Supporting Information for

Synchronous polarization switching at sub-coercive field with noise in ferroelectric thin-film capacitors: unveiling stochastic resonance

Vivek Dey[1||], *Thejas Basavarajappa*[1||], *Jonnalagadda Nikhila*[2], *…., Arvind Ajoy*[2,*], *Pavan Nukala*[1,*]

[1]*Centre* for Nanoscience and Engineering, Indian Institute of Science, 560012, India

[2]Electrical Engineering, Indian Institute of Technology Palakkad, 678623, India

[||] These authors contributed equally to this work

*Corresponding authors: pnukala@iisc.ac.in, arvindajoy@iitpkd.ac.in

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Supplementary Note 1: Mathematical description of stochastic polarization switching dynamics in a double-well potential in the presence of noise using Time Dependent Ginzberg-Landau (TDGL) formulation

The Landau free energy density of a ferroelectric system is given by,

$$F = \alpha P^2 + \beta P^4 - P.E \quad (1)$$

Since, we want to model the polarization switching in presence of noise so the corresponding TDGL equation [1] in presence of the Langevin force *ξ(t),* that reflects the thermodynamic fluctuations due to thermal noise or external driving noise is,

$$\rho \frac{\partial P}{\partial t} = -\frac{\partial F}{\partial P} + \xi(t) \quad (2)$$

where, ρ and ξ are the internal resistivity (a dissipative term) and white Gaussian noise respectively.

ξ(t) has the following properties:

1. $< \xi(t) >= 0$
2. The autocorrelation is $< \xi(t)\xi(t+\delta t) >= 0 \; if \; \delta t > \tau$, where τ is the maximum time over which the gaussian noise has any correlations.
3. The rate at which correlation decays is independent of time such that $< \xi(t)\xi(t+\delta t) >$ only depends on δt.

The behaviour of the system depends more on the cumulative effect of the random forcing ξ(t) i.e., integral over some time period that is long compared to τ rather than the instantaneous value. We can break up that integral into many pieces, each covering a span of length τ:

$$\int_0^t \xi(t')dt' = \int_0^\tau \xi(t')dt' + \int_\tau^{2\tau} \xi(t')dt' + \int_{2\tau}^{3\tau} \xi(t')dt' + \cdots \quad (3)$$

This integral is a sum of independent terms, each drawn from the same distribution. So according to the Central limit theorem, the integral obeys a normal distribution with mean 0 (because ⟨ξ(t)⟩=0), and whose standard deviation scales with √t.

The integral (3) has the properties of a Weiner process and hence the stochastic process of polarization switching in presence of a Langevin force $\xi(t)$ can be related to an underlying Brownian or Weiner process $W(t)$ as:

$$\xi(t) = B\frac{dW(t)}{dt}\text{, where } B \text{ is a constant} \tag{4}$$

$W(t)$ has the following properties:

1. $W(t=0) = 0$ i.e., it has probability of 1.
2. $W(t)$ has independent increments i.e., for all $r < s \leq t < u$, $W(u)$- $W(t)$ is independent of $W(s)$ - $W(r)$.
3. Increments of $W(t)$ are normally distributed, i.e., $W(t)$ - $W(s) \sim N(0, t\text{-}s)$.
4. $W(t)$ is a continuous function of t.

Now according to fluctuation-dissipation theorem we can write,

$$< \xi(t)\xi(t') > = \frac{2k_B\mathrm{T}}{V}\rho\delta(\mathrm{t}-\mathrm{t}') = \frac{2k_B\mathrm{T}}{t_F A_F}\rho\delta(\mathrm{t}-\mathrm{t}') \tag{5}$$

where t_F is thickness and A_F is area of the PZT ferroelectric capacitor.

