A microscopic `toy' model of ferroelectric negative capacitance

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EDTM 2020, April 6, 2020

Is a negative capacitance is a <u>real, physical phenomenon</u> or an <u>unphysical, artificial construct</u> of the phenomenology of the Landau Theory?

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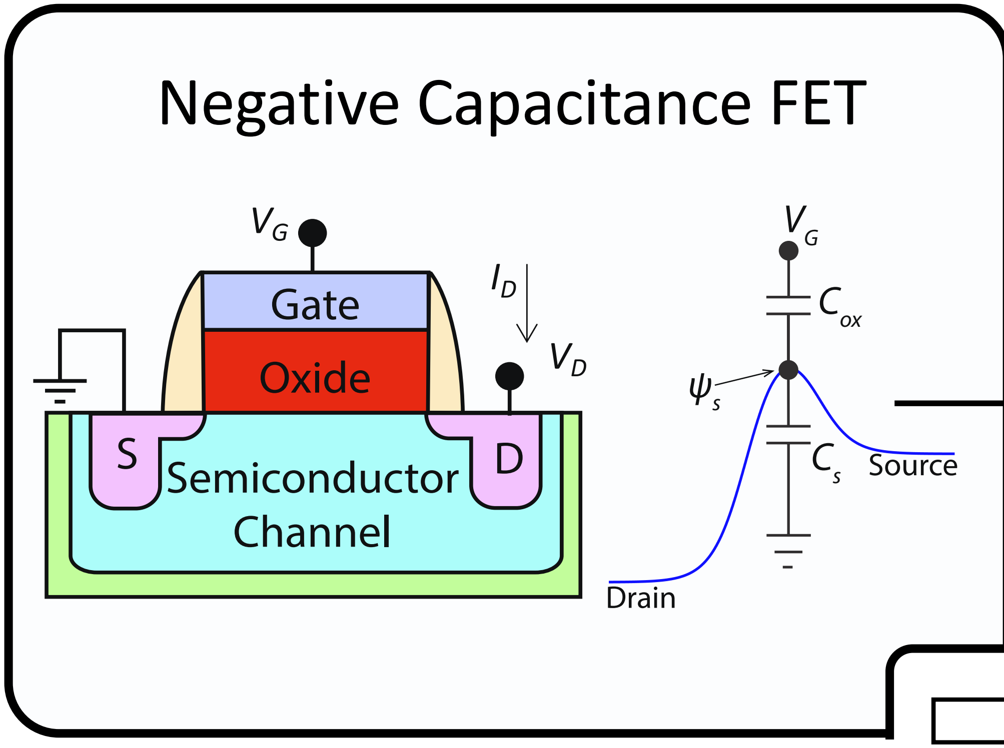
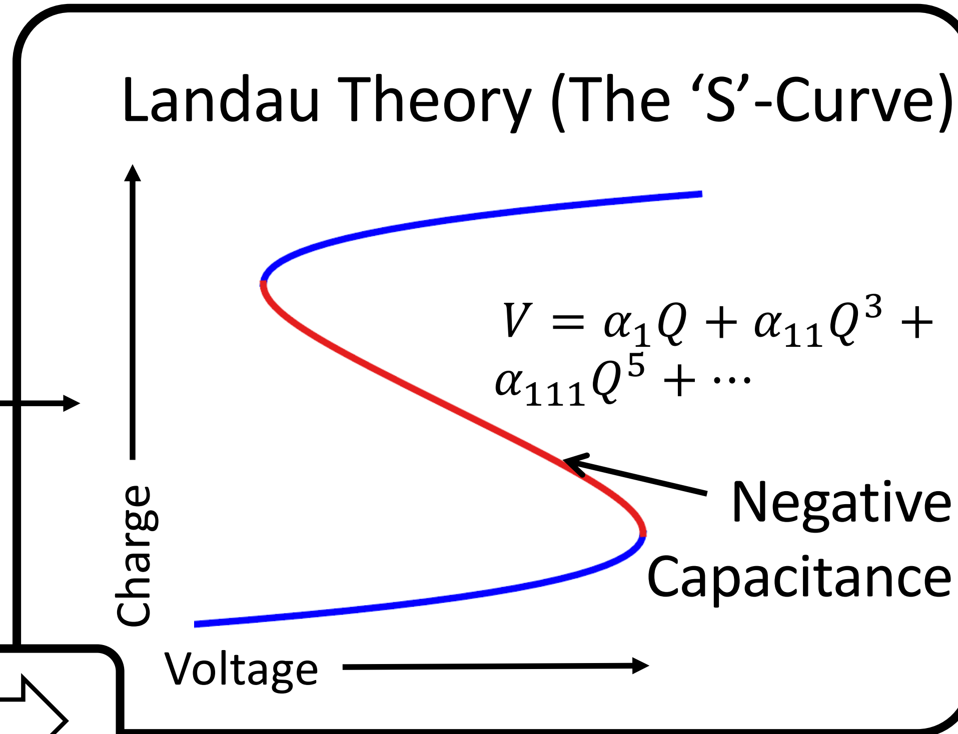


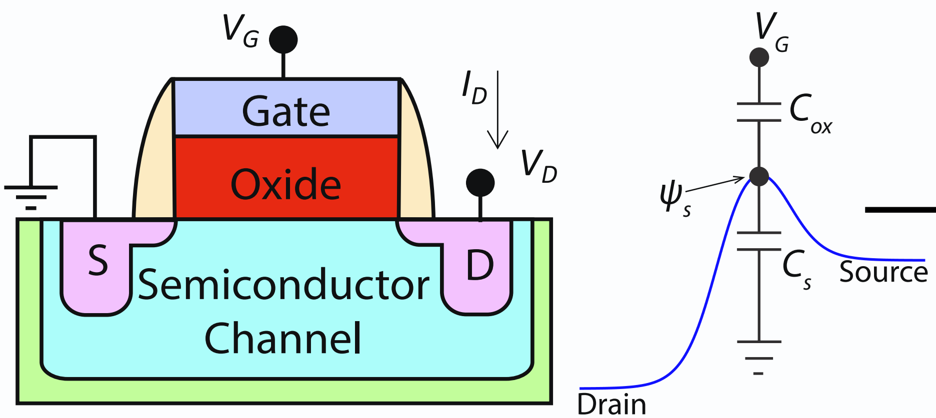


Where does the `S'-shape come from?

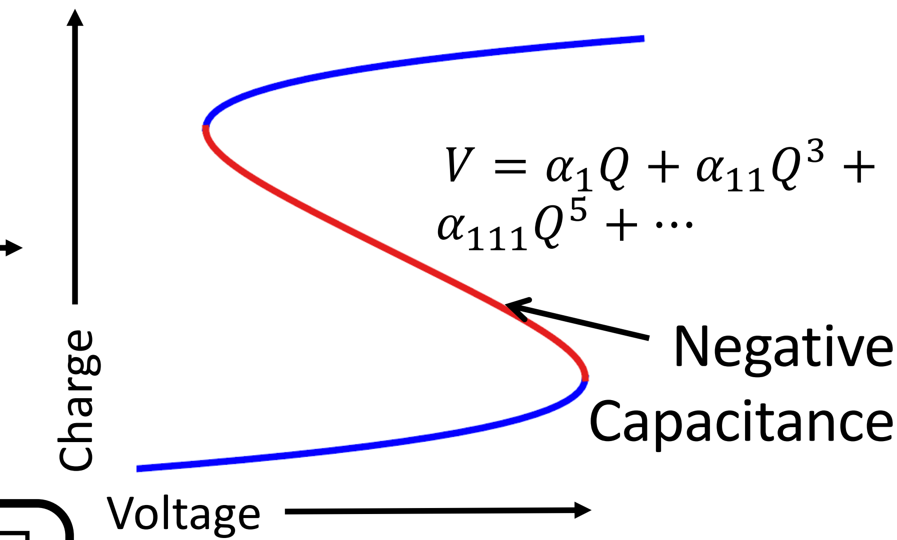
-Microscopically

(Landau theory is phenomenological.)

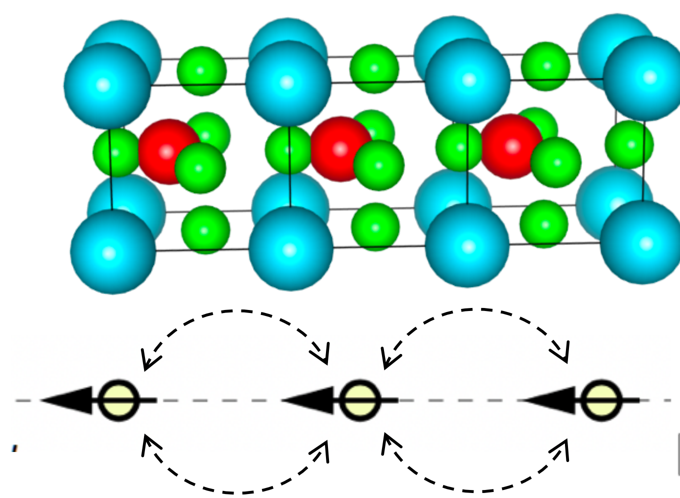
Negative Capacitance FET



Landau Theory (The 'S'-Curve)



Microscopic Origin



Where does the `S'-shape come from?

-dipole-dipole interaction

(atomic scale positive feedback)

Is a negative capacitance is a real, physical phenomenon or an unphysical, artificial construct of the phenomenology of the Landau Theory?

