

NONLINEAR OPTICS OF FERROELECTRICS MATERIALS

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ABSTRACT

Due to the existence of spontaneous polarization below transition temperature T_c , ferroelectrics materials like BaTiO₃ have been known to possess very high linear electric susceptibility. It is expected that these materials also have very high nonlinear susceptibility coefficients for second- and third-order nonlinear processes like second-harmonic generation (SHG) and third-harmonic generation (THG). In principle, they would be able to produce SHG signal for example and have the potential to be used as a frequency doubler at terahertz frequency range. Here, we explore these possibilities. Based on the Landau-Devonshire theory and the Landau-Khalatnikov dynamical equation, we formalize a theory to calculate all the linear and nonlinear electric susceptibility coefficients for bulk ferroelectrics materials by assuming that they have tetragonal symmetry at temperature below T_c . Based on experimentally obtained parameter values for BaTiO₃ we estimate the magnitude of one of the nonlinear susceptibility coefficients for SHG. Finally, using the theory of nonlinear wave propagation we estimate the amount of SHG signal that can be transmitted from a ferroelectric Fabry-Perot resonator.

INTRODUCTION

Ferroelectrics (FE) are advanced materials and have a lot of technological applications. Presently, they are used as memory devices (FDRAM) in cellular phones, smart cards, and computers. PbTiO₃ and BaTiO₃ are examples of ferroelectric perovskite inorganic crystals, the most known and studied ferroelectrics materials. In the last 20 years several preparation routes have been developed for fabrication of FE thin films, single crystals and epilayers. Technological applications of FE materials are possible because they have useful dielectric properties. All FE materials have very high relative dielectric permittivities, for example the relative permittivity for PZT is ~20,000. They undergo structural phase transition at a transition temperature, T_c , where the phase is more ordered below this temperature called the FE phase. In this phase, FE materials have spontaneous polarizations, $+P_s$ or $-P_s$, even in the absence of applied electric field

and the polarization can be switched with the application of electric field. They exhibit very fast rate of switching in the order of nanoseconds.

FE materials are transparent to the far infrared (FIR) or the terahertz (THz) frequencies. It is expected that when intensity of an incident infrared beam is high (in the excess of 10^{12} W/m^2), the response of FE material to this beam becomes nonlinear and the occurrence of nonlinear optical interactions in the material is inherent. Intensive research efforts in studying the nonlinear optical properties of FE materials have been undertaken lately [1-7] because many applications of NL optics have become practical for technological uses and many more have been demonstrated to have the potential in technological applications. Examples of applications are in nonlinear optical switching devices for use in photonics, real-time coherent optical signal processors, as a laser and terahertz radiation source [8-9]. As a consequence, there is a continuing need to find nonlinear (NL) optical materials with large NL coefficients and fast response. FE materials have long been demonstrated to be the candidate materials. It is in this spirit, the works as reported here have been carried out.

The hypothesis is since FE materials have extremely high dielectric permittivities, they should have, according to optical nonlinear principles, high nonlinear dielectric susceptibilities. Based on our earlier work [10,11], we present here the formalism to derive the nonlinear optical susceptibility coefficients for FE material assumed to have tetragonal symmetry in the FE phase, $T < T_C$. We focus only on the second-order nonlinear optical interactions in particular second-harmonic generation (SHG) which is schematically illustrated by Fig. 1. One main application of this effect is as a frequency doubler. If a FE material is used say in a Fabry-Perot resonator, it may be capable of producing waves at twice the frequency of the input waves. We use the results of our calculation in finding the nonlinear optical susceptibility coefficients for SHG to investigate this possibility.

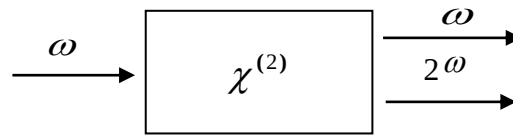


Figure 1: Schematic diagram representing second-harmonic Generation (SHG). $\chi^{(2)}$ represents the response function for SHG.