Also,

$$< \xi(t)\xi(t') > = B^2 < \frac{\partial W(t)}{\partial t}\frac{\partial W(t')}{\partial t'} > = \frac{2k_B\mathrm{T}}{V}\rho\delta(\mathrm{t}-\mathrm{t}')$$

$$\Rightarrow B = \sqrt{\frac{2k_B T\rho}{t_F A_F}} \tag{6}$$

So, expression (4) becomes,

$$\xi(t) = \sqrt{\frac{2k_B T\rho}{t_F A_F}}\frac{dW(t)}{dt} \tag{7}$$

In our problem, the stochastic variable is polarization P. We can represent the system using a stochastic framework by describing how the macroscopic variable P evolves, considering its random fluctuations. To capture these fluctuations, we derive the Fokker-Planck equation [2], which describes how the probability distribution $w(P,t)$ of the polarization fluctuations evolves over time. This equation governs the changes in the distribution of P, incorporating both deterministic dynamics (such as damping or relaxation) and stochastic noise (such as thermal fluctuations).

So, from Fick's law of diffusion the diffusion current is,

$$J_{diff} = -D\frac{\partial w(P,t)}{\partial P} \tag{8}$$

where, D is the diffusion coefficient.

The drift current is,

$$J_{drift} = vw(P,t) \tag{9}$$

where, $v = \frac{\partial P}{\partial t} = -\frac{1}{\rho}\frac{\partial F}{\partial P}$ is the velocity of a representative point *P* along the polarization axis in absence of thermal fluctuation.

From continuity equation we can write as,

$$\frac{\partial w(P,t)}{\partial t} = -\frac{\partial}{\partial t}(J_{diff} + J_{drift})$$

$$= -\frac{\partial}{\partial P}\left[vw(P,t) - D\frac{\partial w(P,t)}{\partial P}\right]$$

$$= \frac{1}{\rho}\frac{\partial}{\partial P}\left[\frac{\partial F}{\partial P}w(P,t) + \rho D\frac{\partial w(P,t)}{\partial P}\right]$$

$$= \frac{1}{\rho}\frac{\partial}{\partial P}\left[\frac{\partial F}{\partial P}w(P,t) + D'\frac{\partial w(P,t)}{\partial P}\right] \tag{10}$$

Now in statistical equilibrium $\frac{\partial w(P,t)}{\partial t} = 0$, *w(P,t)* must reduce to a Boltzmann distribution $w = w_0 \exp(-VF/k_BT)$ where, *V* is the volume of the system, *F* is the free energy density, k_B is Boltzmann constant and *T* is the temperature of the system.

Substituting *w* in equation 10 gives, $D = \frac{k_bT}{V} = \frac{k_bT}{t_FA_f}$, where t_F is the thickness and A_F is the area of ferroelectric PZT capacitor.

Now introducing external noise voltage (rms value V_{noise}) having a power spectral density that is flat over a bandwidth Δf we can write equation 7 as,

$$\xi(t) = \sqrt{2\rho D_{int}}\frac{dW(t)}{dt} + \sqrt{2\rho D_{ext}}\frac{dW(t)}{dt} \tag{11}$$

where, $D_{ext} = \frac{V_{noise}^2}{2\rho t_F^2 \Delta f}$

Commented [VD1]: Need to ask Arvind exactly how this formulation of Dext comes.

In our simulation we assume $D_{int} << D_{ext}$ and we solve the discretized form of the equation 2 using Euler-Maruyama method as,

$$P[i] = P[i-1] - \frac{\Delta t}{\rho} . \frac{\partial F}{\partial P}[i-1] + \sqrt{\frac{2D_{ext}}{\rho}} . \Delta w[i-1] \tag{12}$$

<u>Supplementary Note 2: Resistivity fitting from PE-loop using TDGL equation</u>

