On the Microscopic Origin of Negative Capacitance in Ferroelectric Materials: A Toy Model

Asif Islam Khan
School of Electrical and Computer Engineering, Georgia Institute of Technology, GA 30329, USA
Email: asif.khan@ece.gatech.edu

Abstract**—**We present a simple, physical explanation of underlying microscopic mechanisms that lead to the emergence of the negative phenomena in ferroelectric materials. The material presented herein is inspired by the pedagogical treatment of ferroelectricity by Feynman and Kittel. In a toy model consisting of a linear one-dimensional chain of polarizable units (i.e., atoms or unit cells of a crystal structure), we show how simple electrostatic interactions can create a microscopic, positive feedback action that leads to negative capacitance phenomena. We point out that the unstable negative capacitance effect has its origin in the so called "polarization catastrophe" phenomenon which is essential to explain displacement type ferroelectrics. Furthermore, the fact that even in the negative capacitance state, the individual dipole always aligns along the direction of the local electrical field not opposite is made clear through the toy model. Finally, how the "*S*"-shaped polarization vs. applied electric field curve emerges out of the electrostatic interactions in an ordered set of polarizable units is shown.

I. INTRODUCTION

The negative capacitance effect in ferroelectric materials can be utilized to enable continued performance gains in the complementary metal-oxide-semiconductor (CMOS) platforms. When used in the gate dielectric stack of a metal-oxide-semiconductor field-effect transistor (MOSFET), a ferroelectric oxide owing to its negative capacitance properties can provide a passive voltage amplification of the gate voltage at the oxide-semiconductor interface. This effect can lower the sub-threshold slope below the fundamental Boltzmann limit of 60 mV/decade [1]. Such steep switching in negative capacitance field-effect transistors (NCFETs) can "restart" the aggressive scaling of the power supply voltage, thereby, allowing for significant reduction of power dissipation in the CMOS technology. To date, different aspects of ferroelectric negative capacitance phenomena have been demonstrated in different experimental set-ups such as ferroelectric capacitors, ferroelectric-dielectric heterostructures and superlattices and NCFETs.

The underlying theory of ferroelectric negative capacitance as proposed by Salahuddin *et al.* is based on the Landau's phenomenological theory of phase transitions. The free energy density of a ferroelectric material can be expanded in an even order polynomial of the polarization which through the Landau-Khalatnikov equation, leads to the following relation between the applied electric field E and the polarization P.

$$E = \alpha_1 P + \alpha_{11} P^3 + \alpha_{111} P^5 + \cdots \tag{1}$$

Here, α_1, α_{11}, α_{111} are anisotropy constants. Polarization is equivalent to the surface charge density. Furthermore, $\alpha_1 = a_0(T - T_c)$ where T is the temperature, T_c is the Curie temperature and a_0 is a positive constant. Below Curie temperature, $\alpha_1 < 0$, and the E vs. P curve has an "*S*"-shape where under certain range of E and P, the curve has a negative slope and hence a negative capacitance (*i.e.*, $C = {}^{1}/_{t_F}\left({}^{dP}/_{dE}\right) < 0, t_F$ being the ferroelectric thickness). Between the coercive field ($+E_C$ and $-E_C$), the polarization P is a multiple valued function of E. The negative capacitance states are unstable in a free-standing ferroelectric material; hence in the absence of an applied electric field, the ferroelectric gets spontaneously polarized in either of the stable states ($+P_0$, $-P_0$) indicated by points A and B in fig. 1(a). For a detailed treatment of stability of the negative capacitance state, the readers are referred to Ref. [2].