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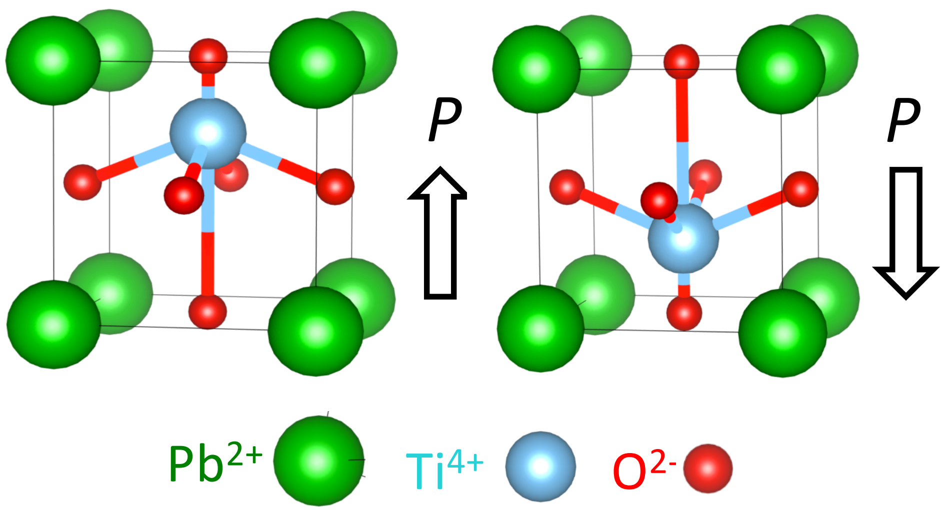
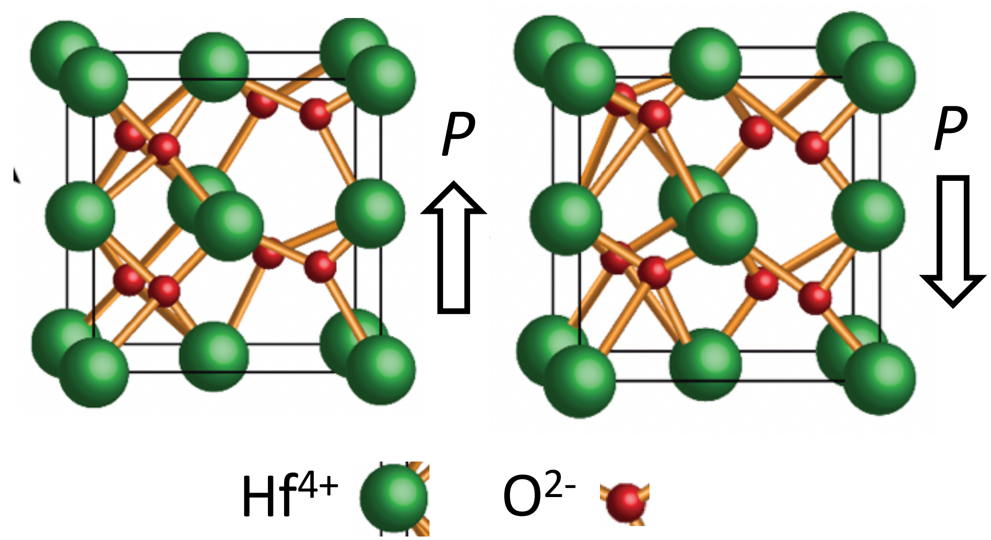
Ferroelectric Oxides

Perovskite Ferroelectric
$PbTiO_3$ ($Pb^{2+}Ti^{4+}O_3^{6-}$)

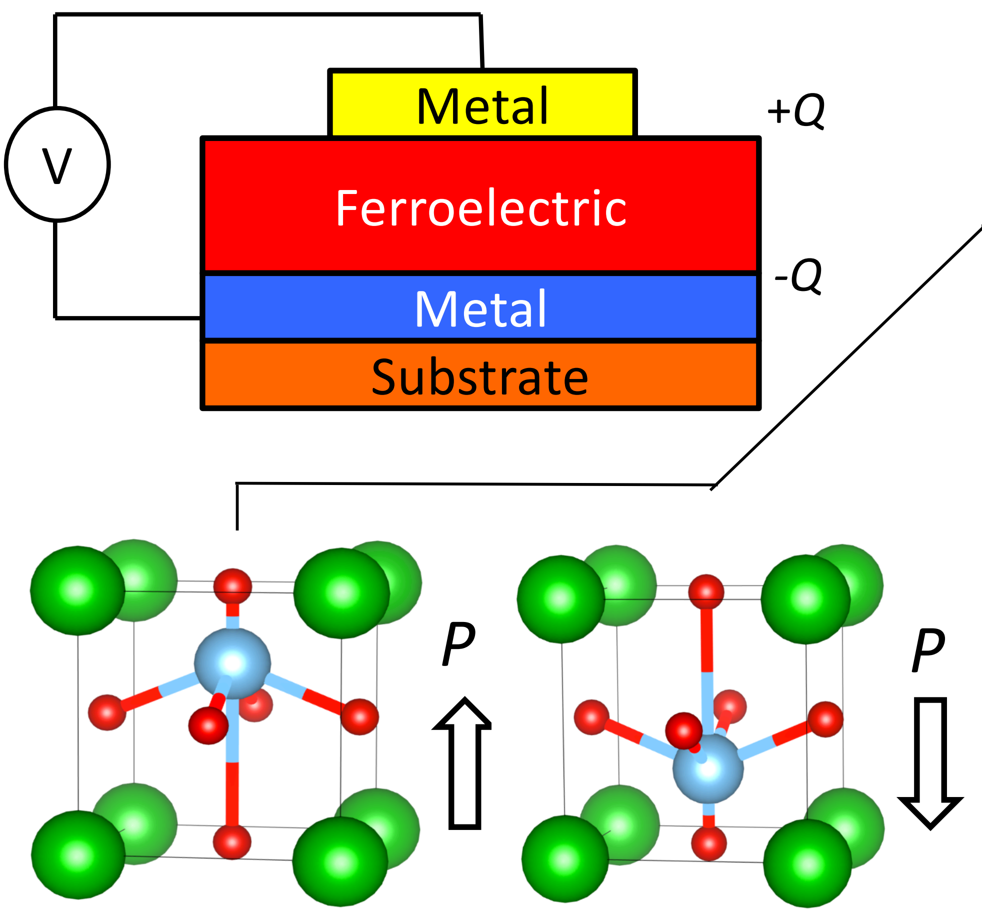


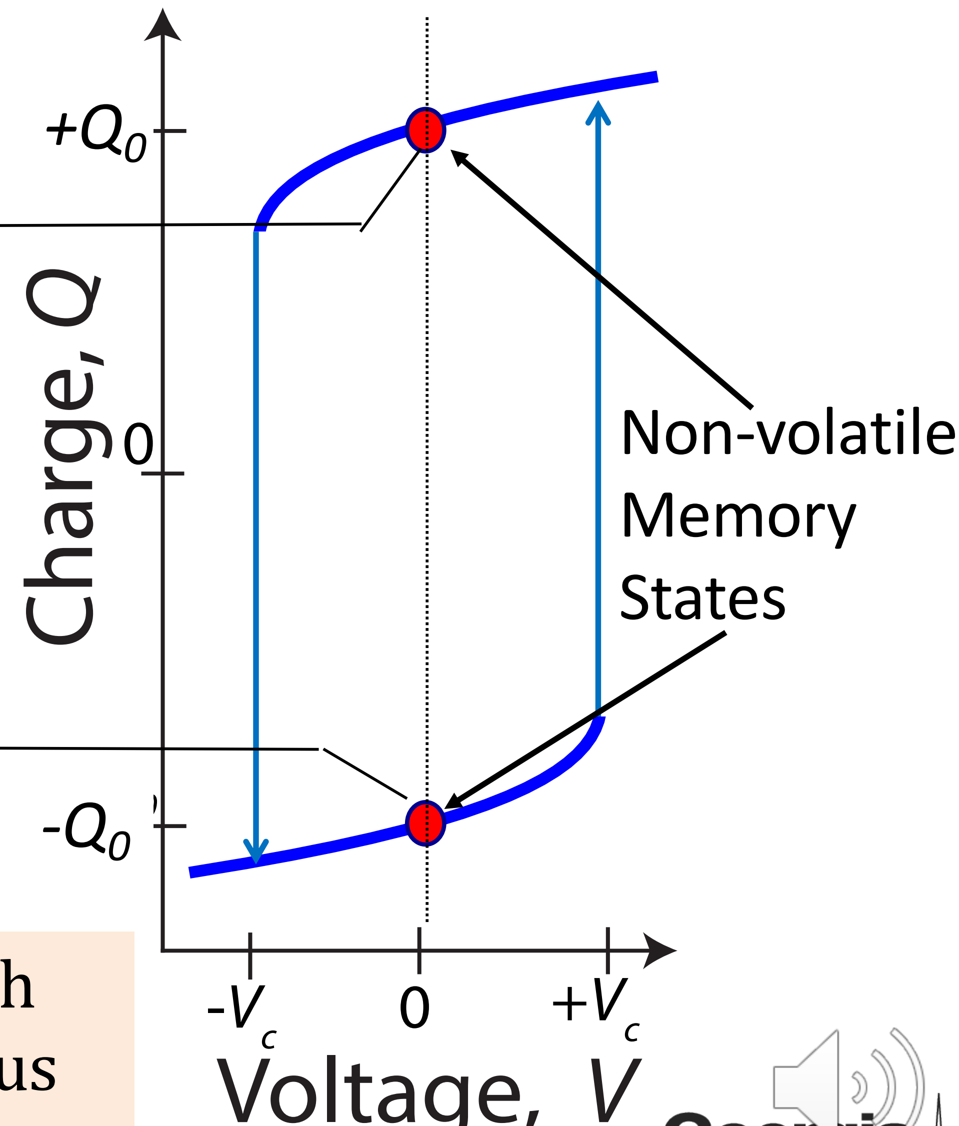
Fluorite-type Ferroelectric
HfO_2 ($Hf^{4+}O_2^{4-}$)



A ferroelectric is an insulator with a spontaneous polarization, which can be switched by an above coercive voltage .

Ferroelectric Oxides

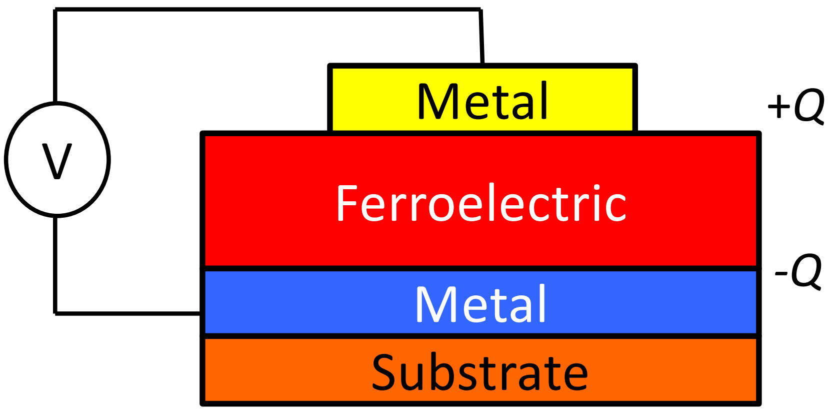




A ferroelectric is an insulator with electrically switchable spontaneous polarization.

