Supplemental Information

S1 Derivation and discussion of Eq.S9

In this section we manipulate the ferroelectric dynamic Eqs.2 in order to derive simplified relations between macroscopic quantities (e.g. average polarization and fields) and microscopic quantities (e.g. μ_{2P} and μ_{3P}), that may help interpret the charge versus ferroelectric field curves obtained by numerical simulations and observed in experiments.

To this purpose we now sum and normalize to n_D the Eqs.2 for all the n_D domains. We start by discussing the domain wall energy term

$$\sum_i \left[\sum_n (P_i - P_n) \right] = 0 \tag{S1}$$

which stems from the fact that, for any domain i, the P_i appears as $+4P_i$ in the equation for domain i, and then as $-P_i$ in the equations corresponding to its four adjacent domains denoted by n in Eq.S1 and in Fig.1. This is straightforward to see for the domains that do not belong to the borders of the device cross-section. In our simulations this is also true for the border domains because we employ periodic boundary conditions for the domain wall coupling, such that the domains at the left border are coupled along the x direction to the domains at the right border and, likewise, the domains at the lower border are coupled along y to those at the top border (see Fig.1). Eq.S1 is a very good approximation also for different boundary conditions when the device area is fairly large, such that the border domains are a small fraction of the overall n_D domain.

We now recall the assumption $\gamma \simeq 0$ (used throughout this paper for simulations of HZO based systems, see Tab.1) and come to the terms $2\alpha_i P_i$ and $4\beta_i P_i^3$ in Eqs.2. Here we simplify the derivations by neglecting the domain to domain variations of α_i, β_i, namely by taking for all domains the average values α, β. The sum and normalization to n_D of the term P_i readily results in P_{AV}, whereas the term with P_i^3 can be evaluated by recalling the definition $\Delta P_i = (P_i - P_{AV})$, which allows us to write

$$\frac{1}{n_D}\sum_{i=1}^{n_D} P_i^3 = \frac{1}{n_D}\sum_{i=1}^{n_D} \left(P_{AV}^3 + 3P_{AV}\Delta P_i^2 + 3P_{AV}^2 \Delta P_i + \Delta P_i^3 \right) = P_{AV}^3 + 3P_{AV}\mu_{2P}^2 + \mu_{3P}^3 \tag{S2}$$

where we have used $\sum_{i=1}^{n_D} \Delta P_i = 0$ and introduced the second, μ_{2P}, and third order P_i momentum, μ_{3P}, defined in Eq.3.

In order to evaluate the term involving the capacitances $C_{i,j}$ we need to recall their definition. In this respect we argue that in the MFIM structure illustrated in Fig.1 the calculation of the ferroelectic and dielectric field is a three-dimensional problem. By invoking the superimposition of effects of the V_T and P_i, which are the sources of the electric field in the linear electrostatic problem, we can write the potential $V_D(\bar{r})$ (with $\bar{r} = (x,y)$) at the ferroelectric-dielectric interface as [20]

$$V_D(\bar{r}) = \sum_{i=1}^{n_D} P_i G_{D,i}(\bar{r}) + \frac{C_F}{C_0} V_T \tag{S3}$$

where $G_{D,i}(\bar{r})$ is the Green's function for $V_D(\bar{r})$ of a unitary charge in domain i. By using Eq.S3 we can express the average V_D in any domain i as

$$V_{D,i} = \frac{1}{d^2}\int_{D_i} V_D(\bar{r})d\bar{r} = \sum_{j=1}^{n_D}\frac{1}{C_{i,j}}P_j + \frac{C_F}{C_0}V_T \tag{S4}$$

which stems from the definition of the capacitances $C_{i,j}$ as

$$\frac{1}{C_{i,j}} = \frac{1}{d^2}\int_{D_i} G_{D,j}(\bar{r})d\bar{r}\,. \tag{S5}$$

In consideration of the Neumann boundary conditions used for the electric field at the edges of the MFIM structure, the capacitances $C_{i,j}$ fulfill the sum rules [20]

$$\sum_{j=1}^{n_D}\frac{1}{C_{i,j}} \simeq \sum_{i=1}^{n_D}\frac{1}{C_{i,j}} = \frac{1}{C_0}\,. \tag{S6}$$