In section 2, we present the formalism on the derivation of nonlinear susceptibility coefficients, tabulate expressions for all nonvanishing tensor elements for SHG, and numerically estimate the value of SHG coefficient. In Section 3, we focus on the SHG reflection and transmission from a Fabry-Perot cavity and numerically calculate the SHG transmissivity for BTO. Finally, some conclusions are given in the final section. Consider a bulk ferroelectrics material with tetragonal symmetry in the FE phase at temperature $T < T_C$. For example for BaTiO_3 , the structural phase transition into this symmetry group happens at 0°C . The presence of spontaneous polarization, P_0 , in the

tetragonal-symmetry FE phase, along the tetragonal axis (the z-axis here) leads to $P_x = Q_x, P_y = Q_y, P_z = P_0 + Q_z$. The high-temperature free energy expression for the tetragonal symmetry is

$$\begin{aligned} f = f_0 + \frac{\alpha}{2\varepsilon_0}(Q_x^2 + Q_y^2 + Q_z^2) + \frac{\beta_1}{4\varepsilon_0^2}(Q_x^4 + Q_y^4 + Q_z^4) + \\ \frac{\beta_2}{2\varepsilon_0^2}(Q_x^2 Q_y^2 + Q_y^2 Q_z^2 + Q_z^2 Q_x^2) + \frac{\alpha}{2\varepsilon_0}(P_0^2 + 2Q_z P_0) + \\ \frac{\beta_1}{4\varepsilon_0^2}(6Q_z^2 P_0^2 + 4Q_z^3 P_0 + 4Q_z P_0^3 + P_0^4) + \frac{\beta_2}{2\varepsilon_0^2}[(2Q_z P_0 + P_0^2)(Q_x^2 + Q_y^2)] \end{aligned} \quad (1)$$

where cyclic permutations and the usual summation convention are employed in (1). $\alpha = a(T - T_0)$ with a as the inverse of the Curie constant. For second-order phase transition, $T_c = T_0$, the bulk critical temperature. Based on the published data for BaTiO₃ [12], $|\alpha| \sim 5.22 \times 10^{-4} \text{ deg}^{-1}$ at room temperature [13]; β_1 and β_2 are material-dependent parameters, with estimated magnitudes of $1.913 \times 10^{-13} \text{ m}^3 \text{ J}^{-1}$ and $5.74 \times 10^{-13} \text{ m}^3 \text{ J}^{-1}$ respectively [14]. Magnitude of P_0 is given by the condition for minimum f , $\partial f / \partial P_z = 0$ evaluated at $P_z = P_0$, which is

$$\alpha + (\beta_1 / \varepsilon_0) P_0^2 = 0 \quad (2)$$

Under a high intensity incident FIR radiation with a single frequency ω , the free energy becomes $F_E = F - \mathbf{E} \cdot \mathbf{P}$, where $\mathbf{E}$ is an applied field due to the incident FIR radiation. Under this condition, the Landau-Khalatnikov (LK) equation of motion for P is

$$\hat{O}P_i = -\partial F / \partial P_i + E_i = f_i(\mathbf{P}) + E_i \quad (3)$$

where the operator $\hat{O}$ is $\hat{O} = m(d^2/dt^2) + \gamma(d/dt)$ for systems with oscillatory dynamics. Equation (3) defines the function $f_i(\mathbf{P})$ and is the dynamical equation from which the NLO coefficients are derived. The NLO coefficients are the coefficients of the Taylor series for P as a function of E , which may be written as

$$Q_i = Q_i^{(1)} + Q_i^{(2)} + Q_i^{(3)} = \varepsilon_0 \chi_{il}^{(1)} E_l + \varepsilon_0 \chi_{ilm}^{(2)} E_l E_m + \varepsilon_0 \chi_{ilmn}^{(3)} E_l E_m E_n \quad (4)$$