Ferroelectric Oxides: Landau Theory

Ferroelectric Capacitor



$$V = \alpha_1 Q + \alpha_{11} Q^3 + \alpha_{111} Q^5 + \cdots$$

$\alpha_1 = a_0(T - T_c) < 0$ below T_c

T = temperature

T_c =Curie temperature

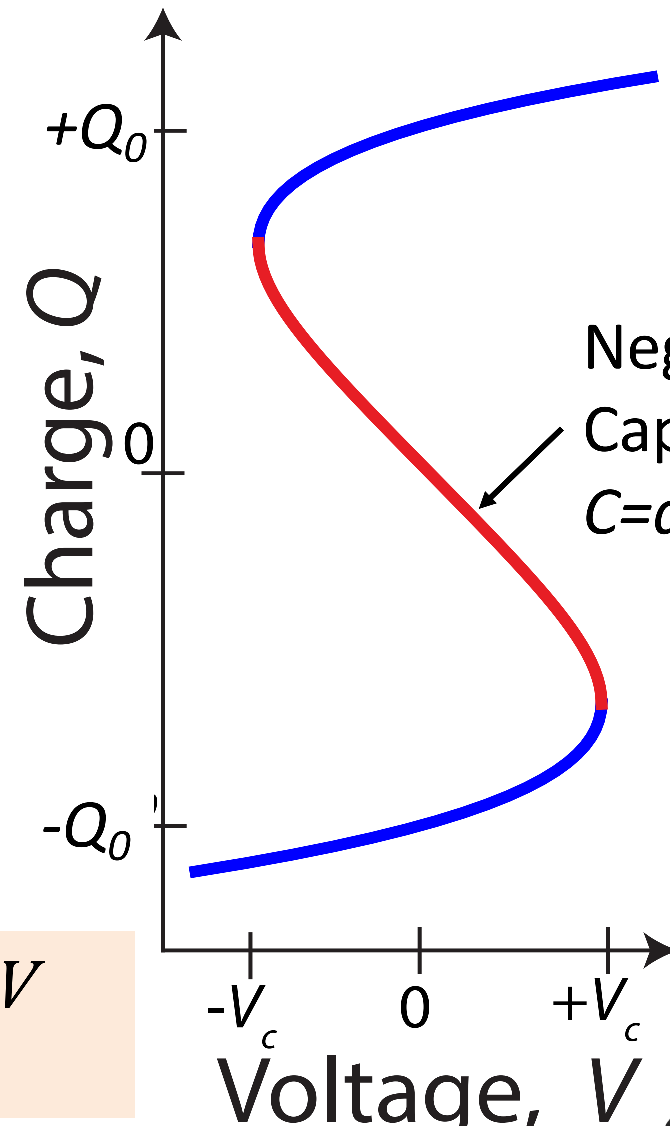
a_0 =positive constant

α_{11}=positive (2nd order) or negative (1st order)

α_{111}=positive

Landau theory: In certain Q and V range, $C=dQ/dV$ is begative

Charge-Voltage Characteristics



History of Landau theory of ferroelectrics

1937: Lev Landau first proposes the framework for phase transition

L. D. Landau, Phys. Z. Sowjun, vol. 11, pp. 545, 1937.; "On the theory of phase transitions. II," Zh. Eksp. Teor. Fiz. Vol. 7, pp. 627, 1937

Late 1940s: Ginzburg and Devonshire applies Landau theory to ferroelectrics after discovery of ferroelectricity in $BaTiO_3$

V. L. Ginzburg, Zh. Eksp. Teor. Fiz., vol. 15, pp. 739, 1945; V. L. Ginsburg, J. Phys. USSR, vol. 10, pp.107, 1946.

A. F. Devonshire: Philos. Mag., vol. 40, pp. 1040, 1949.

Landau framework is <u>phenomenological</u> mean-field theory based on symmetry to analyze phase transitions.

-Connects microscopic models to macroscopic behavior

<u>But does not talk about underlying microscopic origin!</u>

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History of Landau theory of ferroelectrics

'S'-shape in polarization-electric field curve appeared in early papers in 1950s on the Landau Theory of Ferroelectrics!

But negative capacitance region in ferroelectric was <u>not explicitly discussed</u> except for asserting that this region is "thermodynamically unstable."

Merz
Physical Review, 1953.

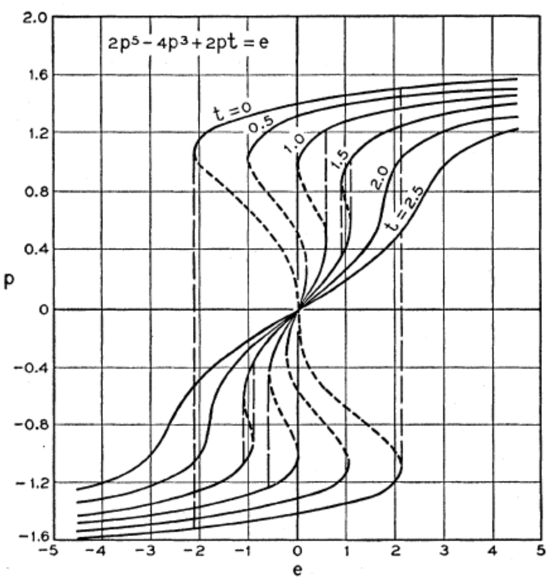
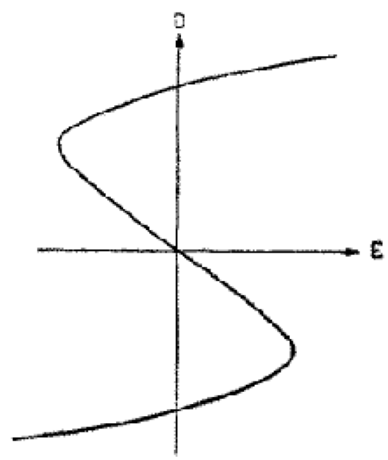


FIG. 3. Plot of function $2p^5-4p^3+2pt=e$.

R. Landauer
J. Appl. Phys, 1957.

FIG. 4. General form of relationship between D and E for barium titanate at room temperature. D and E are taken to be along the c axis and the crystal is assumed to be clamped to the dimensions that it has in the absence of a field. D and $4\pi P$ are almost equal and this curve has essentially the same shape as the P vs E relationship for a "clamped" crystal.



Janovec
Czech. fiz. zurnal, 1959.

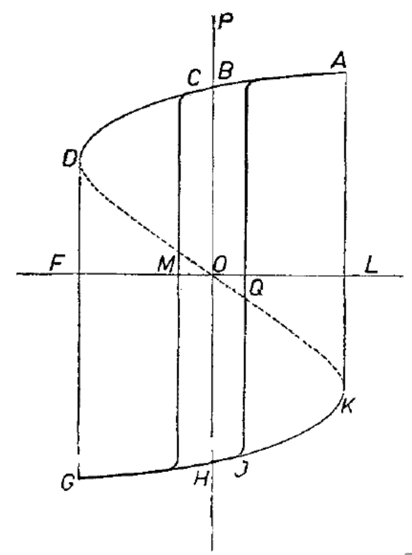
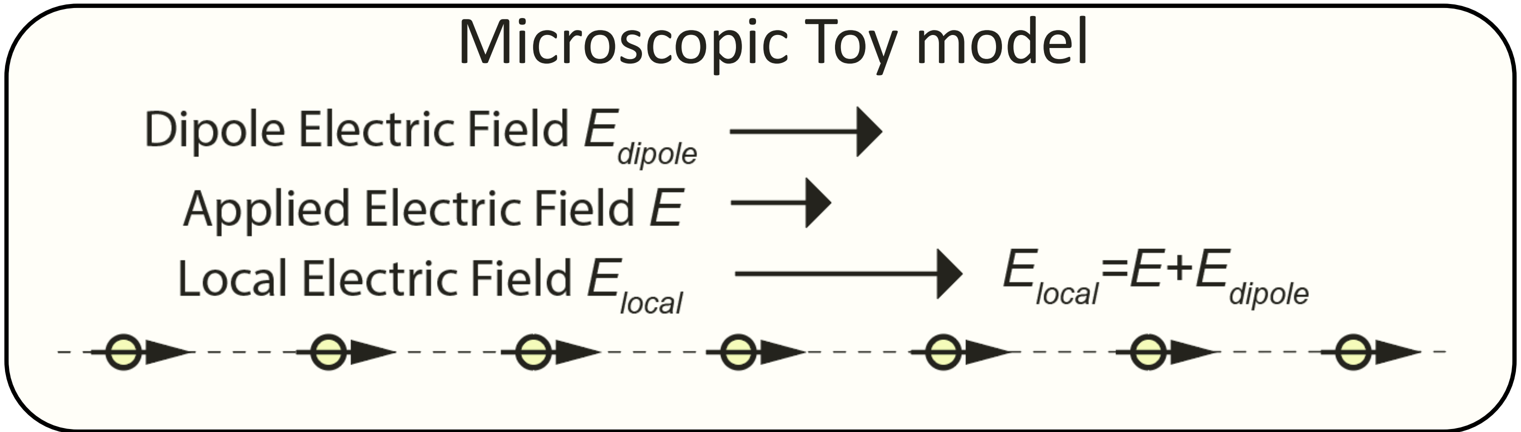
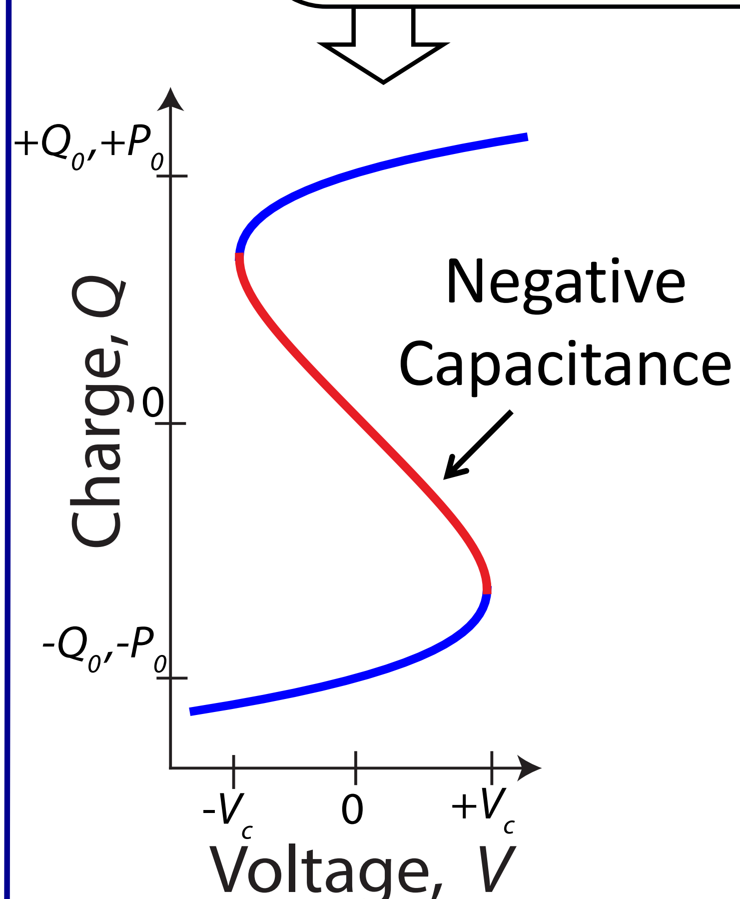


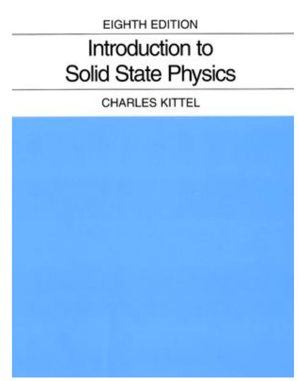
Fig. 1. Hysteresis dependence of polarisation P on intensity of electric field E.

