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Far-infrared optical transmission through asymmetrical ferroelectric films

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Abstract

Investigation of far-infrared (FIR) spectroscopy for symmetrical ferroelectric (FE) films are discussed theoretically based on the basis of the Tilley-Zeks (TZ) model using Landau free energy expansion, Landau-Khalatnikov (LK) equation of motion, and electromagnetic wave equation for the second [1] and first order [2, 3] respectively. Here by using the same model we scrutinized a preliminary result of non-surface effect of FIR spectra behaviour for asymmetrical FE films in both phase transitions incorporating with the effect of refractive index.

Keywords: Ferroelectric films, far-infrared, reflectivity.

Introduction

The unique properties of ferroelectric (FE) materials i.e spontaneous polarization and high permittivity may differ it from normal dielectrics [4]. This characteristic makes FE suitable for memory applications such as capacitor, FE DRAM and non-volatile FE memory. Thin film FE devices are being considered in numerous electronic and opto-electronic devices i.e non-volatile semiconductor memories, optical waveguide devices, spatial light modulators, switching capacitors for integrated circuitry, pyoroelectric devices and imaging sensors [5]. Recently, in the DRAM technology, the conventional SiO_2 film plane structure cannot maintain a sufficient capacitance with decreasing element area and also with the small dielectric constant [6]. FE gives a good alternative for DRAM technology with the high permittivity, i.e almost FE materials have dielectric constant > 1000 compared to the conventional dielectric such as SiO_2, Si_3N_4, Ta_2O_5, and ZrO_2 [6, 7].

Since a lot of FE thin film such as $PbZr_xTi_yO_3$ (PZT), $BaTiO_3$, $PbTiO_3$ and $SrTiO_3$ have been widely studied for memory devices purposes, many experimentalist investigate on the depositions technique between FE and Si because it may affect the performance of FE thin film capacitors [8]. The problems of surface-effect in FE is one of a common study, since the technology is heading for the extremely thin films memory devices, thus, the area is greater than the volume, and gives domination of surface effect. Ishibashi [9] and Chew et al. [10] recently have given in-deep theoretical work of second-order FE thin film based on Tilley-Zeks (TZ) model [11] from symmetrical and asymmetrical case. In this paper, we give a theoretical preliminary optics study of far-infrared (FIR) spectroscopy of asymmetrical FE film in a special case without surface-effect $\delta^{-1} = 0$. As we know, FE and Si are transparent to FIR waves, and here we incorporate the effect of FIR wave's propagation in the asymmetrical FE film with refractive index of Si. This premier-study is highly important as a theoretical-guide to extend this work on more realistic case with surface-effect.

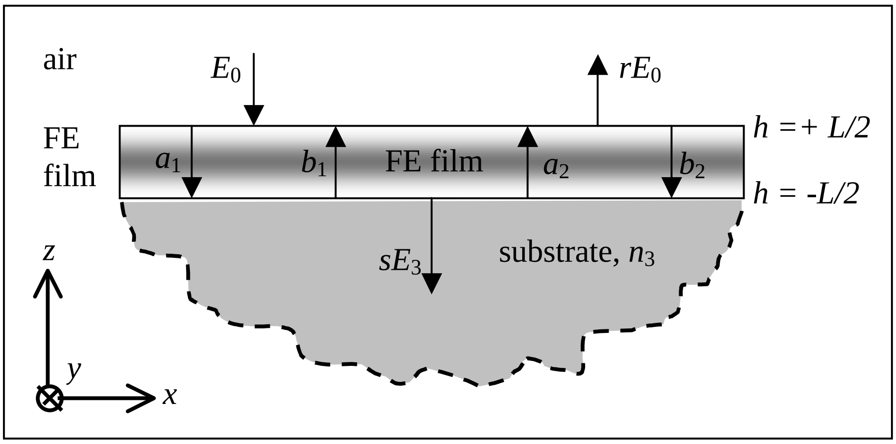


Figure 1: Schematic illustration of asymmetrical ferroelectric films with thickness L on a Si substrate.

