

## Introduction

A model to simulate polarization switching in large sliding ferroelectric systems is proposed in this work. By supplementing the model with established molecular dynamics methods, the effect of interlayer electric potential energy differences in the motion of polarization domain walls can be observed. Following the recent experimental observation of hysteresis in bilayer hexagonal boron nitride (hBN) systems within graphene cavities, a simple mathematical model is proposed to explain this phenomenon.

## Charge exchange model

A simple registry-dependent atom-resolved charge exchange model is deployed in order to replicate both the polarization of different hBN stackings and the change in energies and forces on each atom due to interlayer electric potential differences.

The model is fitted to DFT data and replicates the out-of-plane dipole moment of hBN systems at different temperatures.

$$\Delta q_i = \begin{cases} \sum_{\alpha_i \neq \alpha_j, l_i \neq l_j} q_{0,\alpha_j} \text{Tap}(r_{ij}) e^{-r/d_{0,\alpha_j}}, & l_i = 1 \\ \sum_{\alpha_i \neq \alpha_j, l_i \neq l_j} -q_{0,\alpha_j} \text{Tap}(r_{ij}) e^{-r/d_{0,\alpha_j}}, & l_i = 2 \end{cases} \quad (1)$$

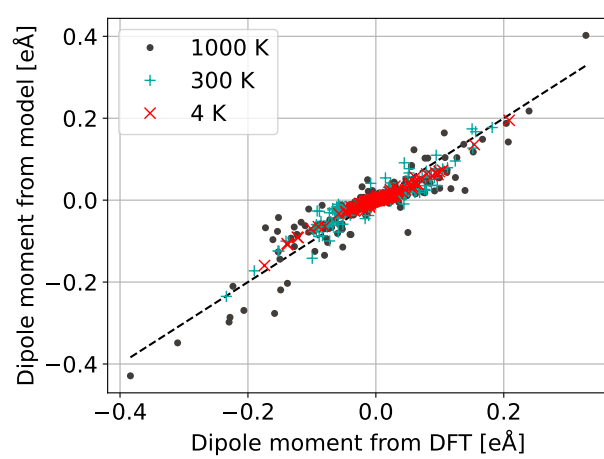


Figure 1. Model accuracy comparison

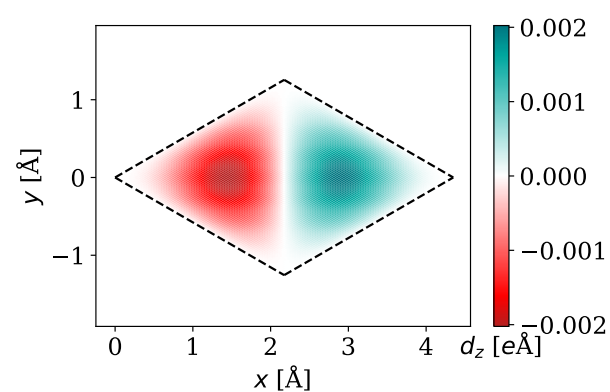


Figure 2. Model prediction of out-of-plane dipole moment in configuration space

The resulting energy difference and forces from interlayer electric potential differences can be written as

$$\Delta E_{el} = \sum_i V_i \Delta q_i, \quad \Delta F_{el,j} = - \sum_i V_i \nabla \vec{r}_j \Delta q_i \quad (2)$$

## Molecular dynamics

Interlayer van der Waals interactions and intralayer covalent bonds are calculated through well established methods in LAMMPS. Specifically, the ILP potential and a Tersoff potential are deployed.