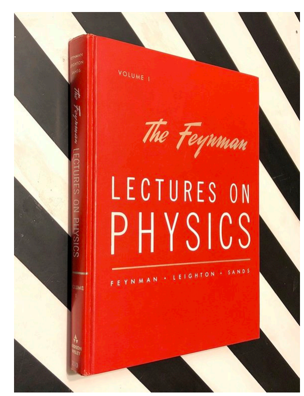
Toy model of Negative Capacitance (‘S’-curve)





-Simple, physical explanation of underlying microscopic mechanisms of ferroelectric negative
-Electrostatic dipole-dipole interactions
-Atomic scale positive feedback mechanism





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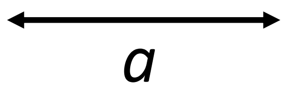
Toy model of Negative Capacitance (‘S’-curve)

Dipole Electric Field E_{dipole}

Applied Electric Field E

Local Electric Field E_{local}

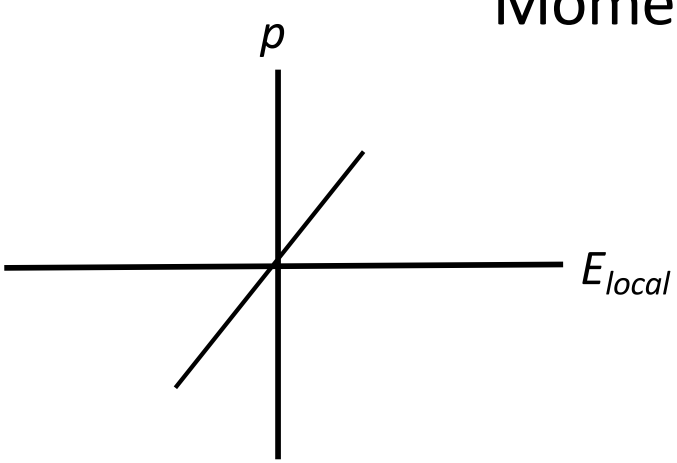
$E_{local}=E+E_{dipole}$



Polarizability

Dipole Moment $p = \alpha \epsilon_0 E_{local}$

Vacuum Permittivity



Toy model of Negative Capacitance ('S'-curve)

Dipole Electric Field E_{dipole}

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Local Electric Field E_{local}

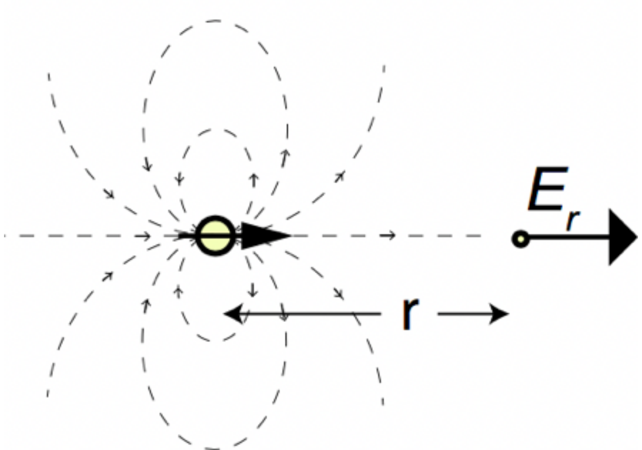
$E_{local} = E + E_{dipole}$

a

Polarizability

Dipole Moment $p = \alpha \epsilon_0 E_{local}$

Vacuum Permittivity



$$E_r = \frac{1}{4\pi\epsilon_0}\frac{2p}{r^3}$$

Toy model of Negative Capacitance ('S'-curve)

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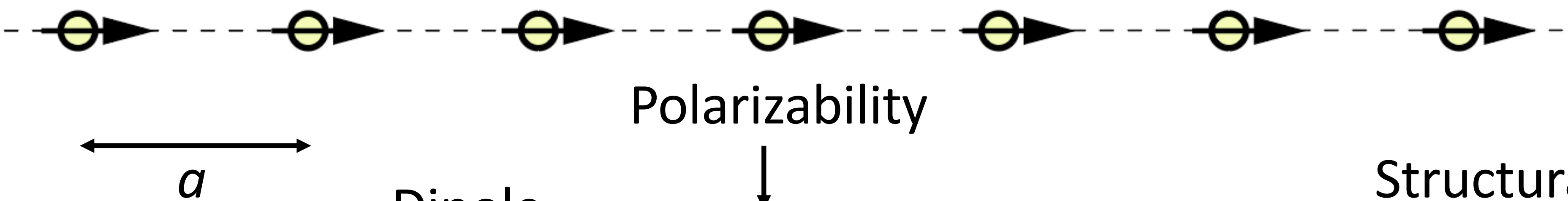
Applied Electric Field E

Local Electric Field E_{local}

$E_{local}=E+E_{dipole}$



Polarizability

Dipole Moment $p = \alpha\epsilon_0 E_{local}$

Structural Factor

$$E_{dipole} = 2\times\frac{1}{4\pi\epsilon_0}\frac{2p}{a^3}\left(1+\frac{1}{2^3}+\frac{1}{3^3}+\cdots\right)=\frac{p}{\epsilon_0}\frac{0.383}{a^3}=\zeta p$$

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Toy model of Negative Capacitance ('S'-curve)

Dipole Electric Field E_{dipole}

Applied Electric Field E

Local Electric Field E_{local}

$E_{local} = E + E_{dipole}$

a

Polarizability

Dipole Moment $p = \alpha \epsilon_0 E_{local}$

Structural Factor

$$E_{dipole} = 2 \times \frac{1}{4\pi\epsilon_0} \frac{2p}{a^3} \left(1 + \frac{1}{2^3} + \frac{1}{3^3} + \cdots \right) = \frac{p}{\epsilon_0} \frac{0.383}{a^3} = \zeta p$$

$$E_{local} = E_{dipole} + E$$

Atomic scale feedback

1. Dipole moment depends on local electric field.
2. Local electric field depends on dipole moment.

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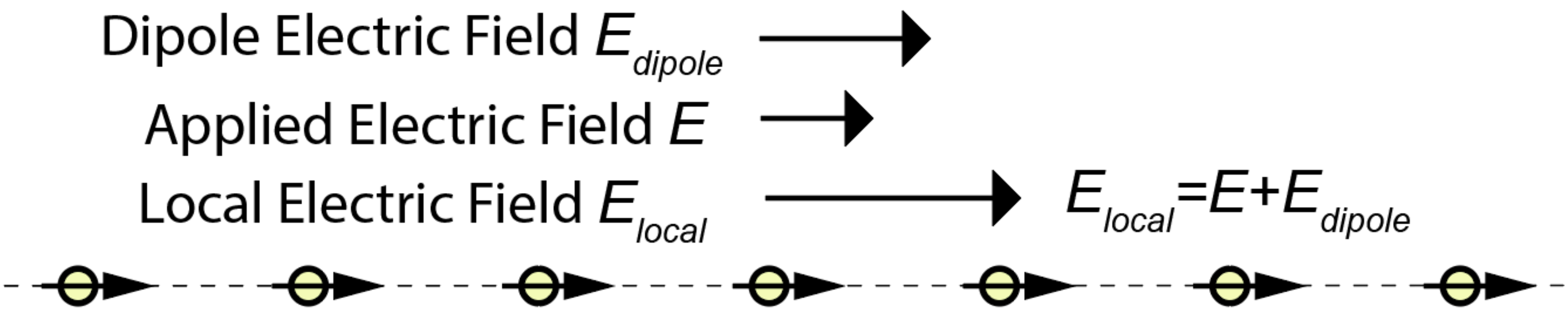
Toy model of Negative Capacitance ('S'-curve)