The TILLEY ZEKS Model

Normally in the Tilley-Zeks (TZ) model for FE film with thickness L as illustrated in Fig. 1, we may start our theoretical calculations from Gibbs free-energy per unit area [1-3, 12]

$$G = \int_{-L/2}^{+L/2} f(p, dp/dh)dh + \frac{\kappa}{2\delta}(p_-^2 + p_+^2) \tag{1}$$

Eq. (1) is in dimensionless form using conventional scaling [1 - 4] to perform a universal result without any materials dependent. The extrapolation length δ gives the conditions of presence of boundaries in thin films where polarization at the bottom and top of film represented by p_-^2 and p_+^2 respectively. This condition may represented by boundary conditions $dp/dh \pm p/\delta = 0$. $\kappa^2 = (4DC/B^2)$ states the existence of spin wave of optical mode as an additional to the normal polariton wave. The first term is the Landau-Devonshire (LD) free-energy density [1- 3, 12]

$$f\left(p, dp/dh\right) = Mp^2 - Np^4 + Up^6 + (1/2)(\nabla p)^2 - Ep \qquad (2)$$

with $M = ½ (T_2 – 1)$, $N = ¼$ and $U = 0$, while $M = ½ T_1$, $N = ½$, and $U = 1/6$, for second and first-order phase transition respectively. T_2 and T_1 represent the temperature for second and first-order transitions in dimensionless form using conventional scaling [1-4, 12]. The last term of (2) described the external electric field caused by incident FIR waves and assumed in the x direction. Since we only considered the preliminary results, so-called a non-surface effect case where $\delta^{-1} = 0$, (1) the Gibbs free-energy per unit area is reduced to $\int_{-L/2}^{+L/2} f(p, dp/dh)dh$ with steadfast polarization $dp/dh = 0$. By considering the equilibrium state ($E = 0$), the solution of Euler-Lagrange (EL) from eq. (2) gives a static polarization $p_0(h)$ which leads to the bulk polarization p_B for non-surface term conditions. Second and first-order phase transitions $p_0(h)$ is $p_B^2 = 1 – T_2$, and $p_B^2 = 1 + (1 – T_1)^{1/2}$ respectively [1 - 3, 12].

Optics Formulation

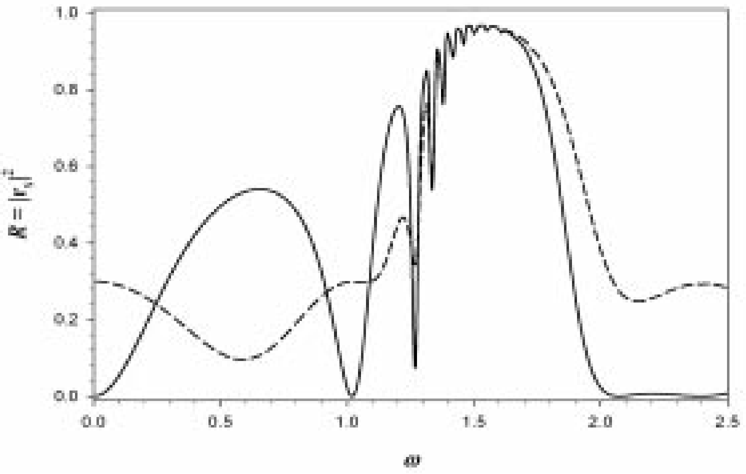
Fig. 1 illustrated the simple 2-D geometry for asymmetrical FE thin film on substrate. For this preliminary study we may focus on silicon (Si) substrate with refractive index $n_3 \approx 3.42$ in FIR ranges. To simplify the calculations, the incident wave e_0 is taken normal to the film surface. a_1, a_2, b_1, and b_2 represented the two mode wave propagation in the films [1 – 3]. r and s is the reflection and transmission coefficient respectively. There are two major equations to describe the optics behind the FIR response of FE thin films [1 - 3]. The first is the Landau-Khalatnikov (LK) equation of motions where the coupling to the electric field from FIR wave is described by $\hat{O}p = -\delta f/\delta p$, where we assumed the external energy from the electric field gives the same effect as damped oscillatory spring, thus $\hat{O} = m\partial^2/\partial t^2 + \gamma\partial/\partial t$. To simplify the calculation by assuming a single frequency input, and then the LK may be rewritten as

$$(d^2 q_i/dh^2) + \left(\omega^2 - \omega_i^2 + i\gamma\omega\right)q_i + E_i = 0 \quad \text{for } i = x \text{ or } y \qquad (3)$$

