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Tunability response in exponentially graded ferroelectrics: A TIM model approach

H. Vivas C.*, J.F. Jurado, C. Vargas-Hernández

Grupo de Propiedades Ópticas de los Materiales, Departamento de Física, Bloque Y, Universidad Nacional de Colombia, Manizales A.A. 127, Colombia

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ABSTRACT

Relative dielectric function response associate to a non-homogeneous layered ferroelectric system is calculated in the framework of the Mean Field Approximation (MFA) for the Transverse Ising Model (TIM). Analytical self-consistent expressions for the average polarization, dielectric susceptibility, and tunability percentage are outlined and solved for different configurations and sizes. It is found that exponentially graded ferroelectrics magnify the tunability response for stronger interlayer coupling and it reaches its saturation value for smaller intensities of the applied electric field.

1. Introduction

Ferroelectric materials possess spontaneous polarization that can be depleted hysteretically by an applied electric field. Ferroelectrics belong to a bigger family of special materials called multiferroics [1], that may be used to couple magnetic materials due to their subtle piezoelectric properties and large polarization fields in tunable microwave devices operating at room temperature [2,3]. Developments in ferroelectric (FE) thin-film technologies have stimulated the study of theoretical models and numerical approaches in arrays of multilayers. Different techniques such as the microscopic Green-de Gennes (GdG) functions [4], the thermodynamic free energy Landau–Ginzburg (LG) approach [5,6] or the Mean Field-Zernike approximation for the Transverse Ising Model (TIM) in layered configurations [7], provide examples for applicability in the calculation of the polarization response, strain effects, transition temperatures and phase diagrams. Variations of LG theories (Ginzburg–Landau–Devonshire, GLD), for instance, have been discussed in tunable-phase configurations of the FE state in a Paraelectric/Ferroelectric/Paraelectric trilayer [8], while the GdG functions have shown to be a powerful method for approaching the microscopic problem of exchange anisotropy in layered (FE) systems. The 2D Transverse Ising Model has also been considered as a promising model for describing proton ordering in systems such as potassium di-hydrogen phosphate or KDP [9], polarization fields fluctuations in $BaTiO_3$ films via temperature gradients [10,11], or FE/AFE bilayer coupling in $BiTiO_3$/$SrTiO_3$ (BST) superlattices [12]. The coherent tunneling hypothesis, the key idea in the framework of the TIM scenario, has found supporting evidence in Neutron Scattering measurements even when phonon transferring mechanism, inferred from Raman Spectroscopy experiments, is still widely accepted [13]. Artificially graded ferroelectrics have gained relevance since novel properties due to interfacial defects and/or induced thermo-electromechanical strain are observed and explained in terms of dopands concentration dependence, the strength of the interlayer coupling, and more recently, by performing *ab initio* calculations in geometrically frustrated systems [14]. Applications for optimization of stable tunability and low losses have also been reported in matching-impedance circuits, built from Barium Strontium Titanate Oxide (BSTO)/Al ceramic materials in multilayers with graded dielectric constant [15], or waveguides with BSTO/Mg composites with low tangent losses and high tunability outcome [16]. Wedge domains of special geometries in compositionally graded BST multilayers might be engineered to design a new class of electromagnetic devices with highly controllable dielectric response at room temperature or stress [17,18]. In this scenario, we study the Transverse Ising Model for the polarization and tunability responses driven by an uniform electrical field in a quasi-2D graded array. The effective interlayer interaction K_{nm}, being n,m the layer indexes, is considered non-homogeneous and dependent on the relative distance between layers n and m along the growth direction, whereas the transverse field parameter remains uniform for all possible configurations analyzed. This model allows elucidate, among other effects, the correlation between the average polarization per layer, the size of the lattice and the interaction strength, which can be indirectly interpreted as a

* Corresponding author. Tel.: +57 6 8879300x55747; fax: +57 6 8742725.
E-mail address: hvivasc@unal.edu.co (H. Vivas C.).

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signature for the depolarization field distribution in graded arrays. In Section 2 we develop the formalism leading towards the quantities of interest such as the average polarization and the coefficient of tunability; in Section 3 we display and discuss the numerical results and Section 4 is devoted to drawing general conclusions and some perspectives.

2. Model

We consider a device consisting of a set of N ferroelectric layers aligned in Z direction. The distance between two any consecutive planes is uniform and equal to a, as shown in Fig. 1. An uniform electrical field e^Z is also applied along Z direction. The microscopical description for the polarization field has been addressed by the Transverse Ising Model Hamiltonian [19]

$$\mathcal{H} = -\frac{1}{2}\sum_{ij} K_{ij} S_i^Z S_j^Z - t\sum_i S_i^X - e^Z \sum_j S_j^Z, \tag{1}$$

where K_{ij} defines the exchange interaction between sites i,j, and t might represent the tunneling constant or transverse field between pseudospin-$\frac{1}{2}$ states $S^X \rightarrow S^Z$. By recasting the MFA-type formalism developed by Blinc and Zěckš [20], we obtain the thermal mean values for the polarization components per layer with index n, $\langle S_n^Z \rangle$ and $\langle S_n^X \rangle$ explicitly as

$$\langle S_n^Z \rangle = \frac{R_n^Z}{2G_n} \tanh\left(\frac{G_n}{2k_BT}\right), \quad \langle S_n^X \rangle = \frac{t}{2G_n} \tanh\left(\frac{G_n}{2k_BT}\right) \tag{2}$$

with $R_n^Z = e^Z + \sum_m K_{nm} \langle S_m^Z \rangle$, and $G_n = (t^2 + (R_n^Z)^2)^{1/2}$. The total mean polarization $\mathcal{P}^Z(T)$ is defined as