Atomic scale feedback

1. Dipole moment depends on local electric field.

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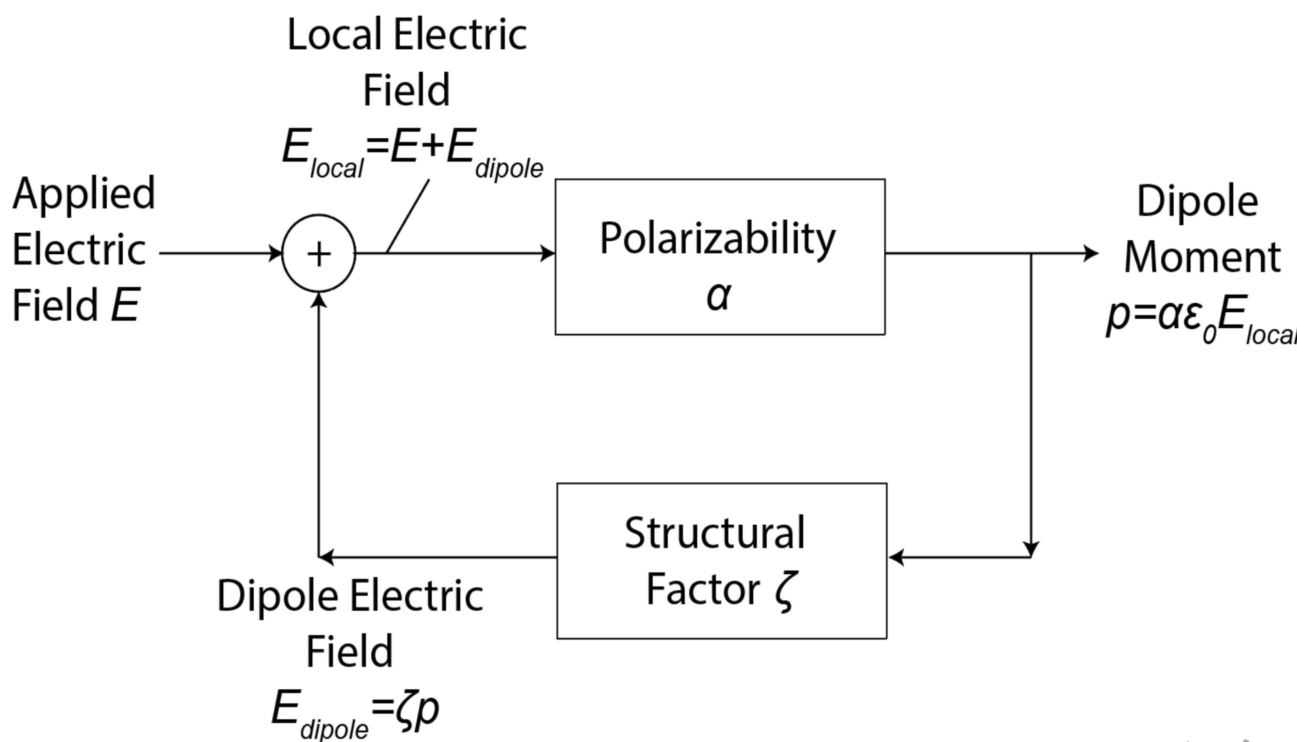


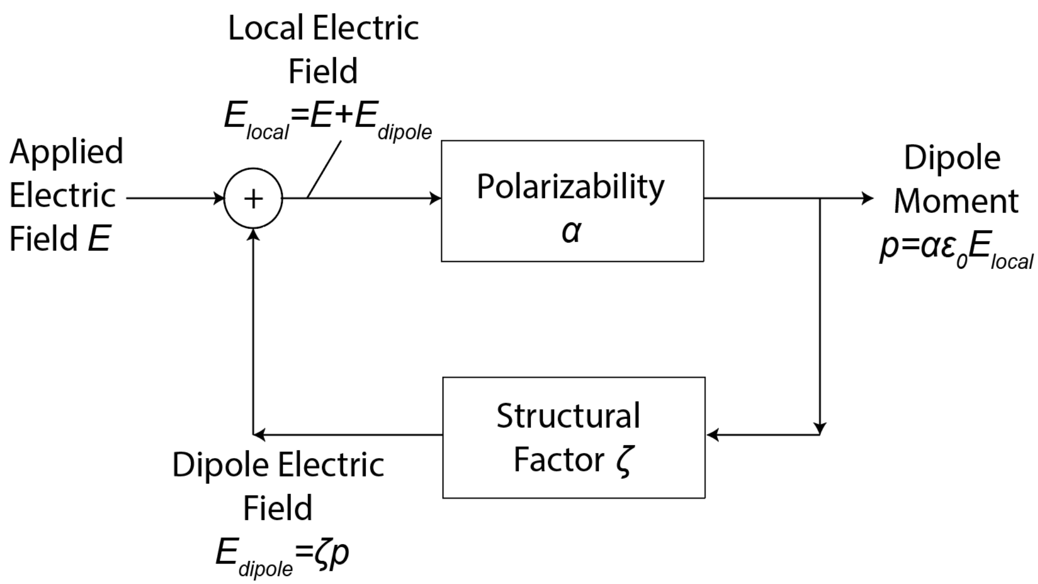
Toy model of Negative Capacitance ('S'-curve)

Dipole Electric Field E_{dipole}

Applied Electric Field E

Local Electric Field E_{local}

$E_{local}=E+E_{dipole}$



Dipole moment

$$p = \alpha\epsilon_0 E_{local}$$

Dipole field

$$E_{dipole} = \zeta p$$

Local field

$$E_{local} = E_{dipole} + E$$

$$p = \alpha\epsilon_0(\zeta p + E)$$

Dipole moment: $p = \dfrac{\alpha\epsilon_0}{1-\alpha\epsilon_0\zeta}\mathrm{E}$

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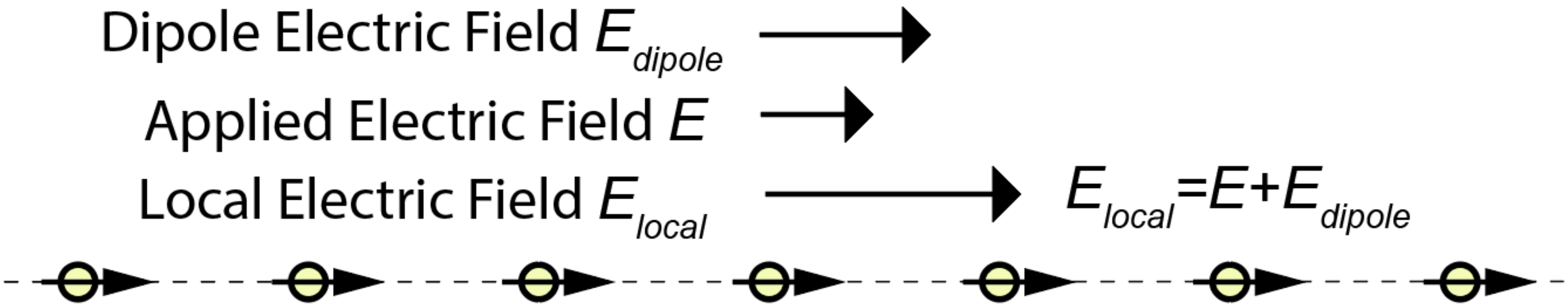
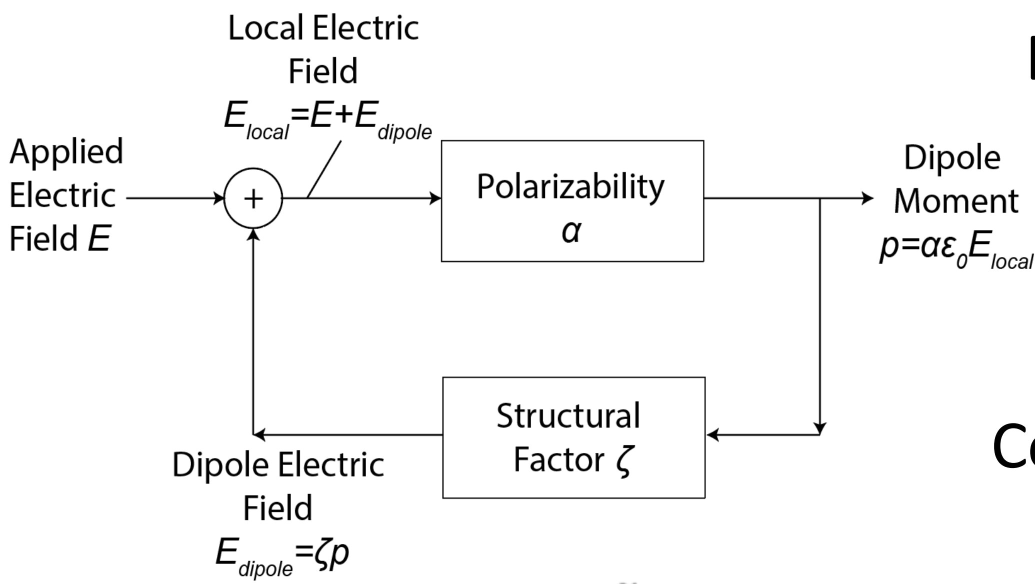
Toy model of Negative Capacitance ('S'-curve)

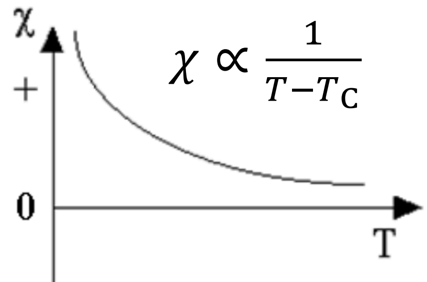




Dipole moment: $p = \frac{\alpha\epsilon_0}{1-\alpha\epsilon_0\zeta}\mathrm{E}$

$$\alpha = \frac{1}{\gamma\epsilon_0 T} \qquad \zeta = \gamma T_C$$

Constant   Temp.   Curie Temp.



$$\frac{dp}{dE} = \frac{\alpha\epsilon_0}{1-\alpha\epsilon_0\zeta} \propto \frac{1}{T-T_C}$$

Curie-Weiss Law

Toy model of Negative Capacitance ('S'-curve)

Dipole Electric Field E_{dipole}

Applied Electric Field E

Local Electric Field E_{local}

$E_{local}=E+E_{dipole}$

Dipole moment: $p = \frac{\alpha\epsilon_0}{1-\alpha\epsilon_0\zeta}\mathrm{E}$

$$\frac{dp}{dE} = \frac{\alpha\epsilon_0}{1-\alpha\epsilon_0\zeta}$$

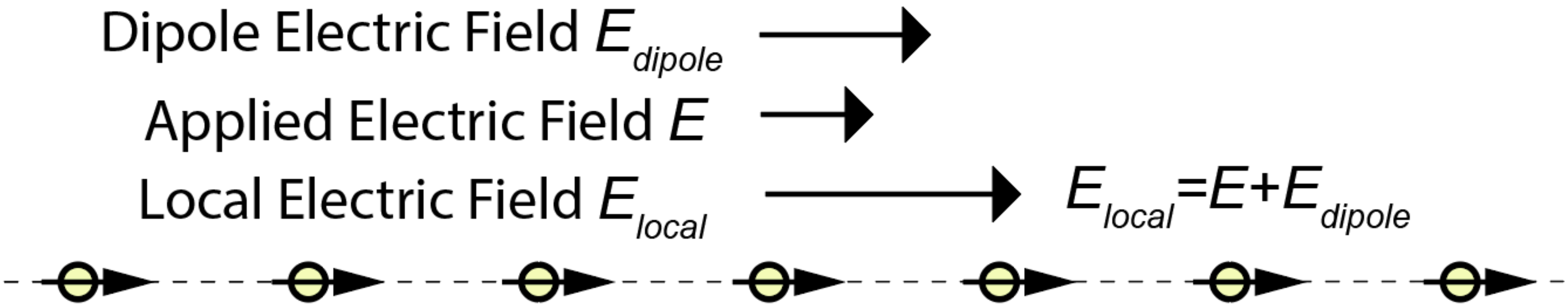
If $\alpha\epsilon_0\zeta > 1$ (if the material is highly polarizable),