First appeared in the classic 1937 paper [3,4], the Landau framework takes a phenomenological, mean field and symmetry-based approach to analyze phase transitions. Right after the discovery of ferroelectricity in $BaTiO_3$ in early 1940s, the Landau theory was first applied to ferroelectric oxides led

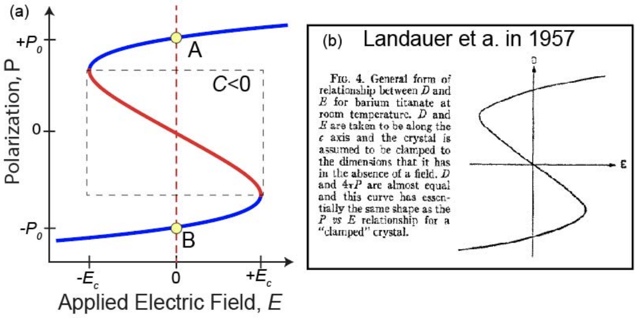

FIG. 4. General form of relationship between D and E for barium titanate at room temperature. D and E are taken to be along the c axis and the crystal is assumed to be clamped to the dimensions that it has in the absence of a field. D and $4\pi P$ are almost equal and this curve has essentially the same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

Fig. 1. (a) The "S"-shaped polarization P vs. applied electric field E curve of a ferroelectric material. (b) One of the earliest illustrations of the "S"-shaped P vs. E curve appeared in Landauer et al. in Ref. [7]. Similar illustration appeared in Ref. [8]. In these reports, the properties of the negative capacitance region were not explicitly discussed except for saying that this region is "thermodynamically unstable," and hence needs to be replaced by hysteretic jumps. Reproduced with permission from Ref. [7].

978-1-7281-1987-8/18/$31.00 ©2018 IEEE

the works of Ginzburg and Devonshire [5, 6]. However, in none of these early works and the ones that followed, the negative capacitance region in ferroelectric was explicitly discussed except for asserting that this region is “thermodynamically unstable.” Furthermore, while the Landau theory serves as a reliable, conceptual bridge between microscopic models and observed macroscopic phenomena, the theory itself leaves out the physical, microscopic details of the phenomenon it describes. These two facts in fact has created multitudes of the confusion whether the negative capacitance effect is a real, physical phenomenon or an unphysical, artificial construct for the convenience of the phenomenology (the Landau theory neither implicitly nor explicitly implies the latter). To date, all of the theoretical analysis of negative capacitance effects and modeling and simulation of NCFETs starts with Eq. 1 or its variants all based on the Landau framework. What is missing in the current discussion of negative capacitance is a physical picture that explains the microscopic origin of the negative capacitance in ferroelectric materials. As such, in this invited article, we take on this task to present a simple, atomic scale model that elucidates the underlying physical mechanisms responsible for this phenomenon. We point out that the unstable negative capacitance effect has its origin in the so called “polarization catastrophe” phenomenon which is generally used in the ferroelectric literature to explain the emergence of ferroelectricity of displacement type.

Another commonly held misconception is that, in the negative capacitance state even when it is stabilized by putting a positive capacitor in series, the ferroelectric dipoles align opposite to the electric field they feel thereby “violating” the fundamentals of thermodynamics. To resolve this issue, we make the point that it is important to distinguish between the applied electric field and the local electric field (the field that the dipoles feel). The local electric field is, in fact, the sum of the applied electric field and the effective dipole field (mean field) created by all the other dipoles. It is only when the polarizability of the ferroelectric dipoles attains a large enough value to create an atomic scale positive feedback mechanism such that the local field overcompensates the applied field and the unstable negative capacitance phenomena and the “polarization catastrophe” ensue. Even in such a case, the polarizability of the dipoles always remains positive and the dipoles always remain in the same direction of the local electric field—the dipole aligns opposite only to the applied electric field. Building upon the pedagogical treatment of ferroelectricity by Feynman, we show how an “*S*”-shaped dipole moment p vs. applied electric field E curve emerges from a toy model consisting of a linear one-dimensional chain of polarizable units.

