A Numerical Approach of Polarization Profile for First-Order Ferroelectric Film System Using Finite-Difference Method

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***Abstract*- A detail account of numerical calculations using Finite-Difference Method (FDM) to calculate the polarization profile in first-order ferroelectric (FE) film system is given. For film, both symmetric and asymmetric cases are considered. The formalism is based on Tilley and Žekš (TZ) model using Landau-Devonshire (LD) equations with the boundary conditions. The numerical technique is described using Central and Forward Finite-Difference Approximation.**

I. INTRODUCTION

With the continued demand for portability in consumer electronics, it is becoming increasingly important to understand the effects of miniaturization on the properties of the active components in electronic devices. In many cases, however, the basic physics of such size reduction is poorly understood and can be difficult to characterize, because competing effects such as surface properties, strain effects from substrates, and fundamental size quantization complicate the behavior [1].

In the past few years, studies on ferroelectric (FE)-semiconductor devices are gaining importance, especially for future non-volatile memory devices, and would be a major focal point of future research activities [2]. FE non-volatile memories is attractive as replacement for conventional EEPROM and flash EEPROM because of lower write voltages, faster write speeds, and potentially fewer processing steps [3]. An important possible application of FE-semiconductor devices is the microminiaturization of a large number of adaptive transistors in an integrated circuit. Taylor [4] reported that in FE-semiconductor devices fabrication, the method of fabrication strongly influences the electrical stability of the devices. FE random access memory (FRAM) is one of semiconductor memory devices which have attracted a lot of attention, where the dielectric material in DRAM capacitor cell is replaced with a FE thin film [5]. In principle, FRAM should have a lower requirement, a faster access time, and potentially lower cost than many other many semiconductor devices, but the commercialization has been quite slow because of materials issues [6, 7]. FE materials should satisfy the following criteria in order to realize a practical FRAM which is compatible to the current DRAM technologies, large remnant polarization P_r, and good reliabilities. The FE memories have more advantages as a memory embedded product than other conventional memories. This is because of the non-volatile behavior, high speed operation, low power supply, high endurance and good CMOS process compatibility [7].

The size effect of FE has been studied extensively in recent years since there is a rapid progress in FE films and composite materials. It is well known that the FE polarization profile $p(z)$ of semi-infinite and film for surface-effect study are changed near the surface. Tilley and Zeks [8] showed that the explicit expressions for $p(z)$ in the second-order FE films using Jacobi elliptic functions and was later improved by Ong et al.[9] The extrapolation length δ was introduced in FE size effect study in order to understand the behavior of $p(z)$ between bulk and surface area. The $p(z)$ enhanced and depressed near the surface for $\delta < 0$ and $\delta > 0$ respectively. Recently, Halif et al.[10, 11] give the overview solution of far-infrared first-order FE transmission model using Finite-Difference Method (FDM). The same approach of numerical method to solve surface effects and size effect on first-order FE using Ising model is given by Wang et al.[12].

II. GENERAL FORMALISM

To perform the results in universal overview, all the formalism may be written in dimensionless form using conventional scaling [1, 9-11]. In the FE systems, the Gibbs free energy are given by

$$G = \int_i^j f\left(p, dp/dz\right)dz + \left(\phi/2\delta\right)\left(p_-^2 + p_+^2\right) \tag{1}$$

where

$$f\left(p, dp/dz\right) = \alpha T p^2 + \beta p^4 + \gamma p^6 + (1/2)\left(dp/dz\right)^2 - Ep \tag{2}$$

is the Landau-Devonshire (LD) free energy per unit volume. For first-order FE, $\alpha = 1/2$, $\beta = -1/2$ and $\gamma = 1/6$, while for second-order, $\alpha = 1/2$, $\beta = 1/4$ and $\gamma = 0$. From (1), $i = 0$ and $j = \infty$ for semi-infinite system and, $i = -l/2$ and $j = +l/2$ for film system. T and E represent the electric field and temperature in dimensionless form respectively. The last term in (1) shows the polarization at the top and bottom of films, while for semi-infinite only one polarization plays a role for one boundary condition. As mentioned earlier, extrapolation length δ is the term to compare the $p(z)$ between the surface and the bulk region. This term leads to the boundary conditions (BC) that have to be satisfied namely

$$(dp/dz) \pm p/\delta = 0 \tag{3}$$

where the "+" and "-" sign applies for films case at top ($z = l/2$) and bottom ($z = -l/2$) of film surface. For semi-infinite medium, the BC may be written as $(dp/dz) = p/\delta$. By considering the equilibrium state (minimum of f) with the final term of (2) omitted, thus the static polarization, $p_0(z)$, profile is the solution of Euler-Lagrange (EL) equation, and expressed by

$$(d^2 p_0 / dz^2) + 2\alpha t p_0 - 4\beta p_0^{\,3} + 6\gamma p_0^{\,5} = 0 \qquad (4)$$