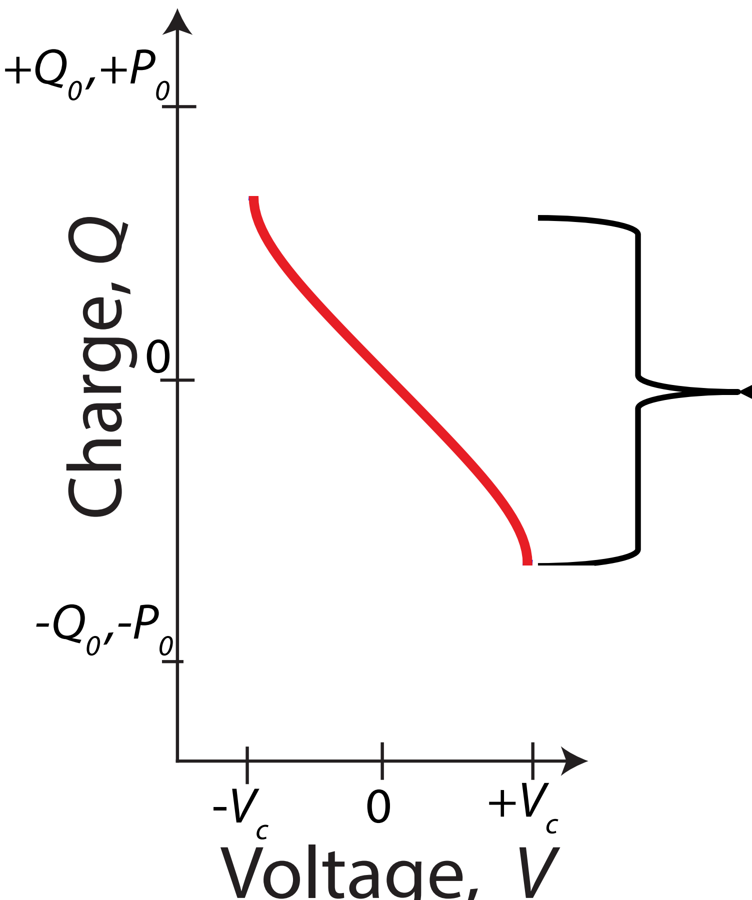
$$\frac{dp}{dE} < 0$$

<u>Polarization Catastrophe</u>

The negative dielectric negative capacitance that sets off the polarization catastrophe and leads to the emergence of the spontaneous polarization in ferroelectric material.

Toy model of Negative Capacitance ('S'-curve)





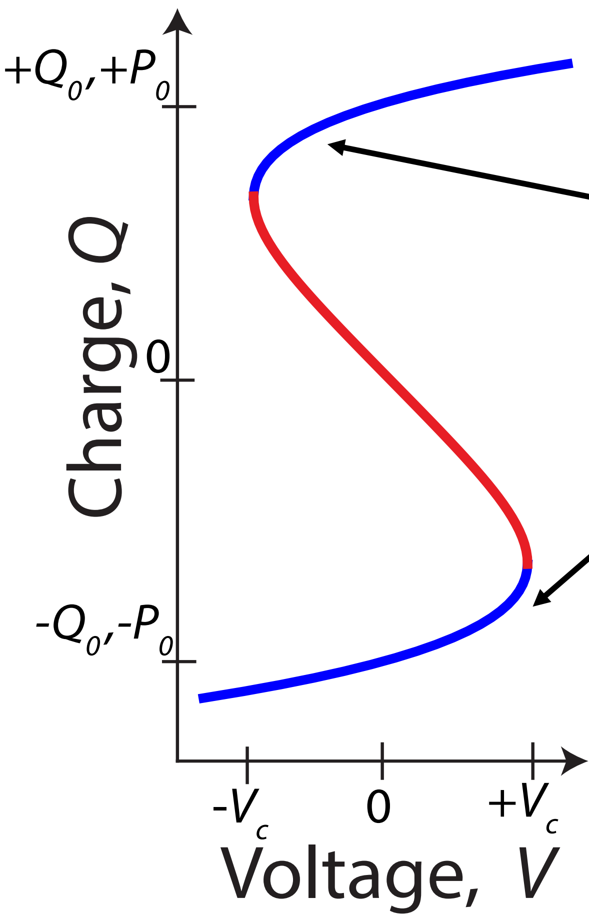
$T<T_c$: : Negative Capacitance and Initiation of the Polarization Catastrophe

Toy model of Negative Capacitance ('S'-curve)

Dipole Electric Field E_{dipole}

Applied Electric Field E

Local Electric Field E_{local} $\quad E_{local}=E+E_{dipole}$



How to get the turn-back feature?

Toy model of Negative Capacitance ('S'-curve)

Dipole Electric Field E_{dipole}

Applied Electric Field E

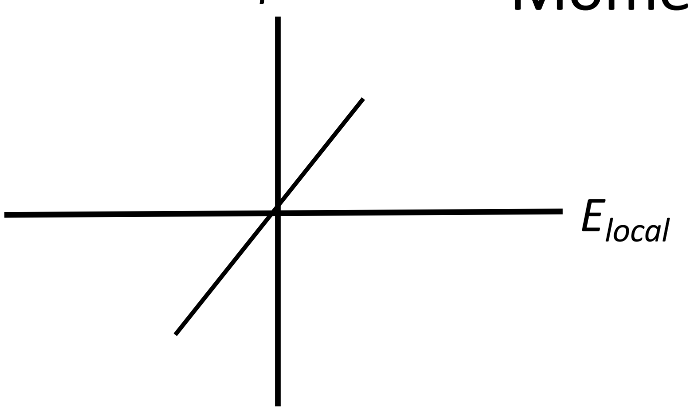
Local Electric Field E_{local}

$E_{local}=E+E_{dipole}$

Polarizability

Dipole Moment $$p = \alpha\epsilon_0 E_{local}$$

Vacuum Permittivity



Toy model of Negative Capacitance ('S'-curve)

Dipole Electric Field E_{dipole}

Applied Electric Field E

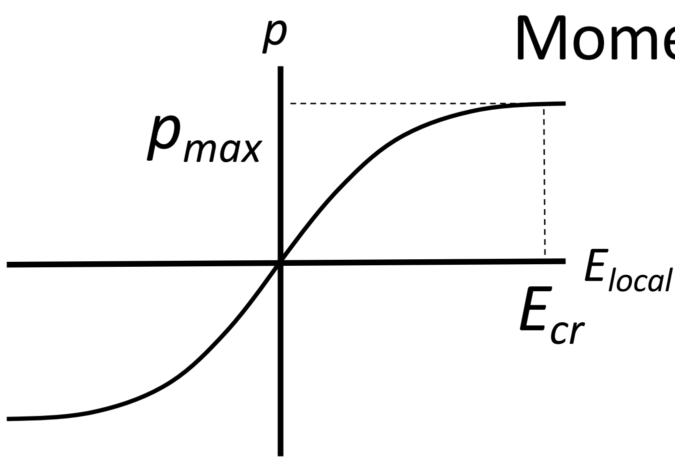
Local Electric Field E_{local}

$E_{local}=E+E_{dipole}$

Polarizability

Dipole Moment

$$p=\alpha\epsilon_0 E_{local}$$



Because dipoles are not infinitely polarizable.

Dipole moment: $p=p_{max}\tanh\frac{E_{local}}{E_{cr}}$

Maximum moment: $p_{max}=\alpha\epsilon_0 E_{cr}$

Critical field: E_{cr}

Toy model of Negative Capacitance ('S'-curve)

Dipole Electric Field E_{dipole}

Applied Electric Field E

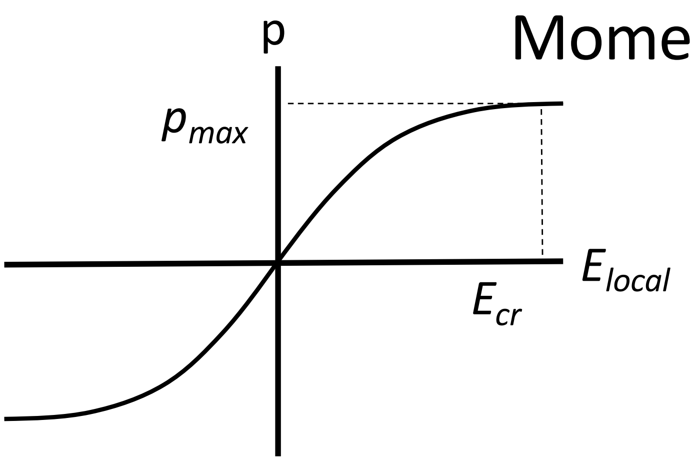
Local Electric Field E_{local}

$E_{local}=E+E_{dipole}$

Dipole moment: $p = p_{max} \tanh \frac{E_{local}}{E_{cr}}$

Maximum moment: $p_{max} = \alpha \epsilon_0 E_{cr}$

Dipole Moment



Polarizability

$$\alpha = \frac{1}{\underbrace{\gamma}_{\text{Constant}} \epsilon_0 \underbrace{T}_{\text{Temperature}}}$$

Structural Factor

$$\zeta = \gamma \underbrace{T_C}_{\text{Curie Temperature}}$$

Toy model of Negative Capacitance ('S'-curve)

