The dynamics of polarization and magnetization: Susceptibilities of magnetoelectric multiferroic heterostructure

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Abstract. The dynamic susceptibility is the crucial parameter of a material, especially when the material is needed to be applied in the electromagnetic applications. In this report, we discuss the magnetic and the electric susceptibilities of a magnetoelectric (ME) multiferroic heterostructure comprised of ferromagnetic (FM) and ferroelectric (FE) materials. Since the magnetoelectric couplings in multiferroic heterostructure have only existed at the interface between FM and FE, then the equation of motions in each lattice in every layer of FM and FE should be considered. The equations of motion in each lattice are solved simultaneously by employing entire-cell effective medium approximation. In numerical calculation, the parameters appropriate for Fe and BaTiO$_3$ are used. We found that interfacial ME couplings shift down the resonance frequency for both FM and FE susceptibilities. We also noticed that the addition of the number of FE or FM layer, which raises the number of interface between FE and FM affects the susceptibility curves.

1. Introduction
Magnetoelectric multiferroic is a kind of material that possesses magnetic and electric properties simultaneously[1]. The magnetization and electric polarization as the representation of ferromagnetism and ferroelectricity are interconnected through a magnetoelectric coupling. However, the magnetoelectric coupling in single-phase multiferroics is relatively weak[2]. Then, the artificial magnetoelectric multiferroics composite (heterostructure) comprised of ferromagnet and ferroelectric, which have much stronger magnetoelectric coupling are proposed[3] and extensively studied in the last two decades[4-7]. These multiferroics heterostructure are received enormous attention due to its potential application in memory devices, sensors and electromagnetic devices[7,8].

In the application related to the electromagnetic waves, the determination of the dynamics of the materials is required. The study of susceptibilities in single-phase magnetoelectric BaMnF$_4$ had performed[9,10]. Those studies found that magnetoelectric interaction shift both magnon and phonon resonance frequencies. However, the conventional derivation method in calculating susceptibilities for a single-phase could not be used in determining susceptibility components for a sample with multi-structure geometry. The conventional effective medium approximation in the previous study[11] can only be employed to obtain susceptibility when the parameters in each component are homogeneous. In that study, it should be noticed that there is no interaction between constituent components of multi-layers materials[11].
Magneto-electric multiferroic heterostructure comprises of the series of ferroelectric (FE) and ferromagnetic (FM) components. Since the FM and FE components are in the form of film, then the parameters such as electric polarization in FE and magnetization in FM components are inhomogeneous. Also, the magneto-electric interactions exist only in the interfaces between FE and FM. Hence, the conventional effective medium approximation cannot be applied. Entire-cell effective medium approximation was employed successfully to calculate the susceptibility of antiferromagnetic films or superlattices with the interface exchange interaction was considered [12]. In entire-cell effective medium method, the components of multiferroics heterostructure are sliced into several layers. Then, by applying a suitable equation of motion in each layer and considering the continuity of the fields across every layer, the susceptibility components are obtained by solving those equations of motion simultaneously.

Motivating by the fact that the entire-cell effective medium approximation can describe surface or interface effect, we employ this method to calculate susceptibility components in magnetoelectric multiferroic heterostructure. The effect of interface magnetoelectric interaction is also studied.

2. The Geometry and The Method

The geometry of multiferroic heterostructure in this study is illustrated in Figure 1. The multiferroic is composed of the series of ferromagnet (FM) and ferroelectric (FE) components in the $\hat{y}$ direction. The magneto-electric interaction appears in the FM-FE interfaces. The planes of the interface are placed in the $x$-$z$ plane. The static magnetization of ferromagnet M and the static electric polarization of ferroelectric P are directed parallel to the $\hat{z}$ direction. Here, both the ferroelectric and the magnetic components are sliced into several layers. Red arrows represent the magnetizations in FM layers while blue arrows symbolize the polarization in FE components. The black arrows at the FM-FE interfaces illustrate that polarization and magnetization exist simultaneously at the site.

![Figure 1](image)

Figure 1. The geometry of the multiferroic heterostructure. The FM-FE interfaces are set in the $x$-$z$ plane. The magnetization of ferromagnet and the electric polarization of ferroelectric are arranged in the $z$-direction.