## 1D domain walls

Domain walls arise from non-trivial solutions of Euler-Lagrange equations that take the intralayer elasticity and interlayer van der Waals interaction into account.

$$E = \int_{-\infty}^{\infty} dx \left[ \frac{\lambda_{1D}}{2} \left( \frac{\partial \phi}{\partial x} \right)^2 + V(\phi) \right] \quad (3)$$

$$\phi_{DW}(x) = \pm a_0 \tanh \left( \frac{2(x - x_0)}{w} \right), \quad w = \frac{a_0}{2} \sqrt{\frac{\lambda_{1D}}{V_0}} \quad (4)$$

There exist 4 types of 1D domain walls in bilayer hBN, they are labeled depending on the deformation direction. Their formation energies  $\gamma = E[\phi_{DW}(x)] - E[\pm a_0]$  and related parameters can be calculated from molecular dynamics minimization.

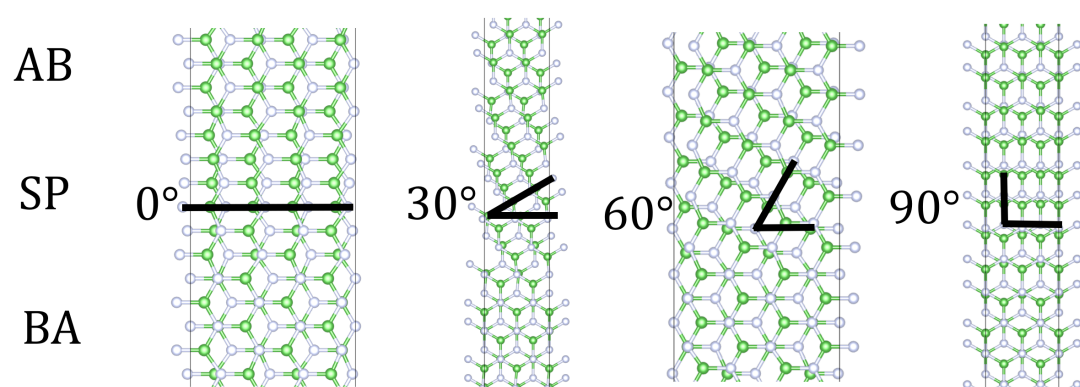


Figure 3. 4 different types of 1D domain walls

DW type	$\gamma$ [eV/Å]	$\lambda_{1D}$ [eV/Å <sup>2</sup> ]	$v_e = \sqrt{\lambda_{1D}/\rho}$ [km/s]
0°	0.0763	1.710	9.155
30°	0.0880	2.277	10.564
60°	0.1119	3.682	13.434
90°	0.1244	4.550	14.934

Table 1. Properties of the different domain walls

## Domain wall annihilation and 2D domain walls

The formation energies of the different domain walls can be calculated from molecular dynamics energy minimization. Opposing domain walls annihilate in a system where an interlayer electric potential difference is applied. The domain walls reach a maximum speed which can be calculated from the effective elastic parameter  $\lambda_{1D}$  and the density of monolayer hBN. Time-dependent simulations are performed at 0 K with a Velocity-Verlet algorithm as implemented in the ASE Python library.

The formation energy of a relaxed twisted hBN bilayer can be inferred from the formation energy of the 0° domain wall.

$$E(\theta) = \varepsilon_{AB} A(\theta) + 3\gamma_{0^\circ} L(\theta) + E_{AA} \quad (5)$$

