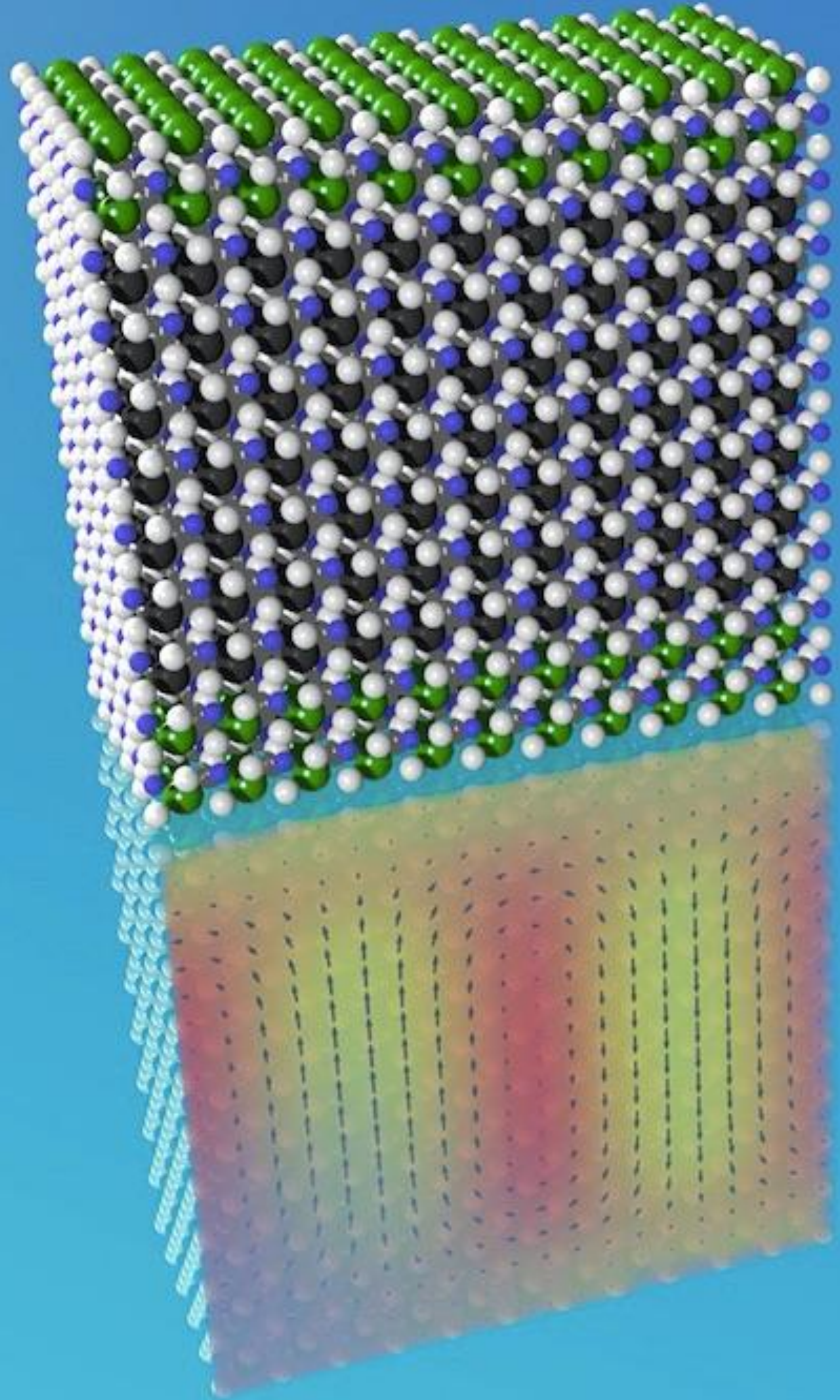
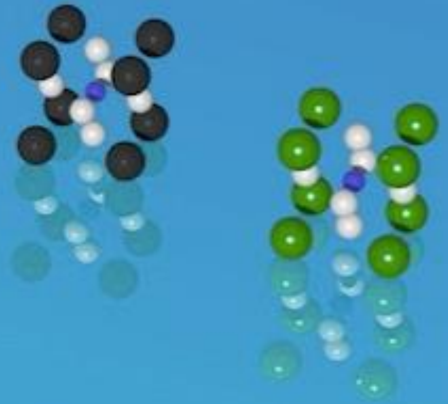


Ferroelectric negative capacitance



Pavlo Zubko

ISOE 2023, Cargèse

J. Iñiguez, P. Zubko, I. Luk'yanchuk & A. Cano
Nature Reviews Materials 4, 243 (2019)

What this tutorial is NOT about

It is **NOT** about:

- Inductive artefacts
- Negative capacitance in Schottky diodes, electrolyte-electrode interfaces and other non-equilibrium effects
- High frequency response or dielectric permittivity of metals
- Negative capacitance in correlated electron systems

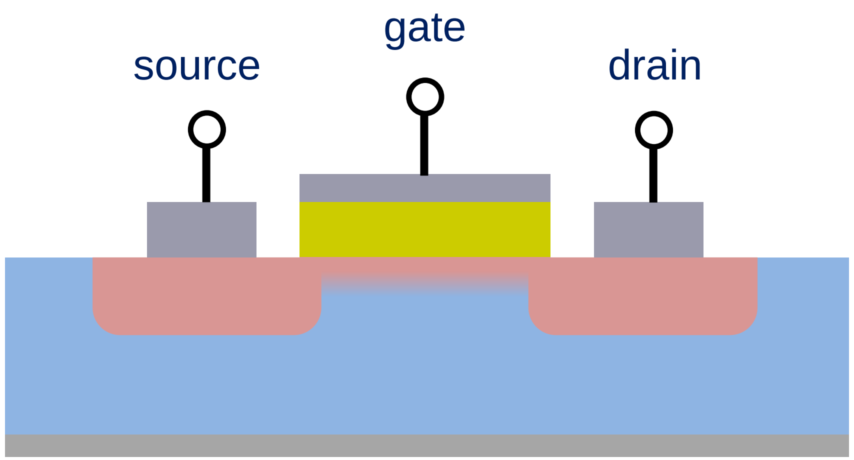
It **IS** about:

- Static permittivity of ferroelectrics
- Transients during ferroelectric switching

Outline

- Motivation
- What we mean by negative capacitance
- Basics of NC in homogeneous ferroelectrics
- NC in multidomain ferroelectrics
- Experimental evidence
- Effect of screening by free carriers
- Transient NC
- NC devices

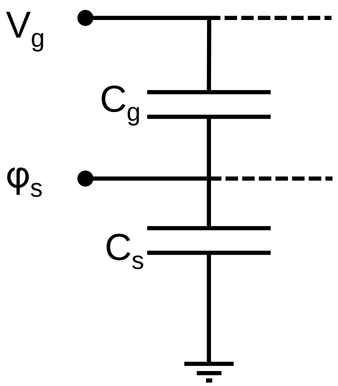
Motivation



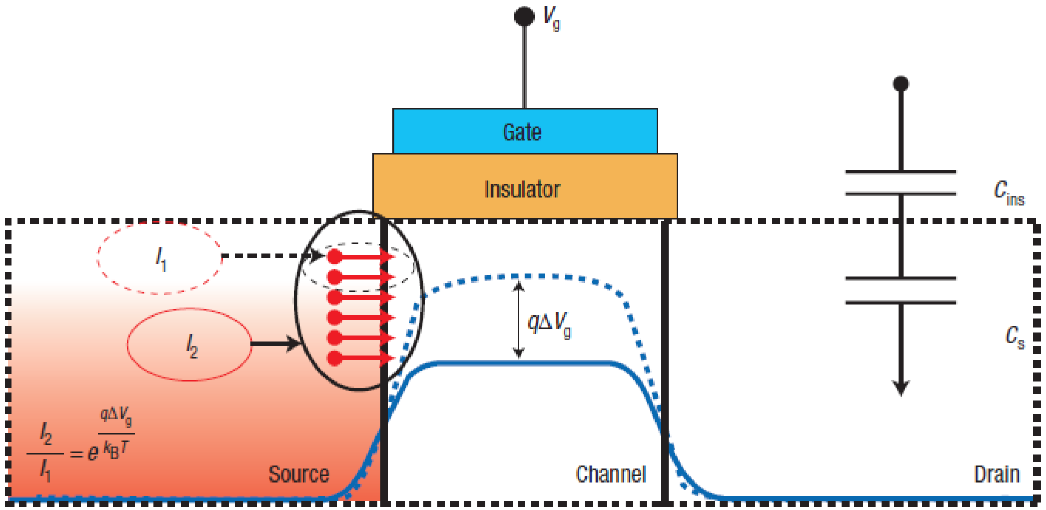
Scaling of transistors increases power dissipation – huge bottleneck for making faster processors!

Important parameter: subthreshold swing – change in gate voltage required for an order of magnitude change in current

$$S \equiv \frac{\partial V_g}{\partial(\log_{10} I)} = \frac{\partial V_g}{\partial \phi_s} \cdot \frac{\partial \phi_s}{\partial(\log_{10} I)}$$



Boltzmann tyranny



Zhirnov & Cavin, Nature Nano. (2008)

$$S \equiv \frac{\partial V_g}{\partial(\log_{10} I)} = \frac{\partial V_g}{\partial \phi_s} \times \frac{\partial \phi_s}{\partial(\log_{10} I)}$$

Body factor

$$\frac{\partial V_g}{\partial \phi_s} = 1 + \frac{C_s}{C_g} \quad > \mathbf{1}$$

Electrons follow Boltzmann distribution

$$I \propto \exp(q\phi_s / k_B T)$$

$$\frac{k_B T}{q} \ln 10 = 60 \text{ mV}$$ @ 300 K

⇒ S > 60 mV/.decade for conventional transistor

Beating Boltzmann

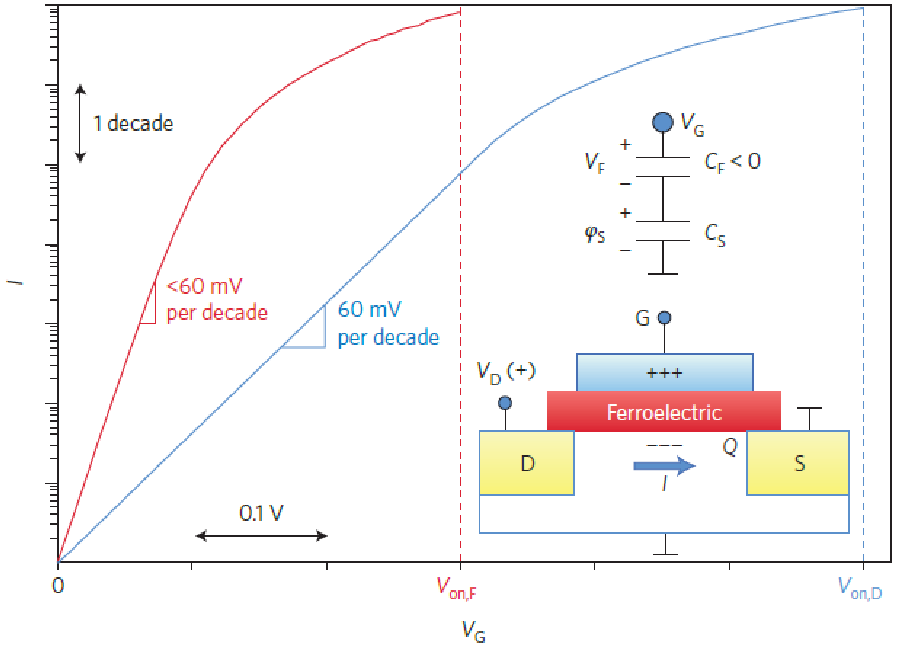
Change method of conduction

- e.g. tunnel injection

Improve body factor

- Salahuddin-Datta transistor: replace gate dielectric with negative capacitance

Catalan et al. Nature Mater. 2015



Use of Negative Capacitance to Provide Voltage Amplification for Low Power Nanoscale Devices

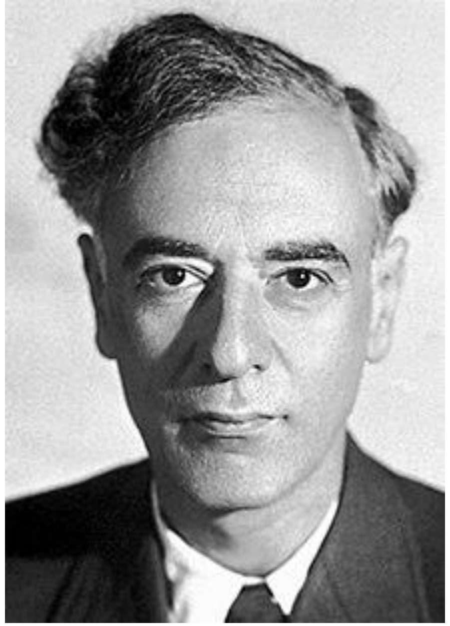
Sayeef Salahuddin* and Supriyo Datta†

School of Electrical and Computer Engineering and NSF Center for Computational Nanotechnology (NCN), Purdue University, West Lafayette, Indiana 47907

Received July 24, 2007; Revised Manuscript Received October 3, 2007

NANO LETTERS

2008
Vol. 8, No. 2
405–410



Lev Landau

ELECTRODYNAMICS OF CONTINUOUS MEDIA

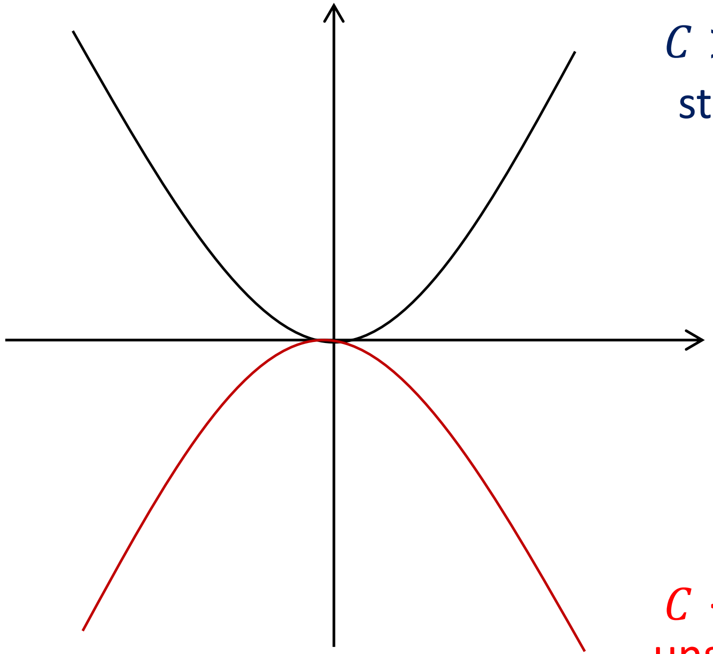
by

L. D. LANDAU and E. M. LIFSHITZ

Institute of Physical Problems, USSR Academy of Sciences

§14. The sign of the dielectric susceptibility

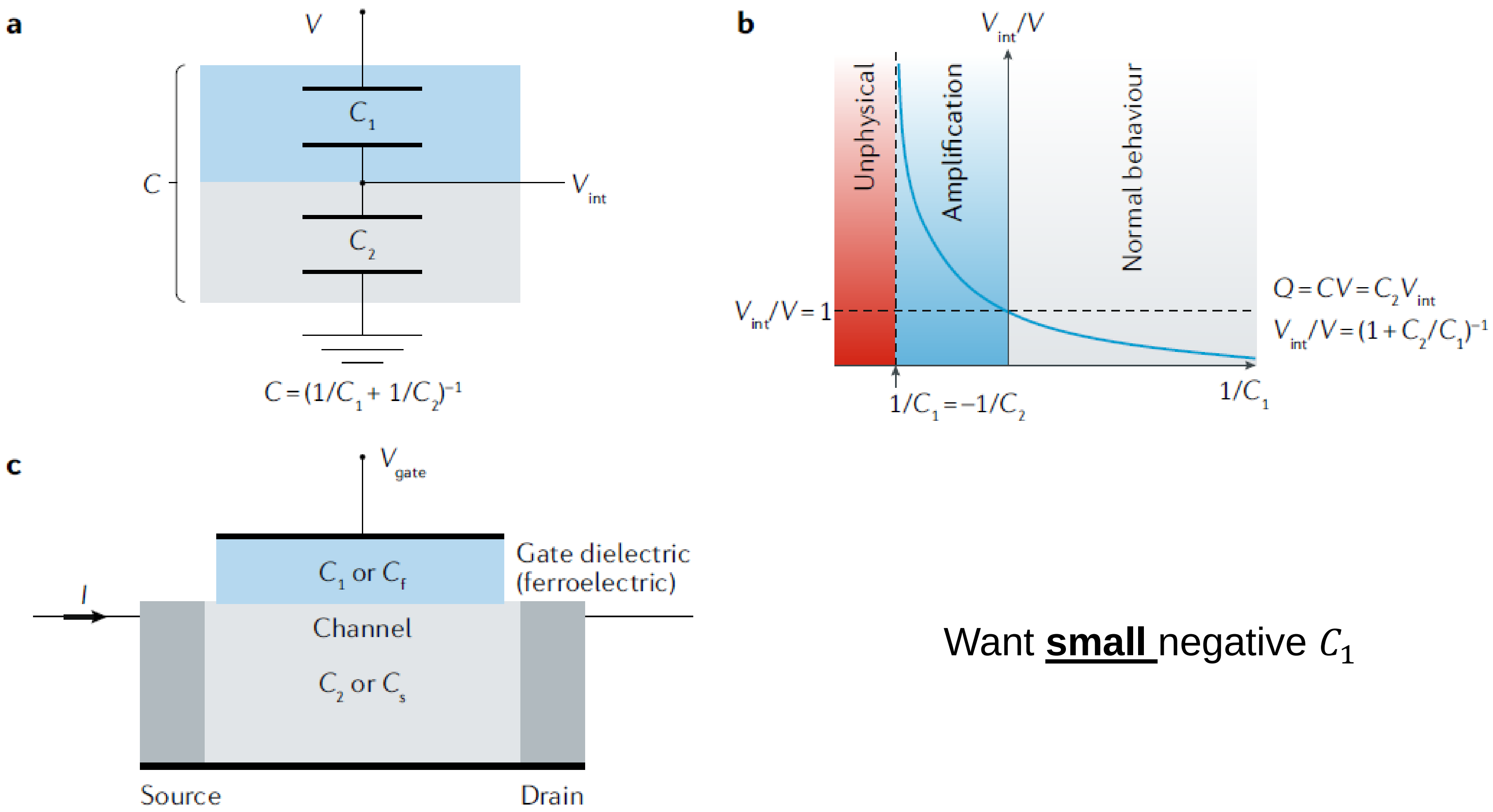
This leads to the conclusion mentioned in §7 and already made use of, namely that the permittivity of all bodies exceeds unity, and the dielectric susceptibility $\kappa = (\varepsilon - 1)/4\pi$ is therefore positive.



$$Q = CV \qquad E = \frac{Q^2}{2C} \qquad C = \frac{\epsilon\epsilon_0 A}{d}$$

A negative capacitor is unstable!