We start by defining the energy density of the system. Here, the energy density of multiferroic heterostructure comprises of the FM, FE and magnetoelectric (ME) energy densities which can be written as [4, 6, 10]

$$F = \sum_n \frac{\alpha_0 T^2}{z} \left( \frac{T}{T_c} - 1 \right) P_n^2 + \frac{\beta}{4} P_n^4 + \frac{\kappa}{2} \left( \frac{\partial P_n}{\partial y} \right)^2 - E \cdot \vec{P}_n + \frac{2A}{M_0^2} \left( \frac{\partial \vec{M}_n}{\partial y} \right)^2 - \frac{\kappa_0}{2} \left( \vec{M}_n \cdot \hat{z} \right)^2 \tag{1}$$

Here, the first four terms are the energy density from ferroelectric with $\alpha_0$ and $\beta$ represent ferroelectric stiffness constants. Here, $\kappa$ is a constant describing interaction of the polarization at a site with the other polarization at the nearest neighbour. The symbol $E$ represents an external electric field. The
fourth to sixth terms of Equation (1) describe the ferromagnet energy density, where \( A \) is an exchange constant and \( K_\alpha \) represents anisotropy constant. Here, parameter \( H_0 \) describes an external magnetic field. The last term of Equation (1) illustrates the magneto-electric interaction with \( \alpha \) determines a magneto-electric coupling which relates the electric polarization of ferroelectric to the magnetization of the ferromagnet. Here, the ME coupling is in bi-quadratic type since it is suitable for most of the crystal symmetry.

The next step is deriving the equations of motion of both the electric polarization and the magnetization. Using Landau-Khalatnikov (LKh) equation of motion for the polarization as[13,14]

\[
\frac{\partial^2 \vec{P}}{\partial t^2} = -f \frac{\partial F}{\partial \vec{P}},
\]

and Landau-Lifshitz (LL) torque equation for magnetization as[15,16]

\[
\frac{\partial \vec{M}}{\partial t} = -\gamma \vec{M} \times \frac{\partial F}{\partial \vec{M}},
\]

where the parameters \( f \) and \( \gamma \) represent the inverse of phonon mass and gyromagnetic ratio. We derive the equations of motion for each involved layers, including the interfaces. We are using magnetization and polarization in the form

\[
\vec{M} = (m_x, m_y, M_z), \quad \text{and} \quad \vec{P} = (p_x, p_y, P_z, p_z).
\]

In Equation (4) above, \( M_z \) and \( P_z \) are the static magnetization and static polarization. The parameters \( m_x \) and \( m_y \) are the dynamic magnetization components while the dynamic of polarization components are represented by \( p_x \), \( p_y \) and \( p_z \). The dynamic parts are much smaller than the static parts, i.e.: \( m_i \ll M_z \) and \( p_i \ll P_z \).

The dynamic equations of motion of both polarization and magnetization in each involved layers are obtained by substituting Equation (4) into Equation (2) and Equation (3) using density energy Equation (1). Next, Maxwell boundary conditions are applied, which are the continuity of tangential dynamic electric field \( \mathcal{E}_x \), continuity of tangential magnetic field \( h_x \) and continuity of normal induced magnetic field \( b_y \) across the layers. These conditions can be written as

\[
h_1^x = h_2^x = \cdots = h_{n-1}^x = h_n^x = C_x,
\]

\[
h_1^y + m_1^y = h_2^y + m_2^y = \cdots = h_{n-1}^y + m_{n-1}^y = h_n^y + m_n^y = C_y,
\]

\[
\mathcal{E}_1 = \mathcal{E}_2 = \cdots = \mathcal{E}_{n-1} = \mathcal{E}_n = C_z.
\]

After dynamic linearization, we get the dynamic equation for the layer \( n \) inside FE components excluding interfaces as

\[
(A_0 \omega_f^2 - \omega^2)p_n - \omega_k^2 p_{n+1} - \omega_k^2 p_{n-1} = \omega_f^2 C_z
\]

while for magnetic layers inside FM components (without interfaces) can be written as

\[
i \omega m_n^y + (\Omega_m + \omega_s)m_n^y - \omega_{ex} m_{n+1}^y - \omega_{ex} m_{n-1}^y = \omega_s C_y
\]
\[ i \omega m_n^y - \Omega_m m_n^x + \omega_{ex} m_{n+1}^x + \omega_{ex} m_{n-1}^x = -\omega_s C_x. \]  \hfill (6c)

Here, \( \omega_f^2 \) is the frequency form of the inverse of phonon mass \( f \). The parameter \( A_0 \) is determined as \( A_0 = 2a_0 T_c \left( 1 - \frac{T}{T_c} \right) + \frac{2\kappa}{d^2} \) with \( d \) is the nearest neighbor layer distance. Frequency \( \omega_f^2 = \omega_f^2 K_1 \) is interaction frequency between the nearest neighbor electric layer in FE components. The frequency \( \Omega_m = \gamma (H_a + H_0 + 2H_{ex}) \) with anisotropy field \( H_a = K_a M_s \), static external field \( H_0 \) and exchange field \( H_{ex} = K_{ex} M_s \), where the exchange constant is defined as \( K_{ex} = 2A/(M_s^2 d^2) \).