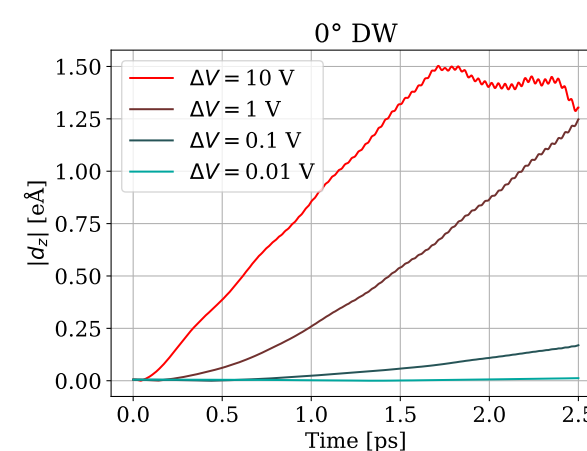


Figure 4. Polarization switching under different potential differences

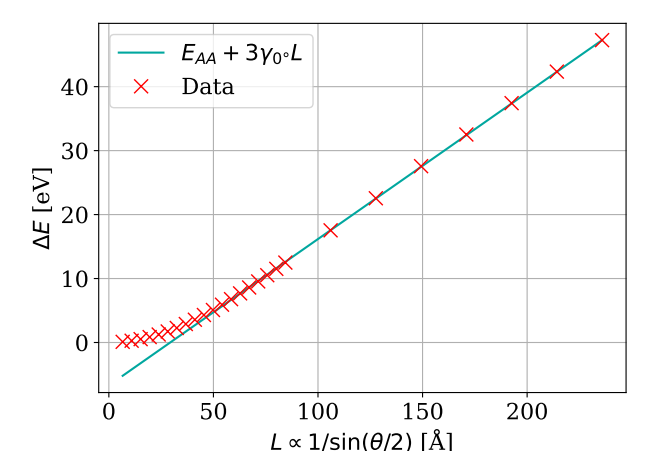


Figure 5. Formation energies of twisted bilayer hBN

## Mathematical argument for hysteresis

Given a width function  $f(x)$  for a graphene cavity, the energy of a domain wall being present along  $x$  can be written as

$$E(x) = \gamma f(x) + p_z E_z \left( \int_{x_1}^x f(s) ds - \int_x^{x_2} f(s) ds \right), \quad (6)$$

where the first term is the formation energy of the domain wall and the second term is the energy due to the interaction with an electric field. From this simple mathematical argument, the energy barrier to switch from one side of the cavity to the other as a function of the applied electric field can be calculated.

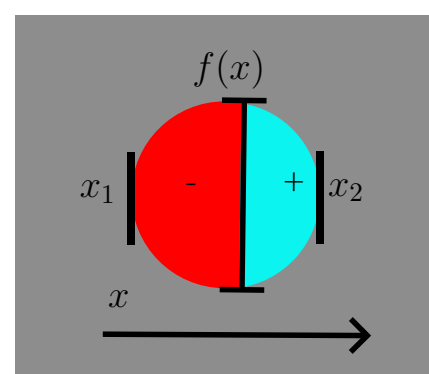


Figure 6. Parameters of the cavity model.

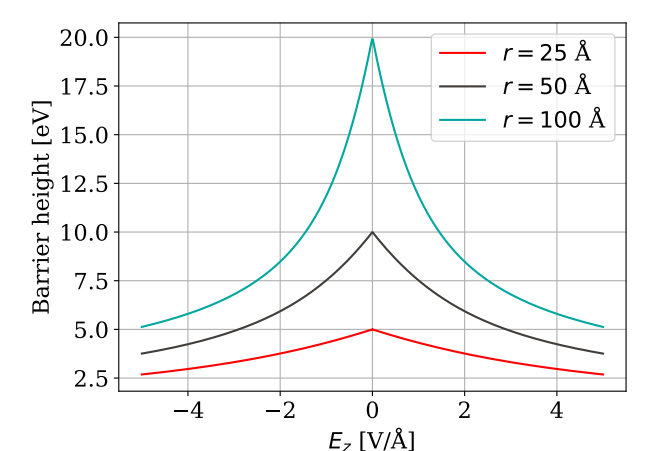


Figure 7. Energy barrier in the cavity model

## Conclusions

- A simple charge exchange model can be used to predict the polarization of sliding ferroelectric systems and simulate motion of domain walls under interlayer electric potential differences.
- Different types of domain walls have different speeds of sound that can be predicted from their formation energies.
- The formation energies of 1D domain walls can help predict the formation energies of 2D domain walls.
- The mechanism through which graphene cavities allow for the possibility of a polarization hysteresis in bilayer hBN can be mathematically explained through the interplay of domain wall formation energies and electric field interactions. More computational simulations are needed to determine the validity of the mathematical argument.

## References