‘Local’ negative capacitance



Want **small** negative C_1

Iniguez et al, Nature Rev. Mater. 4, 243 (2019)

Some definitions

Linear dielectric: $C = \frac{Q}{V}$

$|D| = \frac{Q}{A}$

$D = P + \epsilon_0 E = \epsilon \epsilon_0 E$

$P = \chi \epsilon_0 E$

$\epsilon = \chi + 1$

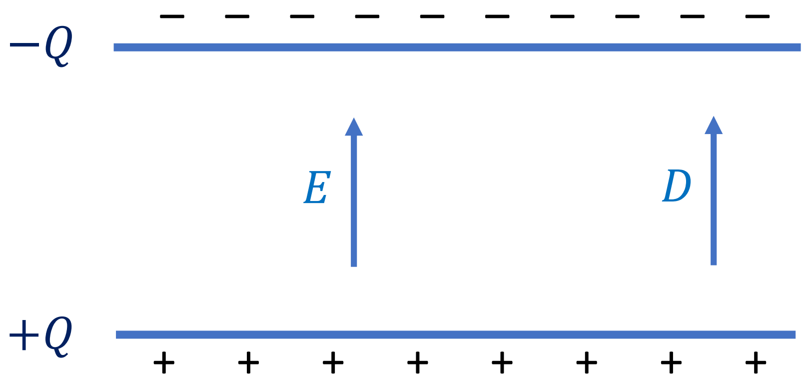
Non-linear dielectric: $C = \frac{dQ}{dV}$

$C = \frac{\epsilon \epsilon_0 A}{d}$

$\epsilon_{tot} = \frac{1}{\epsilon_0} \frac{dD_{ext}}{dE_{ext}} > 1$

'Local' permittivity (∝ 'local capacitance')

$\epsilon(r) = \frac{1}{\epsilon_0} \frac{dD(r)}{dE(r)}$



Some definitions

Linear dielectric: $C = \frac{Q}{V}$

$|D| = \frac{Q}{A}$

$D = P + \epsilon_0 E = \epsilon\epsilon_0 E$

$P = \chi\epsilon_0 E$ $\epsilon = \chi + 1$

Non-linear dielectric: $C = \frac{dQ}{dV}$

$C = \frac{\epsilon\epsilon_0 A}{d}$

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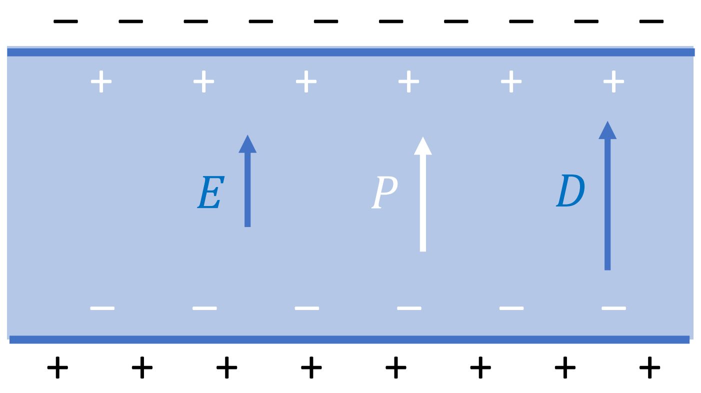


Positive permittivity: $D > P$

Some definitions

Linear dielectric: $C = \frac{Q}{V}$

$|D| = \frac{Q}{A}$

$D = P + \epsilon_0 E = \epsilon\epsilon_0 E$

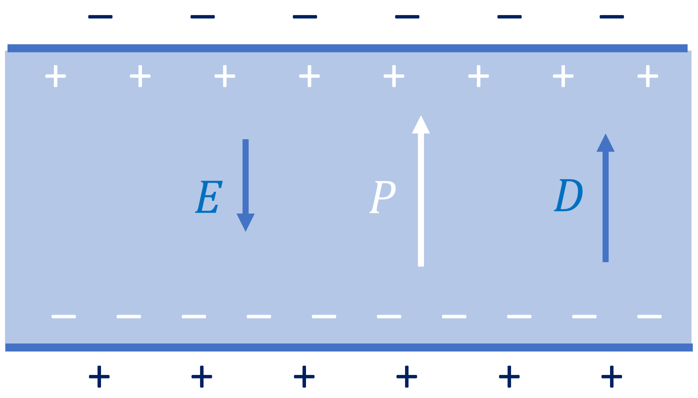
$P = \chi\epsilon_0 E$

$\epsilon = \chi + 1$

Non-linear dielectric: $C = \frac{dQ}{dV}$

$C = \frac{\epsilon\epsilon_0 A}{d}$

$\epsilon_{ext} = \frac{1}{\epsilon_0}\frac{dD_{ext}}{dE_{ext}} > 1$



‘Local’ permittivity (∝ ‘local capacitance’)

$\epsilon(r) = \frac{1}{\epsilon_0}\frac{dD(r)}{dE(r)}$

Positive permittivity: $D > P$

Negative permittivity: $\boldsymbol{P > D}$ (‘overscreening’)

Collective Phenomena
1976, Vol. 2, pp. 167–170

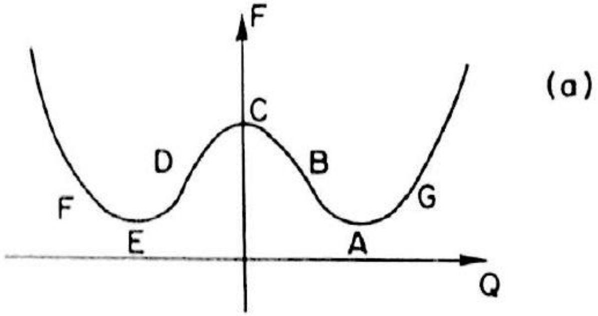
CAN CAPACITANCE BE NEGATIVE?

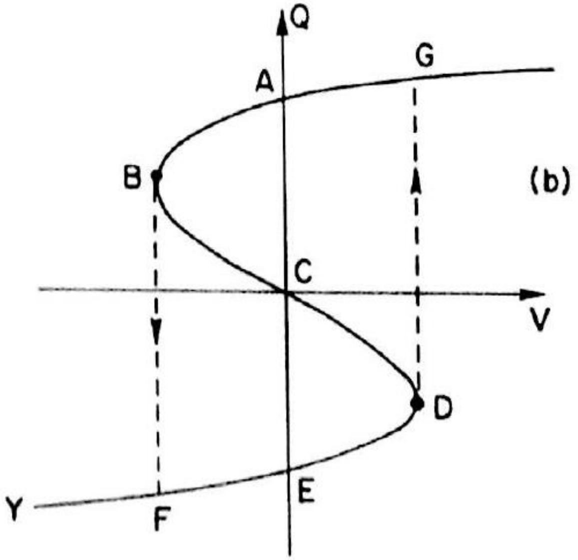
ROLF LANDAUER

IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598

(Received July 25, 1975)

Prigogine and Glansdorff in their analysis of the stability of the steady state have invoked the fact that compressibilities must be positive. The ferroelectric analog of that proposition is analyzed with the probable conclusion that negative differential dielectric constants can exist in the stable steady state.



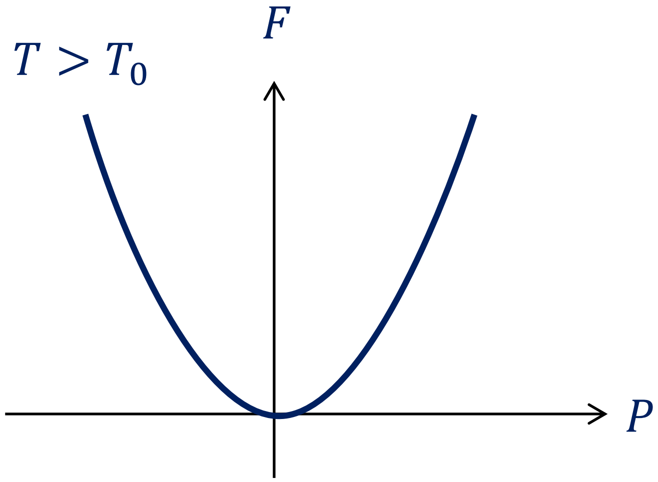


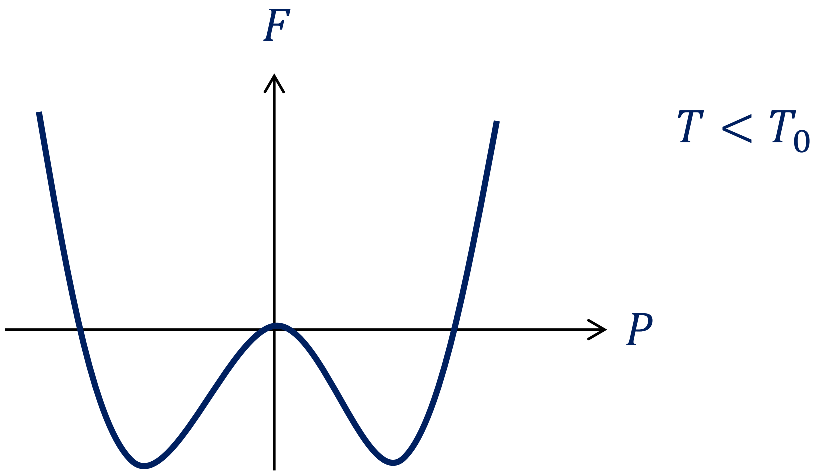
Ferroelectrics: the ‘forbidden’ region

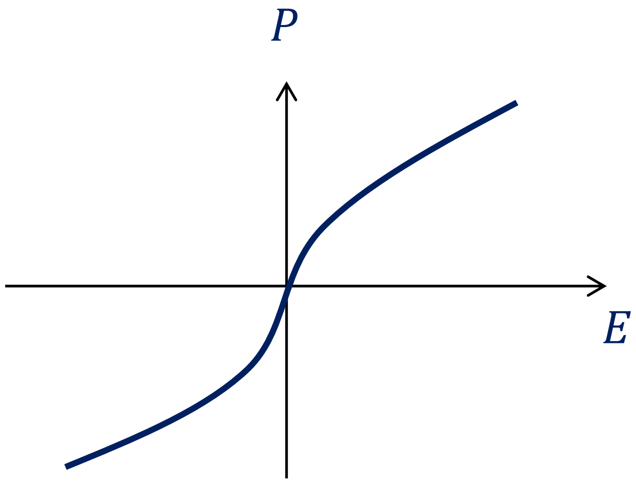
$$F = \frac{\alpha}{2}P^2 + \frac{\beta}{4}P^4 - EP$$

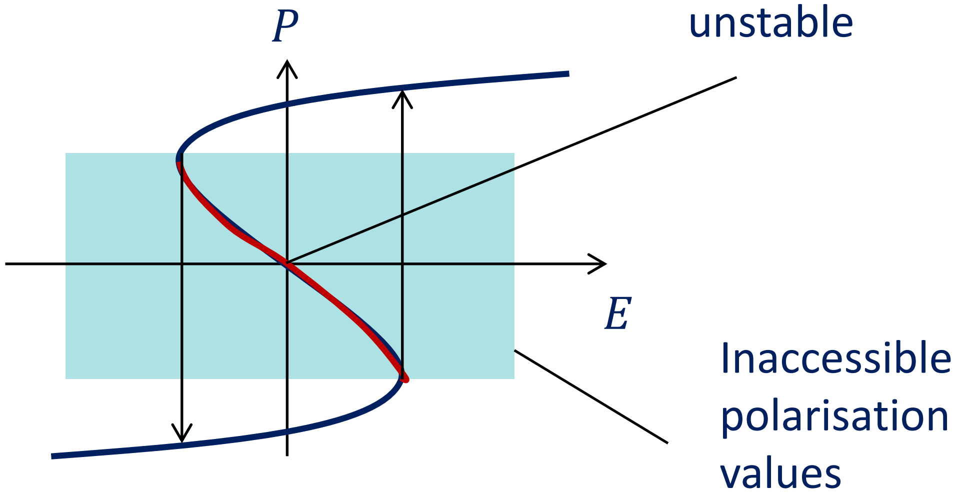
$$\alpha = \frac{T - T_0}{C}$$

$$\alpha P + \beta P^3 = E$$



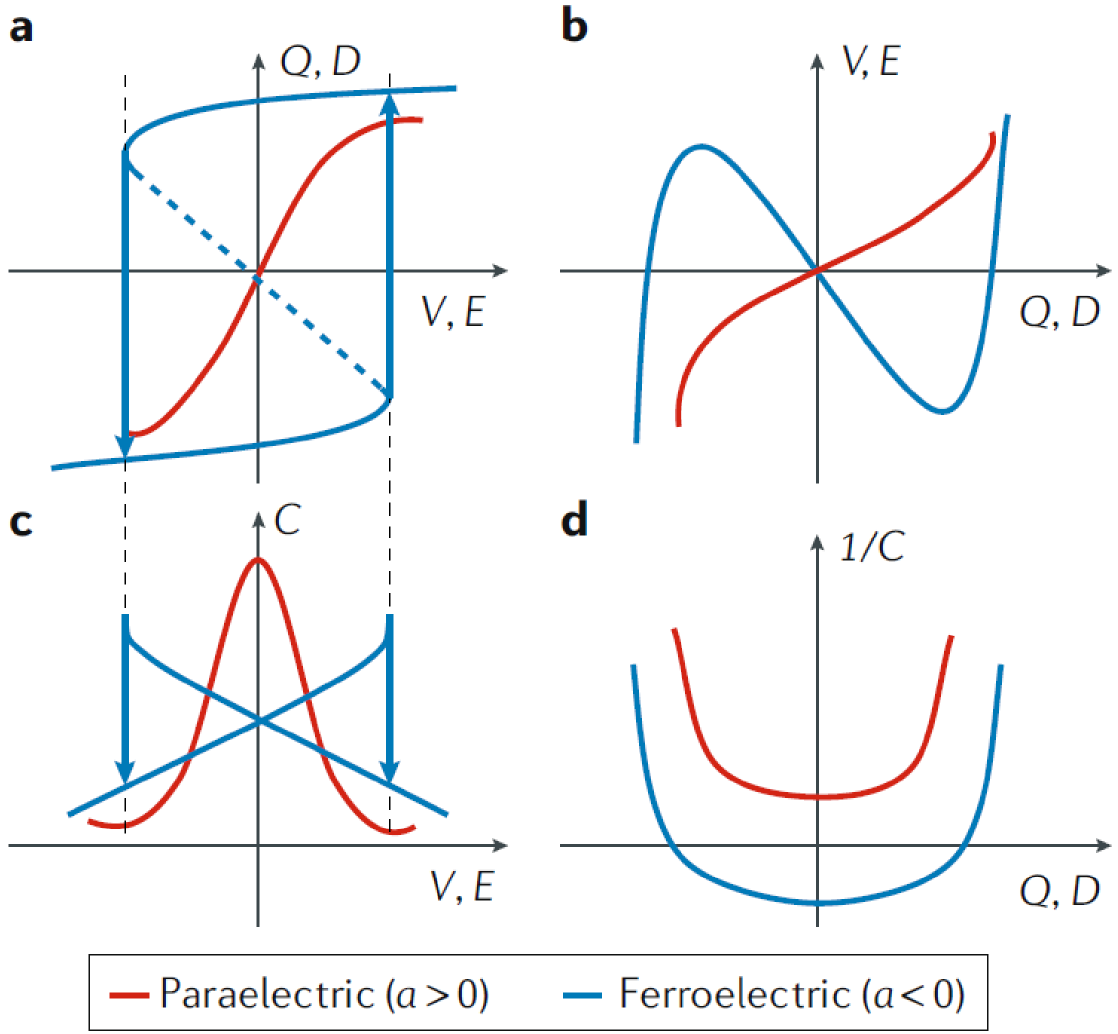






Controlling charge vs voltage

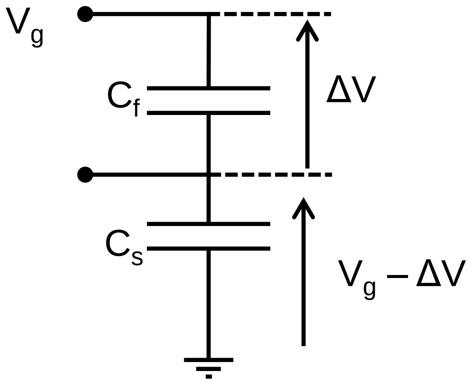
$$C = \frac{dQ}{dV}$$



Key point:
Need to control **charge**

Iniguez et al, Nature Rev. Mater. 4, 243 (2019)

Ferroelectric + series capacitance



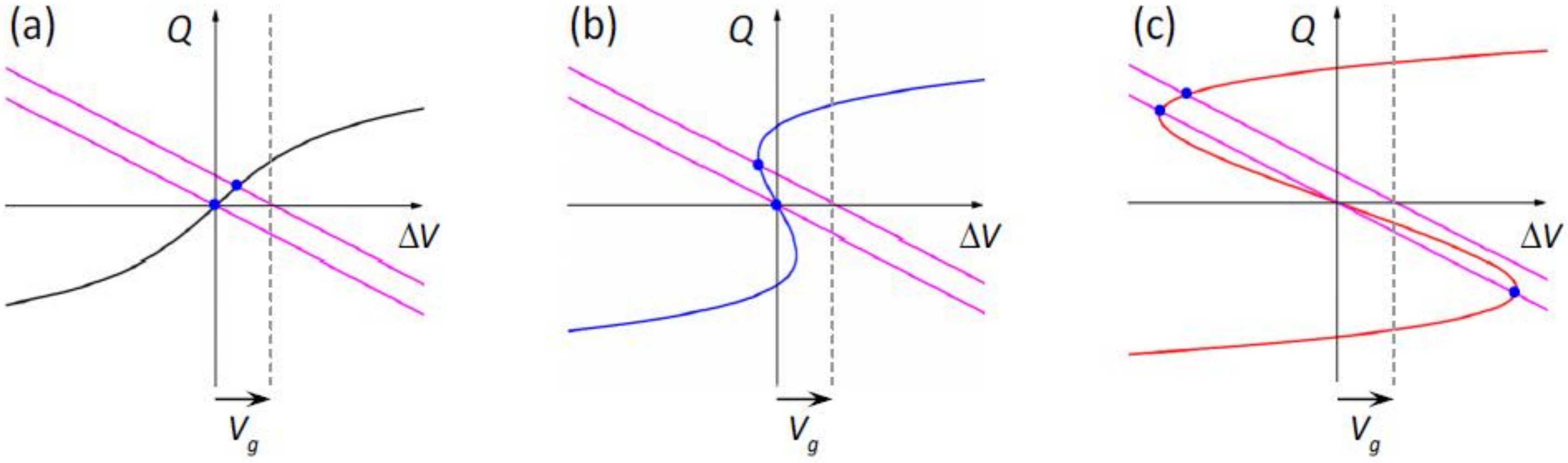
$$Q_f = Q_f(\Delta V)$$

$$Q_s = C\,(V_g - \Delta V)$$

$$Q_s = Q_f = Q$$

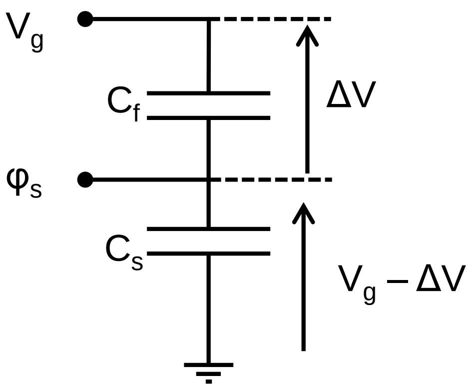
- FeFET
- Sawyer-Tower circuit

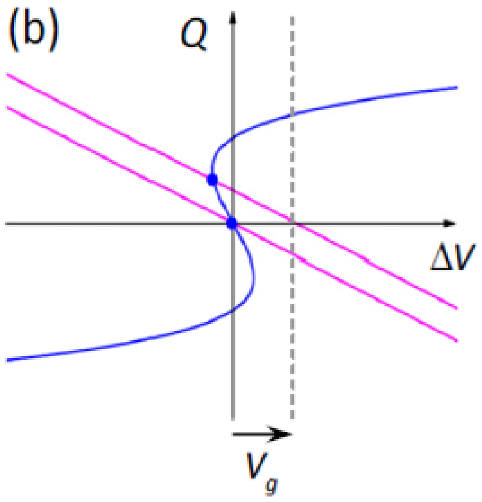
A. Cano and D. Jiménez Appl. Phys. Lett. **97**, 133509 (2010)



$$T > T_C \rightarrow \frac{\Delta Q}{\Delta V} > 0 \qquad T < T_C \text{ (small } C_S) \rightarrow \frac{\Delta Q}{\Delta V} < 0 \qquad T \ll T_C \text{ (large } C_S) \rightarrow \frac{\Delta Q}{\Delta V} > 0$$

Ferroelectric + series capacitance





Note: $\frac{dQ}{dV_g} > 0$ – overall system capacitance positive → system is stable

$T < T_C$ (small C_S) → $\frac{\Delta Q}{\Delta V} < 0$

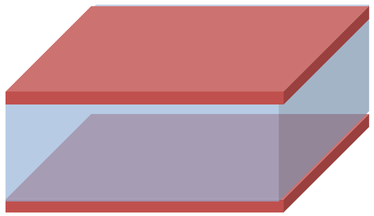
BUT $\phi_s > V_g$ - voltage amplification at the interface!

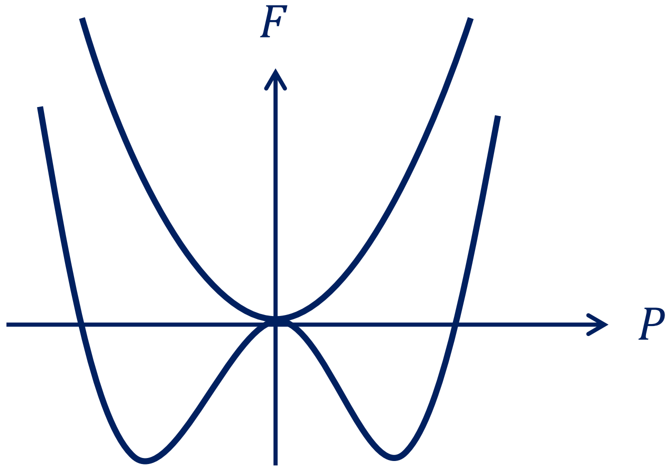
$$\frac{1}{C_{tot}} = \frac{1}{C_f} + \frac{1}{C_s} < \frac{1}{C_s}$$

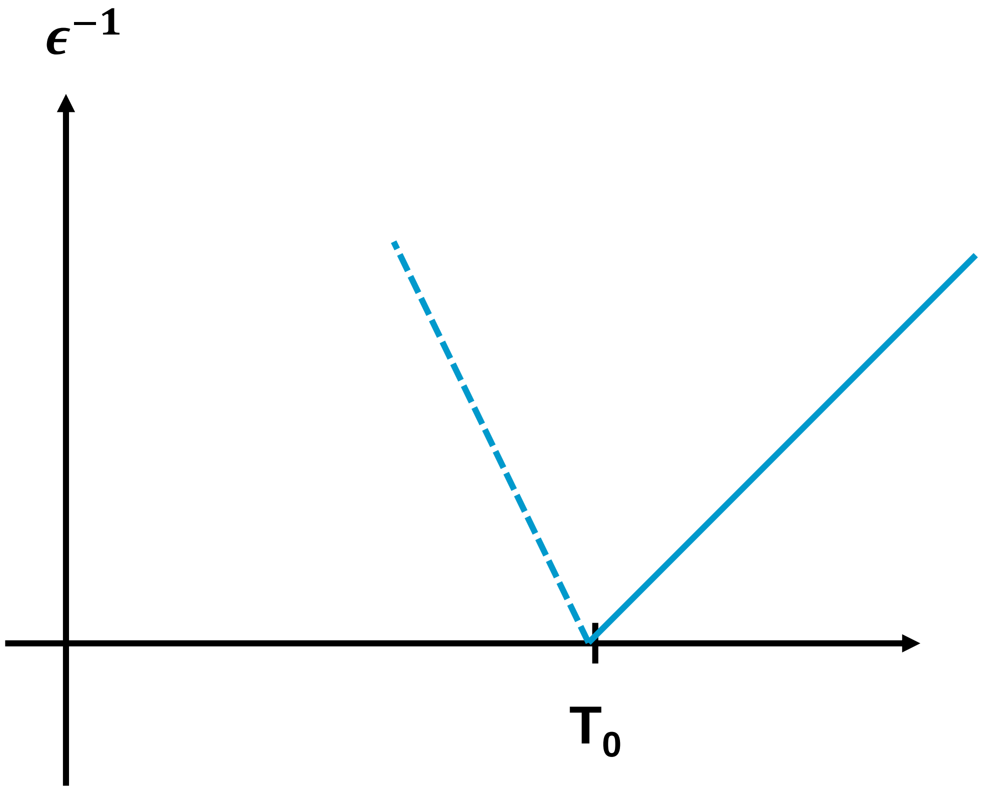
When added in series, the negative capacitance increases the overall capacitance → Signature of NC!