with q represented the optical polarization upshot from electromagnetic wave representing the dynamic part. ω is the dimensionless frequency [1 – 3]. For the special case $\delta^{-1} = 0$, ω_x and ω_y are constant, where $\omega_x^2 = 2(1 – T_2)$, and $\omega_x^2 = 4\{1 – T_1 + (1 – T_1)^{1/2}\}$ for second and first-order case respectively. For y-polarization, both cases give $\omega_y^2 = 0$. The second equation called Maxwell’s electromagnetic wave equation and gives a complementary relation between E that appeared in (2) and q (both in x direction) in the variational derivative and described by

$$(d^2 e_x/dh^2) + \alpha\varepsilon_\infty\omega^2 e_x + \alpha\beta\omega^2 q_x = 0 \qquad (4)$$

Eq. (3) and (4) may be solved together by considering the propagation of E and q in exponential function of $\exp(ik\sqrt{\alpha}h)$ to generate the dispersion equation [1 -3]. Chew et al. [1], Halif et al. [2, 3] and Tilley and Zeks [6] give a detailed explanation of propagation of two existence mode $k_{1,2}$ in bulk FE. Works of Chew et al. [1] and Halif et al. [2, 3] implement that for the dispersion curves of k_1 has the usual polariton shape and have a pure imaginary region, so-called reststrahl region in the reflectivity curves. The only comparison between second and first-order FE phase transitions in FIR optics case is the frequency ω of dispersion ranges, where second-order appeared in the ranges of $0 < \omega < 2.5$, while the first-order is 10 times greater. Using experimental data of $BaTiO_3$ [13, 14], from conventional scaling [1-3], thus $\alpha = D/(m\varepsilon_0 c^2) \approx 10^{-3}$, $\beta = (aT_0)^{-1} \approx 5$, and $\gamma = (g^2\varepsilon_0 m/aT_0)^{1/2} \approx 0.01$ for second-order case, while $\alpha = D/(m\varepsilon_0 c^2) \approx 10^{-3}$, $\beta = 4C/B^{\,2} \approx 10^3$, and $\gamma = 2(g^2\varepsilon_0 C/B^{\,2} m)^{1/2} \approx 0.1$ is for first-order parameters. ε_∞ is the permittivity constant for high frequency and we used $\varepsilon_\infty = 3$ [1 – 3].



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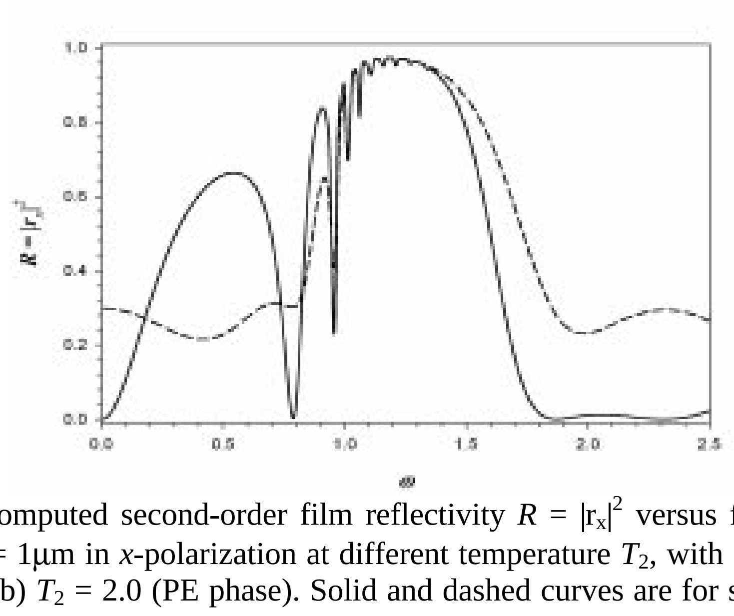

Figure 2: Computed second-order film reflectivity $R = |r_x|^2$ versus frequency ω for thickness $L = 1\mu m$ in x-polarization at different temperature T_2, with (a) $T_2 = 0.1$ (FE phase), and (b) $T_2 = 2.0$ (PE phase). Solid and dashed curves are for symmetrical and asymmetrical (with Si substrate, $n_3 \approx 3.42$) FE films respectively. Second-order FE parameters are $\alpha_2 = 10^{-3}$, $\beta_2 = 5$, $\gamma_2 = 0.01$, and $\varepsilon_\infty = 3.0$.