$Q_i^{(n)}$ is the nth-order polarization induced by $E_i(t) = (1/2)[E_{oi} \exp(-i\omega t) + E_{oi}^* \exp(i\omega t)]$ written in terms of complex amplitudes E_{oi} . We define $Q_i^{(n)}$ as

$$\left(Q_i^{(n)} \right)_{\omega_\sigma} = \varepsilon_0 K(-\omega_\sigma; \omega_1, \dots, \omega_n) \chi_{i\alpha_1 \alpha_2 \dots \alpha_n}^{(n)}(-\omega_\sigma; \omega_1, \dots, \omega_n) (E_{\alpha_1})_{\omega_1} (E_{\alpha_2})_{\omega_2} \dots (E_{\alpha_n})_{\omega_n} \quad (5)$$

where $\omega_\sigma = \omega_1 + \omega_2 + \dots + \omega_n$ [14,15]. The definition given by (5) leads to $\chi^{(n)}$ being invariant under all permutations of $(\alpha_1\omega_1), (\alpha_2\omega_2), \dots, (\alpha_n\omega_n)$ called the intrinsic permutation symmetry [15]. The factor K in (5), defined explicitly in Ref. 14 and 15, is a product of a combinatorial factor arising from the symmetrization of $\chi^{(n)}$. In order to find the NLO coefficients, we expand $f_i(P)$ in Taylor series by obtaining the derivatives of free energy, f in (1), with respect to polarization evaluated at P_0 . Expansion of $f_i(P)$ about P_0 gives

$$f_i(P) = f_i(P_0) + f_{il}(P_0)Q_l + \frac{1}{2} f_{ilm}(P_0)Q_l Q_m + \frac{1}{6} f_{ilmn}(P_0)Q_l Q_m Q_n \quad (6)$$

where the relevant derivatives are detailed[14]. Substituting (6) into (3) and collecting terms of the same order in E , we obtain the linear response as

$$[O - f_{il}(P_0)\delta_{il}]Q_l^{(1)} = E_l \quad (7)$$

where the derivatives explicitly are

$$f_{xx} = f_{yy} = -\left(\frac{\alpha}{\varepsilon_0} + \frac{\beta_2}{\varepsilon_0^2} P_0^2\right); \quad \text{and} \quad f_{zz} = -\frac{2\beta_1}{\varepsilon_0^2} P_0^2 \quad (8)$$

Using (7) and (8), and writing $Q_i^{(1)}$ as $Q_i^{(1)} = (1/2) \left[B_{ii}(\omega) E_{oi} \exp(-i\omega t) + B_{ii}^*(\omega) E_{oi}^* \exp(i\omega t) \right]$ we find the linear susceptibility coefficients as

$$\chi_{xx}^{(1)}(\omega) = \chi_{yy}^{(1)}(\omega) \equiv \sigma(\omega) / \varepsilon_0; \quad \chi_{zz}^{(1)}(\omega) \equiv s(\omega) / \varepsilon_0 \quad (9a)$$

where

$$\sigma(\omega) = 1/\left[\Theta(\omega) + (\alpha / \varepsilon_0) + (\beta_2 / \varepsilon_0^2) P_0^2 \right]; \quad s(\omega) = 1/\left[\Theta(\omega) - 2(\alpha / \varepsilon_0) \right] \quad (9b)$$

with $\Theta(\omega) = -i\gamma\omega - m\omega^2$. The second-order term in E is

$$\hat{O}Q_i^{(2)} = f_{il}(P_0)Q_l^{(2)} + \frac{1}{2} f_{ilm}(P_0)Q_l^{(1)}Q_m^{(1)} \quad (10)$$

Substitution of the expression for $Q_i^{(1)}$ yields

$$\begin{aligned} \left[\hat{O} - f_{il}(P_0)\delta_{il} \right] Q_i^{(2)} = & \frac{1}{2} f_{ilm}(P_0) \frac{1}{2} [B_{ll}(\omega)E_{ol} \exp(-i\omega t) + B_{ll}^*(\omega)E_{ol}^* \exp(i\omega t)] \\ & \times \frac{1}{2} [B_{mm}(\omega)E_{om} \exp(-i\omega t) + B_{mm}^*(\omega)E_{om}^* \exp(i\omega t)] \end{aligned} \quad (11)$$

From (11), the coefficients for SHG are determined. Table 1 gives the results of the explicit evaluation for the tensor elements of SHG. For SHG, $K=1/2$ as defined in (5) and there are 3 independent elements and a total of 7 nonvanishing tensor elements, which are in agreement with Ref [16] and the elements are symmetric on exchange of the indices lm .

