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Bonding and Electronic Properties of $BiFeO_3$ Multiferroic Material

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Abstract: We present a framework for modeling the boundary-value problem (frequency driven) for homogeneous multiferroic material bismuth ferrite under the validity of post constraint and apply it to fundamental differential equation of electromagnetism in valid new regimes (boundary conditions for material with simultaneous electric and magnetic order). The only multiferroic material exhibiting the unambiguous magneto-electric effect at room temperature is bismuth ferrite. We illustrated the presence of Tellegen parameter as magneto-electric tensor or magneto-electric quadrupole, which is not related to the position of crystal. We developed the model to enhance the magneto-electric effect of the bismuth ferrite.

Key words: Bismuth ferrite, Tellegen parameter , post constraint, electromagnetism, boundary conditions.

1. Introduction

The discussion of the magneto-electric effect of bismuth ferrite at room temperature is itself interesting as a rich variety of physical mechanism plays an instrumental role in bringing about the effect. The only multiferroic material exhibiting the unambiguous magneto-electric effect at room temperature is bismuth ferrite [1-3]. The density functional theory of single phase BFO has evaluated the small ME (magneto-electric) coupling effect at about ~ 0.067 $mV\cdot cm^{-1}\cdot Oe^{-1}$ at ~ 9.5 kOe [4, 5]. Kimura et al. [6] have demonstrated the enhancement of ME coupling of BFO on RT by destroying the long-range spin cycloid and/or shifting the transition Curie temperature or Neel temperature towards the room temperature. In this paper, we extend to characterize the magneto-electric tensor of magneto-electric effect of bismuh ferrite. We obtain an axial scalar, which was expected to be disappeared in the electrodynamics theory, as per Post. This is referred as the Post constraint. The validity and vindication of Post constraint have always been strenuously discussed.

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2. Boundary Problems at Surface States

We know that the magneto-electric tensor has 36 components.

For linear, anisotropic, non-dispersive medium

$$\mathcal{D}^{\beta} = \varepsilon^{\beta\alpha}E_{\alpha} + \varrho^{\beta\alpha}B_{\alpha} + \varpi B_{\beta\alpha} \quad (1)$$

$$\mathcal{H}^{\beta} = \nu^{\beta\alpha}B_{\alpha} + \varsigma^{\beta\alpha}E_{\alpha} - \varpi E_{\beta\alpha} \quad (2)$$

where $\varepsilon^{\beta\alpha}$ is the dielectric tensor, $\nu^{\beta\alpha}$ is the impermeability tensor; $\varrho^{\beta\alpha}$ and $\varsigma^{\beta\alpha}$ are a magneto-electric tensor.

$$\text{Trace } (\varrho^{\beta\alpha} - \varsigma^{\beta\alpha}) = 0$$

ϖ is the pseudoscalar, the Tellegen parameter. It is antisymmetric and evaluated as

$$\varpi = \frac{\chi^{[\alpha\beta\gamma\delta]}}{\tilde{\epsilon}_{\alpha\beta\gamma\delta}} \quad (3)$$

where in $\tilde{\epsilon}_{\alpha\beta\gamma\delta}$ is Levi-Civita symbol which is 1 or -1 depending on permutation of $\alpha\beta\gamma\delta$ is odd or even.

$$\varpi = \frac{\chi^{\alpha\beta\gamma\delta}}{4!} = \frac{1}{6}(\mathcal{E}_{ij} - \mathcal{M}_{ij}) \quad (4)$$

It shows ϖ is assimilated by the magneto-electric tensor under external electric and magnetic field. It is proposed by Post, that it is the constraint that eventually vanishes in electrodynamics theory and referred as Post constraint.

According to Maxwell's equations:

$$\nabla_{[i}B_{jk]} = 0 \tag{5}$$

$$\nabla_{[i}E_{j]} + \dot{B}_{ij} = 0 \tag{6}$$

$$\nabla_{[i}B_{jk]} = \frac{1}{c^2}\dot{E} - \mu_0({J_f}^j + {J_b}^j) \tag{7}$$

Let us consider a region R having uniaxial anisotropic BFO is separated from the matter free space O by boundary surface. This is illustrated in Fig. 1. The unit vector perpendicular to the surface is $\hat{u}$.

