Phonon polaritons in a bilayer ferroelectrics structure with a polarization coupling at the interface

V Gunawan, A Subagio, N A K Umiai
Physics Department, Faculty of Sciences and Mathematics, Diponegoro University, Jl. Prof. Soedarto, Tembalang, Semarang, Indonesia
Corresponding author: goenangie@lecturer.undip.ac.id

Abstract. In this paper, we discuss the optical properties of a bilayer ferroelectrics structure comprised of two different types of ferroelectric materials. The interface interaction between these two ferroelectric materials is considered. This interaction requires the entire-cell effective medium treatment where each layer of ferroelectric is sliced into several latticed. Then, by applying Landau-Khalatnikov (LKh) equation of motion in each lattice and solving it simultaneously, the dynamic susceptibility of the bilayer system can be determined. By employing Maxwell equations, we derive the dispersion relation, which can be solved numerically using the root-finding technique. Since here we use two ferroelectric with different resonant frequencies, then the polaritons can also be found around these two frequencies. Hence, there are two leading bulk bands in the dispersion relation. The existence of polarization coupling between two ferroelectrics at the interface generates the narrow additional bulk bands in the dispersion relation. We found that these additional bulk bands are affected by the strength of interface coupling and the number of lattices in each involved layer. We also found that the increase of the thickness of the layers weaken the additional bulk bands.

1. Introduction
Phonon polaritons are basically electromagnetic waves which are modified by lattice vibrations in polarized materials such as polar dielectrics or ferroelectrics. This type of polaritons draws many attentions due to its properties, such as good containment [1] and a long-distance of propagation [2] with the potential application in data storage [3], nano-optical devices [4], lensing devices [5] and photonics [6]. The studies are also involving various types of materials, such as dielectric SiC [7], hexagonal-Boron nitride h-BN [8], semiconductor [9] and Ferroelectrics [10,11].

In order to obtain better properties of the polaritons, the involved materials are developed from only a single material into multilayer structure which is involving more than one material. Hence, the studies are also involving bilayer [12] using two isotropic materials, multilayer using algebraic manipulation approximation [13] and transfer matrix method [14] and also superlattices structures employing interface response theory [15]. In studying polaritons in the layered systems, the effective medium approximation can also be used where the multilayer structure can be treated as one unity material [16]. The effective susceptibility represents this effective material.

However, when the involved materials are no longer isotropic; for example, electric polarization in ferroelectric films resulted from depolarization and interlattices interaction, then the above methods do not appropriate. One method that can be used to treat anisotropy materials is the entire cell effective medium which is proposed in Ref. [17] to treat the magnetic system involving interlattice exchange interaction. This method are also successfully to study additional magnetization in composite...
multiferroics [18]. Since in this paper, we study the polaritons generated in bilayer ferroelectrics which is comprised of two different ferroelectric materials with the polarization interaction at the surface, then the entire-cell effective medium is employed to obtain the effective susceptibility. This effective susceptibility is then used in the derivation of the polariton’s dispersion relation.

2. Theory

2.1. The effective susceptibility

We consider the geometry of the research, as illustrated in figure 1. The material is a bilayer system structure which is consisted of two different ferroelectrics with interface is represented by the red dotted lines. The surface of the bilayer is placed at the x-z plane with y = 0. The static polarizations which are denoted by the arrows are set parallel to the z-axis.

Figure 1. The geometry of the bilayer structure. The bilayer consists of ferroelectric A, and ferroelectric B with static polarizations are illustrated by the arrows. Here, the polarizations are set in the z-direction. The interface between ferroelectric A and B is set in the y-z plane and denoted by red dotted lines. The surface of the bilayer is arranged in x-z plane at z = 0.

In, order to apply entire cell effective medium approximation, each layer of ferroelectrics are divided into several lattices which are shown by black dotted lines. The electric susceptibility is then used in the derivation of the entire-cell effective medium approximation, each layer of ferroelectrics are divided into several lattices which are shown by black dotted lines. The electric susceptibility is then used in the derivation of the entire-cell effective medium is employed to obtain the effective susceptibility. This effective susceptibility is then used in the derivation of the polariton’s dispersion relation.