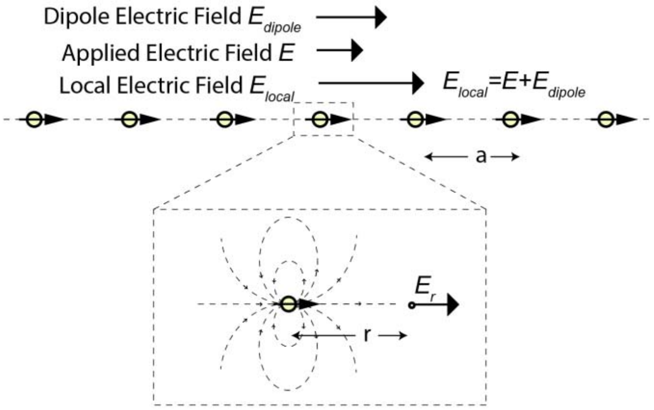

Fig.2. A linear, one dimensional chain of polarizable units with a spacing of a. The electric field created by a polarized dipole is also shown.

II. A MICROSCOPIC TOY MODEL OF FERROELECTRICS AND NEGATIVE CAPACITANCE

Let us consider a linear, one dimensional chain of polarizable units with a spacing of a. on which an electric field E is applied along the chain axis (Fig. 2). The treatment presented herein is influenced by the pedagogical writing of Feynman in Ref. [9]. A more rigorous treatment along the same lines based on Clausius-Mossotti relation available in Ref. [10]. The polarizable unit here can represent an atom or a bond or a unit cell of a crystal. For the sake of simplicity, we first assume that these units have a linear polarizability α such that the dipole moment p and the local electric field E_{local} is relation by the following relation.

$$p = \alpha\epsilon_0 E_{local} \tag{2}$$

Here, ϵ_0 is the vacuum permittivity. The field created by a dipole at a distance r from the along its axis is given by $E_r = \frac{1}{4\pi\epsilon_0}\frac{2p}{r^3}$. Hence, at a given dipole, the electric field due to the interaction with all the other dipoles in the chain is

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

This particular calculation of E_{dipole} is repeated from Ref. [9] Here, ζ is a structural factor. In the calculations that follow, we will not pay attention to the value of ζ, rather treat it as a variable that depends on the arrangement of the units. ζ will have a different value if the dipoles have a different arrangement (e.g. three dimensional cubic, tetragonal, or orthorhombic lattice). Note that the local electric field is the sum of the applied electric field E and the electric field created by all the other dipoles E_{dipole}, *i.e.*,

$$E_{local} = E_{dipole} + E \tag{4}$$

It is interesting to note that, the dipole moment p depends on the local electric field E_{local} (Eq. 2), which in turn depends on p through Eq. 4 thereby creating a microscopic positive feedback. A block diagram representation is shown in Fig. 3. Combining Eq. (2), (3) and (4), we obtain the following expression.

$$p = \alpha\epsilon_0(\zeta p + E) \tag{5}$$

Rearranging this equation, we obtain,

$$p = \frac{\alpha\epsilon_0}{1-\alpha\epsilon_0\zeta} E \tag{6}$$

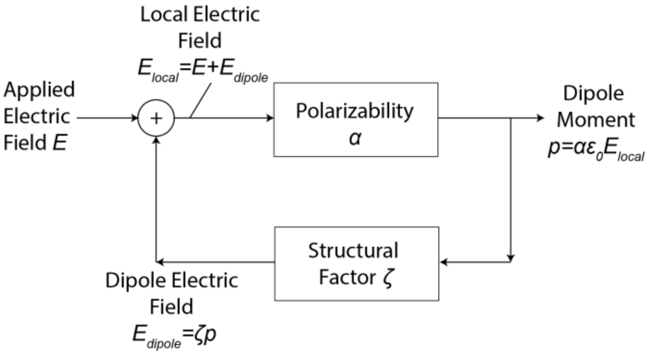

Fig.3. The positive feedback mechanism at the atomic scale.

and scaled dielectric constant, ϵ_r is given by the following relation.

$$\epsilon_r = \frac{1}{\epsilon_0}\frac{dp}{dE} = \frac{\alpha}{1-\alpha\epsilon_0\zeta} \quad (7)$$

The polarizability α is generally inversely proportionate to the temperature T (i.e., $\alpha \propto {}^{1}/_{T}$). At Curie temperature T_c, $\alpha = 1/\epsilon_0\zeta$ for which the scaled dielectric constant ϵ_r shows a singularity. For $T>T_c$, $\alpha < 1/\epsilon_0\zeta$, and p=0 is stable solution of Eq. 5 for E=0. Physically, it means that in the absence of an applied electric field, the dipoles are not polarized and the dipole moment and the local electric field are both zero. It can be shown that Eq. 7 leads to the Curie-Weiss dependent of ϵ_r on temperature (i.e., $\epsilon_r \propto {}^{1}/_{T-T_c}$).