Vary V_g but effectively control charge

Phase transition in a screened ferroelectric



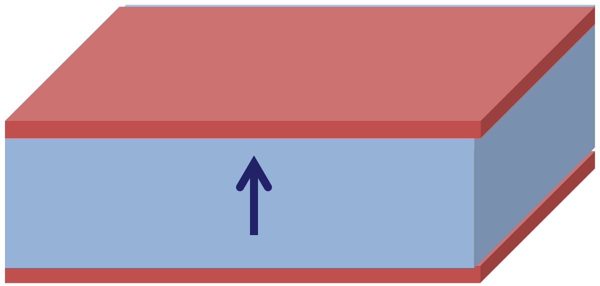




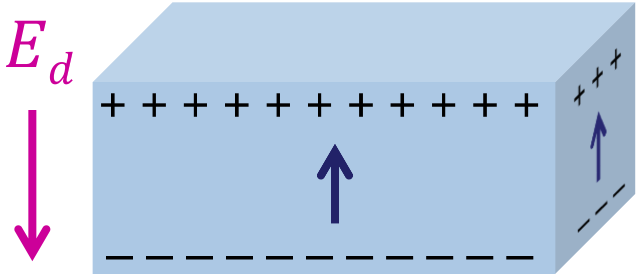
$$F = \left(\frac{\alpha_f}{2}P^2 + \frac{\beta_f}{4}P^4\right) - EP$$

$$\alpha_f = \frac{T - T_0}{C}$$

$$\frac{1}{\epsilon} \approx \frac{1}{\chi} = \left(\alpha_f + 3\beta_f P^2\right)$$



$E = 0$

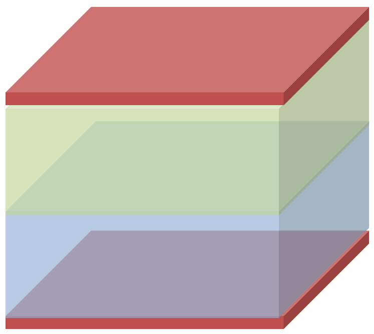


Perfect screening

No screening

(assuming no domain formation…for now)

Phase transition in a poorly screened FE



$$F = l_f\left(\frac{\alpha_f}{2}P_f^2 + \frac{\beta_f}{4}P_f^4 - E_f P_f\right) + l_d\left(\alpha_d P_d^2 - E_d P_d\right)$$

$$D = P_f + \epsilon_0 E_f = P_d + \epsilon_0 E_d$$

$$l_f E_f = -l_d E_d$$

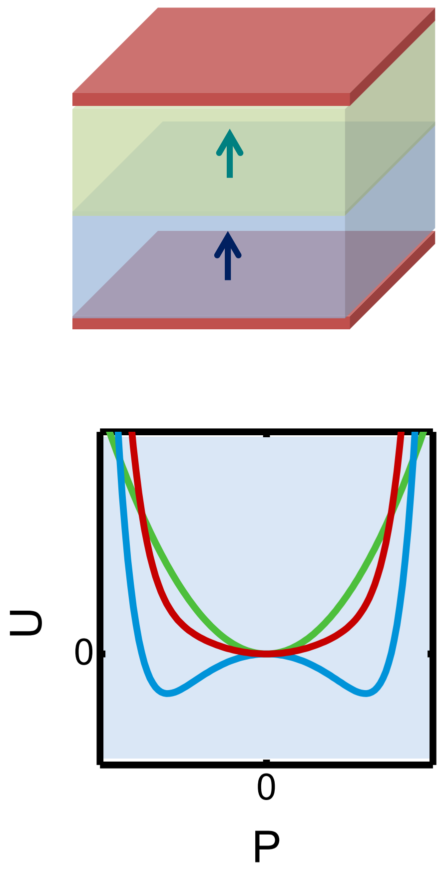
$$F = l_f\left(\frac{\alpha_f}{2}P_f^2 + \frac{\beta_f}{4}P_f^4\right) + l_d \alpha_d P_d^2 + \frac{l_f l_d}{\epsilon_0\left(l_f + l_d\right)}\left(P_f - P_d\right)^2$$

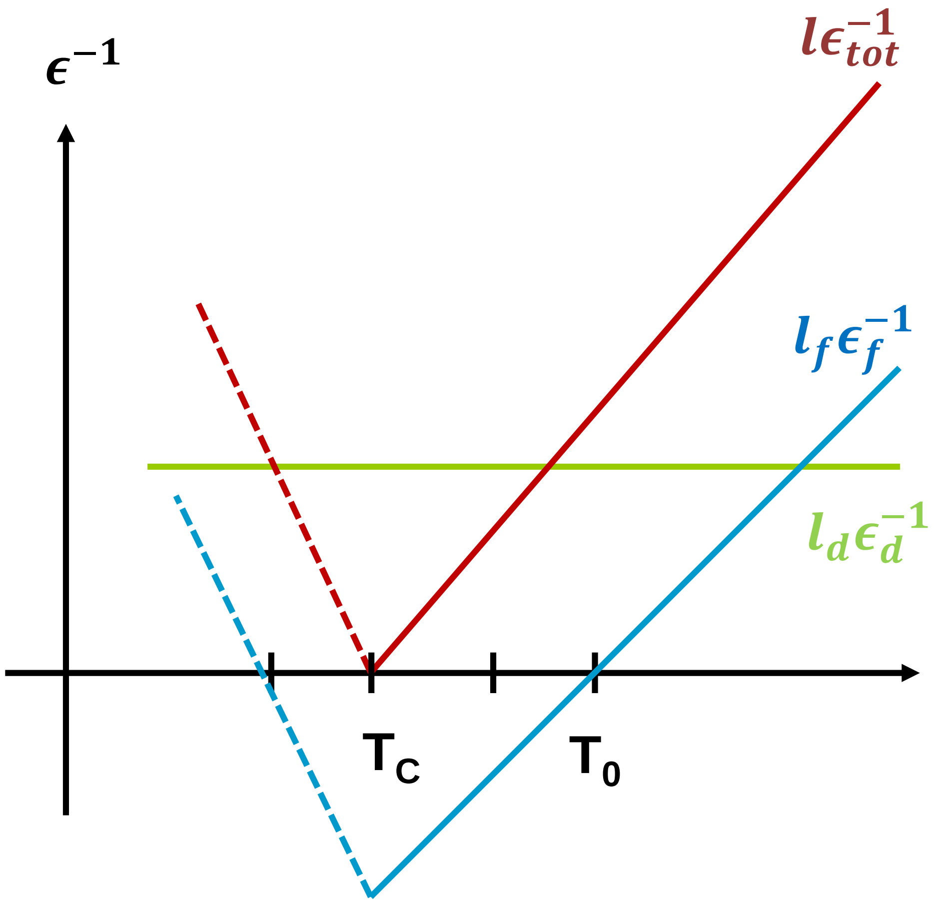
Layers electrostatically coupled!
(~ constant P throughout)

Very large unless $P_f \approx P_d$

$$F \approx l_f\left(\frac{\alpha_f}{2}P^2 + \frac{\beta_f}{4}P^4\right) + l_d \alpha_d P^2$$

Negative capacitance – homogeneous case





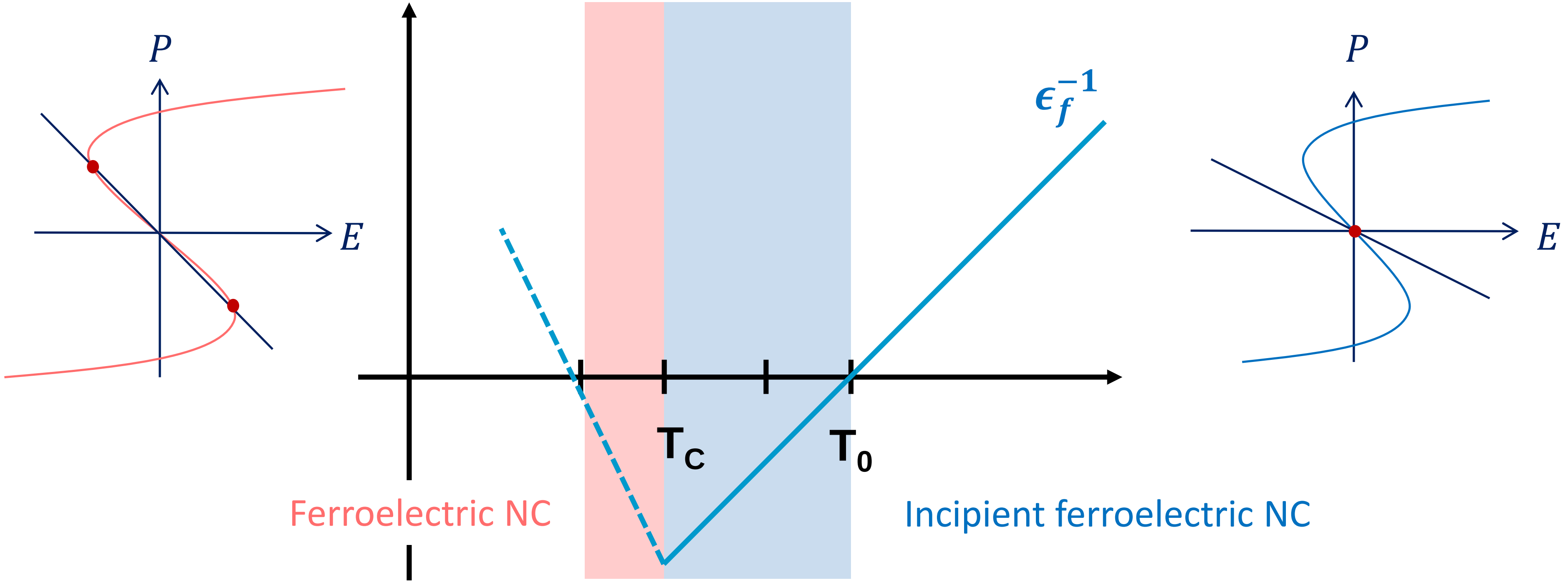
$$F = l_f\left(\frac{\alpha_f}{2}P^2 + \frac{\beta_f}{4}P^4\right) + l_d\alpha_d P^2$$

$$\alpha_f = \frac{T - T_0}{C}$$

$$\frac{l}{\chi} = l_d\alpha_d + l_f\left(\alpha_f + 3\beta_f P^2\right)$$

$$= \frac{l_d}{\chi_d} + \frac{l_f}{\chi_f}$$

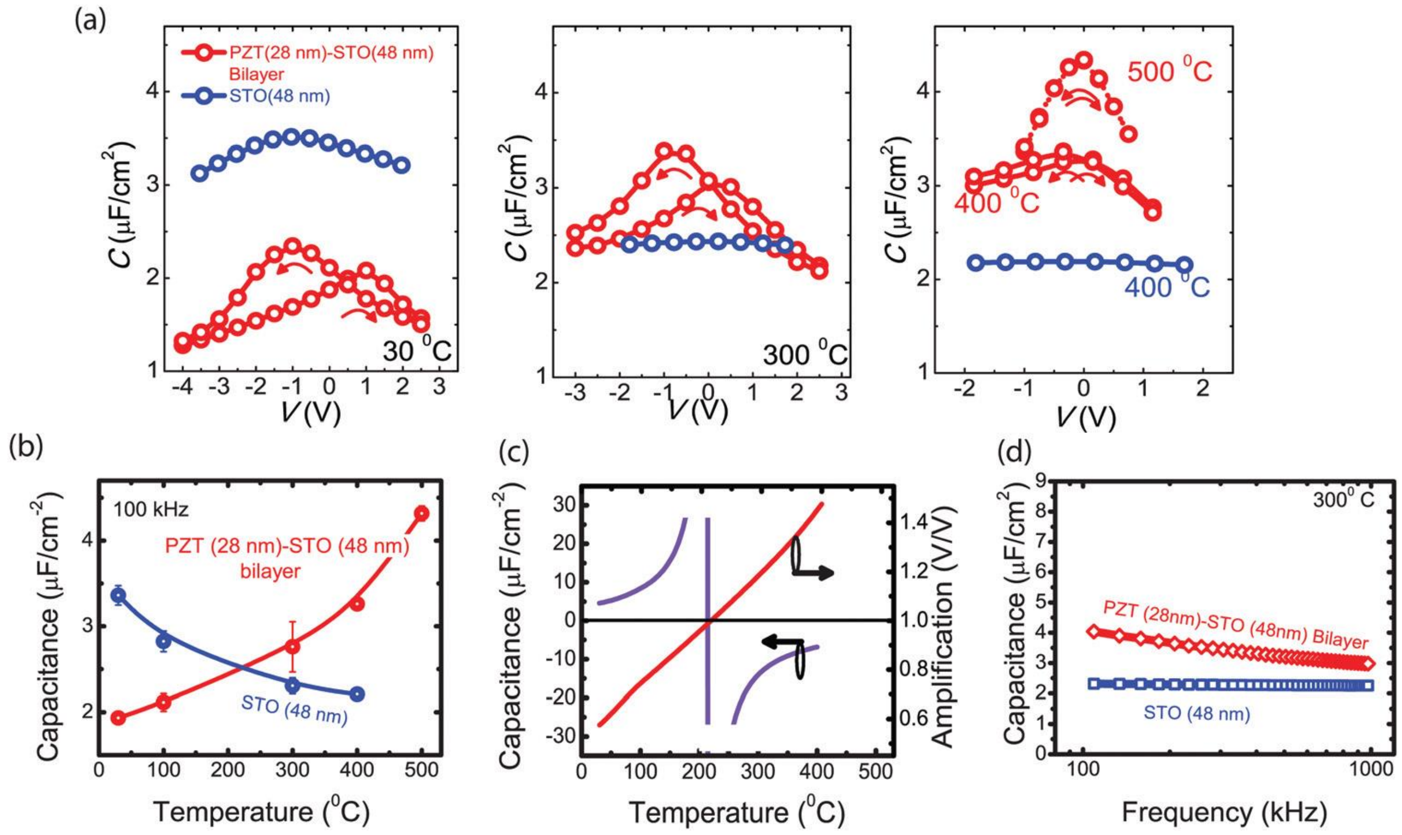
Negative capacitance – homogeneous case



- Main idea – destabilise ferroelectricity (electrostatically)
- Will happen in **any** system with imperfect screening
- Two temperature regimes – incipient ferroelectric & ferroelectric
- Overall capacitance positive
- Experimental signature – capacitance boost when added in series

Experimental evidence

Capacitance enhancement

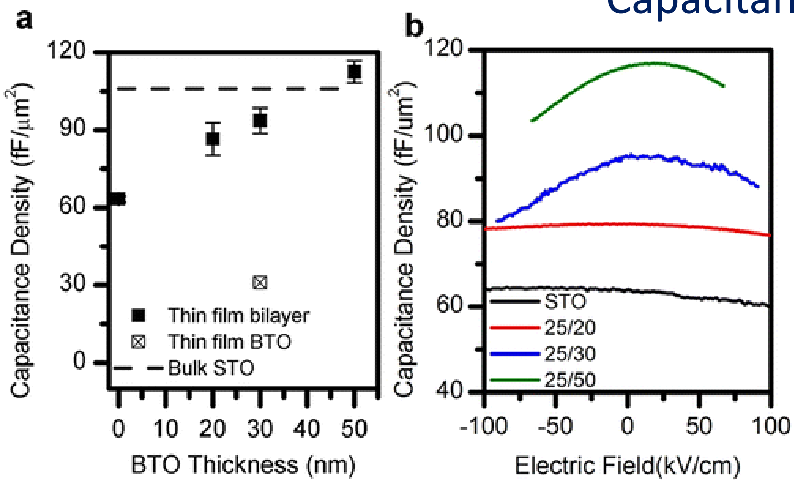


Capacitance smaller in Au/STO (48 nm)/SRO than in Au/PZT/STO (48 nm) /SRO

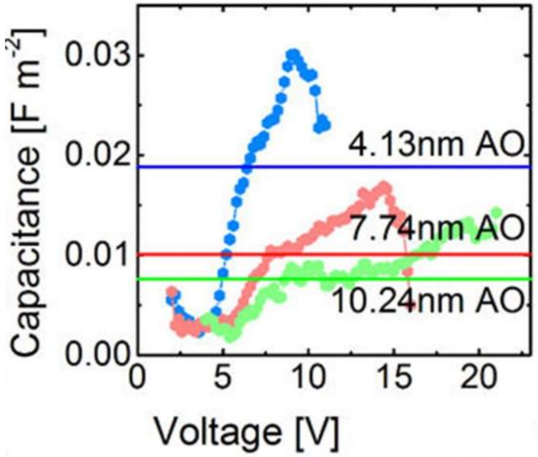
Khan et al. APL 99, 113501 (2011)

Experimental evidence

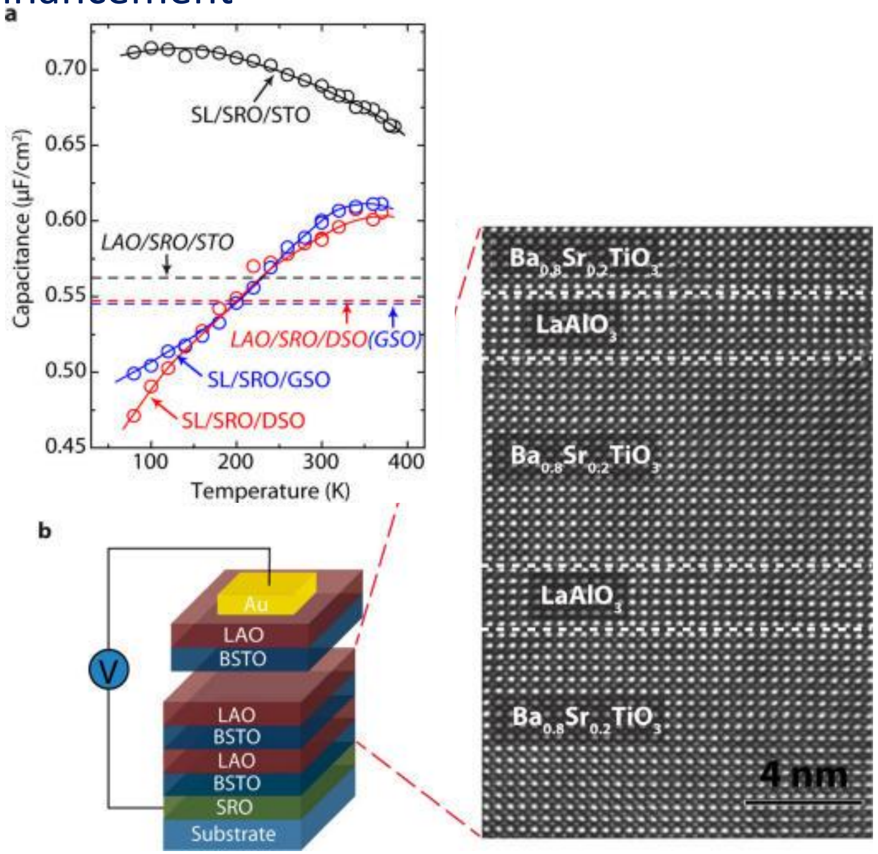
Capacitance enhancement



$BaTiO_3/SrTiO_3$ bilayers with Pt/$SrRuO_3$ electrodes
Appleby et al. Nano Letters 2014