Dipole Electric Field E_{dipole}

Applied Electric Field E

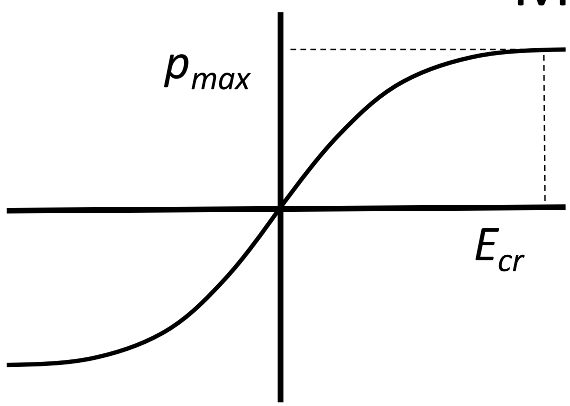
Local Electric Field E_{local}

$E_{local}=E+E_{dipole}$

Dipole moment: $p = p_{max} \tanh \frac{E_{local}}{E_{cr}}$

Maximum moment: $p_{max} = \alpha\epsilon_0 E_{cr}$

Dipole Moment



$$\alpha = \frac{1}{\gamma\epsilon_0 T} \qquad \zeta = \gamma T_C$$

Dipole field

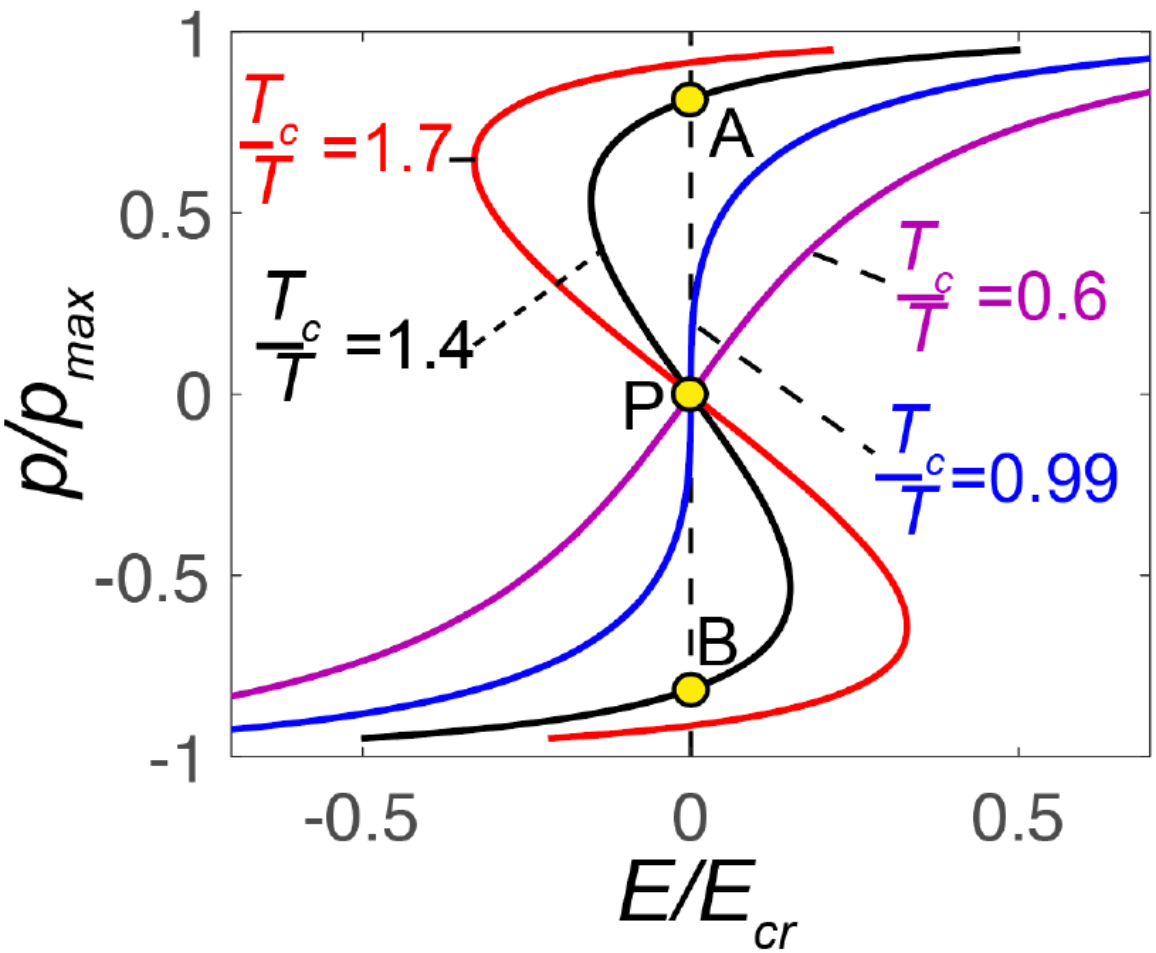
$$E_{dipole} = \zeta p$$

$$E_{local} = E_{dipole} + E$$

$$\frac{E}{E_{cr}} = -\frac{T}{T_c}\frac{p}{p_{max}} + \frac{1}{2}\log\frac{1+{}^{p}/_{p_{max}}}{1-{}^{p}/_{p_{max}}}$$

Toy model of Negative Capacitance ('S'-curve)

$$\frac{E}{E_{cr}} = -\frac{T}{T_c}\frac{p}{p_{max}} + \frac{1}{2}\log\frac{1 + {}^{p}/_{p_{max}}}{1 - {}^{p}/_{p_{max}}}$$



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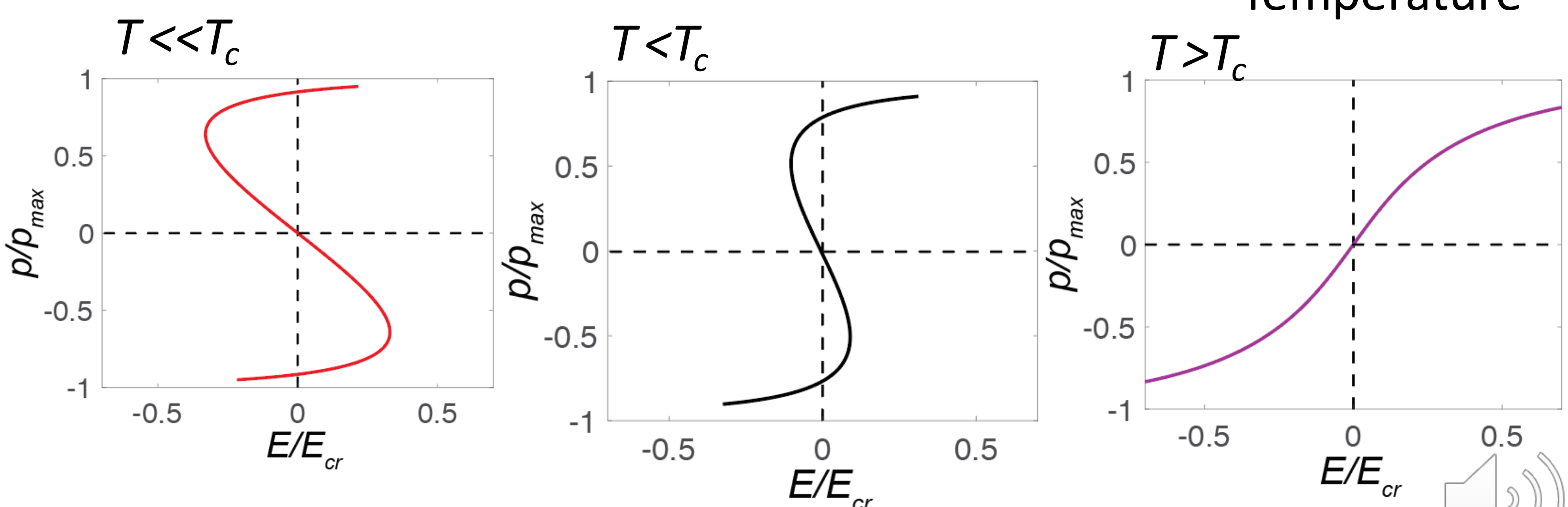
Toy model of Negative Capacitance ('S'-curve)

$$\frac{E}{E_{cr}} = -\frac{T}{T_c}\frac{p}{p_{max}} + \frac{1}{2}\log\frac{1 + {}^{p}/_{p_{max}}}{1 - {}^{p}/_{p_{max}}}$$

Maximum moment: $p_{max} = \alpha\epsilon_0 E_{cr}$

Critical field: E_{cr}

$$\alpha = \frac{1}{\gamma\epsilon_0 \underbrace{T}_{\text{Temperature}}} \qquad \zeta = \gamma \underbrace{T_C}_{\text{Curie Temperature}}$$



Toy model of Negative Capacitance (‘S’-curve)

$$\frac{E}{E_{cr}} = -\frac{T}{T_c}\frac{p}{p_{max}} + \frac{1}{2}\log\frac{1 + {}^{p}/_{p_{max}}}{1 - {}^{p}/_{p_{max}}}$$

?

How to get to the Landau expression?

$$E = \alpha_1 P + \alpha_{11} P^3 + \alpha_{111} P^5 + \cdots$$

Toy model of Negative Capacitance ('S'-curve)

$$\frac{E}{E_{cr}} = -\frac{T}{T_c}\frac{p}{p_{max}} + \frac{1}{2}\log\frac{1+{}^{p}/_{p_{max}}}{1-{}^{p}/_{p_{max}}}$$

$$\log\frac{1+x}{1-x} = x + \frac{x^3}{3} + \frac{x^5}{5} + \cdots$$

$$E = \left(\frac{E_{cr}}{p_{max}} - \zeta\right)p + \frac{E_{cr}}{3{p_{max}}^3}p^3 + \frac{E_{cr}}{5{p_{max}}^5}p^5 + \cdots$$

$$\alpha = \frac{1}{\gamma\epsilon_0 T}$$

$$\zeta = \gamma T_C$$

$$\alpha_1 = \frac{E_{cr}}{p_{max}} - \zeta = {}^{1}/_{\alpha\epsilon_0} - \zeta = \varsigma(T - T_C),$$

$$\alpha_{11} = {}^{E_{cr}}/_{3p_{max}^3},\ \alpha_{111} = {}^{E_{cr}}/_{5p_{max}^5}$$

$$E = \alpha_1 p + \alpha_{11}p^3 + \alpha_{111}p^5 + \cdots$$

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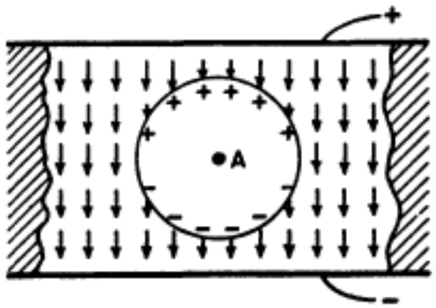
Similar treatment: Clausius-Mossotti Equation

The Clausius Mossoti Picture



$$E_{local} = E_{applied} + E_{Lorentz} + E_{cavity}$$

$$E_{Lorentz} = \frac{P_0}{3\varepsilon_0}, E_{cavity} = 0$$

$$E_{local} = E_{applied} + \frac{P}{3\varepsilon_0}$$

$$P = N\alpha E_{local}$$

E_{local}=Local electric field, $E_{applied}$=Applied electric field, $E_{Lorentz}$=Lorentz cavity Field, E_{cavity}=E-field due to dipoles in the cavity, α = Electronic polarizability, N = Dipole density