Of particular interest to our analysis is the case when $\alpha > 1/\epsilon_0\zeta$ and the scaled dielectric constant ϵ_r becomes negative. As the ferroelectric material is cooled down from a temperature higher than T_C to below that, the resulting negative value of ϵ_r amplifies small thermal fluctuations in dipole moments through the positive feedback mechanism in the absence of an applied electric field E. As soon as a dipole electric field E_{dipole} however small emerges due to the fluctuations of p, it regeneratively increases p. In the ferroelectric literature, this situation is typically referred to as the "polarization catastrophe". This means that in the absence of an applied electric field, any thermal fluctuation of dipole moment sets up a local electric field that spontaneously polarizes the dipoles.

The point to note here is that it is the negative dielectric constant or the negative capacitance at $T<T_C$ that sets off the polarization catastrophe and leads to the emergence of the spontaneous polarization in ferroelectric material. By introducing time dependent, kinetic terms in Eq. (2) and (5), it can be shown that p=0 is no longer a stable solution of the system when $T<T_C$ *and* $\alpha > 1/\epsilon_0\zeta$.

III. THE EMERGENCE OF THE SPONTANEOUS POLARIZATION AND THE "*S*"-SHAPED *P-E* CURVE

Now that we have seen at $T<T_C$, the negative capacitance sets off the positive feedback mechanism that tends to increase the dipole moment p in an unbounded fashion, we now address what stops this run-way process such that p settles down to stable spontaneous polarization states indicated by points A and B in fig. 1 (in fig. 1, the polarization P is a scaled version of the dipole moment p). To explain the emergence of the stable spontaneously polarized state, we need to add the next level of details by considering the non-linearity in the polarizability in the dipole moment. An electric field stretches a dipole—however, a dipole is not infinitely stretchable. With the increase of the local electric field beyond a critical value, the dipole moment is not expected to increase any further. We assume that the saturation dipole moment is $p_{max} = \alpha\epsilon_0 E_{cr}$, E_{cr} being the critical saturation local electric field. For the sake of simplicity, we assume the following relation between p and E_{local}:

$$p = p_{max}\tanh\frac{E_{local}}{E_{cr}} \quad (8)$$

where $p_{max} = \alpha\epsilon_0 E_{cr}$. When $E_{local} \ll E_{cr}$, we get back the linear relation between p and E_{local} as in Eq. (2). Combining Eq. 2, 3, 4 and 8, we obtain

$$p = p_{max}\tanh\frac{\zeta p+E}{E_{cr}} \quad (9)$$

Using the identity: $\tanh^{-1}x = \frac{1}{2}\log\frac{1+x}{1-x}$ in Eq. (9), the following relation is obtained.

$$E = -\zeta p + \frac{E_{cr}}{2}\log\frac{1+{}^{p}/_{p_{max}}}{1-{}^{p}/_{p_{max}}} \quad (10)$$

Assuming $\alpha = {}^{1}/_{\varsigma\epsilon_0 T}$, ς being a positive constant, and $\zeta = \varsigma T_C$, Eq. 10 can be simplified as follows.

$$\frac{E}{E_{cr}} = -\frac{T}{T_c}\frac{p}{p_{max}} + \frac{E_{cr}}{2}\log\frac{1+{}^{p}/_{p_{max}}}{1-{}^{p}/_{p_{max}}} \quad (11)$$

Fig. 4 plots p/p_{max} vs. E/E_{cr} curves using Eq. 11 for T_c/T=0.6, 1.01, 1.4 and 1.7. The "*S*"-shaped p-E curve clearly emerges for $T<T_c$. In fig. 4, the stable p=0 at $T>T_c$ is indicated point P. As the temperature reduces below T_c, the point P no longer remains stable due to the instability of negative capacitance. At this point, as soon as thermal fluctuations cause the dipole moment p to attain a small value, the dipole moment p traverses a path through the "S"-shaped curve in a transient path and settles at one of the stable spontaneously polarized states (for example, at points A or B for T/T_c=1.4 in fig. 4).