$Al_2O_3/BaTiO_3$
Kim et al. Nano Letters 2016

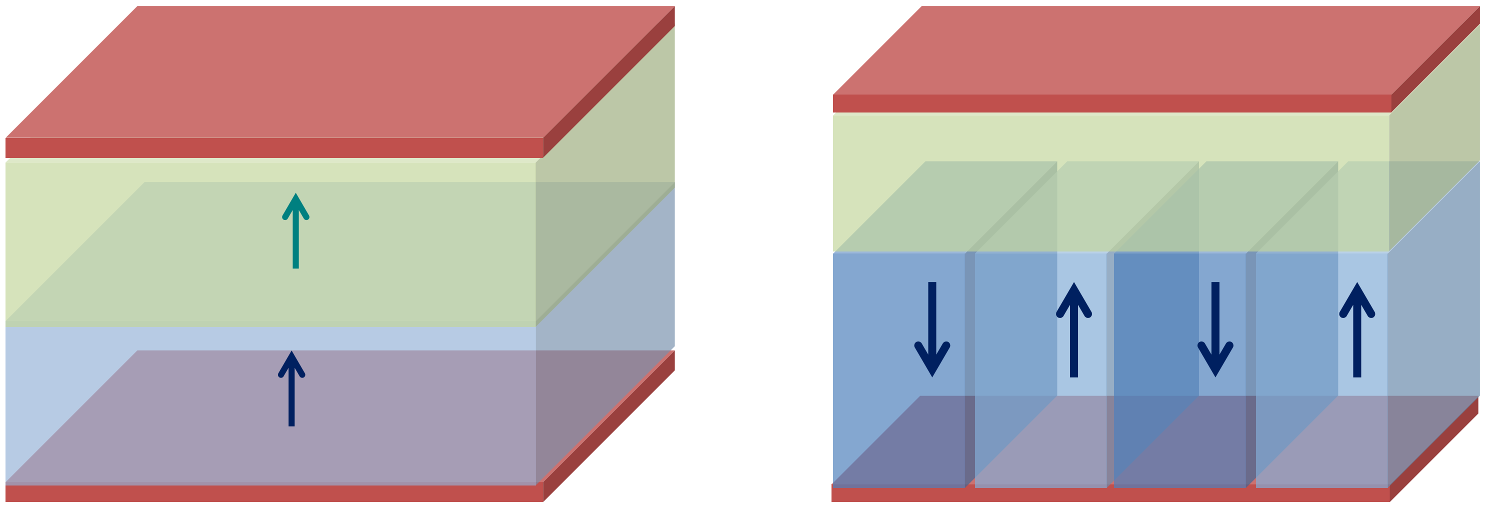


BST/$LaAlO_3$ superlattices
Gao et al. Nano Letters 2014

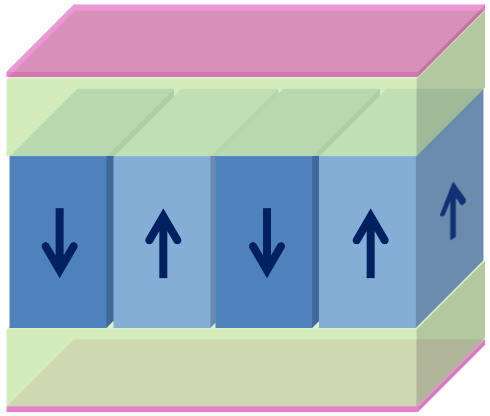
Capacitance enhancement reported by a number of groups – generally in the ferroelectric (i.e. hysteretic) phase

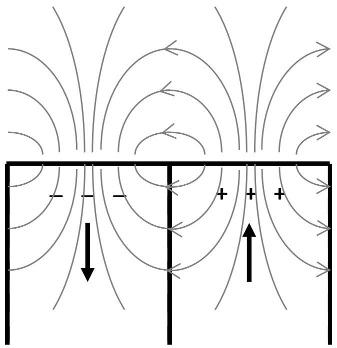
Domain formation

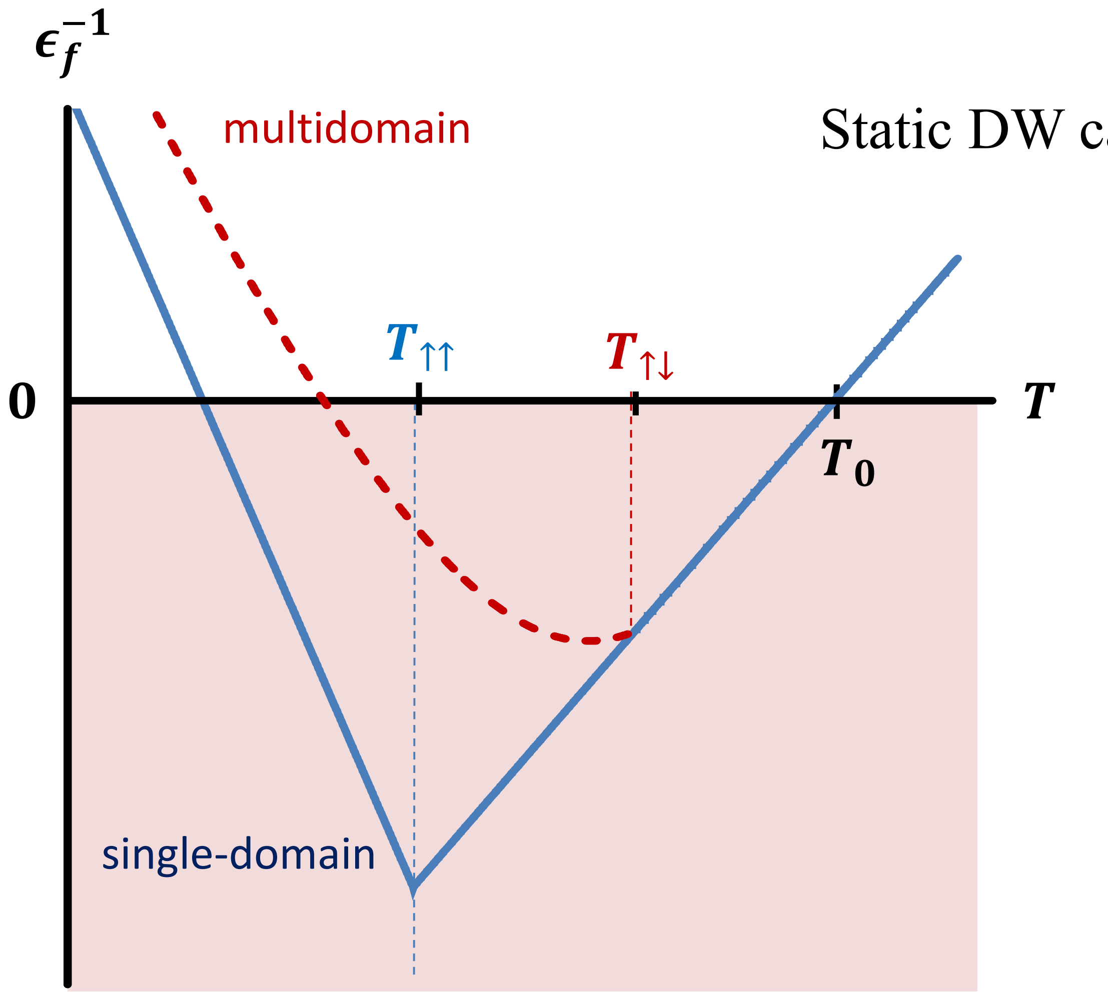
Usually ferroelectric will try to reduce the depolarising field via domain formation



Negative capacitance with domains?



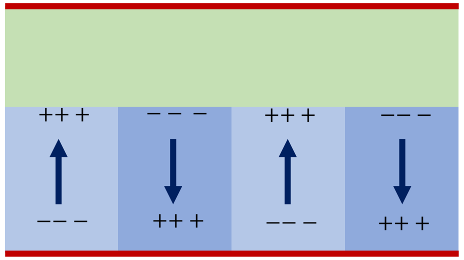




Sene (PhD thesis) & Luk'yanchuk
Cano & Jimenez, APL 97, 133509 (2010)

Domain wall motion contribution

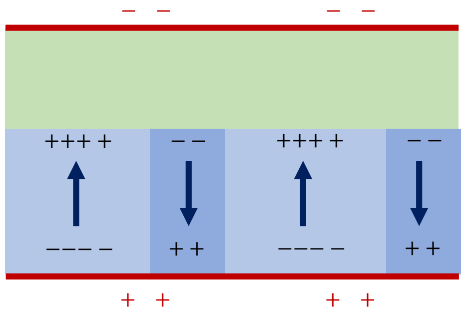
$Q = 0, D = 0$



$P_f = 0$

Ideal case
(no stray fields,
ideal DWs)

$Q > 0, D > 0$



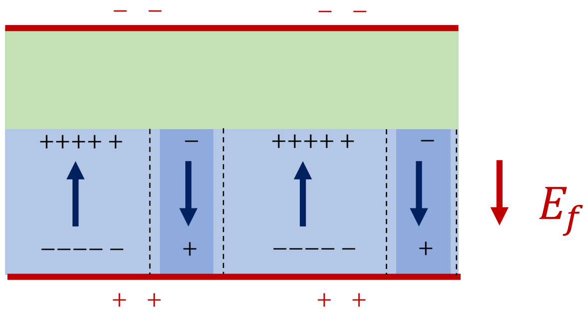
$P_f = D, E_f = 0$

'perfect screening'

(infinite permittivity)

Real case
(stray fields, real
DWs)

$Q > 0, D > 0$

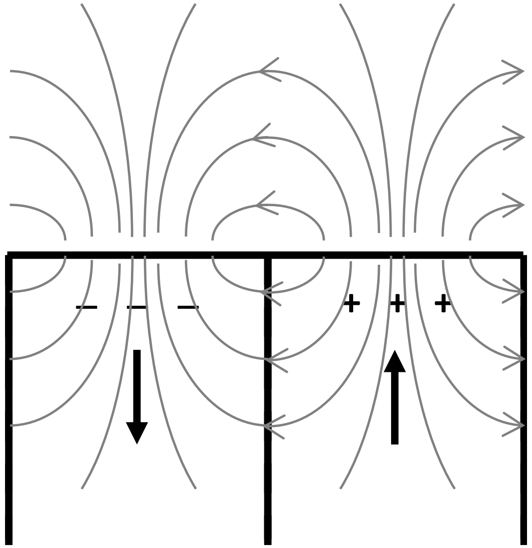


$P_f > D, E_f < 0$

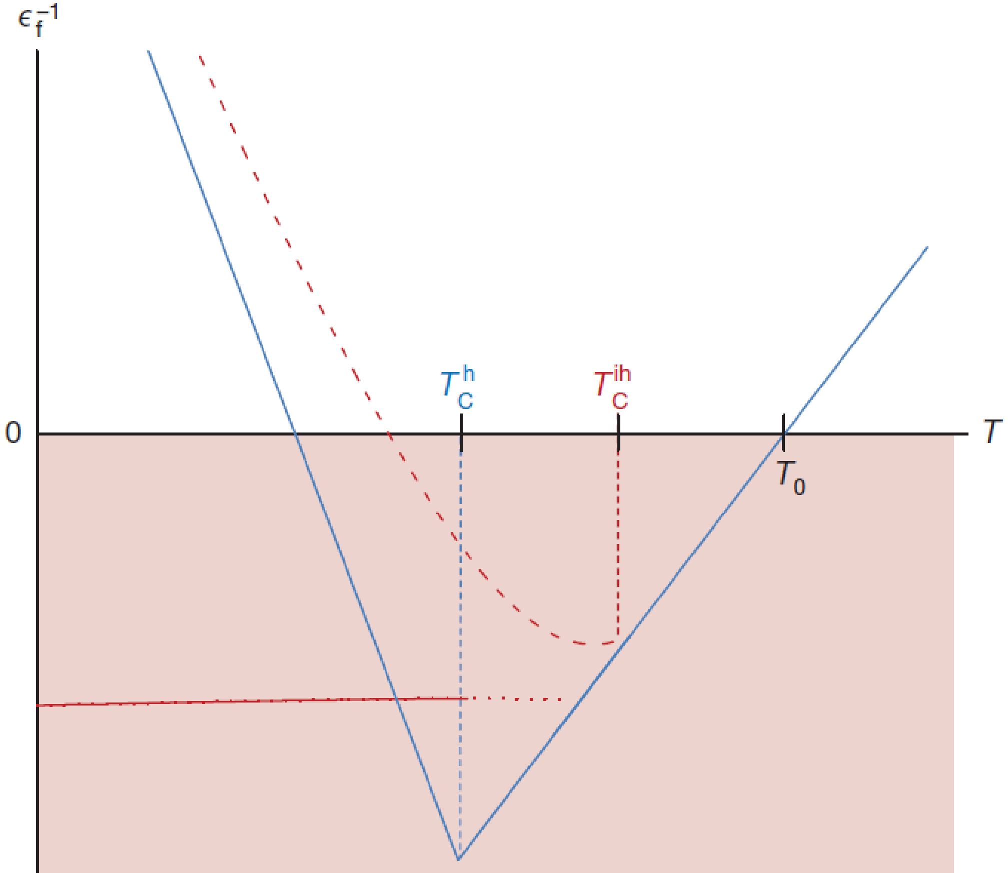
'overscreening'

(negative permittivity)

Negative capacitance with domains



$$\epsilon_f = \epsilon_\parallel - \frac{\pi}{4\ln 2}\sqrt{\frac{\epsilon_\perp}{\epsilon_\parallel}\frac{l_f}{w}}\,\epsilon_\parallel$$



Zubko et al. Nature 534, 524 (2016)

Kopal et al. Ferroelectrics 223, 127 (1999)
Bratkovsky & Levanyuk, PRB 63, 132104 (2001)
Luk'yanchuk et al. PRB 98, 024107 (2018)
Iniguez et al. Nature Rev. Mater. 4, 243 (2019)

Mono- vs multidomain NC

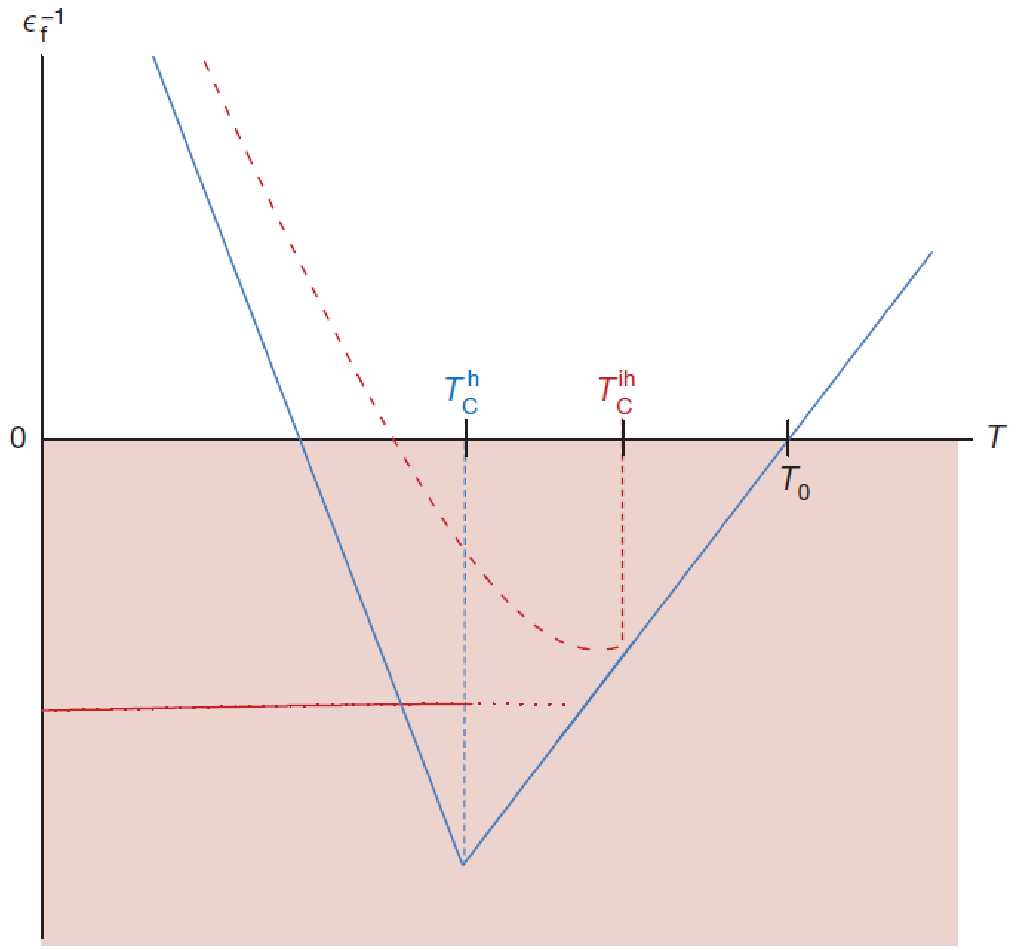
Monodomain NC:

- Large magnitude effect

Multidomain NC

- Hysteresis-free
- Temperature stability
- Fast response time

- Ideally, want system with high DW energy



Experimental evidence

- Measure capacitance of the whole structure

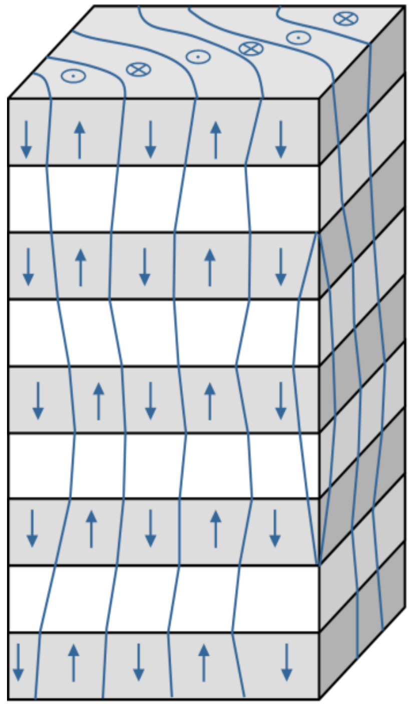
$$C_{meas} = \frac{\epsilon\epsilon_0 A}{d}$$

- If consider as separate capacitors in series

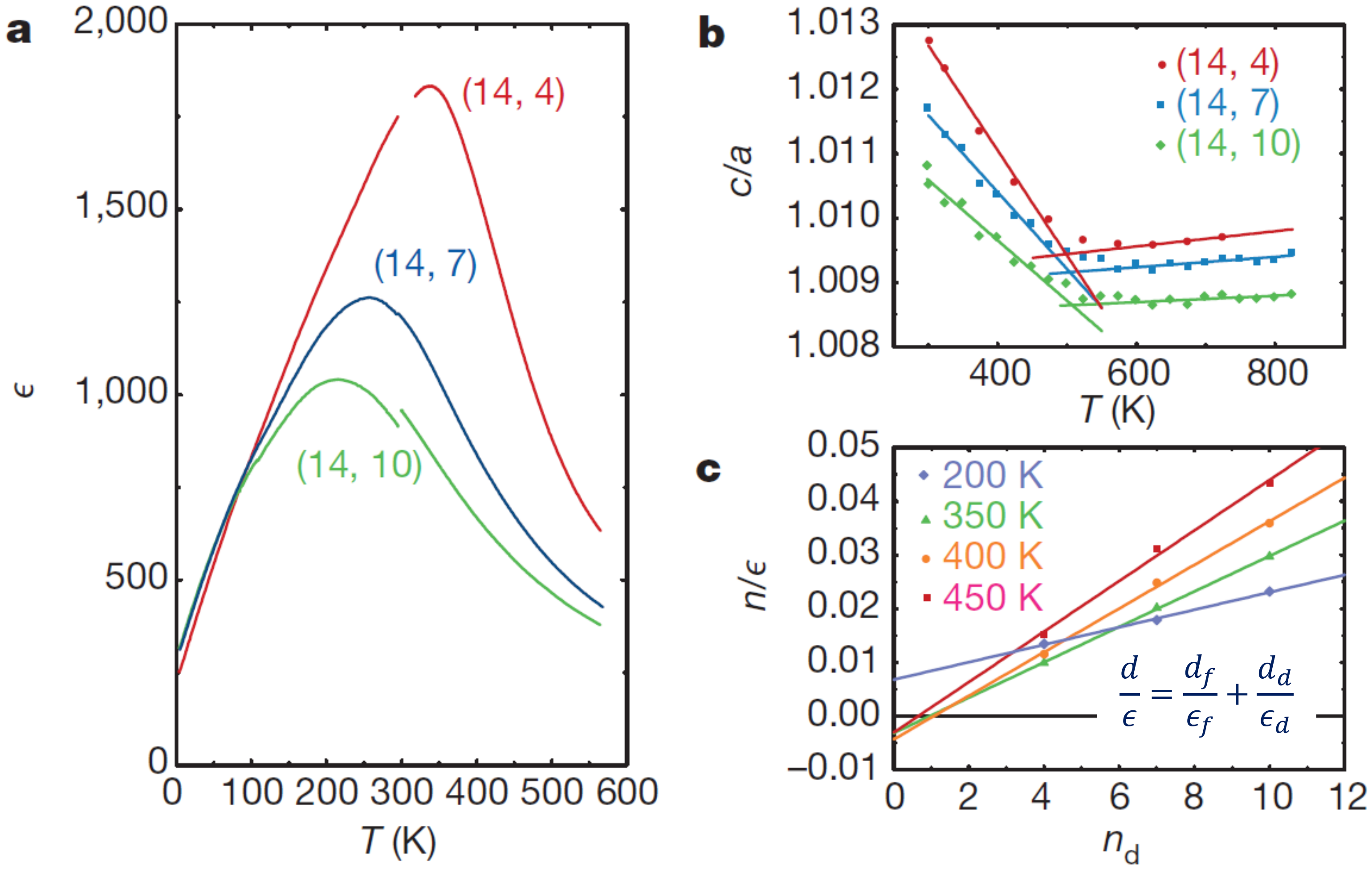
$$\frac{d}{\epsilon} = \frac{d_f}{\epsilon_f} + \frac{d_d}{\epsilon_d}$$