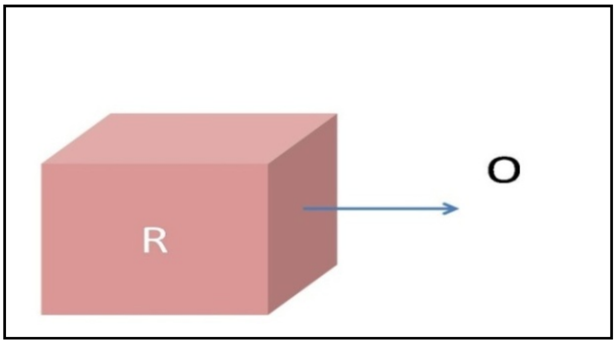

Fig. 1 The analysis of surface states.

We use non relativistic semi-classical electrodynamics of linear media interacting with harmonic planar waves $F = F_0 e^{[i(k\cdot x - \omega t)]}$, where $\boldsymbol{F} = \boldsymbol{B}\ or\ \boldsymbol{E}$. The behavior of the medium R in the frequency domain relations is expressed as

$$\boldsymbol{D}(\boldsymbol{x},\omega) = \overrightarrow{\overrightarrow{\varepsilon(\omega)_{ani}}} \cdot \boldsymbol{E}(\boldsymbol{x},\omega) + \overrightarrow{\overrightarrow{\varrho(\omega)}} \cdot \boldsymbol{B}(\boldsymbol{x},\omega) + \overrightarrow{\overrightarrow{\varpi(\omega)}} \cdot \boldsymbol{B}(\boldsymbol{x},\omega) \tag{8}$$

$$\boldsymbol{H}(\boldsymbol{x},\omega) = \overrightarrow{\overrightarrow{\varsigma(\omega)}} \cdot \boldsymbol{E}(\boldsymbol{x},\omega) + \overrightarrow{\overrightarrow{\nu(\omega)_{ani}}} \cdot \boldsymbol{B}(\boldsymbol{x},\omega) - \overrightarrow{\overrightarrow{\varpi(\omega)}} \cdot \boldsymbol{E}(\boldsymbol{x},\omega) \tag{9}$$

$\overrightarrow{\overrightarrow{\varepsilon(\omega)}}_{ani}$ and $\overrightarrow{\overrightarrow{\nu(\omega)}}_{ani}$ are 3 × 3 permittivity and permeability dyadic with 3 × 3 diagonal matrices.

$\overrightarrow{\overrightarrow{\varsigma(\omega)}}$ and $\overrightarrow{\overrightarrow{\varrho(\omega)}}$ are 3 × 3 magneto-electric dyadic with trace $[\overrightarrow{\overrightarrow{\varrho(\omega)}} - \overrightarrow{\overrightarrow{\varsigma(\omega)}}] = 0$.

The uniaxial anisotropic medium is determined by single axis with unit vector $\boldsymbol{i}$.

$\overrightarrow{\overrightarrow{\varepsilon(\omega)}}_{ani}$ and $\overrightarrow{\overrightarrow{\nu(\omega)}}_{ani}$ are dyadic di-electric tensor and impermeability.

$$\overrightarrow{\overrightarrow{\varepsilon(\omega)}}_{ani} = \varepsilon(\omega)\left(\overrightarrow{\overrightarrow{I}} - \boldsymbol{ii}\right) + \overrightarrow{\overrightarrow{\varepsilon(\omega)}}_{ani}\boldsymbol{ii} \tag{10}$$

$$\overrightarrow{\overrightarrow{\nu(\omega)}}_{ani} = \nu(\omega)\left(\overrightarrow{\overrightarrow{I}} - \boldsymbol{ii}\right) + \overrightarrow{\overrightarrow{\nu(\omega)}}_{ani}\boldsymbol{ii} \tag{11}$$

where $\overrightarrow{\overrightarrow{I}}$ is identity dyad.