As an example of numerical estimate based on the derivation just presented, a nonresonant SHG value of $\chi_{zzz}^{(2)}(-2\omega; \omega, \omega)$ is calculated. Using the experimental values of BaTiO₃ [12], the soft-mode frequency is $\omega_T \approx 3 \times 10^{12}$ Hz, and using the estimated values β_1 and β_2 [14], we obtain the magnitude of $\chi_{zzz}^{(2)}(-2\omega; \omega, \omega) \approx 1.36 \times 10^{-11}$ mV⁻¹ at frequency 4.75×10^{13} Hz and temperature 273° K for $\gamma = 3.32 \times 10^{-5}$. These values are reasonably acceptable. To compare the magnitude, the experimental value obtained by Shen [17] is $(1.32 \pm 1.00) \times 10^{-11}$ mV⁻¹ at the input frequency of 2.835×10^{14} Hz and room temperature. This is also comparable to the value calculated by Razak et al. [18].

Table 1: The non-vanishing NL susceptibility tensor elements for SHG

SHG, $K=1/2$	Tensor elements, $\chi_{ilm}^{(2)}$
$\chi_{ilm}^{(2)}(-2\omega; \omega, \omega)$	$\chi_{zzz}^{(2)} = -3\beta_1 P_0 s(2\omega) s^2(\omega) / \epsilon_0^3$
Symmetric on exchange of (l, m)	$\chi_{zyy}^{(2)} = \chi_{zxx}^{(2)} = -\beta_2 P_0 s(2\omega) \sigma^2(\omega) / \epsilon_0^3$
	$\chi_{xzx}^{(2)} = \chi_{yzy}^{(2)} = \chi_{xxz}^{(2)} = \chi_{yyz}^{(2)} = -\beta_2 P_0 \sigma(2\omega) \sigma(\omega) s(\omega) / \epsilon_0^3$

Consider a thin FE film of thickness L . SHG waves are assumed generated in the film with the input waves as the driving fields, considered undepleted in the weak nonlinearity regime. Here, we assume that the FE medium is anisotropic in its tetragonal phase ($T < T_C$) and has nonzero nonlinear susceptibility tensor element for SHG, $\chi_{zzz}^{SHG}(-2\omega; \omega, \omega)$. In order to generate the SHG we therefore take the input waves polarized in the z direction at normal incident.

The geometry of the system is illustrated in Fig. 1. In principle under the undepleted input field approximation, the problem involves: (1) a linear optics of reflection and transmission problem from the film involving amplitudes aE_0 , bE_0 , rE_0 and tE_0 representing down, up, reflected and transmitted waves respectively. (2) A SHG reflection and transmission problem from the film involving amplitudes $\phi_1 E_0$, $\phi_2 E_0$, ρE_0 and τE_0 where they are the down, up, reflected and transmitted waves of the generated SHG waves respectively. The coefficients a , b , r , t , ϕ_1 , ϕ_2 , ρ and τ each

refers to a fraction of an input field with an assumed amplitude E_0 . The film medium is represented by the number 2, the number 1 is the medium above and 3 below, which in general may be air above and a substrate below.

The solutions of a , b , r and t are standard and can be found in any standard textbook on optics [19]. For completeness and clarity, the results are summarized below, in terms of the Landau-Liftshitz notations τ_{lm} and ρ_{lm} .

$$a = \tau_{12} / \Sigma, \quad b = \tau_{12} \rho_{23} (\delta_2)^2 / \Sigma \quad (12)$$

$$r = \left[\tau_{21} \tau_{12} \rho_{21} - (\delta_2)^2 \rho_{12} \rho_{23} - 1 \right] / \rho_{21} \Sigma, \quad t = [\tau_{23} \tau_{12} \delta_2] / \delta_3 \Sigma \quad (13)$$

where $\Sigma = 1 + (\delta_2)^2 \rho_{12} \rho_{23}$, $\delta_l = \exp(ik_l L)$, $k_l = (\omega/c) \sqrt{\varepsilon_l}$, $\rho_{lm} = (k_l - k_m - \gamma) / (k_l + k_m + \gamma)$ and $\tau_{lm} = 2k_l / (k_l + k_m + \gamma)$ with $l, m = 1, 2$, or 3 . γ represents a mirror term to include the possibility that the film may be coated with a dielectric mirror layer on both sides. ε_1 , ε_2 and ε_3 is the relative permittivity of medium 1, 2 and 3 respectively.