In the regime where $|E| < E_{cr}$ and $|p| < p_{max}$, we expand Eq. 10 to obtain the following relation.

$$E = -\zeta p + E_{cr}\left(\frac{p}{p_{max}} + \frac{1}{3}\left(\frac{p}{p_{max}}\right)^3 + \frac{1}{5}\left(\frac{p}{p_{max}}\right)^5 + \cdots\right)$$
$$= \left(\frac{E_{cr}}{p_{max}} - \zeta\right)p + \frac{E_{cr}}{3{p_{max}}^3}p^3 + \frac{E_{cr}}{5{p_{max}}^5}p^5 + \cdots \quad (12)$$

Thus, from our toy model, we arrive at an odd order expansion of the applied electric field E in terms of p as in Eq. 1. Comparing Eq. 12 with Eq. 1, $\alpha_1 = \frac{E_{cr}}{p_{max}} - \zeta = {}^{1}/_{\alpha\epsilon_0} - \zeta = \varsigma(T - T_C)$, $\alpha_{11} = {}^{E_{cr}}/_{3p_{max}^3}$, $\alpha_{111} = {}^{E_{cr}}/_{5p_{max}^5}$ and so on.

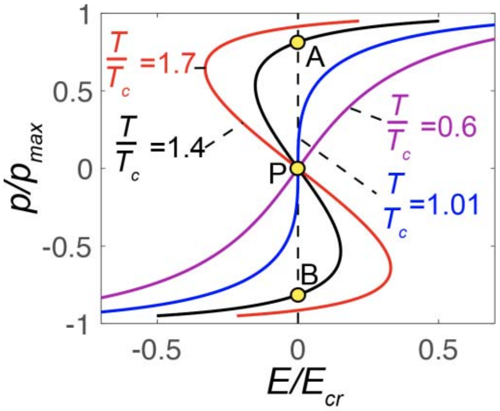

Fig.4. p/p_{max} vs. E/E_{cr} curves for T_c/T=0.6, 1.01, 1.4 and 1.7 plotted using Eq. 11: $\frac{E}{E_{cr}} = -\frac{T}{T_c}\frac{p}{p_{max}} + \frac{E_{cr}}{2}\log\frac{1+{}^{p}/_{p_{max}}}{1-{}^{p}/_{p_{max}}}$. Note that p_{max} is a function of T as well.

An important point to note in this analysis is that the polarizability of the polarizable units is always positive—i.e., the dipole always aligns along the direction of the local electric field. This is because p and E_{local} always have the same sign in Eq. 2 and 8. It is in fact the case, at T<Tc, the polarizability attains a large positive value which leads to the negative capacitance phenomena and the emergence of ferroelectricity.

Given the limited scope of the article, we intentionally excluded a discussion of the stabilization mechanism of the otherwise unstable negative capacitance states which is described in details elsewhere. In a stabilized negative capacitance state in a ferroelectric-dielectric series combination and an NCFET, the polarization and the external electric field in the ferroelectric are in opposite direction—however, the local electric field and the polarization are still in the same direction.

IV. CONCLUSIONS

We have presented a physical, microscopic picture of the emergence of negative capacitance in ferroelectric materials. We have shown that a large dielectric polarizability set off an atomic scale, positive feedback mechanism that aligns the dipoles opposite to the applied electric field but not to the local electric field. This situation is unstable and hence transient. The instability there caused leads to the "polarization catastrophe" and the dipoles become spontaneously polarized even in the absence of an applied electric field. By assuming a non-linear polarizability of the dipoles, we show how an "*S*"-shaped dipole moment p vs. applied electric field E curve emerges from a toy model consisting of a linear one-dimensional chain of polarizable units (i.e., atoms or dipoles or crystal unit cells).