The Maxwell equations for the free space O:

$$\nabla \times \boldsymbol{E}(\boldsymbol{x},\omega) - i\omega\boldsymbol{B}(\boldsymbol{x},\omega) = \boldsymbol{0} \tag{12}$$

$$\nabla \cdot \boldsymbol{B}(x,\omega) = 0 \tag{13}$$

$$\nabla \times \boldsymbol{H}(\boldsymbol{x},\omega) + i\omega\boldsymbol{D}(\boldsymbol{x},\omega) = \boldsymbol{J}(\boldsymbol{x},\omega) \tag{14}$$

$$\nabla \cdot \boldsymbol{D}(\boldsymbol{x},\omega) = \rho\boldsymbol{B}(\boldsymbol{x},\omega) \tag{15}$$

The Maxwell equation for regions R by applying equation (1.1) and (1.2) in general Maxwell equations

$$\nabla \times \boldsymbol{E}(\boldsymbol{x},\omega) - i\omega\boldsymbol{B}(\boldsymbol{x},\omega) = \boldsymbol{0} \tag{16}$$

$$\nabla \cdot \boldsymbol{B}(\boldsymbol{x},\omega) = 0 \tag{17}$$

$$\nabla \times [\overrightarrow{\overrightarrow{\varsigma(\omega)}} \cdot \boldsymbol{E}(\boldsymbol{x},\omega) + \overrightarrow{\overrightarrow{\nu(\omega)_{ani}}} \cdot \boldsymbol{B}(\boldsymbol{x},\omega) - \overrightarrow{\overrightarrow{\varpi(\omega)}} \cdot \boldsymbol{E}(\boldsymbol{x},\omega)\ ] + i\omega[\overrightarrow{\overrightarrow{\varepsilon(\omega)_{ani}}} \cdot \boldsymbol{E}(\boldsymbol{x},\omega) + \overrightarrow{\overrightarrow{\varrho(\omega)}} \cdot \boldsymbol{B}(\boldsymbol{x},\omega) + \overrightarrow{\overrightarrow{\varpi(\omega)}} \cdot \boldsymbol{B}(\boldsymbol{x},\omega)] = \boldsymbol{J}(\boldsymbol{x},\omega) \tag{18}$$

$$or\ \ \nabla \times \left[\overrightarrow{\overrightarrow{\varsigma(\omega)}} \cdot \boldsymbol{E}(\boldsymbol{x},\omega) + \overrightarrow{\overrightarrow{\nu(\omega)_{ani}}} \cdot \boldsymbol{B}(\boldsymbol{x},\omega)\right] + i\omega\left[\overrightarrow{\overrightarrow{\varepsilon(\omega)_{ani}}} \cdot \boldsymbol{E}(\boldsymbol{x},\omega) + \overrightarrow{\overrightarrow{\varrho(\omega)}} \cdot \boldsymbol{B}(\boldsymbol{x},\omega)\right] = \boldsymbol{J}(\boldsymbol{x},\omega) \tag{19}$$

$$\nabla \cdot [\overrightarrow{\overrightarrow{\varepsilon(\omega)_{ani}}} \cdot \boldsymbol{E}(\boldsymbol{x},\omega) + \overrightarrow{\overrightarrow{\varrho(\omega)}} \cdot \boldsymbol{B}(\boldsymbol{x},\omega) + \overrightarrow{\overrightarrow{\varpi(\omega)}} \cdot \boldsymbol{B}(\boldsymbol{x},\omega)] = \rho(\boldsymbol{x},\omega) \tag{20}$$

$$or\ \ \nabla \cdot \left[\overrightarrow{\overrightarrow{\varepsilon(\omega)_{ani}}} \cdot \boldsymbol{E}(\boldsymbol{x},\omega) + \overrightarrow{\overrightarrow{\varrho(\omega)}} \cdot \overrightarrow{\boldsymbol{B}(x,\omega)}\right] = \rho(\boldsymbol{x},\omega) \tag{21}$$

The medium is related to dimensional quantity Υ admittance which is pseudoscalar. It is the reciprocal of resistance. $[\varepsilon_0] = \Upsilon/c$ and $[\nu_0] = \Upsilon c$.