Assuming an input frequency of ω , the E fields in each medium are

$$\begin{aligned} \text{Medium 1: } E_z^{(1)}(y, t) &= E_0 \left[\exp(ik_1^\omega y) + r \exp(-ik_1^\omega y) \right] \exp(-i\omega t) \\ &+ \rho E_0 \exp(-ik_1^{2\omega} y) \exp(-i2\omega t) \end{aligned}$$

$$\text{Medium 2: } E_z^{(2)}(y, t) = (1/2) \left[\begin{aligned} &a E_0 \exp(ik_2^\omega y) \exp(-i\omega t) + a^* E_0^* \exp(-ik_2^\omega y) \exp(i\omega t) \\ &+ b E_0 \exp(-ik_2^\omega y) \exp(-i\omega t) + b^* E_0^* \exp(ik_2^\omega y) \exp(i\omega t) \end{aligned} \right]$$

$$\text{Medium 3: } E_z^{(3)}(y, t) = t E_0 \exp(ik_3^\omega y) \exp(-i\omega t) + \tau E_0 \exp(ik_3^{2\omega} y) \exp(-i2\omega t)$$

In the above equations, $k_1^\omega = (\omega/c) \sqrt{\varepsilon_1^\omega}$ and $k_1^{2\omega} = (2\omega/c) \sqrt{\varepsilon_1^{2\omega}}$ is the propagation constant of the input field and the reflected SHG field respectively. $k_3^\omega = (\omega/c) \sqrt{\varepsilon_3^\omega}$ and $k_3^{2\omega} = (2\omega/c) \sqrt{\varepsilon_3^{2\omega}}$ are the propagation constant of the transmitted input and SHG field respectively. In general, the relative permittivity of medium 1, 2 or 3 is dispersive. Consequently, ε_1^ω and $\varepsilon_1^{2\omega}$ for example, denote the relative permittivity of medium 1 with each evaluated at frequency ω and 2ω respectively; and in general $\varepsilon_1^\omega \neq \varepsilon_1^{2\omega}$ and $\varepsilon_3^\omega \neq \varepsilon_3^{2\omega}$. The respective H field in each medium is obtained from the Maxwell's equations.