We reconfigure the TDGL equation 2 ignoring the noise term and rewriting the equation into terms representing the voltage across capacitor (V_c) and voltage drop across the resistor (V_ρ) as,

$$\rho \frac{\partial P}{\partial t} = -\frac{\partial F}{\partial P} + \xi(t)$$

$$\rho \frac{\partial P}{\partial t} = -2\alpha P - 4\beta P^3 + E_s$$

$$\Rightarrow \rho \frac{\partial P}{\partial t} = -2\alpha P - 4\beta P^3 + E_s$$

$$\Rightarrow E_s = \rho \frac{\partial P}{\partial t} + 2\alpha P + 4\beta P^3$$

$$\Rightarrow E_s = E_\rho + E_C$$

$$\Rightarrow V_s = V_\rho + V_C$$

where $V_\rho = \rho \frac{\partial P}{\partial t} = \rho \frac{I_c}{A}$ and $V_C = \left.\frac{\partial F}{\partial P}\right|_{V_s=0} = 2\alpha P + 4\beta P^3$, ρ is the switching resistivity, A is the area of the device, I_c is the switching current at coercive field.

FigS1a shows the equivalent RC circuit for the PZT ferroelectric device, and the E_C and P_r extracted from PE loop (Fig S1b) are E_{C1}=7.98×10^6 V/m, E_{C2}=-8.1×10^6 V/m and P_{r1}=0.17C/m^2, P_{r2}=0.16C/m^2 respectively. Then ρ is estimated as,

$$\rho_1 = \frac{E_{C1} \times A}{I_{C1}(P = 0)} = 1621.69\Omega m$$

$$\rho_2 = \frac{E_{C2} \times A}{I_{C2}(P = 0)} = 1112.64\Omega m$$

$$\rho_{est} = \frac{\rho_1 + \rho_2}{2} = 1367.16\Omega m$$

<u>Supplementary Note 3: Gaussian white noise generation and experimental measurement details</u>

The mean of white Gaussian noise is 0 and hence the standard deviation is proportional to the noise magnitude i.e., n×noise will scale the standard deviation of the noise linearly by factor of n. Considering this, we generate an array of normally distributed random numbers using *randn* from MATLAB. Then we scale the array with different factors to get white noise of desired standard deviation, here we considered standard deviation of noise ranging from 0.15 to 5.10 with a resolution of 0.15. Fig S2 shows the process of white noise generation using MATLAB and then the noise + signal is fed to the PZT device through radiant ferroelectric analyzer.

<u>Supplementary Note 4: Poling protocols</u>

Before every measurement we pole the device at a voltage > 2*V_c and the poling pulse along with the sub-coercive signal + noise is shown in Fig S6.

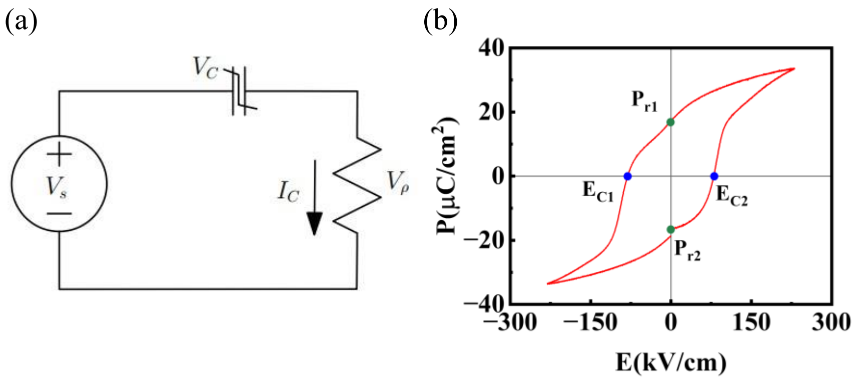

Fig S1. (a) Equivalent RC circuit of PZT ferroelectric device. (b) PE hysteresis loop for extraction of E_c and P_r.