The boundary conditions imposed on this free space are

$$\boldsymbol{B}^n(\boldsymbol{x}^+,\omega) = \boldsymbol{B}^n(\boldsymbol{x}^-,\omega)$$

$$\boldsymbol{E}^t(\boldsymbol{x}^+,\omega) = \boldsymbol{E}^t(\boldsymbol{x}^-,\omega)$$
$$\boldsymbol{D}^n(\boldsymbol{x}^+,\omega) = \boldsymbol{D}^n(\boldsymbol{x}^-,\omega)$$
$$\boldsymbol{H}^t(\boldsymbol{x}^+,\omega) = \boldsymbol{H}^t(\boldsymbol{x}^-,\omega) \quad (22)$$

where $\boldsymbol{x}^-$ is for the region R having BFO while $\boldsymbol{x}^+$ is the for the space surrounding the region; n and t represent normal and tangent components, respectively.

Boundary condition on the region with uniaxial anisotropic medium (BFO).

$$\boldsymbol{B}^n(\boldsymbol{x}^+,\omega) = \boldsymbol{B}^n(\boldsymbol{x}^-,\omega)$$
$$\boldsymbol{E}^t(\boldsymbol{x}^+,\omega) = \boldsymbol{E}^t(\boldsymbol{x}^-,\omega)$$
$$\boldsymbol{D}^n(\boldsymbol{x}^+,\omega) = \boldsymbol{D}^n(\boldsymbol{x}^-,\omega) + \varrho\boldsymbol{B}^n(\boldsymbol{x}^-,\omega)$$
$$\boldsymbol{H}^t(\boldsymbol{x}^+,\omega) = \boldsymbol{H}^t(\boldsymbol{x}^-,\omega) + \varsigma\boldsymbol{E}^t(\boldsymbol{x}^-,\omega) \quad (23)$$

In the matter free space O, $\nu_{ani}(\omega)$ is the absolute impermeability $\nu_0(\omega)$, and $\varepsilon_{ani}(\omega)$ is the absolute permittivity $\varepsilon_0(\omega)$. While the magneto-electric tensor (as it is matter related) is absent.

In the region having BFO material R, the $\varepsilon_{ani}(\omega) = \varepsilon(\omega)$, $\nu_{ani}(\omega) = \nu(\omega)$, magneto-electric tensor is present.

Hence the Post constraint is established when the Maxwell equation is applied to linear anisotropic multiferroic (BFO) material.

3. Space-Time Symmetry and Magneto-Electric Tensors

$BiFeO_3$ has asymmetric components of the secondary ferroic magneto-electrics coefficients. Thus, it has a toroidal magnetic moment at high fields with induced point group 3 m, obtained after destruction of the free-field antiferromagnetic phase (which is magnetically incommensurate) by using high magnetic field. The generating matrixes for its crystal

$A_i = \begin{pmatrix} -1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}$ reflecting in *yz* plane.

And $\begin{pmatrix} -1/2 & -\sqrt{3}/2 & 0 \\ \sqrt{3}/2 & -1/2 & 0 \\ 0 & 0 & 1 \end{pmatrix}$ with 3-fold rotation about *z* axis.

The magneto-electric effect in BFO is due to 3 time even electric quadrupole magneto dipole order.

1) The origin independent electric dipole—electric dipole polariazation $\mathfrak{X}_{ij}$,

2) the electric dipole—magnetic dipole polarization $\mathfrak{F}_{ij}$,

and 3) the electric dipole—electric quadrupole polarization $\mathfrak{L}_{ij}$ [7, 8].

The time dependent perturbation theory explains the magneto-electric tensors as the electric and magnetic polarization parameters induced by macroscopic multipole moment densities originate by the assimilation of molecular multipole moment.

As per the crystallographic axes of uniaxial point group 3 m of BFO, there are no permanent electric and magnetic dipoles, rather $\mathfrak{L}_{ij}$ exists.