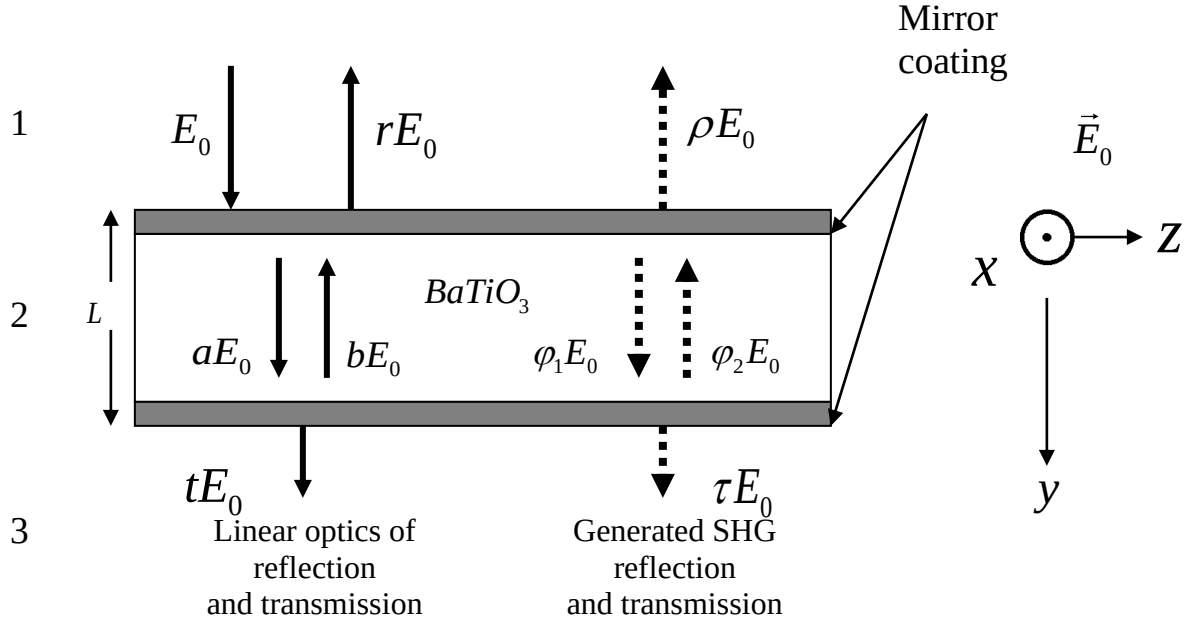


Figure 2: Geometry of the problem showing the cross section of the FE film considered to be BaTiO_3 with thickness L . The input field with amplitude $\vec{E}_0$ is assumed z polarized. In general, the number 1, 2, and 3 represent medium: 1 is the medium above the film, 2 the film medium, and 3 the medium below the film.

In order to solve for ρ and τ , we consider a nonlinear wave equation for the generated SHG wave propagating in the film medium, $\nabla^2 \vec{E} - \mu_0 \partial^2 \vec{D} / \partial t^2 = 0$, where $\vec{D} = \epsilon_0 \vec{E} + \vec{P}^L + \vec{P}^{NL}$ with $\vec{P}^L$ and $\vec{P}^{NL}$ denoting linear and nonlinear polarization induced by the input fields. For z -polarized input field, we have $P_z^L = \epsilon_0 \chi_{zz}^{(1)} E_z^{2\omega}$, and $P_z^{NL} = \epsilon_0 \chi_{zzz}^{SHG} E_z^{(1)} E_z^{(1)}$. $\chi_{zz}^{(1)}$ is the linear and χ_{zzz}^{SHG} the nonlinear susceptibility for SHG respectively. We let the generated SHG field be in the form

$$E_z^{2\omega}(y, t) = (1/2) [\zeta(y) E_0 \exp(-i2\omega t) + \zeta^*(y) E_0^* \exp(i2\omega t)] \quad (14)$$

Substituting into the nonlinear wave equation yields

$$\frac{d^2 \zeta}{dy^2} + 4 \frac{\omega^2}{c^2} \epsilon_{zz}^{2\omega} \zeta(y) + 2 \frac{\omega^2}{c^2} E_0 \chi_{zzz}^{SHG} (a^2 e^{i2k_2^\omega y} + b^2 e^{-i2k_2^\omega y} + 2ab) = 0 \quad (15)$$

where in (15), we use $\varepsilon_{zz}^{2\omega} = 1 + \chi_{zz}^{2\omega}$. $\chi_{zz}^{2\omega}$ and thus $\varepsilon_{zz}^{2\omega}$ denotes the value of $\chi_{zz}^{(1)}$ evaluated at frequency 2ω . Note that $(k_2^{2\omega})^2 = 4\omega^2 / c^2 \varepsilon_{zz}^{2\omega}$ and we consider the case $k_2^{2\omega} \neq 2k_2^\omega$. Using the standard method of undetermined coefficients, we obtain

$$\zeta(y) = \varphi_1 e^{ik_2^{2\omega} y} + \varphi_2 e^{-ik_2^{2\omega} y} + \sigma_{AB}^2 + \sigma_B^2 e^{-i2k_2^\omega y} + \sigma_A^2 e^{i2k_2^\omega y} \quad (16)$$

where

$$\sigma_{AB}^2 = (-2 \frac{\omega^2}{c^2} E_0 \chi_{zzz}^{SHG} ab) (k_2^{2\omega})^{-2} \quad (17a)$$

$$\sigma_A^2 = (-\frac{\omega^2}{c^2} E_0 \chi_{zzz}^{SHG} a^2) ((k_2^{2\omega})^2 - 4(k_2^\omega)^2)^{-1}. \quad (17b)$$

$$\sigma_B^2 = (-\frac{\omega^2}{c^2} E_0 \chi_{zzz}^{SHG} b^2) ((k_2^{2\omega})^2 - 4(k_2^\omega)^2)^{-1}. \quad (17c)$$

