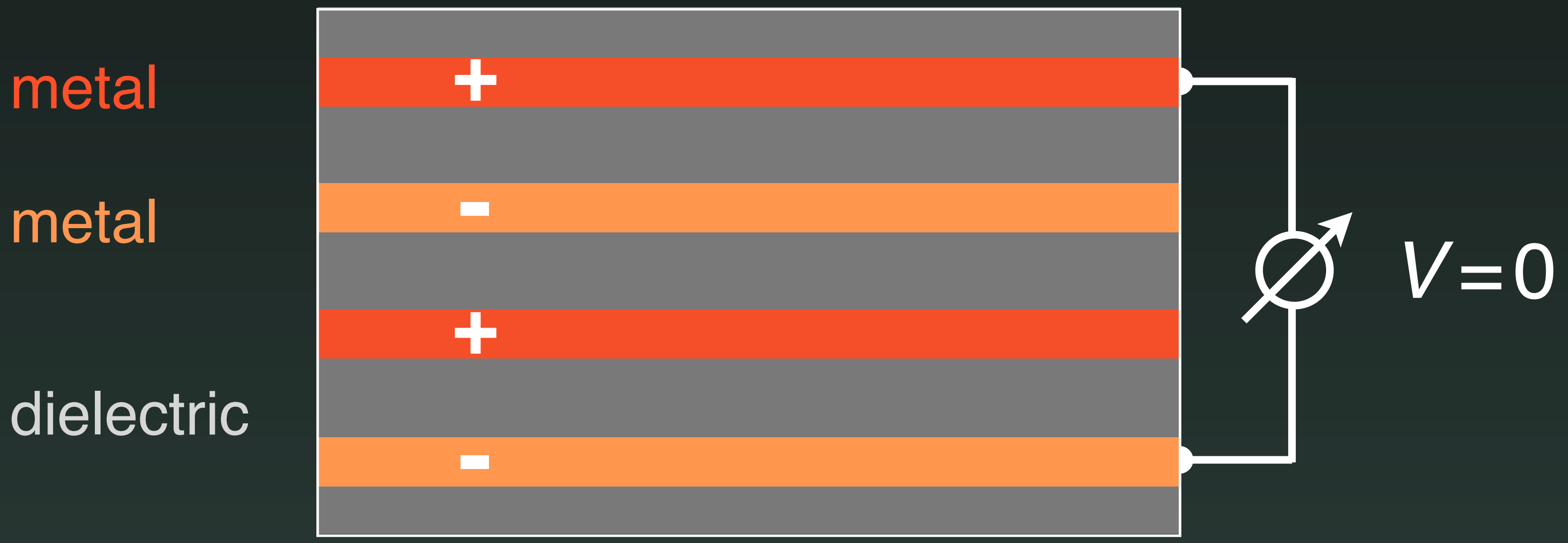
f-dependence

$$Z = i\omega L$$

Artificial Materials Using Negative-*C* Electron Systems

Artificial material with negative effective ε_r

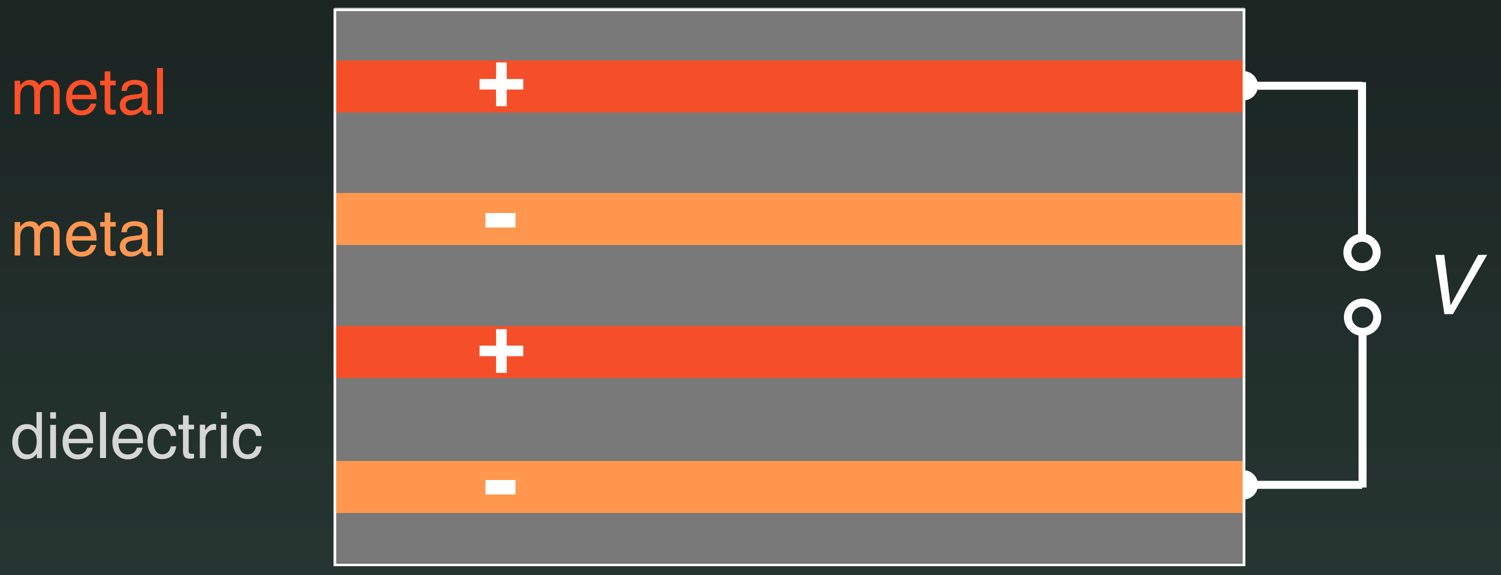
Artificial Materials Using Negative-*C* Electron Systems