- Generally ϵ_f & ϵ_d depend on d_f and d_d due to long range electrostatic interaction…

- …except if FE layers are electrostatically decoupled – can apply series capacitor formula



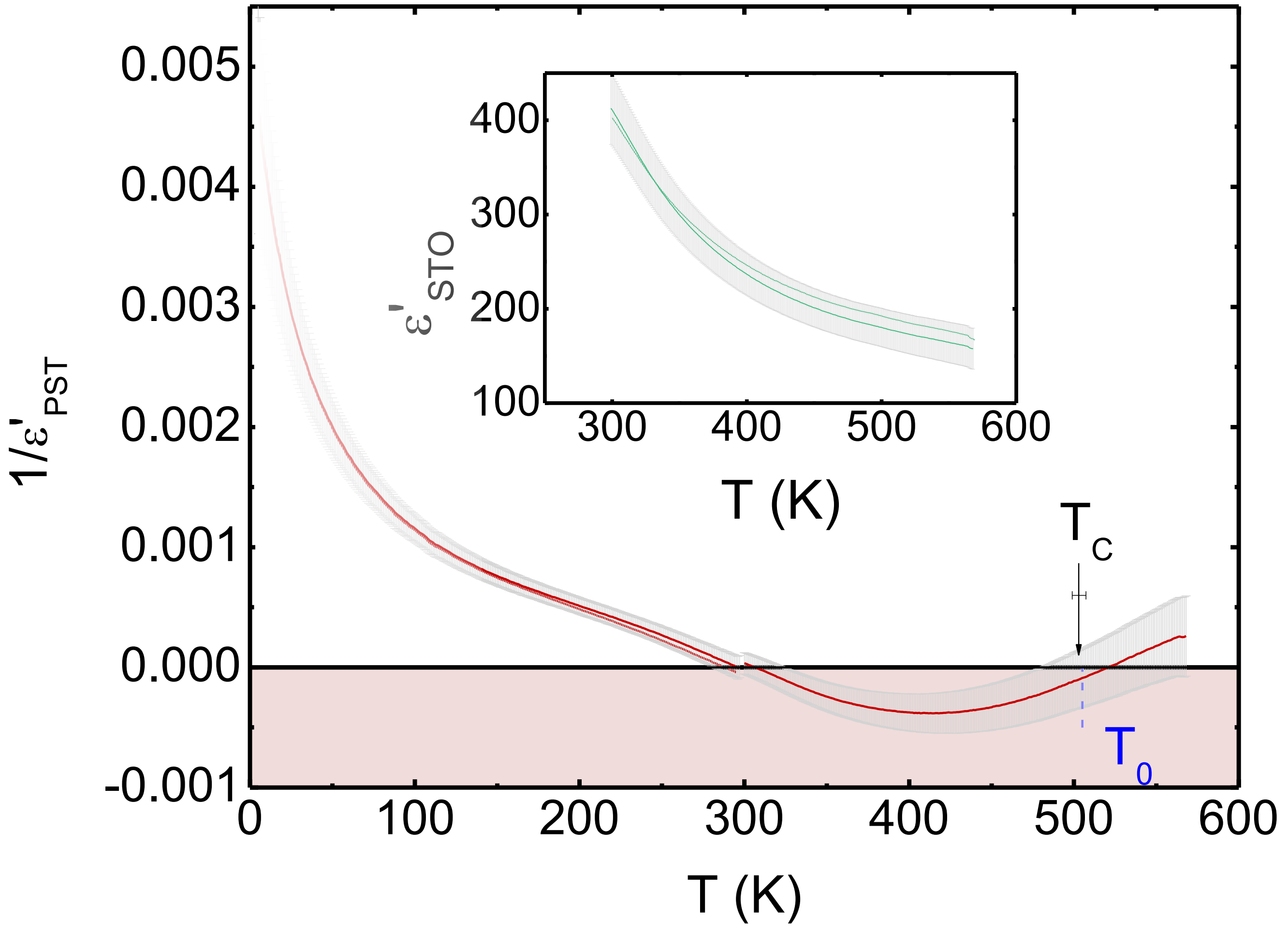
(Pb,Sr)TiO_3-$SrTiO_3$ superlattices



$$\frac{d}{\epsilon} = \frac{d_f}{\epsilon_f} + \frac{d_d}{\epsilon_d}$$

Nature 534, 524 (2016)

PST-$SrTiO_3$ superlattices

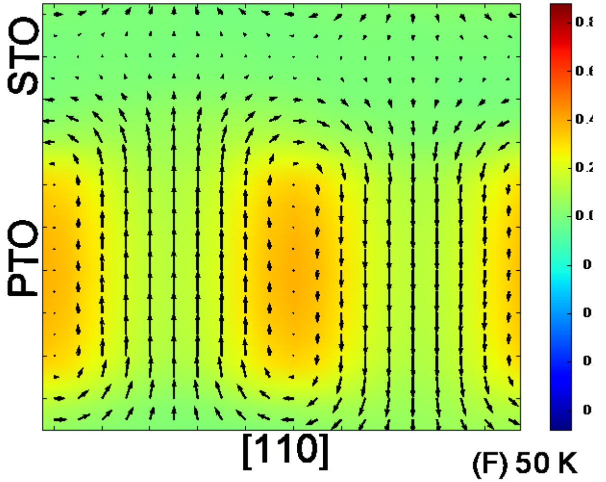


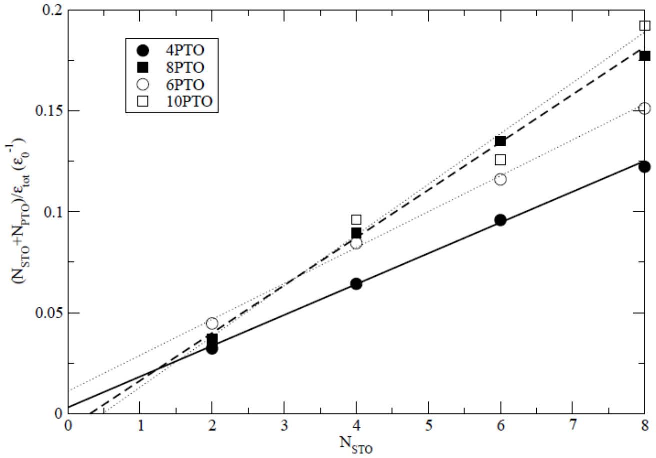
Negative permittivity

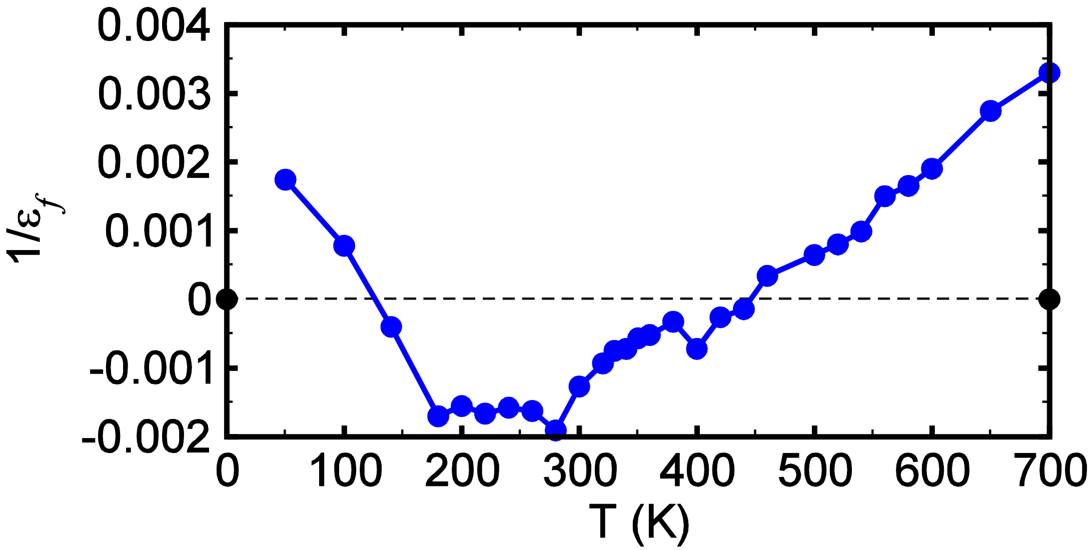
Nature 534, 524 (2016)

Atomistic modelling

Perform experiment on the PC







Jacek Wojdeł & Jorge Íñiguez

Nature 534, 524 (2016)

Local response

Macroscopic quantities:

$$\epsilon = \frac{1}{\epsilon_0}\frac{\Delta D}{E_{ext}} \qquad \chi = \frac{1}{\epsilon_0}\frac{\Delta P}{E_{ext}}$$

—— Externally applied field

Define a local dielectric constant and local 'susceptibility' of a layer $\epsilon_i = \frac{\Delta D_i}{\epsilon_0 \Delta E_i}$

D is continuous across the structure , i.e. $D = P + \epsilon_0 E_{ext} = P_i + \epsilon_0 E_i$ and $\Delta D_i = \Delta D$

$$\epsilon_i = \frac{\Delta D}{\epsilon_0 \Delta E_i} = \frac{\Delta P + \epsilon_0 E_{ext}}{\epsilon_0(\Delta P - \Delta P_i + \epsilon_0 E_{ext})} = \frac{\chi + 1}{\chi - \frac{\Delta P_i}{\epsilon_0 E_{ext}} + 1}{}_0$$

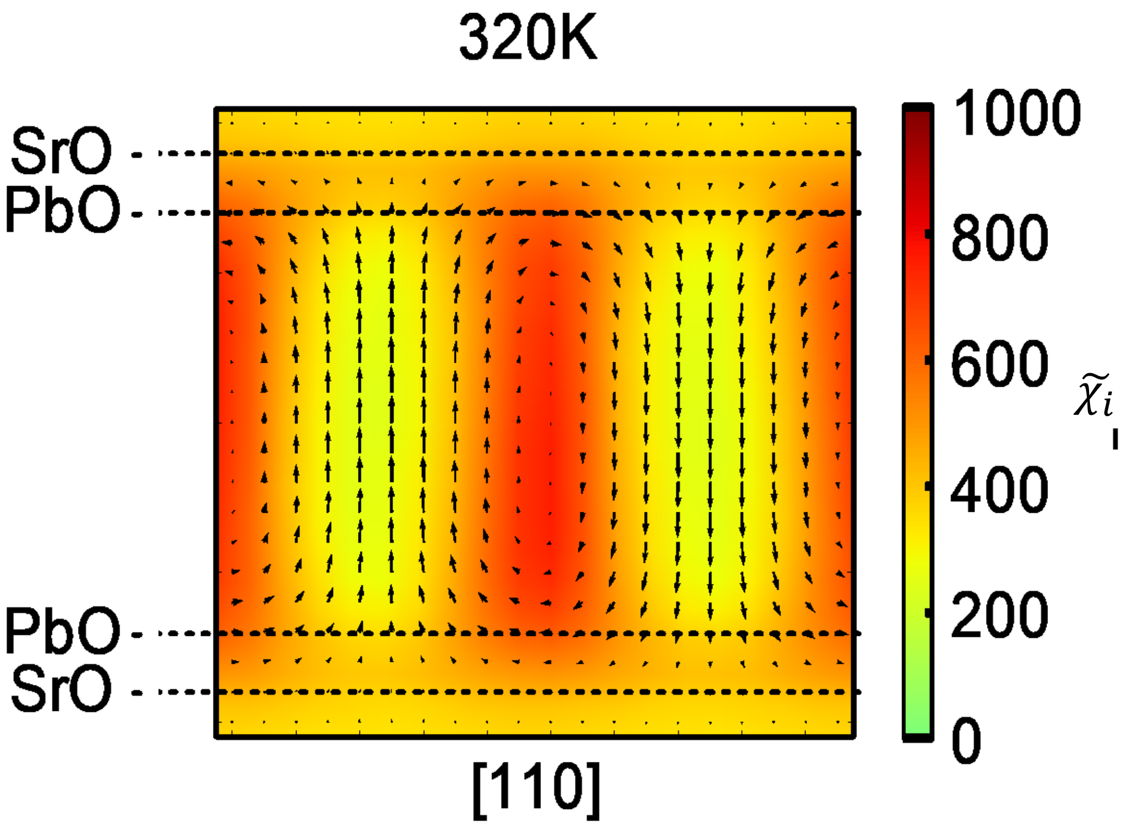
$$\boxed{\epsilon_i = \epsilon_0 \frac{\epsilon}{\epsilon - \epsilon_0 \widetilde{\chi}_i}}$$

where $\widetilde{\chi}_i \equiv \frac{1}{\epsilon_0}\frac{\Delta P_i}{E_{ext}}$ (measure of how responsive that layer is to the applied field)

Layer permittivity will be negative if $\epsilon_0 \widetilde{\chi}_i > \epsilon$, i.e. if locally it is (sufficiently) more responsive than the system as a whole

Nature 534, 524 (2016)

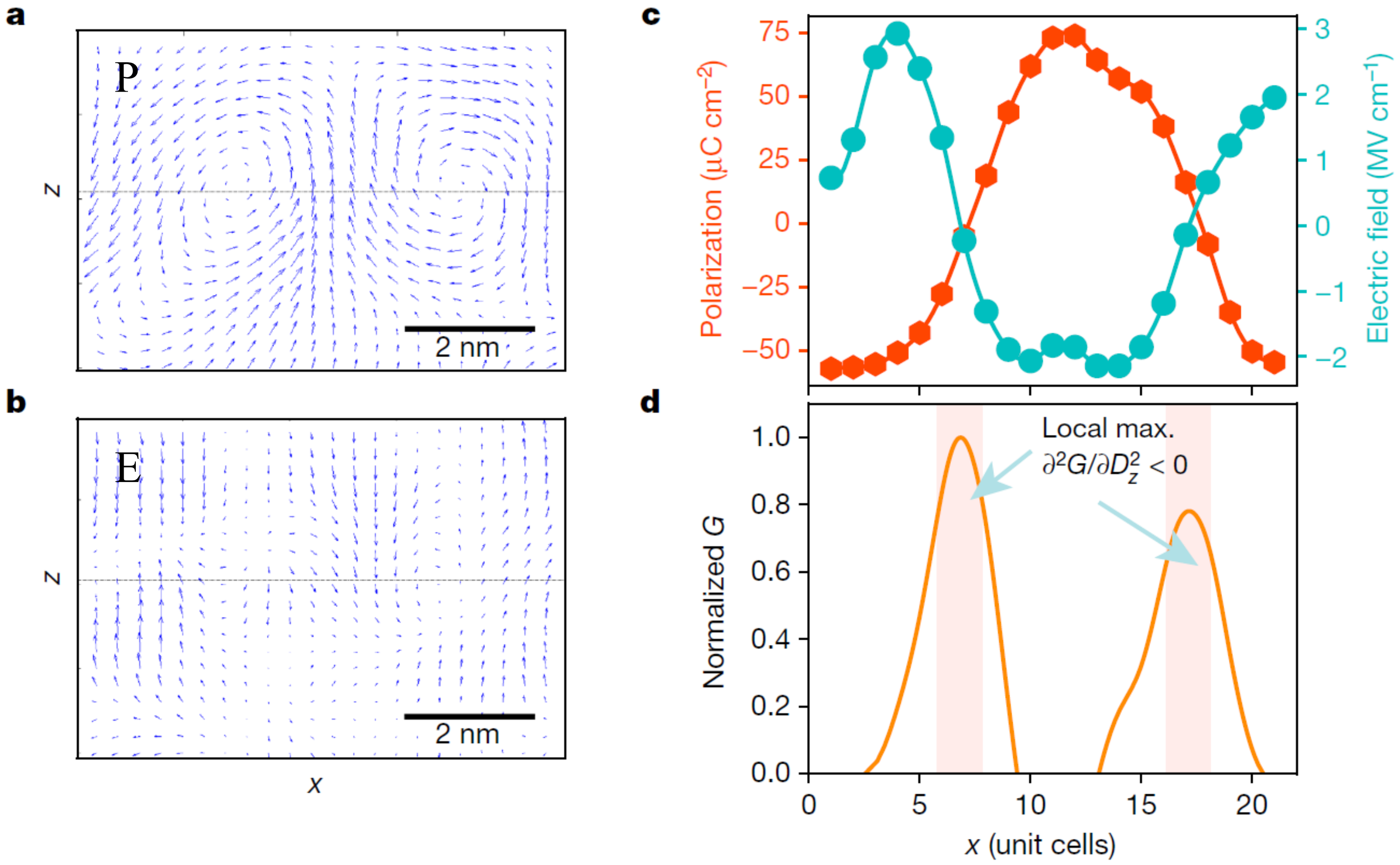
Local response

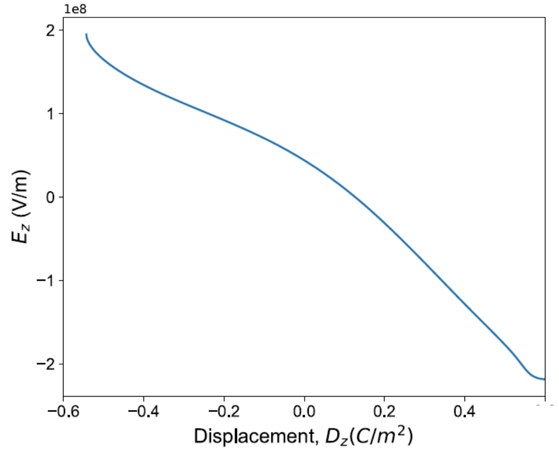


More responsive regions are near domain walls and ferro-para interface

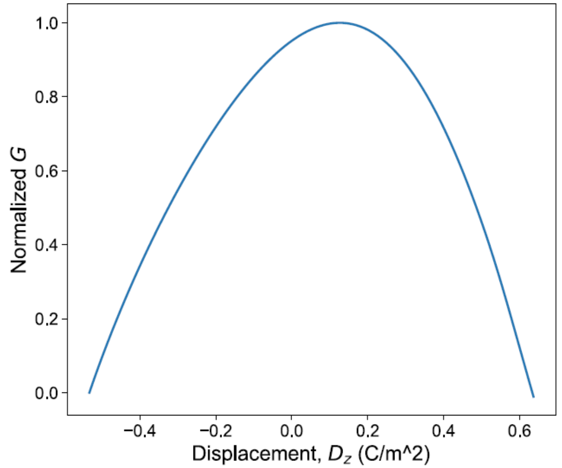
Nature 534, 524 (2016)