These are known parameters with values obtained using (12) and (13). φ_1 and φ_2 are coefficients of the complementary solutions of (16). From (16), all the generated SHG fields within the film may be obtained. Using the fields in each medium and the fields obtained from (16), application of standard electromagnetic boundary conditions at $y=0$ and L gives the following reflection and transmission coefficients for the generated SHG waves:

$$\rho = \frac{(k_2^{2\omega} s_1 + s_2) + 2k_2^{2\omega} \varphi_1}{-(k_1^{2\omega} - k_2^{2\omega})} \quad (18)$$

$$\tau = \frac{(k_2^{2\omega} s_3 + s_4) - 2k_2^{2\omega} \delta_2^{2\omega} \varphi_1}{-(k_1^{2\omega} + k_2^{2\omega}) \delta_1^{2\omega}} \quad (19)$$

where

$$\varphi_1 = \frac{-(\delta_2^{2\omega})^* (k_2^{2\omega} + k_1^{2\omega})(k_1^{2\omega} s_1 + s_2) + (k_1^{2\omega} - k_2^{2\omega})(s_4 - k_1^{2\omega} s_3)}{(\delta_2^{2\omega})^* (k_1^{2\omega} + k_2^{2\omega})^2 - \delta_2^{2\omega} (k_1^{2\omega} - k_2^{2\omega})^2} \quad (20a)$$

$$\varphi_2 = \frac{-(k_1^{2\omega} - k_2^{2\omega})(s_4 - k_1^{2\omega} s_3) - \delta_2^{2\omega} (k_2^{2\omega} - k_1^{2\omega})(k_1^{2\omega} s_1 + s_2)}{(\delta_2^{2\omega})^* (k_1^{2\omega} + k_2^{2\omega})^2 - \delta_2^{2\omega} (k_1^{2\omega} - k_2^{2\omega})^2} \quad (20b)$$

and

$$s_1 = \sigma_{AB}^2 + \sigma_A^2 + \sigma_B^2 \quad (21a)$$

$$s_2 = -2\sigma_B^2 k_2^\omega + 2\sigma_A^2 k_2^\omega \quad (21b)$$

$$s_3 = -\sigma_{AB}^2 - \sigma_A^2 \delta_2^\omega - \sigma_B^2 (\delta_2^\omega)^* \quad (21c)$$

$$s_4 = 2[-\sigma_A^2 \delta_2^\omega + \sigma_B^2 (\delta_2^\omega)^*] k_2^\omega \quad (21d)$$

The above equations contain the following phase factors: $\delta_2^{2\omega} = \exp(ik_2^{2\omega}L)$, $\delta_1^{2\omega} = \exp(ik_1^{2\omega}L)$, and $\delta_2^\omega = \exp(i2k_2^\omega L)$.

RESULTS AND DISCUSSION

The generation of SHG signal from the FE film may be understood by first studying the behavior of $\chi_{zz}^{(1)}$ and χ_{zzz}^{SHG} , with expressions given by (9a) and in Table 1 respectively. We calculate their values based on experimental data of Ref. [12], for BaTiO₃ which are as follows: $m \sim 6.44 \times 10^{-21} \text{ kg m}^3 \text{ s}^{-2} \text{ A}^{-2}$, $\eta \sim 3.32 \times 10^{-5} \text{ kg m}^3 \text{ s}^{-1} \text{ A}^{-2}$, and $P_0 \sim 0.1945 \text{ C m}^{-2}$. Figure 3 and 4 illustrate the behavior of real and imaginary parts of $\chi_{zz}^{(1)}$ and χ_{zzz}^{SHG} at temperature $T=393\text{K}$ versus frequency respectively. As expected for a FE material like BaTiO₃, both $\text{Re}(\chi_{zz}^{(1)})$ and $\text{Re}(\chi_{zzz}^{SHG})$ have very high values, for example at resonance, $\text{Re}(\chi_{zz}^{(1)}) \approx 250$ and the value of $\text{Re}(\chi_{zzz}^{SHG})$ is in the range 10^{-6} mV^{-1} . In comparison, a typical nonlinear optical material like LiNbO₃ has a value of $\text{Re}(\chi_{zz}^{(1)}) < 10$ and $\text{Re}(\chi_{zzz}^{SHG}) \approx 10^{-11} \text{ mV}^{-1}$ [17]. The resonant frequency of $\chi_{zz}^{(1)}$ at $T=393\text{K}$ is calculated to be around $\omega_0 \cong 5 \times 10^{12} \text{ Hz}$ and χ_{zzz}^{SHG} is resonant when the denominator of its expression is zero. Note that the curves are rounded at resonances due to the large damping parameter, which is typical of FE materials, used in the numerical calculation.