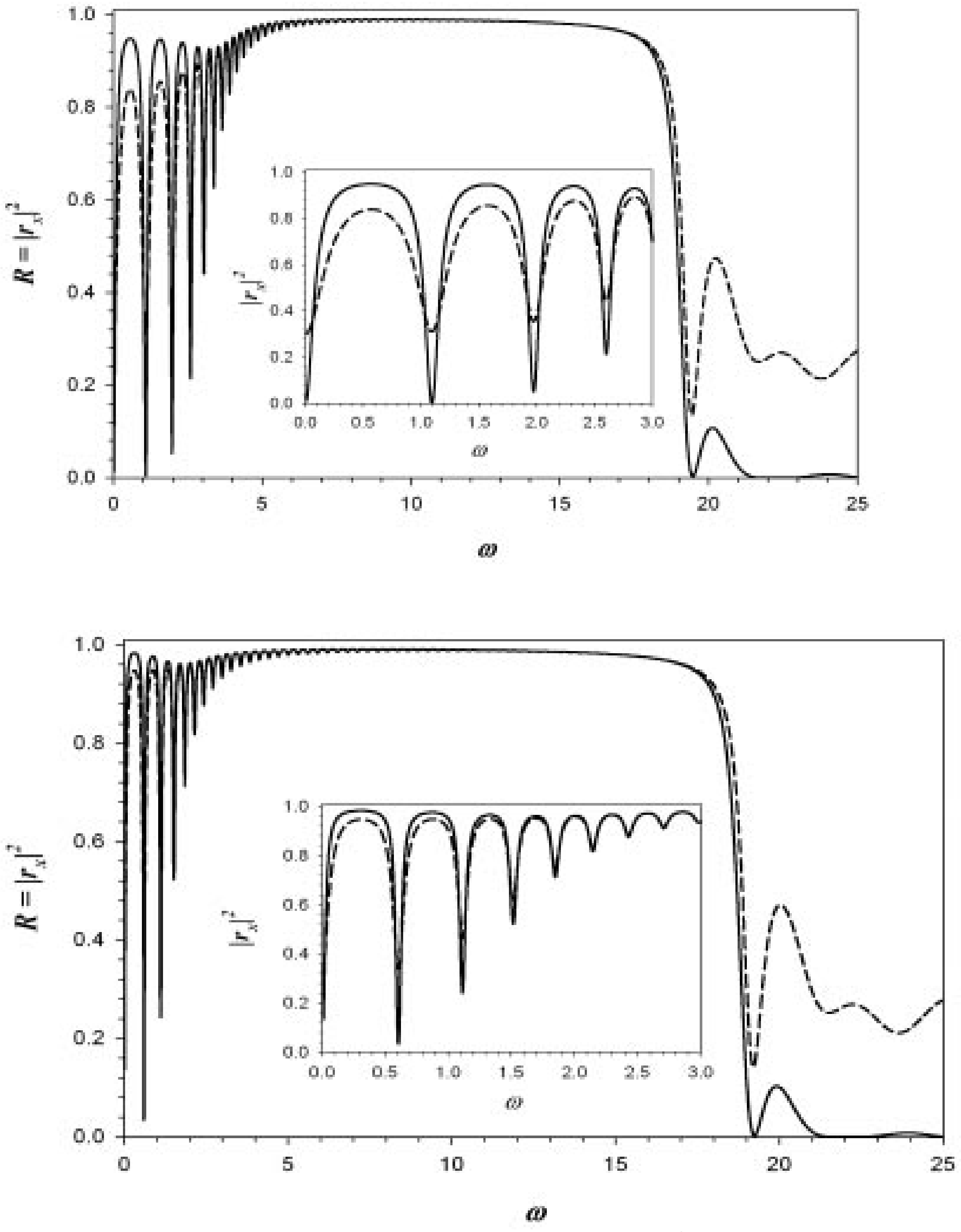

Figure 3: Computed first-order film reflectivity R = $|r_x|^2$ versus frequency ω for thickness L = 1μm in x-polarization at different temperature T_1, with (a) T_1 = -2.0 (FE phase), and (b) T_1 = 3.0 (PE phase). Solid and dashed curves are for symmetrical and asymmetrical (with Si substrate, $n_3 \approx 3.42$) FE films respectively. First-order FE parameters are $\alpha_1 = 10^{-3}$, $\beta_1 = 10^3$, $\gamma_1 = 0.1$, and $\varepsilon_\infty = 3.0$.