“Direct” measurement





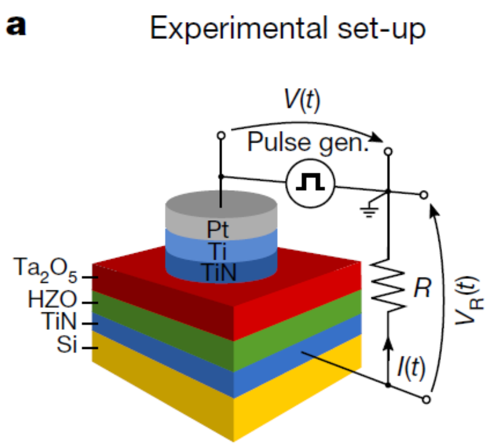
$$G = \int E_z dD_z$$



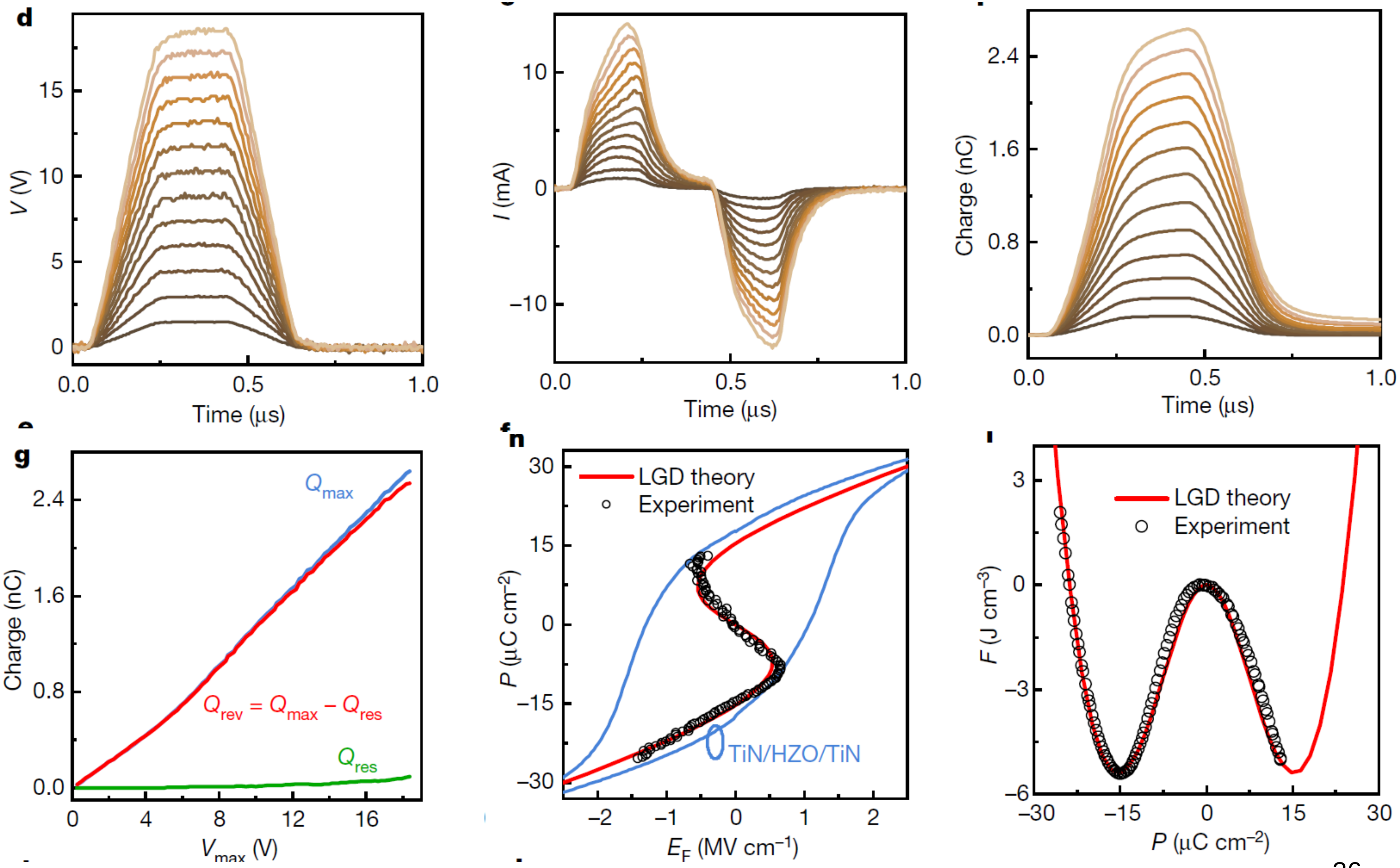
$$\epsilon = \frac{1}{\epsilon_0}\frac{dD}{dE}$$

Yadav et al. Nature 565, 468 (2019)

NC in hafnia: pulsed measurements

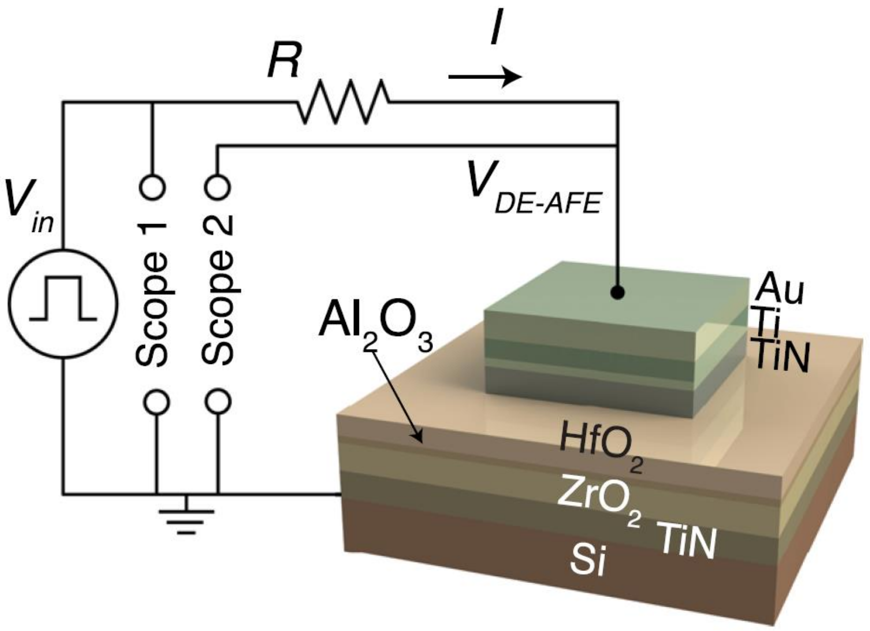


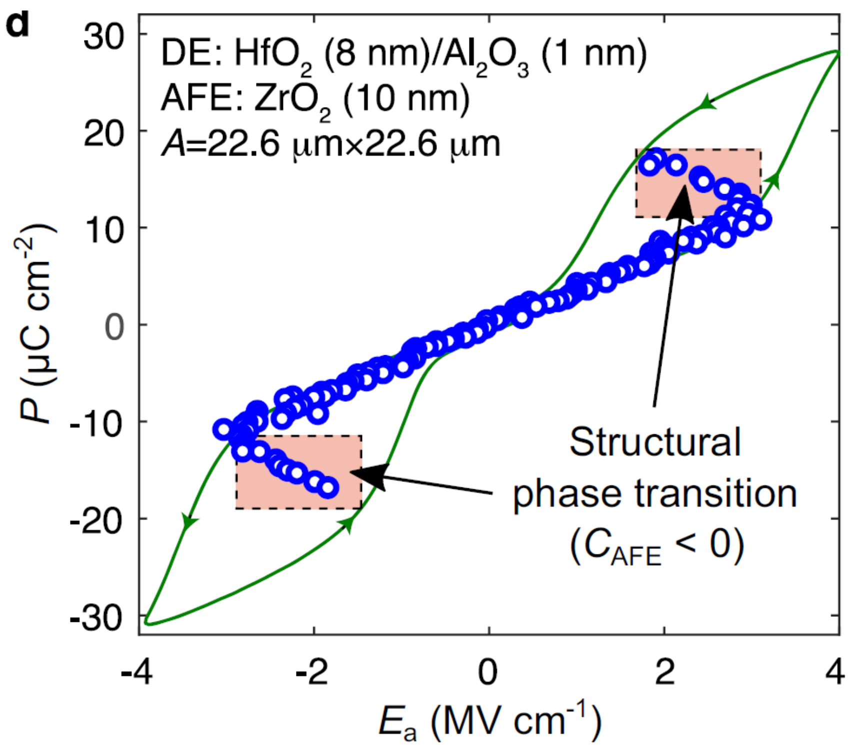
No low-frequency capacitance enhancement but…



Hoffmann et al. Nature (2019)

Antiferroelectric NC



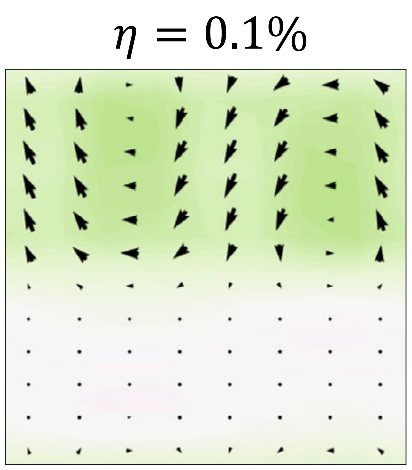


Hoffmann et al. Nature Comms (2022)

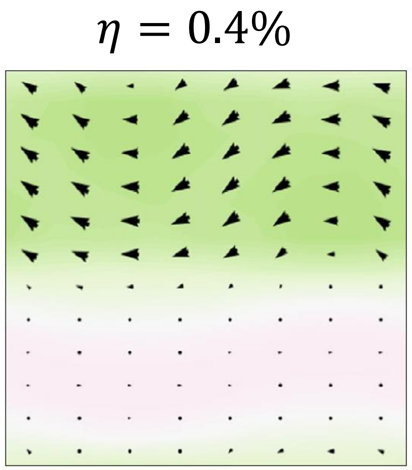
See also: Karda et al. APL 106, 163501 (2015)

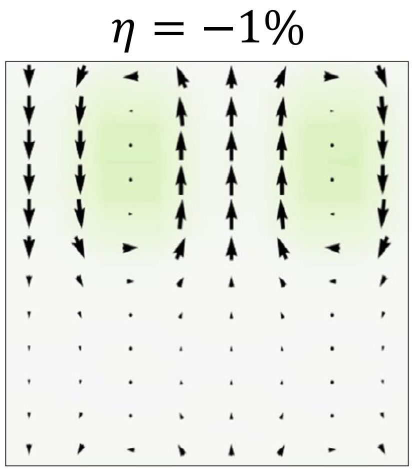
Voltage amplification from 'incipient states'

$PbTiO_3/SrTiO_3$ SLs

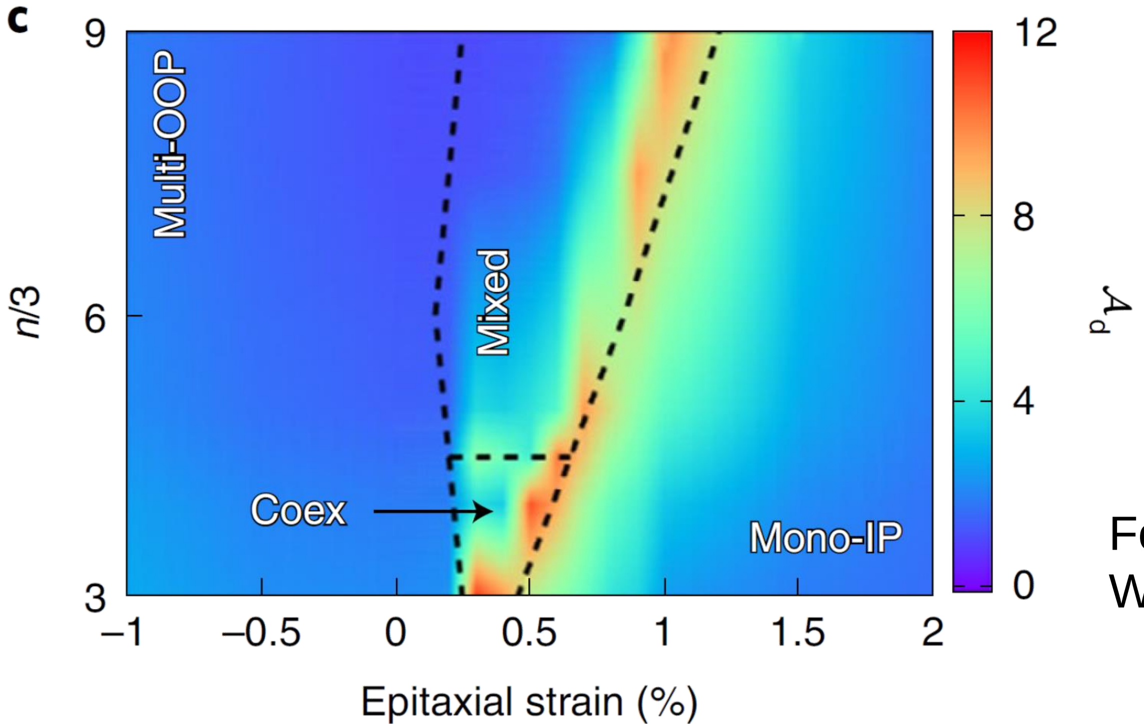


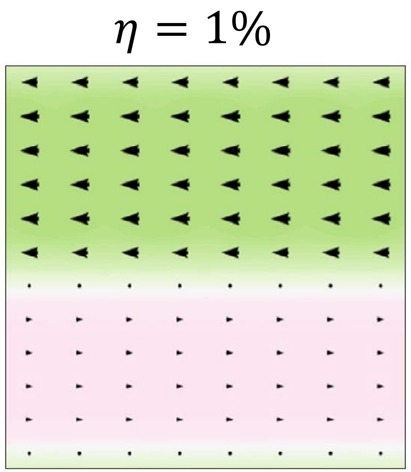
Mixed





Multi-OOP



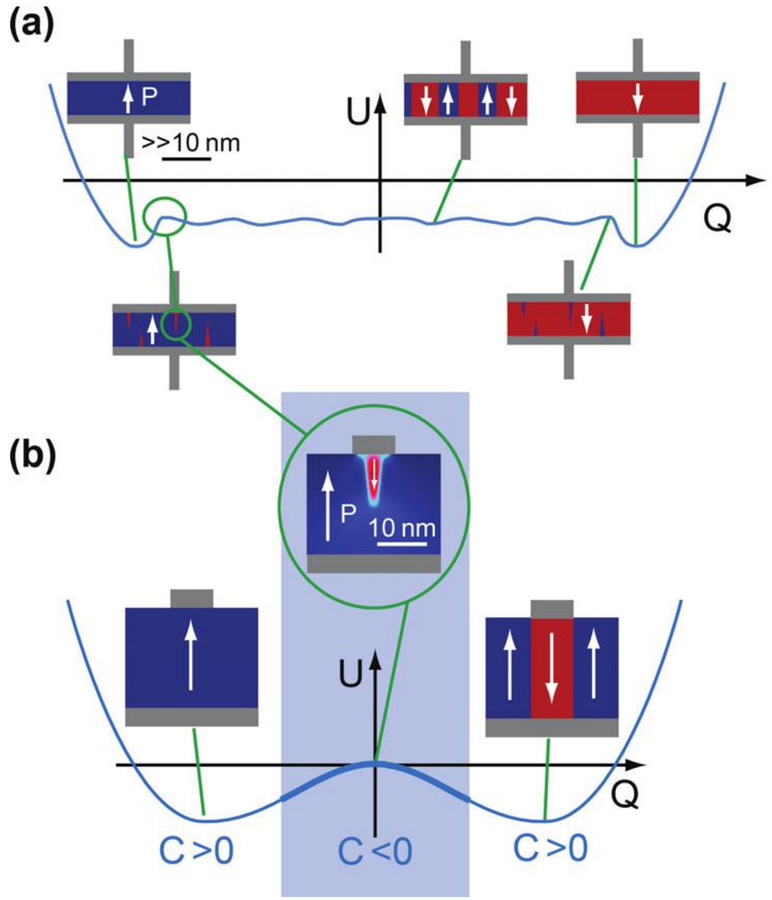


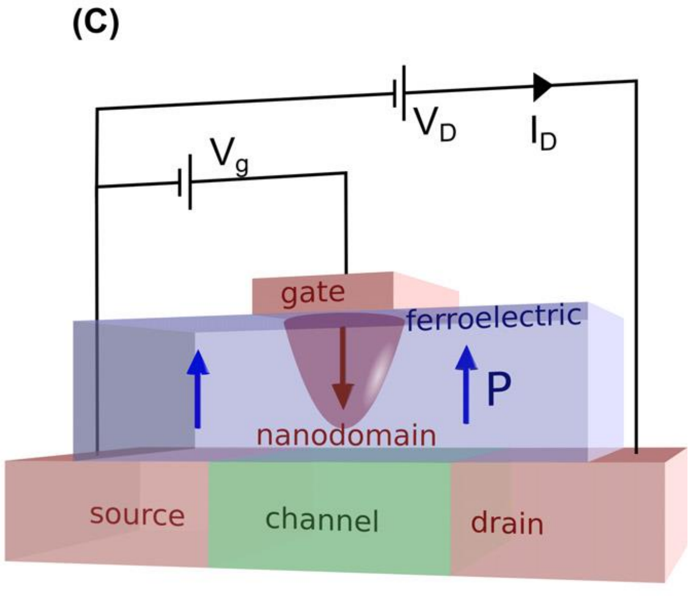
Mono-IP

Graf et al. Nature Materials (2022)

For $BaTiO_3/SrTiO_3$ SLs, see: Walter et al. npj Comp. Mat. 2020

NC in nanocapacitors

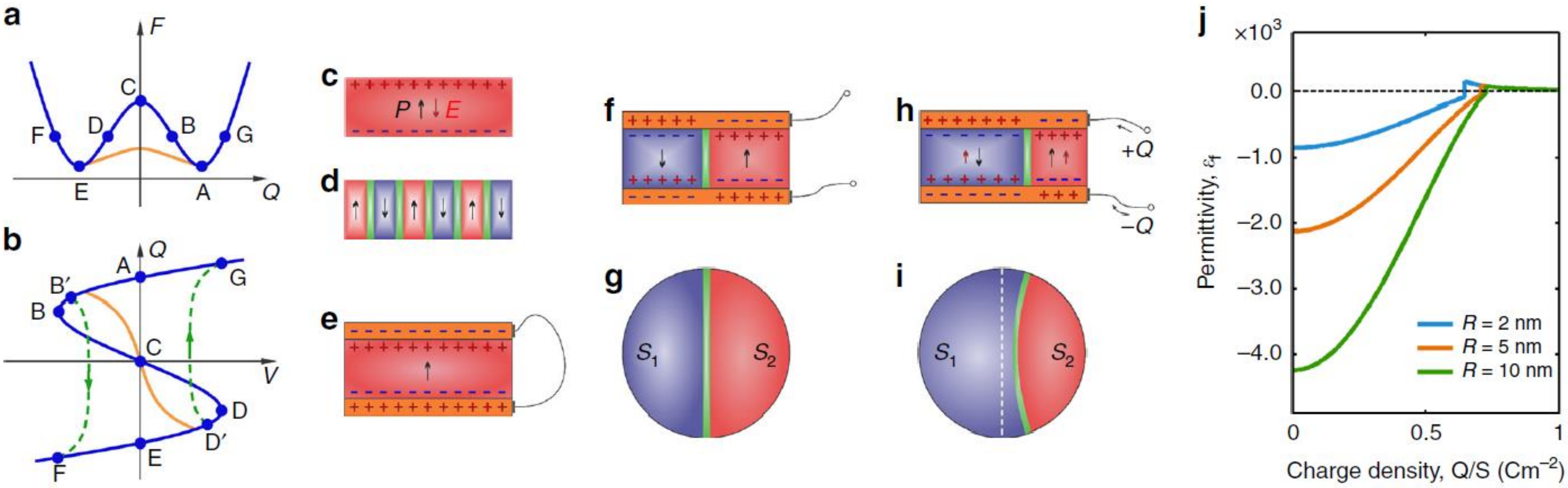




Reduce dimensions of device to match critical nucleus size → large depolarization field & enhanced NC effect

Sluka et al. APL 111, 152902 (2017)

NC in nanocapacitors



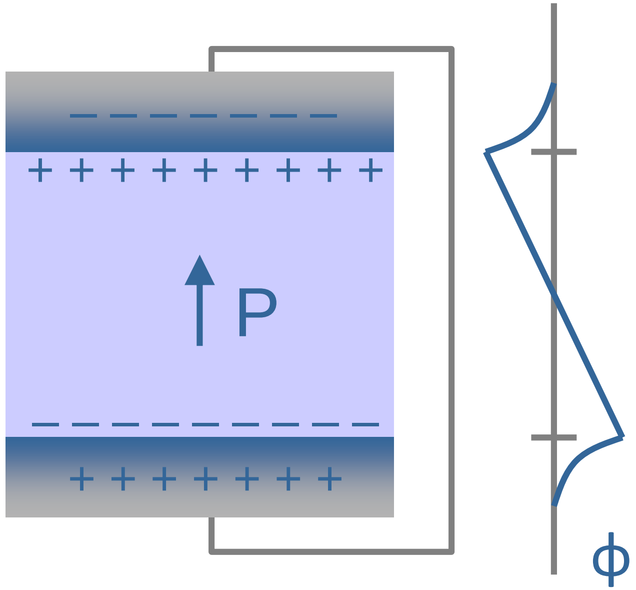
Change in DW length during switching provides additional energy lowering that leads to NC

Luk’yanchuk et al. Comm. Phys. 2, 22 (2019)

Screening at metal-ferroelectric interfaces

What is the effect of free carriers?