By using Eqs.S4, S6 we can also express the average V_D in the overall device area as

$$\begin{aligned} V_{D,AV} &= \frac{1}{n_D}\sum_{i=1}^{n_D} V_{D,i} = \frac{1}{n_D}\sum_{i,j=1}^{n_D}\frac{1}{C_{i,j}}P_j + \frac{C_F}{C_0}V_T \\ &= \frac{1}{n_D}\sum_{j=1}^{n_D} P_j\left(\sum_{i=1}^{n_D}\frac{1}{C_{i,j}}\right) + \frac{C_F}{C_0}V_T = \frac{P_{AV}}{C_O} + \frac{C_F}{C_0}V_T \end{aligned} \tag{S7}$$

We are now ready to evaluate the term involving the capacitances $C_{i,j}$ in the normalized sum of Eqs.2, and we obtain

$$\frac{1}{n_D}\sum_i\left[\frac{1}{2}\sum_{j=1}^{n_D}\left(\frac{1}{C_{i,j}}+\frac{1}{C_{j,i}}\right)P_j\right] = \frac{1}{2n_D}\sum_j P_j\left[\sum_i^{n_D}\left(\frac{1}{C_{i,j}}+\frac{1}{C_{j,i}}\right)\right] \simeq \frac{P_{AV}}{C_O} \simeq V_{D,AV} - \frac{C_F}{C_0}V_T \tag{S8}$$

where we have used again Eq.S6, as well as Eq.S7.

If we sum and normalize to n_D the Eqs.2 for all n_D domains and make use of Eqs.S1, S2 and S8, we readily obtain

$$\rho\frac{dP_{AV}}{dt} = -\left[2(\alpha+6\beta\mu_{2P}^2)P_{AV} + 4\beta(P_{AV}^3+\mu_{3P}^3)\right] + E_{F,AV} \tag{S9}$$

where $E_{F,AV}=(V_T-V_{D,AV})/t_F$ is the average value of the z component of the ferroelectric field. If we now consider a slow transient resulting in a very small value of $\rho(dP_{AV}/dt)$, Eq.S9 simplifies into the quasi static relation

$$E_{F,AV} \simeq 2(\alpha+6\beta\mu_{2P}^2)P_{AV} + 4\beta(P_{AV}^3+\mu_{3P}^3) \tag{S10}$$

Here it should be emphasized that Eq.S9 is not at all an alternative model to Eqs.2, in fact μ_{2P}, μ_{3P} can only be obtained by solving Eqs.2. Eq.S9 and Eq.S10 are instead approximate

relations between macroscopic quantities, such as P_{AV} and $E_{F,AV}$, and microscopic quantities, such as μ_{2P} and μ_{3P}, that may help interpret the charge versus ferroelectric field characteristics.

In particular Eq.S10 can be used to interpret the slope $(dQ/dE_{F,AV})$ in the negative capacitance branches observed in the charge versus E_F curves. To this purpose we focus on the small P_{AV} range where numerical evaluations show that the third order terms in Eq.S10 are much smaller than the other terms, and then derive both sides of Eq.S10 with respect to $E_{F,AV}$ to obtain

$$1 \simeq 2(\alpha + 6\beta\mu_{2P}^2)\,\frac{dP_{AV}}{dE_{F,AV}} \tag{S11}$$

where we used the simplification $P_{AV}(d\mu_{2P}^2/dE_{F,AV}) \ll \mu_{2P}^2(dP_{AV}/dE_{F,AV})$, which is supported by numerical evaluations for the regions of the NC branches in Figs.2, 4 where μ_{2P} is close to its maximum value and P_{AV} is small. If we now write the charge as $Q = P_{AV} + \varepsilon_F\varepsilon_0 E_{F,AV}$ and make use of Eq.S11 for $(dP_{AV}/dE_{F,AV})$, we finally obtain Eq.5 in the main manuscript.