$$\mathcal{P}^Z(T) = \frac{1}{N}\sum_{n=1}^{N} \langle S_n^Z \rangle, \tag{3}$$

where N is the total number of layers. The expression for $\mathcal{P}^Z$ in Ref. (3) has been normalized for simplicity in terms of $n\mu$, where n is the number of pseudospins per volume and μ represents dipole momentum per site. Exchange interaction between two adjacent polarized planes are described through the dependence

$$R_n^Z = e^Z + 4K \exp[-\lambda n]\langle S_n^Z \rangle + K \exp[-\lambda(n+1)]\langle S_{n+1}^Z \rangle + K \exp[-\lambda(n-1)]\langle S_{n-1}^Z \rangle, \tag{4}$$

where K corresponds to the effective *in-plane* interaction, and λ is a normalized parameter (with respect to a) which takes values from 0 to 0.25 in all our calculations. Electrical susceptibility of the graded structure is calculated as $\chi \equiv (\partial \mathcal{P}^Z/\partial e^Z)$, with $\mathcal{P}^Z(T)$ defined in Eq. (3). The susceptibility per layer χ_m is obtained by solving for the tridiagonal matrix system

$$\sum_m \chi_m Q_{mn} = F_n \langle S_n \rangle, \quad F_n = \frac{1}{R_n^Z} + \frac{1}{G_n}\left(\frac{1}{4k_BT \sinh(G_n/2k_BT)} - \frac{1}{G_n}\right) \tag{5}$$

with $Q_{nm} = \delta_{mn} - K_{nm} F_n \langle S_n \rangle$, and δ_{mn} representing the Kronecker delta function. For ferroelectric devices operating in the microwave regime, the tunability is defined as the ratio of the dielectric permittivity of the material at zero electric field to its permittivity at non-zero electric field, explicitly giving by [27]

$$\%\eta = \frac{\varepsilon(e^Z = 0) - \varepsilon(e^Z)}{\varepsilon(e^Z = 0)} \times 100 \tag{6}$$

with $\varepsilon(e^Z) = \varepsilon_0(1 + \chi(e^Z))$. Relationships (3)–(6) constitute a group of self-consistent (non-linear) equations for the percent of tunability ($\%\eta$) as a function of the size of the array, the external field intensity and the characteristic length λ^{-1}.

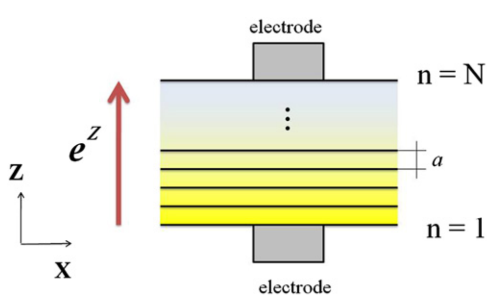

Fig. 1. Array representation of N ferroelectric layers aligned in Z direction.

3. Numerical results

The parameter λ^{-1} in Eq. (4) can be interpreted as a characteristic length intrinsically linked to the cluster geometry of the lattice, and local strength of substituted dopands defects [21]. Exchange interactions models in surface transition layers (STL) have been theoretically proposed as a function of the relative distance between planes and depolarization fields [22,23], or by modifying the *free* carrier density ρ_f with respect to the bound charge density ρ_b through the functional $\xi = 1 - \rho_f/\rho_b$. For two adjacent FE planes, it is shown that the coupling K_{planes} between is proportional to ξ, in the framework of basic theoretical Landau–Maxwell formulation [24,25]. In that model, ξ has two limiting values, namely $\xi = 0$ and $\xi = 1$, which correspond to semiconducting and perfect FE insulating states, respectively. Our *ansatz* (4) might take into consideration these two states under the assumptions $\lambda a \gg 1$, and $\lambda \rightarrow 0$. Fig. 2 (left) shows the mean polarization-external field dependence for different number of

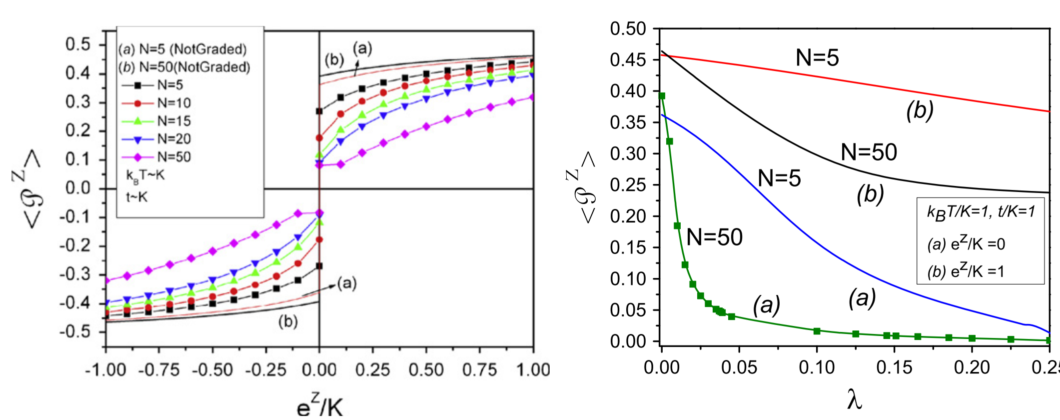

Fig. 2. (Left) Electrical polarization for an exponentially graded multilayer, with $t/K = 1$, $k_BT = K$, $\lambda = 0.05$, and $N = 5, 10, 15, 20, 50$. (Right) Comparative mean polarization behavior for small ($N=5$) and large ($N=50$) systems with and without applied field.