The bounded source densities of Maxwell equation are given by

$$\rho_b = -\mathfrak{X}_{ij}\nabla_i E^j + \frac{i}{\omega}(\mathcal{E}_{ij} + \sum \mathfrak{L}_{ij})\nabla_j \nabla_i E^k \quad (24)$$

$$j_b = -i\omega\mathfrak{X}_{ij}E^j + (\mathcal{M}_{ij} - \sum \mathfrak{L}_{ij})\nabla_j E^k \quad (25)$$

Hence diamagnetic behaviour is deduced as

$$D_i = (\varepsilon_0\delta_{ij} + \mathfrak{X}_{ij} + \sum \mathfrak{L}_{ij})E^j + \mathcal{E}_{ij}B_i \quad (26)$$

Similarly, magnetic behaviour is expressed as

$$H_i = {\mu_0}^{-1}\delta_{ij}B_i - \mathcal{M}_{ij}E^j \quad (27)$$

Comparing with Eqs. (1) and (2)

$$\varrho^{\beta\alpha} = \mathcal{E}_{ij}.$$

And $\varsigma^{\beta\alpha} = -\mathcal{M}_{ij}$.

While $\mathcal{E}_{ij}$ and $\mathcal{M}_{ij}$ are related to the electric dipole-magnetic dipole polarization $\mathfrak{F}_{ij}$.

$$\varpi = \frac{\chi^{\alpha\beta\gamma\delta}}{4!} = \frac{1}{6}(\mathcal{E}_{ij} - \mathcal{M}_{ij}) = \frac{1}{3}\mathfrak{F}_{ij} \quad (28)$$

$\mathfrak{F}_{ij}$, $\mathfrak{L}_{ij}$ and ϖ are scalars that do not change with displacement from crystallographic origin.

Hitherto, the diagonal matrix for anisotropic uniaxial permeability,

$$\mu_{ani}(\omega) = \begin{bmatrix} \mu_{11} & 0 & 0 \\ 0 & \mu_{11} & 0 \\ 0 & 0 & \mu_{33} \end{bmatrix}.$$

And for permittivity $\varepsilon_{ani}(\omega) = \begin{bmatrix} \varepsilon_{11} & 0 & 0 \\ 0 & \varepsilon_{11} & 0 \\ 0 & 0 & \varepsilon_{33} \end{bmatrix}$.

Both are independent and symmetric, having 6 components each.

Hence for determining 36 tensor components, it is sufficient to find scattered field in 6 directions for 12 different incidences.

4. Polarization in Terms of Magneto Electric Tensor

The antiphase domains of $BiFeO_3$ in the full space group symmetry have 96 orientation states, which can be subdivided into four groups with distinct rhombohedral (polar) axes (each group containing 24 states). For each polar axis, the corresponding easy magnetization axis is three fold degenerate; This leads to eight different orientation states with parallel or antiparallel orientation of magnetization and polarization, respectively, which are relevant for the 180° switching of the magnetization.

Henceforth to analyze the polarization, let us consider the z_0- directed incident waves, then the components of polarizability tensors in x_0y_0 planes.

$$\begin{bmatrix} p_1 \\ p_2 \\ m_1 \\ m_2 \end{bmatrix} = \begin{bmatrix} \mathfrak{X}_{ij} \\ \mathfrak{X}_{ij} \\ \mathfrak{F}_{ij} \\ \mathfrak{F}_{ij} \end{bmatrix} \begin{bmatrix} E_{1i} \\ E_{2i} \end{bmatrix} + \begin{bmatrix} \mathfrak{F}_{ij} \\ \mathfrak{F}_{ij} \\ \mathfrak{Y}_{ij} \\ \mathfrak{Y}_{ij} \end{bmatrix} \begin{bmatrix} H_{1i} \\ H_{2i} \end{bmatrix} \tag{29}$$

Similarly, for the waves incident on x_0 and y_0 axes. $\mathfrak{Y}_{ij}$ is for the magnetic-magnetic dipole polarization. Where $E_i = \frac{\Upsilon}{2H_m} x_0$ and $H_i = \pm H_m y_0$, H_m is the magnetitude of incident magnetic field. By substituting, magneto-electric tensor ϖ, we deduced 16 polarization component for z_0- directed incident waves and rest 20 for other 2 axes.