\[
\frac{\partial^2 P_i}{\partial t^2} = -f_{ab} \frac{\partial F}{\partial P_i}
\]

where \( f \) represents the inverse of phonon mass. Here \( F \) is energy density of the system which is determined as [19]

\[
F = \sum_i \left[ \frac{1}{2} a_i T_c^a \left( \frac{T}{T_c^a} - 1 \right) (P_i^a)^2 + \frac{1}{4} \beta_a (P_i^a)^4 + \frac{1}{2} K_a (P_{i+1}^a - P_i^a)^2 - E_i^a P_i^a \right] + \sum_i \left[ \frac{1}{2} a_i T_c^b \left( \frac{T}{T_c^b} - 1 \right) (P_i^b)^2 + \frac{1}{4} \beta_b (P_i^b)^4 + \frac{1}{2} K_b (P_{i+1}^b - P_i^b)^2 - E_i^b P_i^b \right] + \frac{1}{2} \alpha (P_i^a - P_i^b)^2
\]

where parameters \( a \) and \( \beta \) are dielectric stiffness constants, \( T_c \) represents Curie temperature and \( K \) is inter lattice interaction constant. The first line in equation (2) above determines the density energy of Ferroelectric A, while the second line explains the density energy of ferroelectric B. The last line represents the energy density from interface interaction between ferroelectric A and B with the interface interaction constant is \( \alpha \). The detail calculation of derivation the susceptibility using entire-cell effective medium can be seen in Ref.[19]. Here, we only review the main steps.

The electric polarization and the electric field consist of static and dynamic parts as \( P = P_s + p \) and \( E = E_s + e \) with the values of static parts are much bigger than the value of the dynamic parts, i.e., \( p \ll P_s \) and \( e \ll E_s \). Here, we assume that the dynamic parts are proportional to \( \exp(i\omega t) \). Then, by
performing linearization of the dynamic terms and also considering the continuation of the tangential component of electric fields across the lattices and interface \( (\varepsilon_1^a = \varepsilon_2^a = \varepsilon_{\text{int}}^a = \varepsilon_{\text{int}}^b = \cdots = \varepsilon_{N-1}^b = \varepsilon_N^b = C_z) \), the LKh equation of motion for the non-interface lattices can be written in the form

\[
\left[ (\omega_0^{a,b})^2 - \omega^2 \right] p_{1}^{a,b} - (\omega_K^{a,b})^2 p_{1+1}^{a,b} - (\omega_K^{a,b})^2 p_{1-1}^{a,b} = f_{a,b} C_z
\]

where \((\omega_0^{a,b})^2 = f_{a,b} \left[ 2a_{a,b}^T \varepsilon_{c}^b \left( 1 + \frac{T}{2T} \right) + 2K_{a,b} \right] \) and \((\omega_K^{a,b})^2 = f_{a,b}K_{a,b} \). For the interface layer, the equation of motion becomes

\[
\left[ (\omega_0^{a,b})^2 - \omega^2 \right] p_{1}^{a,b} - (\omega_K^{a,b})^2 p_{1+1}^{a,b} + (\omega_0^{a,b})^2 p_{1-1}^{a,b} = f_{a,b} C_z
\]

where \((\omega_K^{a,b})^2 = (\omega_0^{a,b})^2 \pm f_{a,b} \alpha \). By setting the value of \( C_z \) and solving the equation of motion in each lattice and in interface simultaneously, the set of \( \{p_i\} \) and \( \{\varepsilon_i\} \) values can be obtained. Then, the electric susceptibility \( \chi_z^e \) is found numerically using relation \( e_z \chi_z^e = p \). The permittivity for the \( z \) component is obtained using relation \( \varepsilon_{zz} = \varepsilon_1 + 4\pi \chi_z^e \).

In this model, the inter-lattices and inter-layers interactions exist only in the \( z \) component. Hence, the susceptibility in \( y \) component can be simply determined as \( \varepsilon_{yy} = \varepsilon_\infty \frac{\omega_1^2 - \omega_\infty^2}{\omega_1^2 - \omega_2^2} \). Here \( \varepsilon_\infty \) is susceptibility in high frequency while \( \omega_1 \) and \( \omega_\infty \) represent the transverse and longitudinal frequency of soft phonon modes.