By considering the propagation of E and q at three different areas: $h > +L/2$, $-L/2 < h < +L/2$ and $h < -L/2$, thus, it produced six simultaneous equations [1 – 3] with six unknowns which are the four coefficients of the up and down waves in the thin film together with the complex reflection r and transmission s amplitudes. From six simultaneous equations, the preliminary study of asymmetrical FE thin film may differ from symmetrical case by continuity of H field govern by Maxwell's equation at interface of FE-Si, $h = -L/2$. This equation may be written as

$$k_1(a_1\psi_1 - b_1\psi_1^*) + k_2(a_2\psi_2 - b_2\psi_2^*) = \omega s n_3 \psi_3 \qquad (5)$$

where for symmetrical case, $n_3 = 1$ (air), while here is the substrate refractive index. The dimensionless term, $\psi_i = \exp(ik_iL)$, $\psi_i^* = \exp(-ik_iL)$, and $\psi_3 = \exp(i\omega n_3 L)$ with $i = 1$ or 2. Another five [1–3] simultaneous linear equations with (5) are numerically solved using LU Decomposition method [15] to generate the intensity of reflection coefficient $R = |r|^2$.

Results And Discussion

The main feature of both transitions reflectivity curve in Fig. 1 and 2 is the existence of fringes at lower-edge restsrahlung region caused by the destructive and constructive interference from different tendency of mode propagated in the film. Fringes only appeared in the low-frequency region due to impedance matching interpenetration wavelength, film thickness and also the spin mode wave that proliferated from D in the free-energy expansion. Fig. 1 and 2 show the x polarize-reflection curves for symmetrical (solid) and asymmetrical (dashed) FE thin film at thickness $L = 1\mu$m for second and first-order phase transitions respectively. For both transitions of FE, we stated that the behaviour of temperature is increased, where for second-order case: (a) at pure FE phase ($T_2 < T_0$; $T_0 = 1$) with $T_2 = 0.1$, and (b) at pure paraelectric (PE) phase with $T_2 = 2.0$ (polarization, $p = 0$), while for first-order case: (a) at pure FE phase ($T_1 < T_C$; $T_C = 0.75$) with $T_1 = -1.0$ (below supercooling temperature), and (b) at pure PE phase with $T_1 = 3.0$. For first-order phase transition, where $T_1 < T_C$, the FE materials are in different types of structure based on temperature; rhombohedral, tetragonal and monoclinic. At $T_1 > T_C$, the crystals are in cubic phase[4]. The comparison of R spectra at different temperatures is important for FE phase and PE phase in term of optical characteristics of FE thin films. Generally, we have a wide restsrahl region when temperature is increased. Means that FIR waves transmit are decreased.

Basically, our preliminary results of asymmetrical FE thin film incorporating with refractive index influence without surface-effect show that the presence of Si substrate changes the R at different regions: increases R at $\omega \rightarrow 0$ and upper-edge and decreases R at lower-edge of restsrahlung region respectively. It is clearly shown that a FIR wave propagates in films differently at lower-edge and upper-edge, while at restsrahlung region remains the same. It may came from interfaces problems called mismatch effect at bottom of film ($h = -L/2$) between FE-Si interface. Since there are major different between symmetrical and asymmetrical FE films for second and first-order transitions, thus it gives a strong theoretical evidence that reflectivity spectra could be used as an optical probe to investigated mismatch effect between FE-Si. Theoretically, mismatch effect may occur at the FE phase based on the different types of structure of FE as mentioned above. Fig. 1(b) and 2(b) indicated the non-mismatch effect conditions since both materials (FE-Si) are in cubic type-structure. More in-depth theoretical study as well as experimental work is needed before the phenomenological theory of spectroscopy that we proposed here can be used to study the asymmetrical FE film case. We are currently study this behaviour for first-order FE film using the same model by considering the surface-effect in term of asymmetric-surface polarization profile [16] and hope to publish the results soon.

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