The polarization profile $p(z)$ may be evaluated numerically using (4) and BC (3). For the special case (no surface effect) $\delta^{-1} = 0$, $p_0(z)$ is equal to the bulk polarization p_B. The bulk value may be directly calculated from minimum stage of (1) by ignoring the last term in equilibrium state ($E = 0$). p_B for first order case may be written as

$$p_B^2 = 1 + \sqrt{1 - t} \qquad (5)$$

This value may be used as a final value in the numerical calculation. A detailed explanation is described in section III.

III. NUMERICAL APPROACH

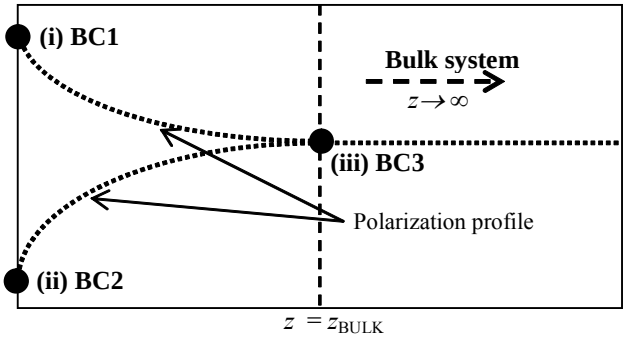

Figure 1: Polarization profile schematic illustration for semi-infinite ferroelectric system based on Brent's Method [13].

Finite-Difference Method (FDM) is a simple and efficient method for solving ordinary differential equations (ODEs) in problem regions with simple boundaries. The method requires the construction of a mesh defining local coordinate surface. For each node of this mesh, the unknown function values are found, replacing the differential equations by difference equations, i.e $dy = h(x,y)dx$ is replaced by $\Delta y = h(x + \Delta x/2 , y + \Delta y/2)\Delta x$, where Δx and Δy are steps in an iterative procedure. The FDM is also a method for solving partial differential equations (PDEs). For example a PDE will involve a function $h(x)$ defined for all x in the domain with respect to some given boundary condition. The purpose of the method is to determine an approximation to the function $h(x)$. The method requires the domain to be replaced by a grid. The central finite difference approximations for second-order derivative may be written as

$$h''(x) = \frac{h(x + \Delta x) - 2h(x) + h(x - \Delta x)}{(\Delta x)^2} \qquad (6)$$

which is generated from Taylor Series. By substituting (6) into EL equation and assuming that $h(x + \Delta x) = h_{n+1}$, $h(x - \Delta x) = h_{n-1}$, and $h(x) = h_n$, thus

$$p_{n+1} = 2p_n[1 + T(\Delta z)]^2 - p_{n-1} - 4\beta p_n^{\,3}(\Delta z)^2 + 6\gamma p_n^5(\Delta z)^2 \qquad (7)$$

From Fig. 1, the numerical procedure of $p(z)$ using FDM method is started from ferroelectric surface towards bulk, where $dp/dz = 0$. Thus, by using forward different approximation for first-order derivative, BC (3) may be expressed by

$$p^*_{n+1} = p_{n-1} \pm 2(\Delta z) p_n / \delta \qquad (8)$$

Eq. (7) and (8) are used to generate $p(z)$ for ferroelectric films. From previous results [10, 11], the polarization profile enhanced and depressed at surface for $\delta < 0$ and $\delta > 0$ respectively. This corresponds to the BC1 and BC2 in Fig. 1. For surface enhancement, $p_B < p_S$, (S : surface) and for surface depressed, $p_B > p_S$. By assuming $p_n = p_{n-1}$ for a initial value, p^*_{n+1} may be used as temporary values for (7). Here, we only give the preliminary result for the surface depressed conditions. The overall polarization profile is generated using (7) until the last two values of $p(z)$ satisfies BC3; $p_{n-1} = p_{n+1}$ at bulk region. For symmetrical film case, the polarization profile is the combination of two semi-infinite cases [11]. While for asymmetrical case the extrapolation length is $\delta_+ \neq \delta_-$. Here we only highlight the results for positive-positive surface conditions.