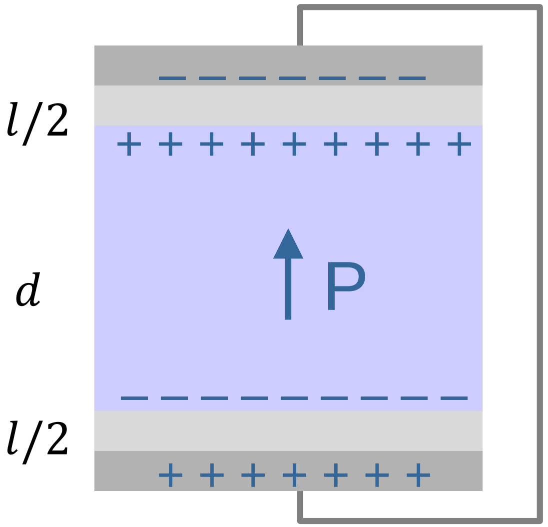
Real electrodes
(imperfect screening)



$\lambda_{\mathrm{eff,SRO}} \approx 15\ \mathrm{pm}$

$$\lambda_{\mathrm{eff}} = \frac{l}{\epsilon_r}$$

Perfect electrodes +
dead layer



11 u.c. of $SrTiO_3$ ($\epsilon = 300$)
$\rightarrow\ \lambda_{\mathrm{eff}} = 14\ \mathrm{pm}$

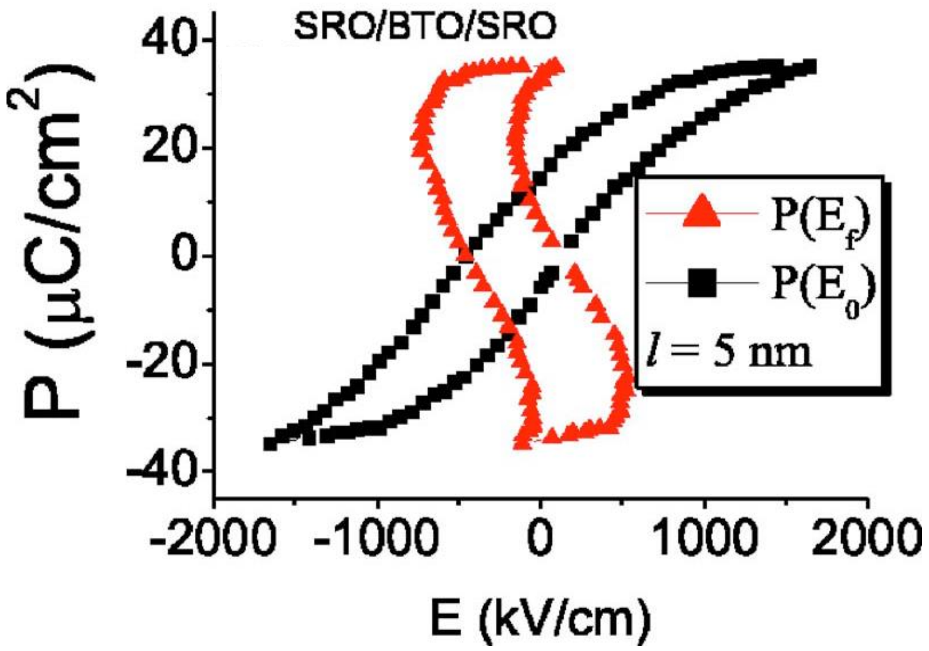
Domains form just as readily in capacitors with metallic $SrRuO_3$ as in superlattices with insulating $SrTiO_3$!

APPLIED PHYSICS LETTERS 89, 253108 (2006)

Depolarizing field and "real" hysteresis loops in nanometer-scale ferroelectric films

Bratkovsky & Levanyuk

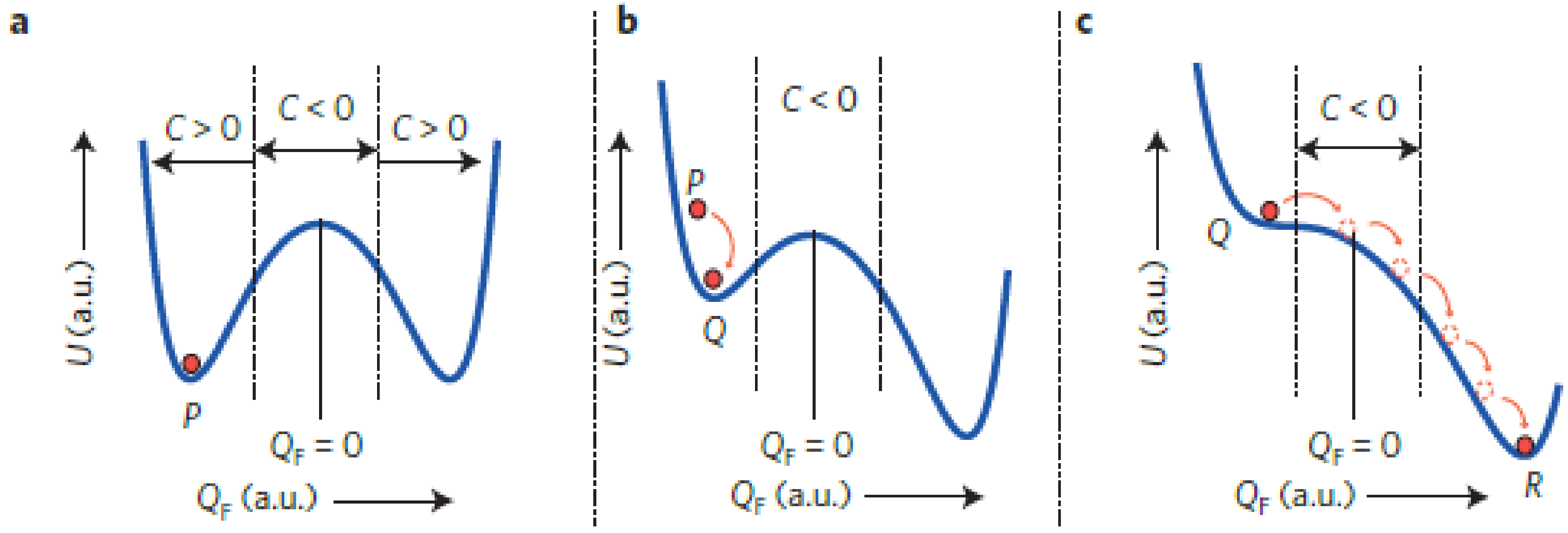
- Model as imperfectly screened ferroelectric capacitor
- Estimate screening length λ_{eff} theoretically → interface capacitance C_i
- Calculate voltage drop across C_i and thus actual field in ferroelectric (E_F)



→ Hysteresis loops have negative slope

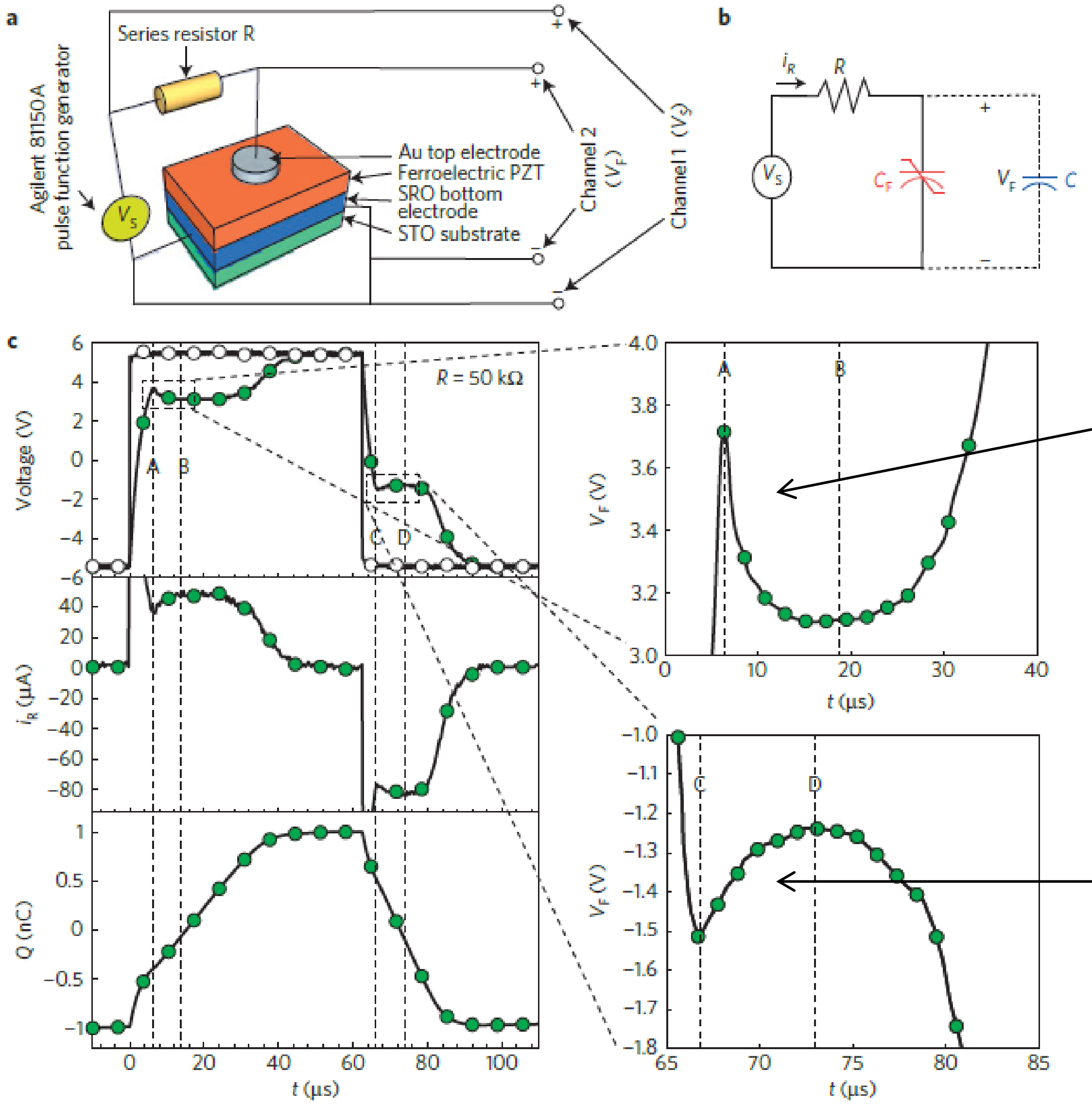
‘Transient’ negative capacitance

Ferroelectric switching



Khan et al. Nature Mater 2015

‘Transient’ negative capacitance



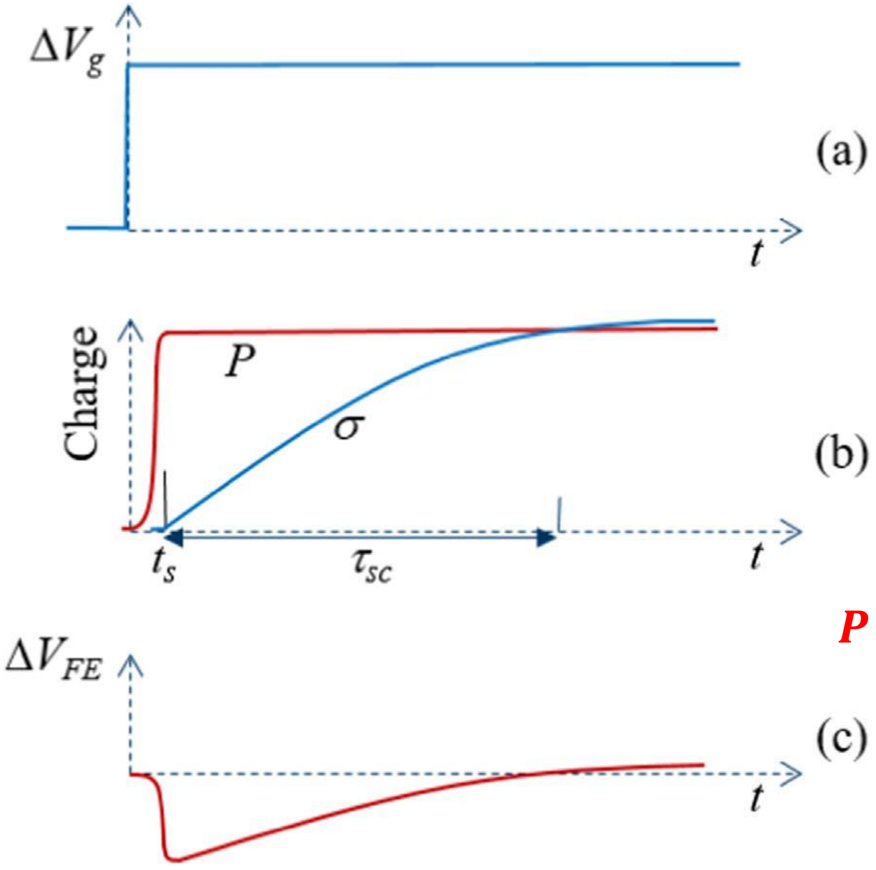
Slow down delivery of screening charge during switching via series resistance

V decreases while Q increases

V increases while Q decreases

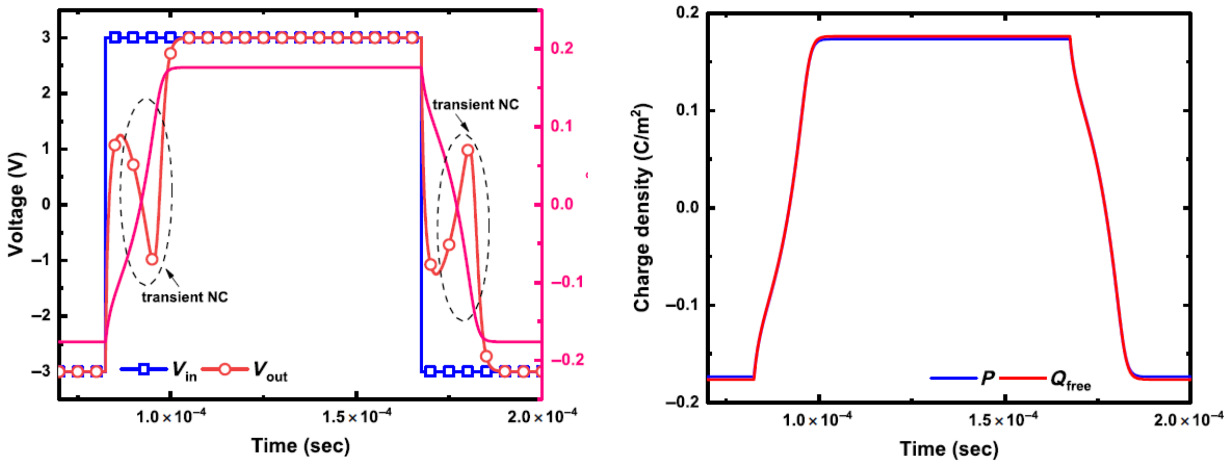
Khan et al. Nature Mater 2015

‘Transient’ negative capacitance



$P > D$

Ng et al. Solis State Commun. 265, 12 (2017)

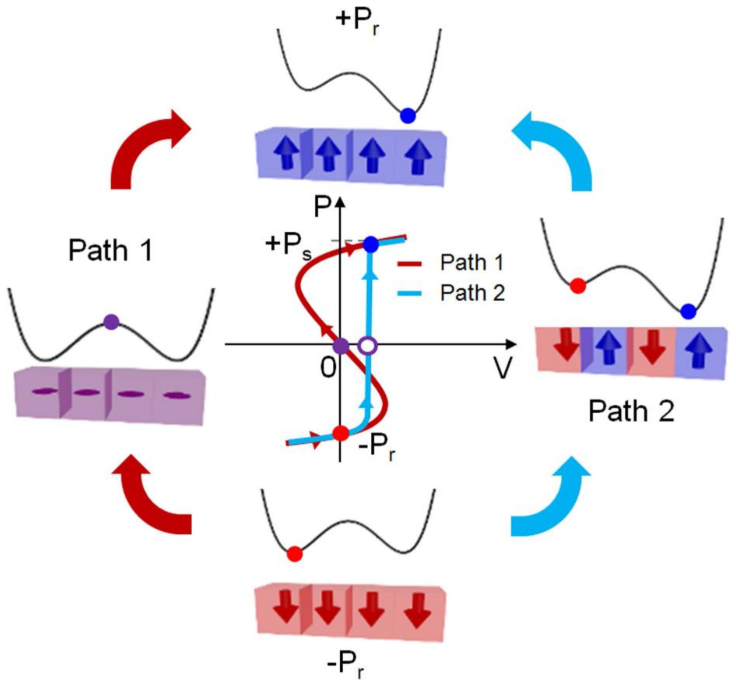


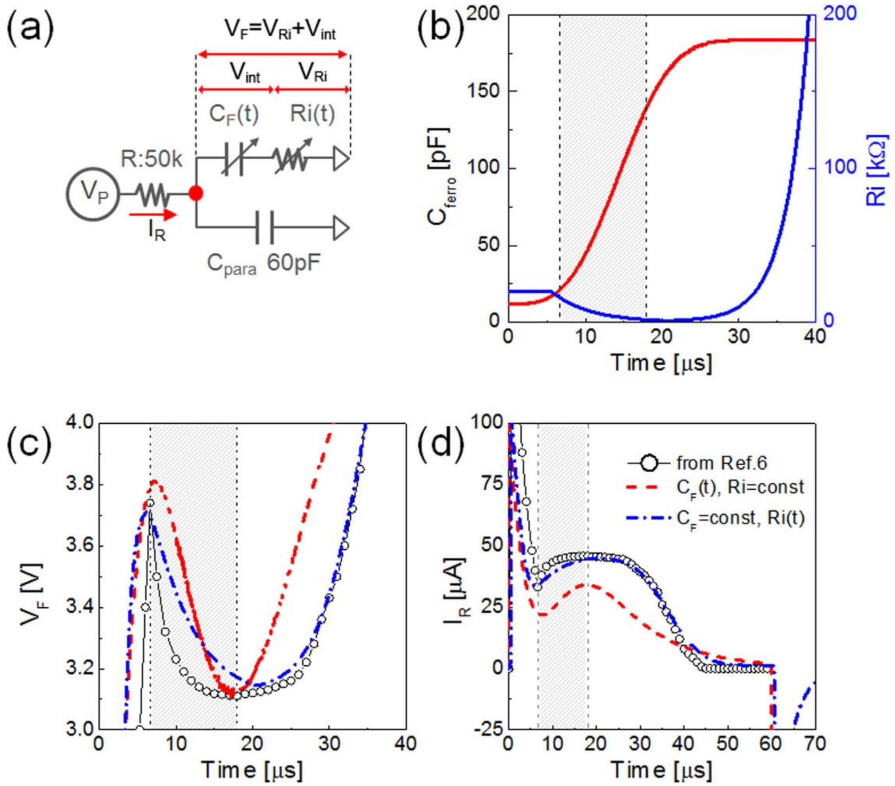
Chang et al. Phys Rev Appl. 9,014010 (2018)

Depolarizing field will slow down the actual switching process; P only slightly exceeds D at any time

Homogenous vs domain mediated switching

Are we really traversing the Landau energy maximum?



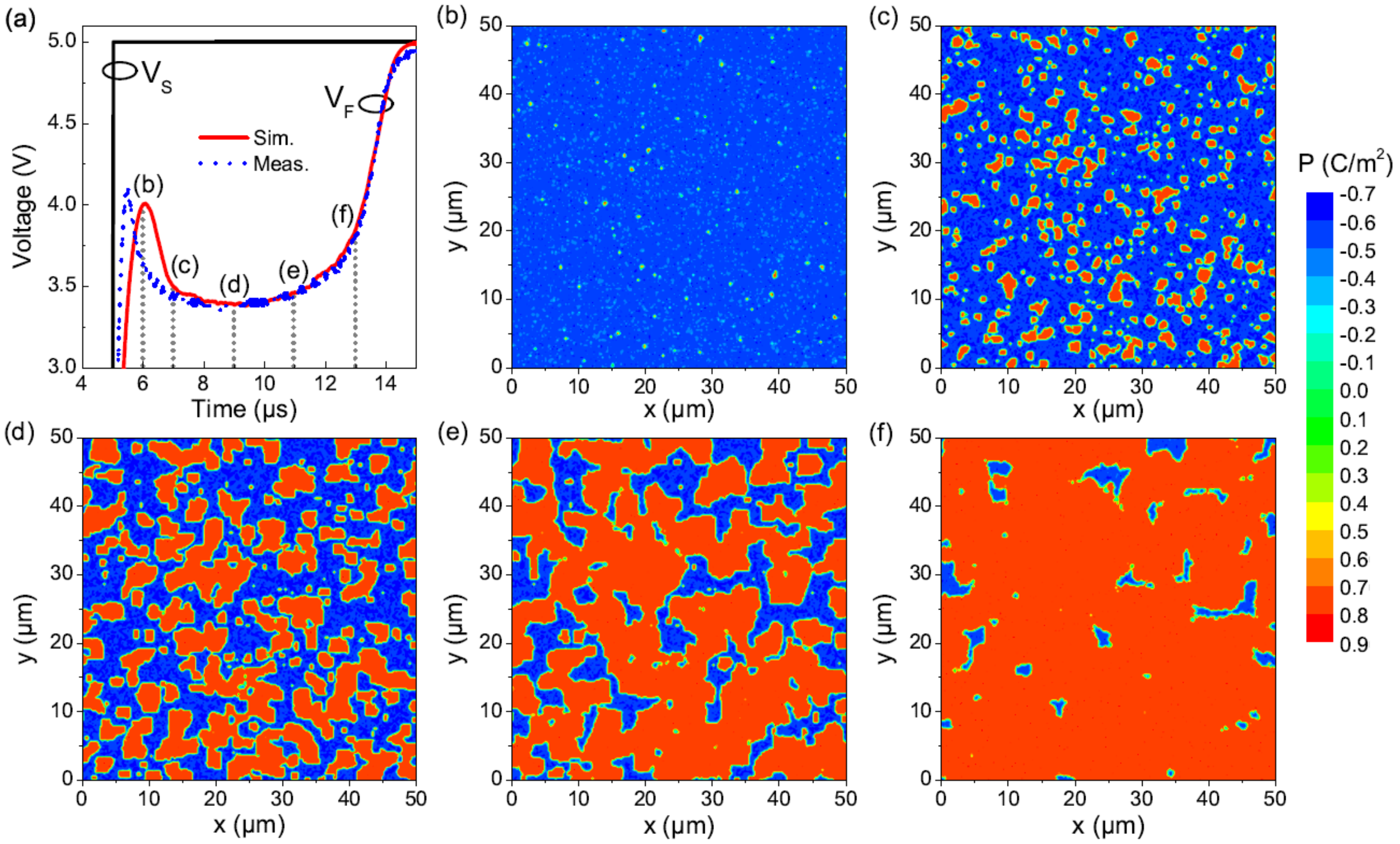


Song et al. Sci. Rep . 2016

See also Saha et al. JAP 123, 105102 (2018)