### 2.2 The bulk polaritons

Consider the electromagnetic waves propagate in the \( y-z \) plane with the magnetic component is perpendicular to the plane of incident. Then, the bulk modes of polaritons can be assumed in the form

\[
\vec{H} = \vec{\varepsilon} H_0 \exp\left( i (\vec{k} \cdot \vec{r} - \omega t) \right).
\]

Using this form or electromagnetic waves into the cross of Maxwell equations, \( \nabla \times \vec{E} = -\frac{1}{c} \frac{\partial \vec{B}}{\partial t} \) and \( \nabla \times \vec{H} = \frac{\varepsilon}{c} \frac{\partial \vec{D}}{\partial t} \), the matrix equation can be obtained as

\[
\begin{pmatrix}
\frac{\omega}{c} & k_z & -k_y \\
k_z & \varepsilon_{yy} \frac{\omega}{c} & 0 \\
k_y & 0 & -\varepsilon_{zz} \frac{\omega}{c}
\end{pmatrix}
\begin{pmatrix}
H_x \\
E_y \\
E_z
\end{pmatrix} = 0.
\]

Matrix equation (6) is solved by setting the determinant of constant matrix to zero. It yields the bulk dispersion relation as

\[
\varepsilon_{yy} k_y^2 + \varepsilon_{zz} k_z^2 = \mu_{xx} \varepsilon_{yy} \varepsilon_{zz} \left( \frac{\omega}{c} \right)^2.
\]

Here, the dispersion relation contains the even order of \( k \), means that the bulk dispersion relation in equation (7) is reciprocal with \( \omega(\vec{k}) = \omega(\vec{-k}) \).

### 3. Results and discussions

In the numerical calculation, we use parameters for ferroelectric A as [20]: \( T_c^a = 391 \text{K} \), the dielectric stiffness constant \( a_a = 6 \times 10^{-6} \text{K}^{-1} \) and the phonon mass \( m = 7.1 \times 10^{-31} \text{g cm}^3 \text{statC}^{-2} \) which represent ferroelectric BaTiO₃. This phonon mass results the inverse phonon mass in Gaussian system as \( f_a = 1.46 \times 10^{30} \text{statC}^2 \text{g}^{-1} \text{cm}^{-3} \), which is equivalent to \( 1.56 \times 10^{9} \text{cm}^{-2} \). Here we set randomly the inter-lattice interaction constant into \( K_a = 1.4 \times 10^{-4} \). Since it is interesting to discuss the condition where the resonance frequency of ferroelectric B close to the resonance frequency of
ferroelectric A, then we situated the parameters for ferroelectric B as: $T^b_c = 450 K$, $a^b = 4.8 \times 10^{-6}K^{-1}$, the inverse of phonon mass $3 \times 10^4$ cm$^{-1}$ and $K^b = 1.1 \times 10^{-4}$. Here, we use the constant of inter-layer interaction as $\alpha = 1 \times 10^{-4}$. For the permittivity in $y$ component, we use the frequencies $\omega^a_T = 2300$ cm$^{-1}$ and $\omega^b_T = 2500$ cm$^{-1}$ for ferroelectric A. In B, we set the frequencies $\omega^b_T = 1600$ cm$^{-1}$ and $\omega^a_T = 1800$ cm$^{-1}$.

The numerical calculation for bulk modes of bilayer ferroelectrics system are illustrated in figure 2. Here, we use two values of interface interaction constant since we do not have information about this constant value. Because it is used two different ferroelectric to create a bilayer structure, then the dispersion relation contains two main resonance frequencies. The first resonance frequency around 96 THz is contributed from ferroelectric A while the second resonance frequency around 68 THz is provided by the ferroelectric B.

![Figure 2](image-url)

**Figure 2.** The bulk dispersion relation for bilayer ferroelectrics. In (a), with interlayer constant $\alpha = 10^{-4}$ while (b) is illustrating bulk dispersion relation with $\alpha = 5 \times 10^{-4}$. It is shown that there are two main resonance frequencies, near 68 THz and 96 THz. Here, we use the number of lattices in each layer is four.

The pattern of bulk modes in figure 2 for each layer (near frequency 68 THz or 96 THz) of ferroelectrics (near frequency 68 THz or 96 THz) is similar to the previous research [20]. It is also shown in figure 2 that the existence of the interface interaction between the electric polarization at the interface A and B result in the additional resonance frequencies. We believe that this appearance is caused by the value of polarization at the interface which is affected by interfacial interaction as discussed in previous report [21]. As we use the weak interface interaction constant, for example: $\alpha = 10^{-4}$, it only results in the existence of small disturbance in the main resonance frequencies as it is illustrated in figure 2a. It happens because when $\alpha$ is small, then the difference between the resonance frequency of the non-interface lattices $\omega^{a,b}_0$ and the resonance frequency of the interface $\omega^{a,b}_r$ is tiny. However, in the more significant value of interface interaction constant, for example: $\alpha = 5 \times 10^{-4}$, the difference between $\omega^{a,b}_0$ and $\omega^{a,b}_r$ is big enough which result in the small additional bulk band near the main resonance frequencies as it is shown in figure 2b. Then, it can be predicted when the interface interaction is strong, and the additional bulk band will become wide.