III. RESULTS & DISCUSSION

Figure 2(a) and 2(b) show the polarization profile for symmetrical and asymmetrical case respectively. As mentioned before, we represent only the surface depressed conditions as a preliminary result. In Fig. 2(a), three values of extrapolation length δ are selected corresponding to the numerical limit, 1, 10 and 1000. The first value gives the highest surface effect and the last gives the approximation of the bulk behavior. We only choose the pure ferroelectric phase below supercooling temperature to demonstrate the film surface effect. For the symmetric film in Fig. 2(a), we assumed that δ at both surface is similar.

As modern electronic devices consist of films on substrate, films are generally constrained by asymmetric boundary conditions. Thus, the study of polarization profile under the influence of substrate is highly important to give more realistic result compared to the symmetrical case. Fig. 2(b) gives the preliminary result for asymmetric film behavior with positive-positive surface conditions. Here we assumed that the polarization is strongly depressed with $\delta = 1$ at right-hand side because of mismatched effect between ferroelectric film surface and substrate surface. The preliminary result here shows that the numerical approach using FDM technique may be used to solve the polarization profile for both film systems: symmetrical and asymmetrical case within the framework of TZ model. Currently, Chew et al. [14] have presented the mathematical modeling of asymmetric ferroelectric film using quasi-numerical approach.

We are currently pursuing to study the far-infrared spectroscopy [15] of asymmetric ferroelectric film with surface effect with three conditions: (i) positive-positive surface conditions; (ii) negative-negative surface conditions; and (iii) positive-negative surface conditions, and hope to publish the results in near future.

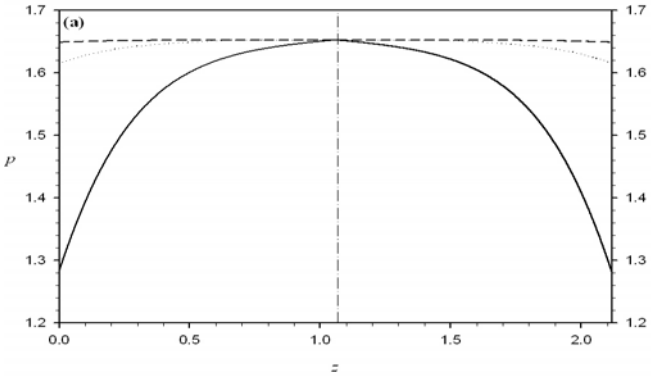

Figure 2(a) Numerical computation of polarization profile $p(z)$ for symmetric ferroelectric thin film with three value of extrapolation length $\delta > 0$ where (____) $\delta = 1$, (…….) $\delta = 10$ and (- - - -) $\delta = 1000$ at below supercooling temperature $T = -2.0$.

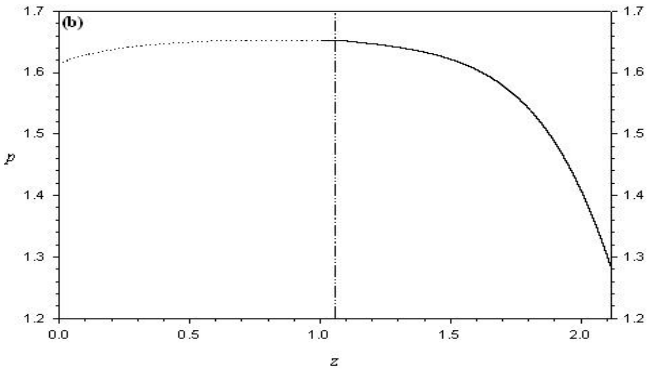

Figure 2(b) Numerical computation of polarization profile $p(z)$ for asymmetric ferroelectric thin film with positive-positive surface conditions: (…….) $\delta = 10$, and (____) $\delta = 1$ at pure FE phase with temperature same as Fig. 2.0. Assume that the right hand-side of the graphs is the ferroelectric film surface with substrate.

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