At the FE-FM interfaces, the ME interaction had to be considered. Hence, the equation of motion for interface layers become

\[ (A_a \omega_f^2 - \omega^2) p_n - \omega_f^2 p_{n+1} - \omega_f^2 p_{n-1} = \omega_f^2 C_z \]  \hfill (7a)

where \( A_a = A_0 - \alpha M_s^2 \). The magnetic equation of motion turns out to be

\[ i \omega m_n^x + (\Omega_m + \omega_s - \omega_{me}) m_n^y - \omega_{ex} m_{n+1}^y - \omega_{ex} m_{n-1}^y = \omega_s C_y \]  \hfill (7b)

and

\[ i \omega m_n^y - (\Omega_m - \omega_{me}) m_n^x + \omega_{ex} m_{n+1}^x + \omega_{ex} m_{n-1}^x = -\omega_s C_x. \]  \hfill (7c)

where \( \omega_{me} = \gamma \alpha g_a^2 M_s \) is frequency from ME interaction.

When the values of constant \( C_x, C_y \) and \( C_z \) are set at certain values, the polarization \( p_x \), magnetization \( m_x \) and \( m_y \), and also the dynamic fields \( h_x, h_y \) and \( E_z \) are obtained by solving Equation (5). Equation (7) simultaneously. It is required two sets of the solutions to get the susceptibility components. For example, two sets of \( \{ C_x, C_y, C_z \} \) lead to the two sets of solutions (for example: \( \{ m_1^x, m_1^y, p_1^y \} \); \( \{ h_1^x, h_1^y, E_1^z \} \) and \( \{ m_2^x, m_2^y, p_2^y \} \); \( \{ h_2^x, h_2^y, E_2^z \} \)). Using these sets of values, we calculate the susceptibility components. For example, susceptibility components \( \chi_{xx} \) and \( \chi_{xy} \) can be determined using relation

\[ \left( \begin{array}{ll}
 h_1^x & h_1^y \\
 h_1^x & h_1^y
\end{array} \right) \left( \begin{array}{ll}
 \chi_{xx} \\
 \chi_{xy}
\end{array} \right) = \left( \begin{array}{ll}
 m_1^y \\
 m_1^y
\end{array} \right) \]  \hfill (8)

3. Results and Discussion

Figure 2. Susceptibility components of FM-FE bilayer in various ME coupling. (a) Magnetic susceptibility component. (b) The electric susceptibility component. Here, the black lines represent the condition without ME coupling at FM-FE interface. The red, blue and green lines illustrate the condition with the ME coupling values are: \( 3 \times 10^{-11} \) cm\(^3\)/erg, \( 5 \times 10^{-11} \) cm\(^3\)/erg and \( 7 \times 10^{-11} \) cm\(^3\)/erg.

In the numerical calculation, we used parameters as \( B = 1.845 \times 10^7 \text{G}^{-1} \text{S}^{-1} \), \( M = 1707 \text{G} \), \( H_a = 628 \text{G} \) and \( A = 2 \times 10^{-6} \text{erg/cm} \) which appropriate for ferromagnet Fe. For ferroelectric, we used \( P = 4.4 \times 10^4 \text{statC/cm}^2 \), \( T_c = 391 \text{K} \), \( a_0 = 6 \times 10^{-6} \text{K}^{-1} \), \( f = 1.56 \times 10^{20} \text{statC}^2 \text{g/cm}^3 \) and \( K = 1.4115 \times 10^{-9} \) which represent ferroelectric \( \text{BaTiO}_3 \). Here, we set the inter-layer distance as \( d = 10^{-4} \text{cm} \). Since we do not have any information about magnetoelectric coupling \( \alpha \), then we calculate using various values of \( \alpha \).

Firstly, we consider the simplest structure, which is a bilayer comprise of one FM and one FE components with only one FM-FE interface. Here both FM and FE layers are sliced into four lattices. It can be noticed from Figure (2) that entire-cell effective medium approximation is successfully employed to obtain susceptibility components of an FM-FE bilayer with the interface interaction. The magnon frequency is around 20 GHz (see Figure (2a)), while the phonon frequency is near 41 THz in Figure (2b). We also try to understand the effect of ME interaction towards the resonant frequency by applying various values of ME coupling: \( 3 \times 10^{-11} \text{ cm}^3/\text{erg} \), \( 5 \times 10^{-11} \text{ cm}^3/\text{erg} \) and \( 7 \times 10^{-11} \text{ cm}^3/\text{erg} \). Since the ME interaction tends to decrease the resonance frequency as it is implicitly stated in Equation (7a) to (7c), then it shifts down both the magnon and phonon resonance frequency (see Figure 2). The resonance frequency shift down further away when the ME coupling increase.