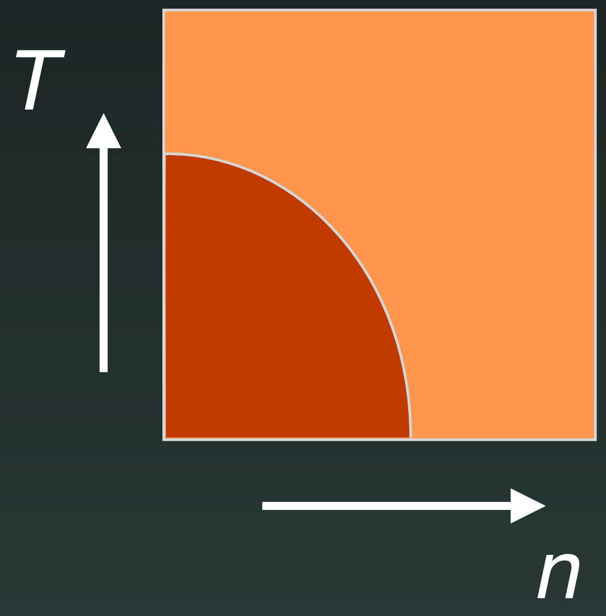


“Stripes”

Artificial material with negative effective ε_r

Artificial Materials Using Negative-*C* Electron Systems

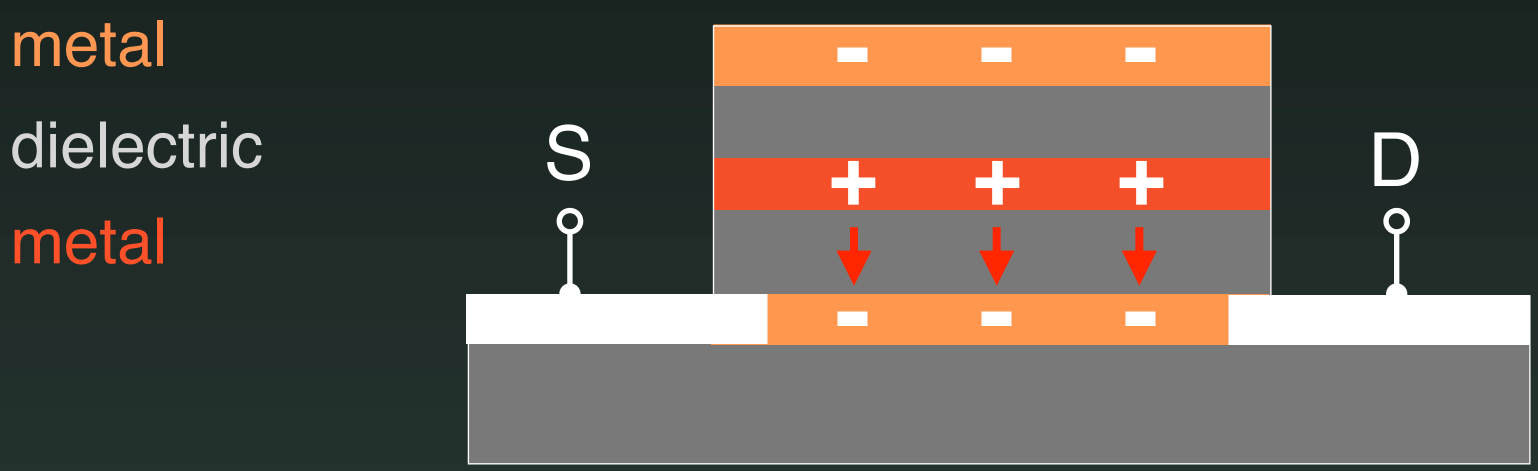




Charge separation and phase transition controllable with *V*,

e.g., magnetic phase transition controlled with *V*

Artificial Materials Using Negative-*C* Electron Systems



$$E = E_{\mathrm{H}} + \sum_i E_{\mathrm{kin},i} + \sum_i E_{\mathrm{xc},i} + \sum_i E_{\mathrm{corr},i} + \sum_i E_{\mathrm{ext},i}$$

- external-parameter (*e.g.*, *H*) alter E_{corr}, charge separation, R_{SD}

  sensor applications?

Outlook

- Energies of integrated circuits dive into the quantum regime
- Correlated materials and interfaces in correlated materials change the rules, great opportunities
- New devices and circuit design possible:
  new game for FET gate stacks and DS-channels,
  new capacitors, low-*C* interconnects

Integrating correlated materials enables new approaches to overcome fundamental limits of miniaturization