In this paper, the effect of the number of lattices in each layer to the bulk dispersion relation is also studied. In this step, the bulk modes with the number of lattices in each layer ($N$) is six compared to that with $N = 16$ lattices (see figure 3). The additional of the number of lattices means the increase of the layer thickness. In figure 3a and 3b, the increase of the number of lattices from six to sixteen is weaken the additional bulk bands. It can be seen by the decrease of the “peak” in additional bulk band near 68 THz from around 2 cm$^{-1}$ for $N = 6$ lattices to around 1.2 cm$^{-1}$ for $N = 16$ lattices. There is also
the decrease of the additional bulk band near 96 THz, from around 3.2 cm$^{-1}$ (figure 3a) to 2 cm$^{-1}$ (figure 3b).

![Graphs showing bulk dispersion relation](image)

**Figure 3.** The bilayer’s bulk dispersion relation with two values of the number of lattices in each layer. In (a), the number of lattices in each layer is six lattices while in (b) is sixteen lattices. Here we use $\alpha = 5 \times 10^{-4}$.

The decrease happens because the addition of the number of lattices in each layer increases only the number of lattices which is not affected by the interface interaction while in bilayer structure there is only one interface which is affected by interface interaction. Then, it is clear that the effect of interface interaction to the all medium will become weaker when the thickness of the layers is increased.

4. **Conclusion**

It had shown that dispersion relation in the layered structured could be obtained by employing the entire-cell effective medium approximation. In bilayer structure comprise of two different ferroelectrics, the polarization interaction at the interface generates the additional bulk bands. The appearance of these additional bulk bands are affected by the value of the interface coupling constant where the strong coupling yields the wide additional bulk bands. The number of lattices in each layer also influences the additional bulk bands. It had shown that the increase in the number of lattices weaken the additional bulk bands.

**Acknowledgments**

We highly appreciate Fakultas Sains Matematika UNDIP for funding this study through the scheme of Penelitian Madya with contract No: 1982/UN7.5.8/PP/2020.

**References**

[1] Gubbin C R, Maier S A and De Liberato S 2017 *Phys Rev B* **95** 035313
[2] Ocelic N and Hillenbrand R 2004 *Nat. Mater.* **3** 606
[3] Yoxall E, Schnell M, Nikitin Y, Txoperena O, Woessner A, Lundeberg M B, Casanova F, Hueso L E, Koppens L and Hillenbrand R 2015 *Nat. Photonics* **9** 674
[4] Anderson M S 2003 *Appl.Phys. Lett.* **83** 2964
[5] Huber A J, Deutsch B, Novotny L and Hillenbrand R 2008 *Appl.Phys. Lett.* **92** 203104
[6] Taubner T, Keilmann F and Hillenbrand R 2004 *Nano Lett.* **4** 1669
[7] Gubbin C R and De Liberato S 2020 *Phys. Rev. X* **10** 021027
[8] Cai M, Wang S, Liu Z, Wang Y, Han T and Liu H 2020 *Nanomaterials* **10** 878
[9] Comas F, Trallero-Giner C and Cardona M 1997 *Phys Rev. B* **56** 4115
[10] Bakker H J, Hunsche S and Kurz H 1998 Rev. Mod. Phys. 70 523
[11] Ikegaya Y, Minami Y, Katayama I and Takeda J 2015 Appl. Phys. Lett. 107 062901
[12] Wendler L 1984 Phys Stat. Sol. (b) 125 K27
[13] Lee S C, Shiong Ng S, Hassan H A and Hassan Z 2011 J. Phys. Soc. Jap. 80 084712
[14] Hamilton A A, Dumelow T, Parker T J and Smith S R P 1996 J. Phys.: Condens. Matter 8 8927
[15] Mendialdua J, Rodriguea A, More M, Akjouj A and Dobrzynski L 1994 Phys. Rev. B 50 14605
[16] Raj N and Tilley D R 1987 Phys. Rev. B 36 7003
[17] Stamps R L and Camley R E 1996 Phys. Rev. B 54 15200
[18] Gunawan V and Stamps R L 2012 Phys. Rev. B 85 104411
[19] Gunawan V, Umiati N A K, Subagio A 2019 JPA 1 53
[20] Chew K H, Ong L H, Osman J, Tilley D R 2001 J. Opt. Soc. Am. B 18 1512
[21] Lim K G, Chew K H, Ong L H, Iwata M 2012 EPL 99 46004