![Figure 3](image-url)  
Figure 3. Susceptibility components of an FM-FE bilayer in various numbers of layers. (a) Magnetic susceptibility component. (b) The electric component. Here, the black lines represent the condition where FM and FE are sliced into four lattices. The blue and red lines illustrate the condition where FM and FE are sliced into six and eight lattices.

We also study the effect of the number of lattices in the FM and FE components on the susceptibility. The susceptibilities of an FM-FE bilayer with the number of lattices in FM or FE component are set to four lattices, six lattices and eight lattices are illustrated in Figure (3a) for magnetic part and Figure (3b) for the electric component. Here, we set the ME coupling at \( 5 \times 10^{-11} \text{ cm}^3/\text{erg} \). In magnetic component, the increase of the number of lattices decrease the ratio of the lattice with ME interaction to the lattice without ME interaction. For example, The increase of lattices from four to six lattices changes the ratio from 1/4 to 1/6. Hence the effect of interface ME interaction are weakened when the non-interface lattices are increased. It is illustrated by the shifting up of the resonance frequency in Figure (3a). In the electric component, the increase of lattice is responded differently. Instead of frequency shift, the increase of lattice decreases the peak of the electric susceptibility at resonant frequency (see Figure 3b).

Lastly, we develop a bilayer structure into the series of FM-FE multilayer. The results are illustrated in Figure (4). In magnetic component, the resonance frequency is shifted up when the structure is...
developed into FM-FE-FM-FE (two bilayers in series with three ME interfaces interaction) layers. The ratio between the number of lattices with ME interaction to the number of lattices without ME interaction is 1/6 in a bilayer while this ratio becomes 3/10 for double bilayer (FM-FE-FM-FE) structure. Since the effect of the ME increase, it reduces the magnetic resonant frequency. Then, the resonant frequency in Figure (4a) is shifted down. When we develop further into three bilayers or more, we found that the ratio between lattices with ME interaction to the lattices without ME interaction is around 0.3 which is almost similar to the ratio in double bilayers. Then the resonant frequency for the structure with three bilayers or more coincides with the resonant frequency of the structure of the double FM-FE bilayers. In the electric component of susceptibility, the increase of the number of bilayers which is arranged in series will increase the effect of ME interaction. Then, this increase weakens the resonant frequency, which is illustrated by the decreased of the peak of the electric susceptibility around the resonant frequency (see Figure (4b)).

Figure 4. Susceptibility components of FM-FE bilayer in various number of layers. (a) Magnetic susceptibility component. (b) The electric component. Here, the black lines represent the condition for the single FE-FM bilayer. The red lines illustrate the condition for double bilayer (FM-FE-FM-FE) structure.

4. Conclusion
The susceptibilities components for multiferroics heterostructure had successfully derived using entire-cell effective medium approximation. The strength of magnetolectric coupling, the number of lattices in each component and the number of FM-FE interfaces affect the susceptibilities.

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References
[1] Schmid H 1994 Ferroelectrics162 317
[2] Harshe G, Dougherty J P and Newham R E 1993 Int. J. Appl. Electromagn. Mater. 4 145
[3] Lopatina I and Lisnevskaya I 1994 Ferroelectrics161 63
[4] Srinivasan G, Srinivasan E T, Gallegos J, Srinivasan R, Bokhan Y I and Laletin V M 2001 Phys. Rev. B 64 214408
[5] Duan C G, Jaswal S and Tsymbal E 2006 Phys. Rev. Lett.97 047201
[6] Gunawan V and Stamps R L 2012 Phys. Rev. B85 104411
[7] Huang W, Yang S and Li X 2015 J. Materiomics1 263
[8] Spaldin N A and Ramesh R 2019 Nat. Mat. 18 203
[9] Tilley D R and Scott J F 1982 Phys. Rev. B 25 3251
[10] Gunawan V and Stamps R L 2011 J. Phys.: Condens. Matter 23 105901
[11] Raj N and Tilley D R 1987 Phys. Rev. B 36 7003
[12] Stamps R L and Camley R E 1996 Phys. Rev. B 54 15200
[13] Gunawan V and Umiati N A K 2018, Int. J. Elec. Comp. Eng. (IJECE 8 4823
[14] Cui L, Chen C Xiang Y 2019 Phys. Lett. A 383 2963
[15] Yamada T, Fujisaka H and Mori H 1973 Prog. Of Theoretical Physics 49 1062
[16] Ares de Parga G, Mares R and Dominguez S 2005 Annales de la Fondation Louis de Broglie 30 283