It is important to note that polarization reversal generally occurs through domain nucleation and growth mechanisms especially in ferroelectric capacitors with lateral dimensions of tens of microns. In fact, that is why an "S"-shaped polarization vs. applied electric curve also had a hysteresis in recent experimental work reported in Ref. [11, 12, 13]. The effects of domain mediated switching in such observation of negative capacitance was analyzed in Ref. [14]. In fact, the topic of intrinsic, homogeneous and single domain switching has appeared in the ferroelectric literature since the 1950 in the context of what is known as the "Landauer paradox." [7, 15, 16]. It is interesting that in nanoscale ferroelectric HfO$_2$ gated field-effect transistors where the dimensions are of a few tens of nanometers, the evidence of single domain switching has been claimed to have been observed in Ref. [17]. Further analysis of such nanoscale devices will elucidate more about the nature of negative capacitance and its stability of associate ferroelectrics.

ACKNOWLEDGMENT

This work was supported in part by the National Science Foundation under grant 1718671, in part by Semiconductor Research Corporation-Global Research Collaboration grant 2018-LM-2829 and in part by ASCENT, one of six centers in JUMP, a Semiconductor Research Corporation (SRC) program sponsored by DARPA.

REFERENCES

[1] S. Salahuddin *et al.*, "Use of Negative Capacitance to Provide Voltage Amplification for Low Power Nanoscale Devices," Nano Lett., vol. 8, pp. 405-410, 2008.

[2] Khan et al. "Negative capacitance behavior in a leaky ferroelectric," IEEE Trans. Electron Dev., vol. 63, pp. 4416-4422, 2016.

[3] 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.

[4] D. ter Haar (Ed.): Collected Papers of L. D. Landau (Pergamon, Oxford 1965) contains Engl. transl. of Ref. [3].

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

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

[7] R. Landauer. "Electrostatic Considerations in BaTi0$_3$ Domain Formation during Polarization Reversal," J. Appl. Phys, vol. 28, pp. 227-234, Feb. 1957.

[8] W. J. Merz, "Double Hysteresis Loop of BaTiO$_3$ at the Curie Point," Phys. Rev., vol. 91, pp. 513-517, Aug. 1953.

[9] R. P. Feynman, The Feynman Lectures of Physics, vol. II. New York: Basic Books, 2011, p. 11-10.

[10] C. Kittel, Introduction to Solid State Physics, New York: John Willey & Sons, 2005.

[11] A. I. Khan et al., "Negative capacitance in a ferroelectric capacitor," Nature Mater. vol. 14, pp. 182-186, Jan. 2015.

[12] M. Kobayashi et al., "Experimental Study on Polarization-Limited Operation Speed of Negative Capacitance FET with Ferroelectric HfO2," Proc. IEDM, 2016.

[13] M. Hoffmann et al., "Direct Observation of Negative Capacitance in Polycrystalline Ferroelectric HfO2", Adv. Func. Mater, Oct. 2016. https://doi.org/10.1002/adfm.201602869

[14] M. Hoffmann et al., "Ferroelectric negative capacitance domain dynamics," J. of Appl. Phys, vol. 123, pp. 184101-10-184101-10, May 2018.

[15] V. Janovec, "On the theory of the coercive field of single-domain crystals of BaTiO3". Cechoslovackij fiziceskij zurnal, vol. 8, pp. 3-15, 1958.

[15] J.F. Scott, "Switching of ferroelectrics without domains". Adv. Mater., vol. 22, pp. 5315-5317, 2010.

[16] Mulaosmanovic et al., "Switching Kinetics in Nanoscale Hafnium Oxide Based Ferroelectric Field-Effect Transistors", ACS Appl. Mater. Interfaces, vol. 9, 3792−3798, 2017.