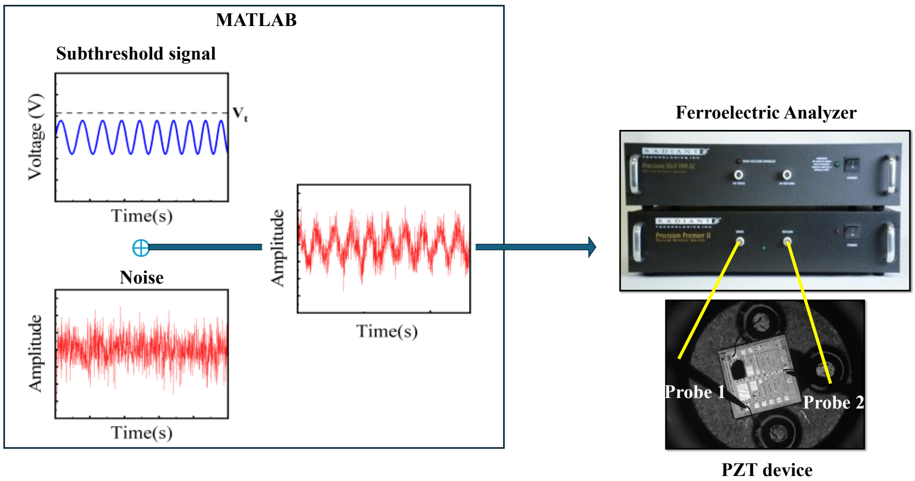

Fig S2. Noise generation and experimental setup for ferroelectric SR measurements.

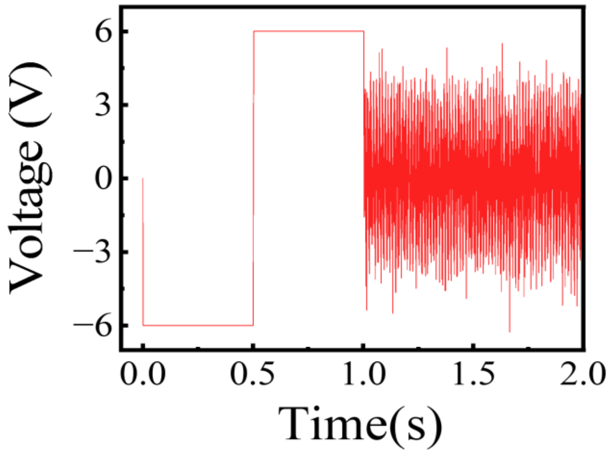

Fig S3. Input total signal containing the poling pulse fed to the PZT device.

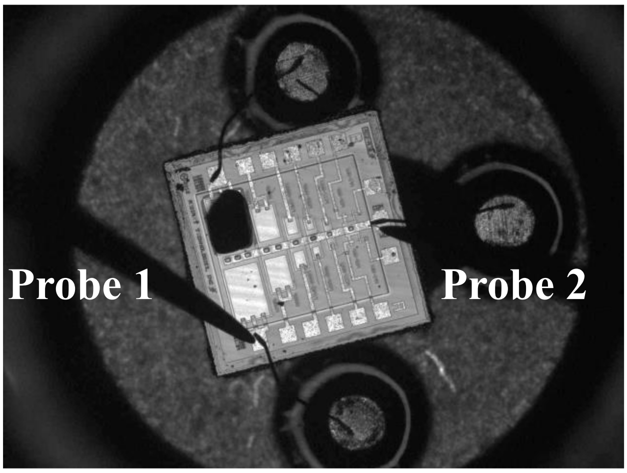

Fig S4. Optical image of the PZT ferroelectric capacitors inside a probe station.