Homogeneous switching is very unlikely

‘Transient’ NC with domains

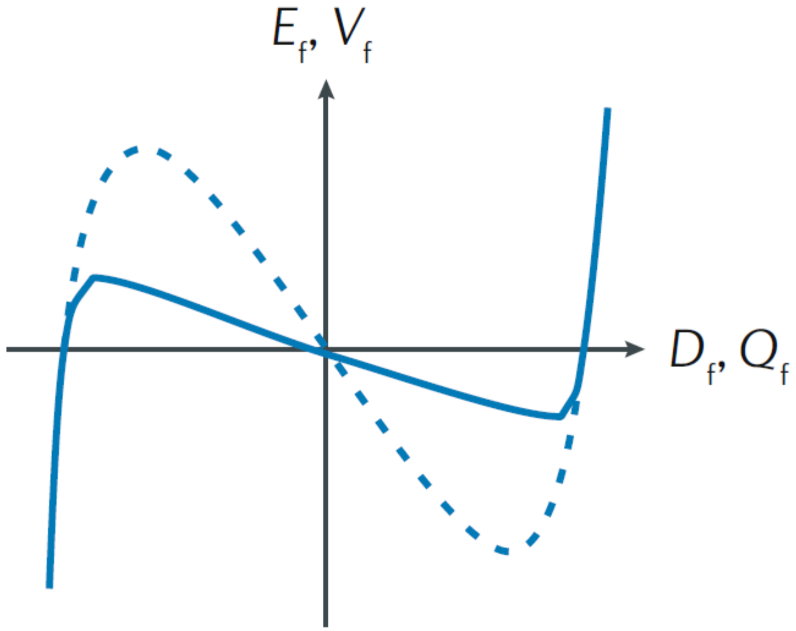


Hoffmann et al. JAP 123, 184101 (2018)

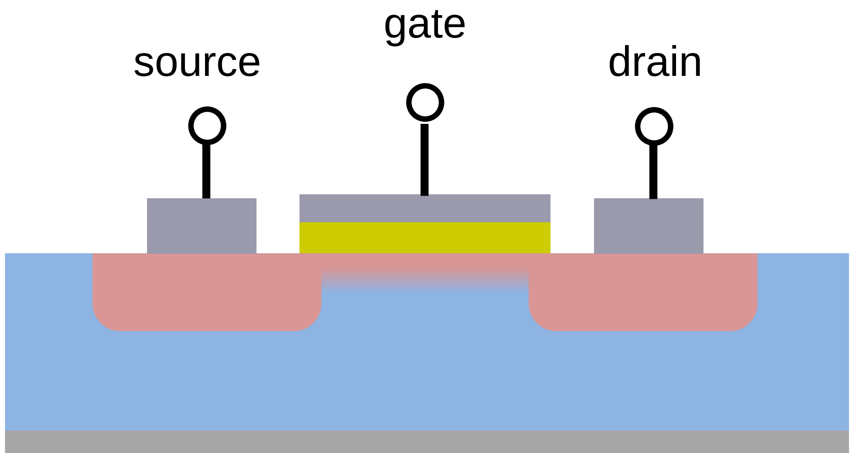
In practice, switching is not homogeneous, but can still observe ‘transient’ NC

‘Transient’ negative capacitance

- Another method of achieving ‘charge control’
- Manifested in opposite signs of $\frac{dQ}{dt}$ and $\frac{dV_f}{dt}$
- Should be present in *any* activated system
- Not (yet?) useful for applications



FeFET vs NCFET



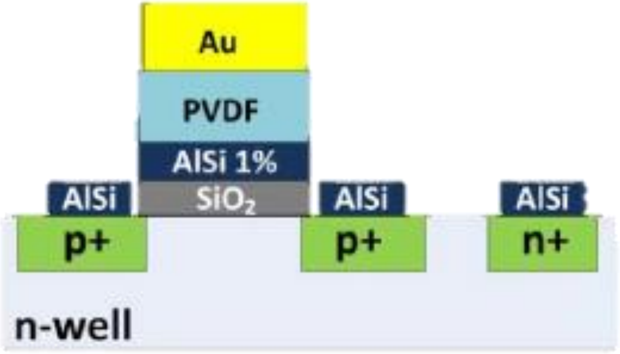
FeFET

- Memory device (requires polarization stability (hysteresis)
- Typically MFIS
- Insulator layer reduces interface traps but destabilizes polarisation

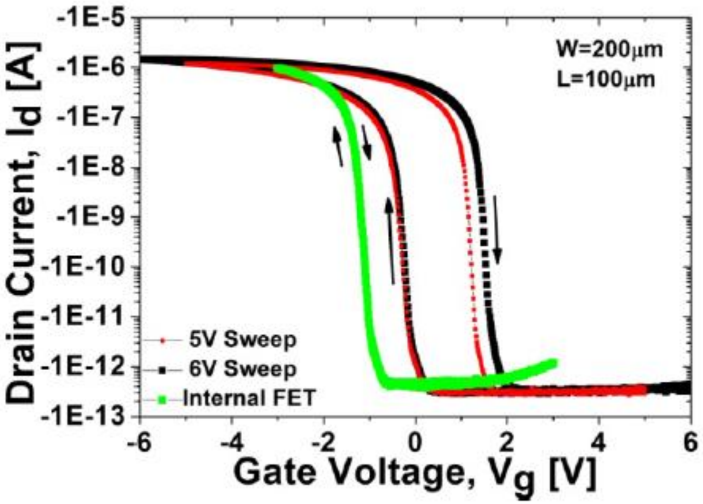
NCFET

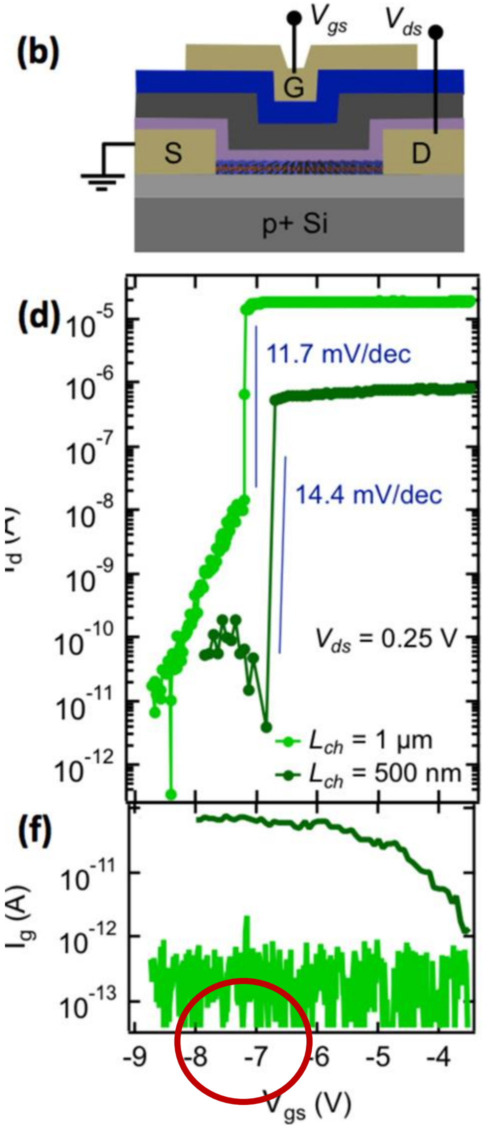
- Logic device
- Should be
  - Hysteresis free
  - Unstable polarization
  - Small, negative ferroelectric capacitance

Early NCFET structures



Rusu et al. IEEE 2010

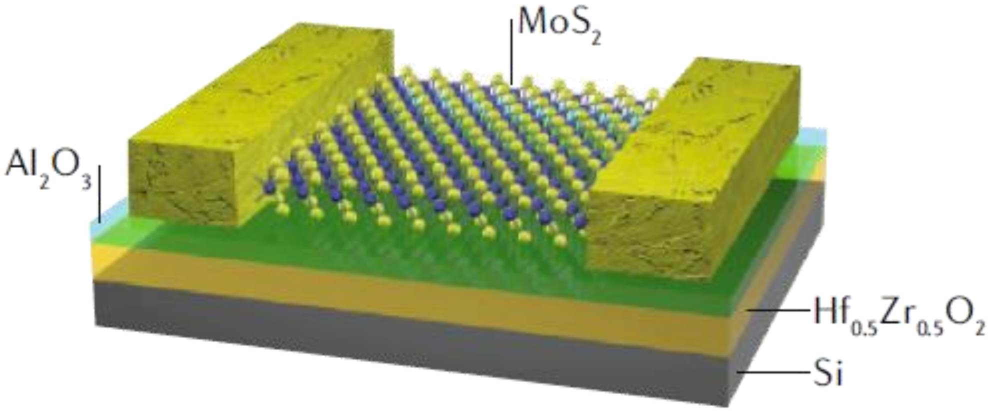


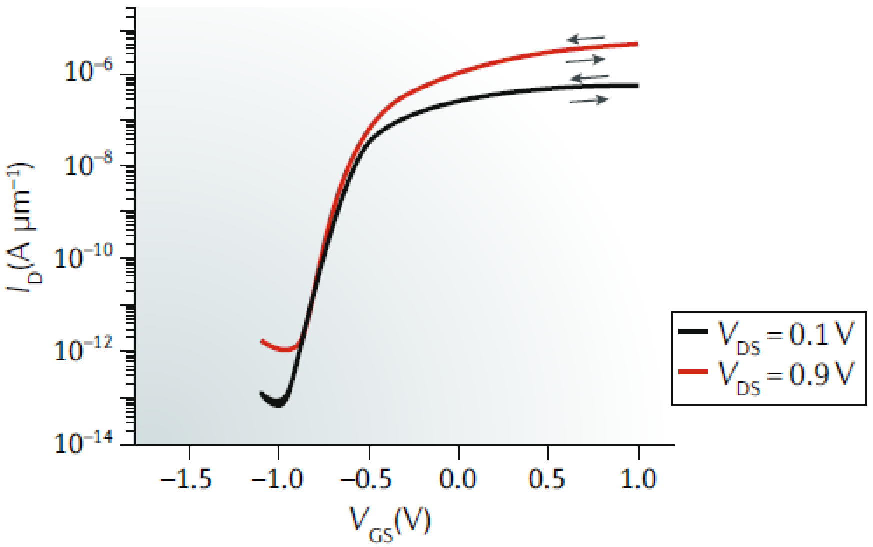


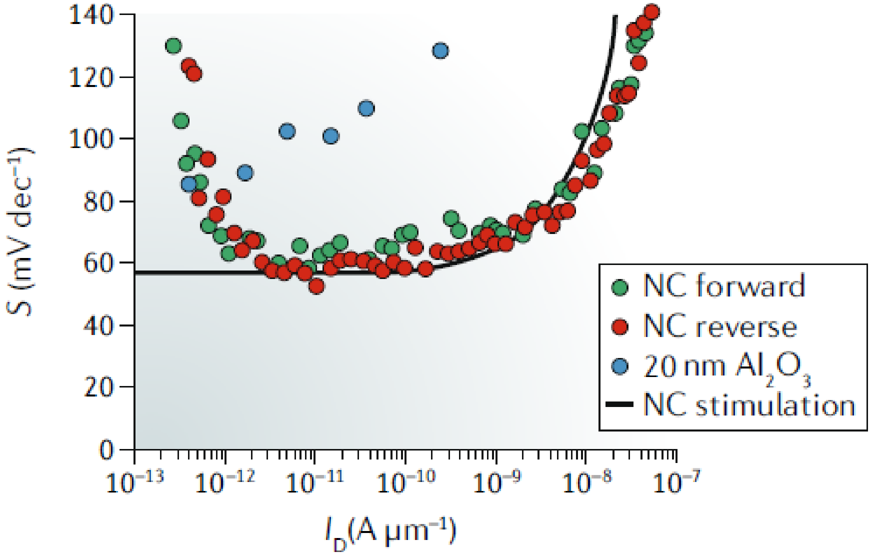
Sharp I_d-V_g characteristics observed but always accompanied by hysteresis and/or large V_g required

McGuire et al. APL 2016

Hafnia-based devices







Si et al. Nature Nano. 13, 24 (2018)

14nm Ferroelectric FinFET Technology with Steep Subthreshold Slope for Ultra Low Power Applications

Z. Krivokapic[3], U. Rana[1], R. Galatage[2], A. Razavieh[2], A. Aziz[1], J.Liu[2], J.Shi[3], H.J. Kim[1], R. Sporer[1], C. Serrao[1], A.Busquet[1], P. Polakowski[4], J. Müller[4], W. Kleemeier[2], A. Jacob[1], D. Brown[2], A. Knorr[2], R. Carter[1], and S. Banna[3]

[1]GLOBALFOUNDRIES, Malta, NY, USA, [2]GLOBALFOUNDRIES, Albany, NY, USA, [3]GLOBALFOUNDRIES, Santa Clara, CA, USA, [4]Fraunhofer IPMS, Dresden, Germany, email: zoran.krivokapic@globalfoundries.com

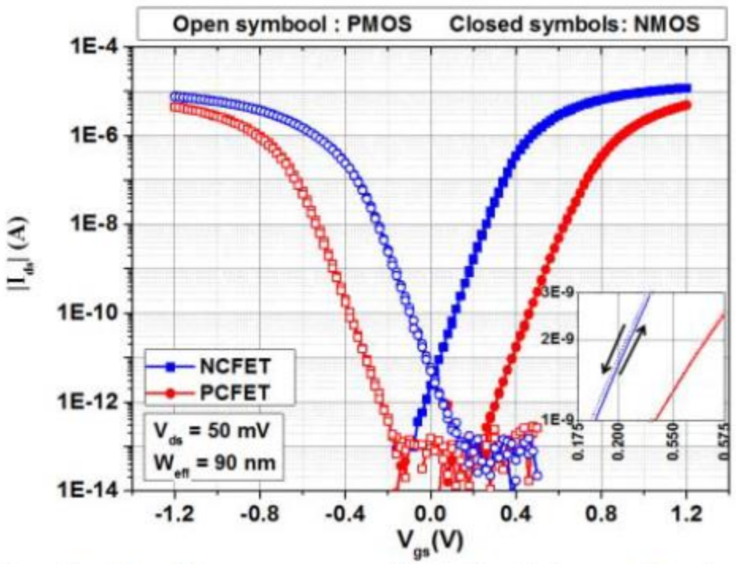
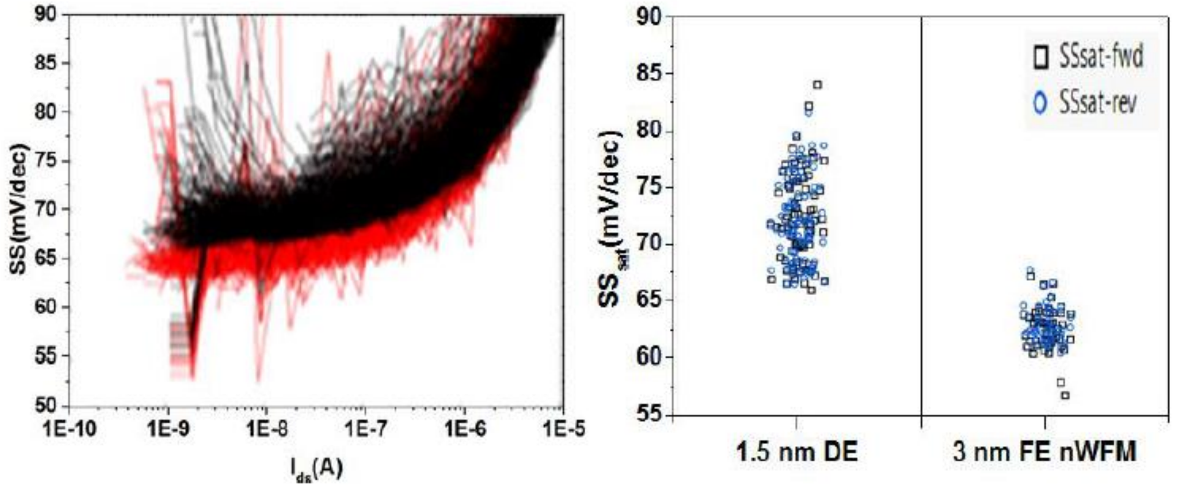


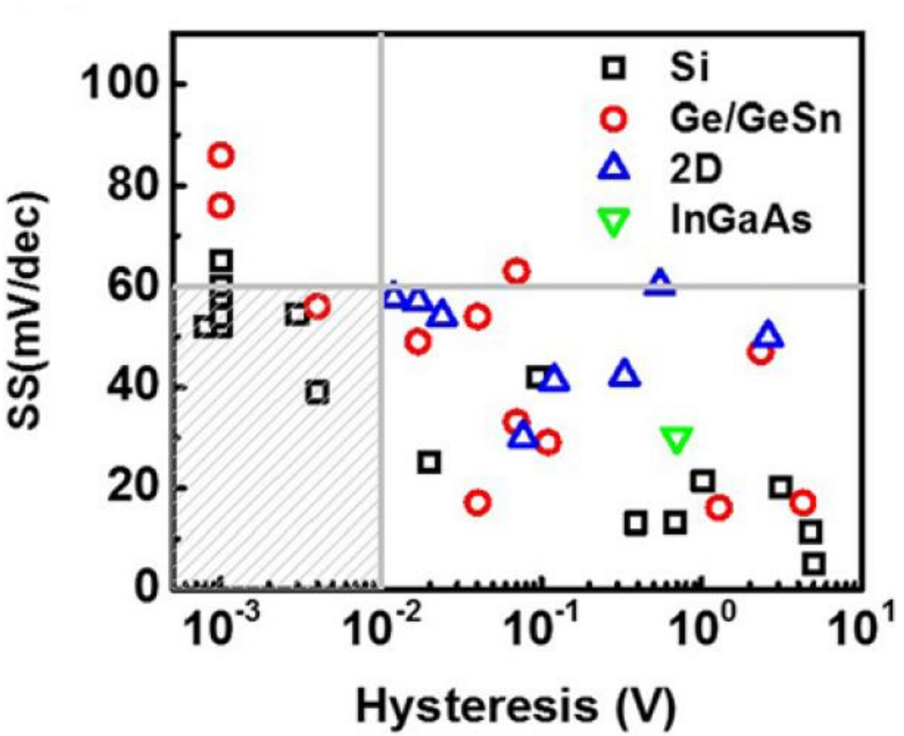
Fig. 8: I_{ds}-V_{gs} curves for, L=14nm device with IL_A and 8nm FE layers. Inset shows slight ferroelectric hysteresis.



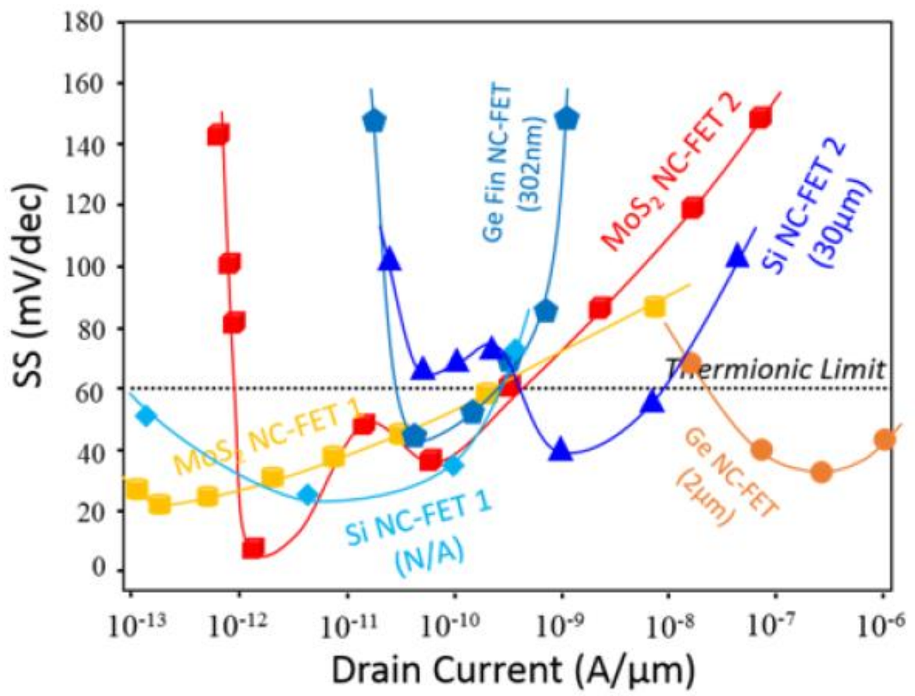
Industry turning to hafnia based ferroelectrics

IEDM17-357 (2017)

Current state



Alam et al. APL (2019)



Cao et al. Nature (2023)

Summary

- Aim: voltage amplification (want a ***small*** negative capacitance/permittivity)
- Negative permittivity is a ‘local’ effect (possible in only part of the system that has positive permittivity overall)
- NC corresponds to “overscreening”
- Occurs when the ferroelectric polarization is suppressed ***electrostatically***
- The key is to control the charge
- Devices development focused on hafnia – we need to better understand ferroelectricity in hafnia