Clausius-Mossotti Equation

$$\frac{\epsilon_r - 1}{\epsilon_r + 2} = \frac{N\alpha}{3\epsilon_0}$$

$$\chi = \varepsilon_r - 1 = \frac{N\alpha/\varepsilon_0}{1 - N\alpha/\varepsilon_0} < 0$$

if $\quad N\alpha/3\varepsilon_0 > 1$

Clausius-Mossotti equation is often used to describe polar dielectrics. [Kittel: Introduction to Solid State Physics]

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Similar treatments of Ferroelectric

PHYSICAL REVIEW VOLUME 78, NUMBER 6 JUNE 15, 1950

The Lorentz Correction in Barium Titanate

J. C. SLATER
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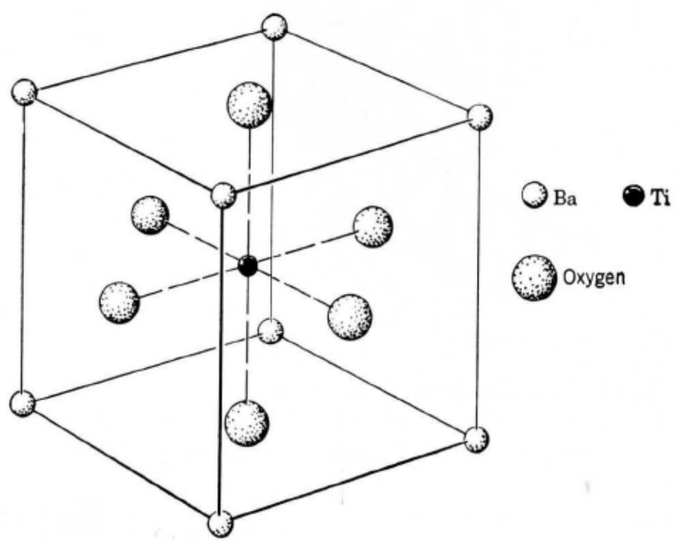

$$\begin{aligned}
E(\mathrm{Ba}) &= P(\mathrm{Ba})/N(\mathrm{Ba})\alpha(\mathrm{Ba}) \\
&= E_0 + q_{11}P(\mathrm{Ba}) + q_{12}P(\mathrm{Ti}) + q_{13}P(\mathrm{O}') + q_{14}P(\mathrm{O}''); \\
E(\mathrm{Ti}) &= P(\mathrm{Ti})/N(\mathrm{Ti})\alpha(\mathrm{Ti}) \\
&= E_0 + q_{21}P(\mathrm{Ba}) + q_{22}(\mathrm{Ti}) + q_{23}P(\mathrm{O}') + q_{24}P(\mathrm{O}''); \\
E(\mathrm{O}') &= P(\mathrm{O}')/N(\mathrm{O}')\alpha(\mathrm{O}') \\
&= E_0 + q_{31}P(\mathrm{Ba}) + q_{32}P(\mathrm{Ti}) + q_{33}P(\mathrm{O}') + q_{34}P(\mathrm{O}''); \\
E(\mathrm{O}'') &= P(\mathrm{O}'')/N(\mathrm{O}'')\alpha(\mathrm{O}'') \\
&= E_0 + q_{41}P(\mathrm{Ba}) + q_{42}P(\mathrm{Ti}) + q_{43}P(\mathrm{O}') + q_{44}P(\mathrm{O}'').
\end{aligned}$$

$$\begin{aligned}
\alpha(\mathrm{Ba}) &= 1.95 \times 10^{-24}\ \mathrm{cm}^3, \\
\alpha(\mathrm{O}') &= \alpha(\mathrm{O}'') = 2.4 \times 10^{-24}\ \mathrm{cm}^3, \\
\alpha(\mathrm{Ti}) &= 0.19 \times 10^{-24} + \alpha_i(\mathrm{Ti});
\end{aligned}$$

$$\epsilon \approx \frac{3T_c}{T - T_c}$$

$$\alpha_i(\mathrm{Ti}) = 0.95 \times 10^{-24}\ \mathrm{cm}^3,$$

$$(4\pi/3)N(\mathrm{Ti})\alpha_i(\mathrm{Ti}) = 0.062,$$

Similar treatment was done in 1950s.

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More details

Khan, Asif Islam. "On the microscopic origin of negative capacitance in ferroelectric materials: A toy model." *2018 IEEE International Electron Devices Meeting (IEDM)*. IEEE, 2018.

Hoffmann, Michael, Prasanna Venkatesan Ravindran, and Asif Islam Khan. "Why Do Ferroelectrics Exhibit Negative Capacitance?." Materials 12.22 (2019): 3743.

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Is a negative capacitance is a

<u>real, physical phenomenon</u>

or

an <u>unphysical, artificial construct</u> of the phenomenology of the Landau Theory?

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Negative capacitance is a real....

physical phenomenon!

Arises from simple dipole-dipole interactions and positive feedback mechanisms!

Without negative capacitance (i.e., static negative dielectric permittivity), ferroelectrics do not exist!

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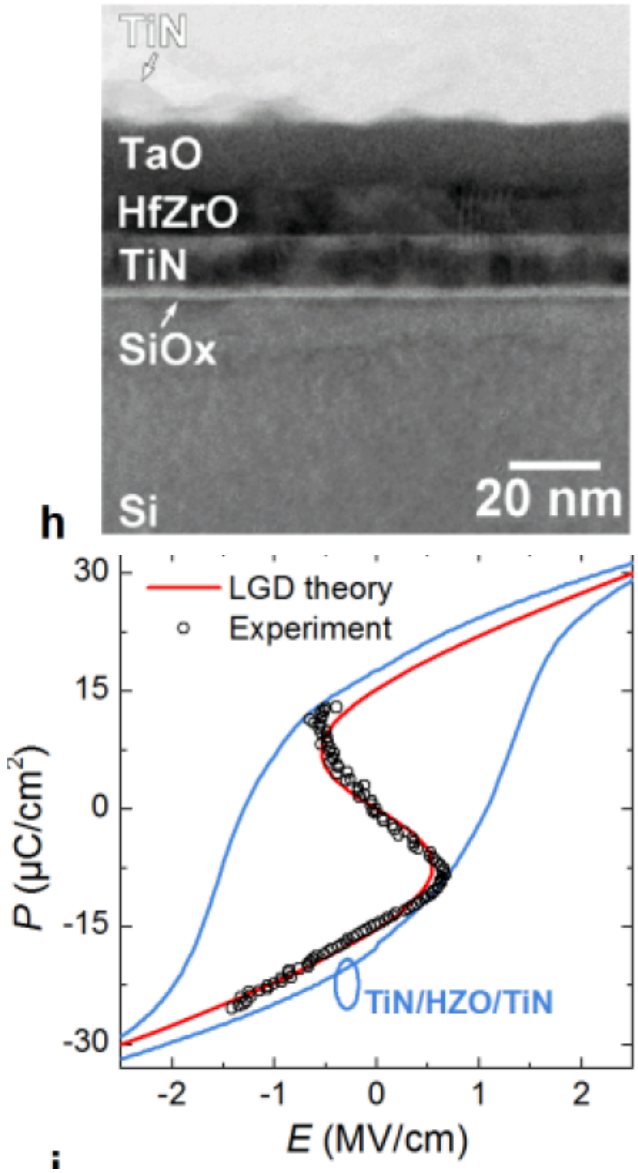
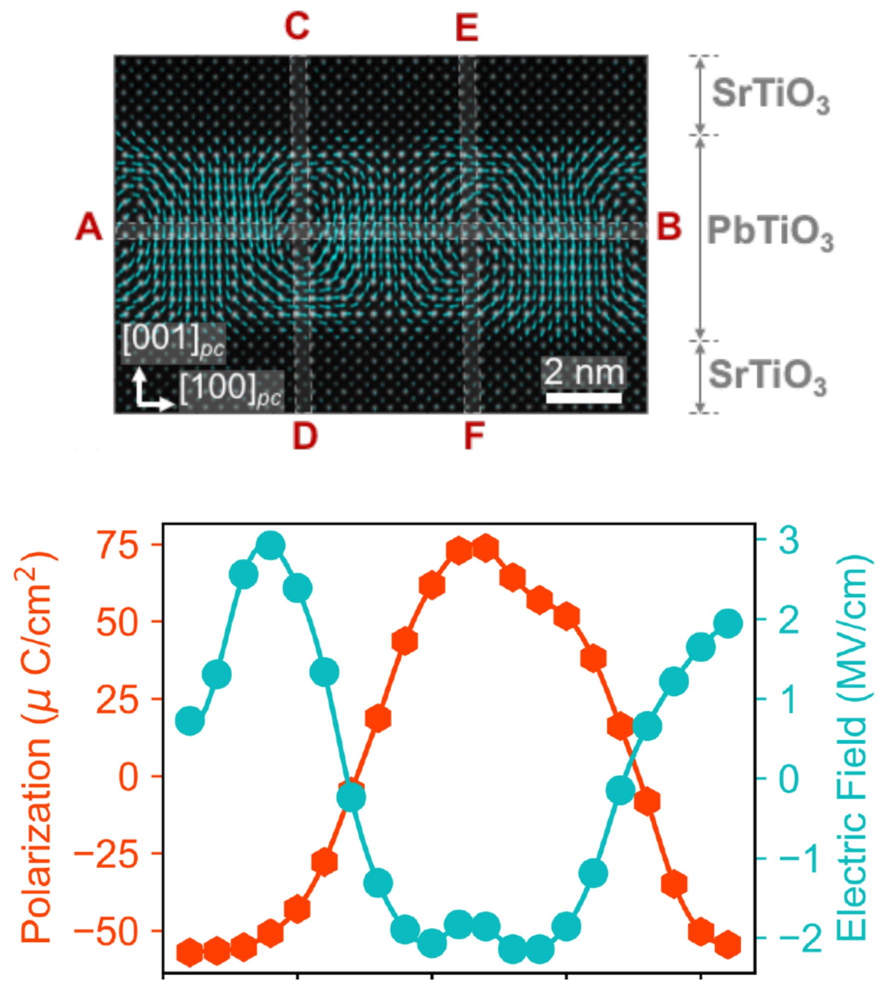
Experimentally Measured ‘S’-curve

Hoffmann et al. Nature 2018

Hoffmann et al. IEDM 2018 p. 31.6

Yadav et al. Nature 2019.





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Microscopic Origin of Negative Capacitance in Ferroelectrics

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Thanks & Questions

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