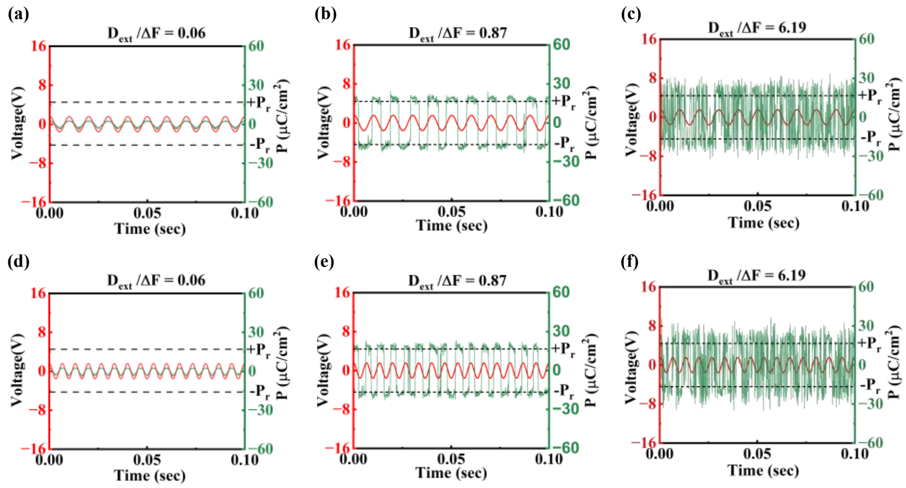

Fig S5. Simulated polarization vs time plots for 100 Hz (a-c) and 150 Hz (d-f) for different noise strengths, $D_{ext}/\Delta F$.

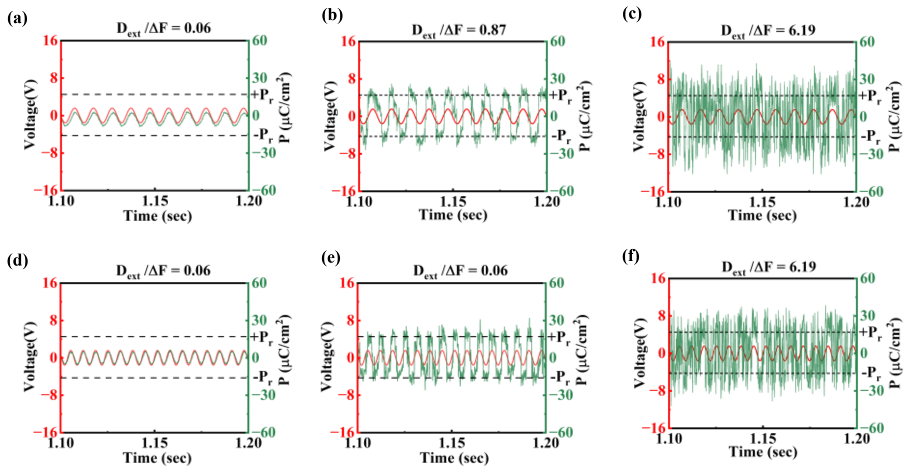

Fig S6. Experimental polarization vs time plots for 100 Hz (a-c) and 150 Hz (d-f) for different noise strengths, $D_{ext}/\Delta F$.

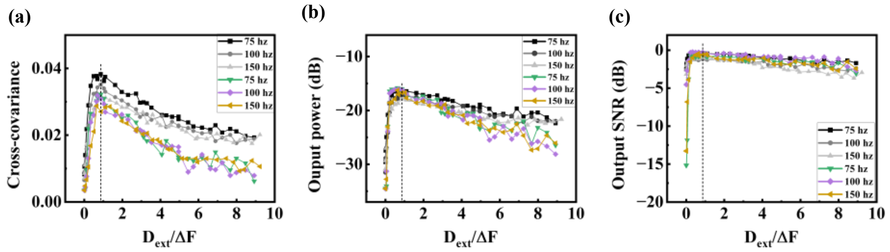

Fig 7. Comparison of all three independent metric (a) covariance, (b) output power, and (c) signal-to-noise ratio (SNR_1) for all frequencies. The colored plots correspond to the simulation result and the grayscale ones are for experiment. All three metrics for both simulation and experiment peak at similar value of $D_{ext}/\Delta F \sim 0.87$.

References:

1. Ramesh, M., Verma, A. & Ajoy, A. Kramers' escape problem for white noise driven switching in ferroelectrics. arXiv Preprint 2112.01373 (2022). https://arxiv.org/abs/2112.01373.
2. Risken, H. The Fokker-Planck Equation: Methods of Solution and Applications (Springer, Berlin, 1996).