For + z_0- directed incident waves, the polarization components in terms of ϖ are

$$\varpi^{11}_{ee} = \frac{\Upsilon}{2H_m} (p_1{}^{+} + p_1{}^{-})$$

$$\varpi^{12}_{em} = \frac{1}{2H_m} (p_1{}^{+} - p_1{}^{-})$$

$$\varpi^{21}_{ee} = \frac{\Upsilon}{2H_m} (p_2{}^{+} + p_2{}^{-})$$

$$\varpi^{22}_{em} = \frac{1}{2H_m} (p_2{}^{+} - p_2{}^{-})$$

$$\varpi^{11}_{me} = \frac{\Upsilon}{2H_m} (m_1{}^{+} + m_1{}^{-})$$

$$\varpi^{12}_{mm} = \frac{1}{2H_m} (m_1{}^{+} - m_1{}^{-})$$

$$\varpi^{21}_{me} = \frac{\Upsilon}{2H_m} (m_2{}^{+} + m_2{}^{-})$$

$$\varpi^{22}_{mm} = \frac{1}{2H_m} (m_2{}^{+} - m_2{}^{-}) \tag{30}$$

For – z_0- directed incident waves, the polarization components in terms of ϖ are

$$\varpi^{11}_{ee} = \frac{\Upsilon}{2H_m} (\overline{p_1}^{+} + \overline{p_1}^{-})$$

$$\varpi^{12}_{em} = \frac{1}{2H_m} (\overline{p_1}^{+} - \overline{p_1}^{-})$$

$$\varpi^{21}_{ee} = \frac{\Upsilon}{2H_m} (\overline{p_2}^{+} + \overline{p_2}^{-})$$

$$\varpi^{22}_{em} = \frac{1}{2H_m} (\overline{p_2}^{+} - \overline{p_2}^{-})$$

$$\varpi^{11}_{me} = \frac{\Upsilon}{2H_m} (\overline{m_1}^{+} + \overline{m_1}^{-})$$

$$\varpi^{12}_{mm} = \frac{1}{2H_m} (\overline{m_1}^{+} - \overline{m_1}^{-})$$

$$\varpi^{21}_{me} = \frac{\Upsilon}{2H_m} (\overline{m_2}^{+} + \overline{m_2}^{-})$$

$$\varpi^{22}_{mm} = \frac{1}{2H_m} (\overline{m_2}^{+} - \overline{m_2}^{-}) \tag{31}$$

Similarly we can deduced for induced oscillating electric and magnetic field multipoles radiating in all directions.

Apart from the magneto-electric effect and spontaneous magnetization, the magnetic symmetry of bismuth ferrite also allows the magnetic ordering of special (toroidal) type in BFO.

The toroidal moment is given by

$$\mathbf{T} = [\mathbf{E} \times \mathbf{H}]. \tag{32}$$

Hence its vector components are proportional to antisymmetric linear magneto-electric tensor.

$$\begin{bmatrix} \boldsymbol{T}_1 \\ \boldsymbol{T}_2 \\ \boldsymbol{T}_3 \end{bmatrix} \sim \begin{bmatrix} \varpi^{23} - \varpi^{32} \\ \varpi^{31} - \varpi^{13} \\ \varpi^{12} - \varpi^{21} \end{bmatrix} \tag{33}$$

This results in to inhomogeneous magneto-electric effect in bismuth ferrite.

5. Conclusion

The Tellegen parameter or axionic tensor ϖ is material related. It disappears from mathematical formulation of Maxwell equations and boundary conditions application in multiferroic uniaxial anisotropic material. In the electrodynamic theory the Post constraint is established in case of BFO.

We illustrated the presence of Tellegen parameter as magneto-electric tensor or magneto-electric quadrupole, which is not related to the position of crystal. We developed the model to enhance the magneto-electric effect of the bismuth ferrite. Henceforth we elucidated the polarization components in terms of linear magneto-electric effect BFO that displays linear magneto-electric tensor, toroidal moment and inhomogeneous magneto-electric effect.

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