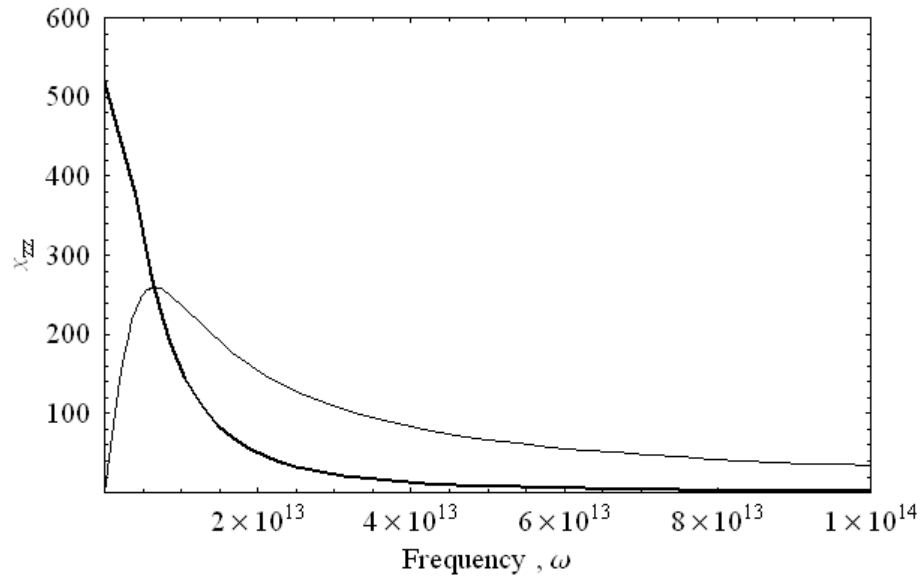


Figure 3: The graph of $\text{Re}(\chi_{zz})$ and $\text{Im}(\chi_{zz})$ versus frequency, ω , at temperature $T=393\text{K}$ using parameters for BaTiO₃. (Thicker line: $\text{Re}(\chi_{zz})$. Thinner line: $\text{Im}(\chi_{zz})$).

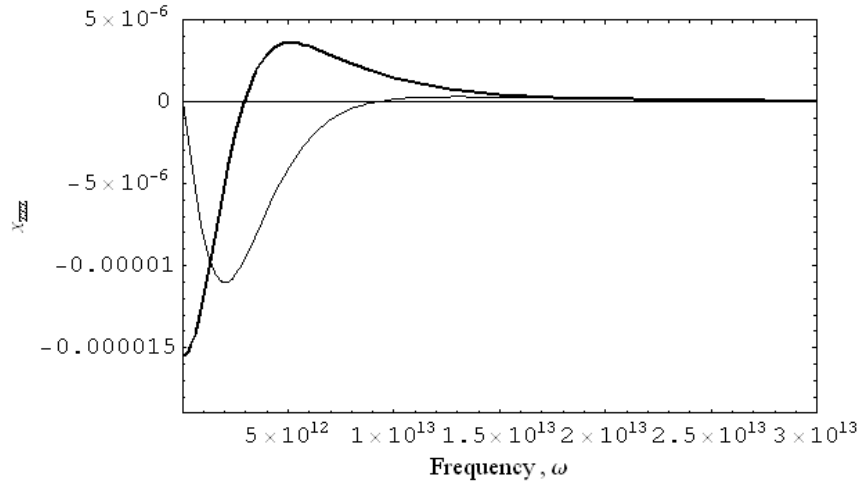


Figure 4: The graph of $\text{Re}(\chi_{zzz})$ and $\text{Im}(\chi_{zzz})$ versus frequency, ω , at temperature $T=393\text{K}$ using parameters for BaTiO_3 . (Thicker line: $\text{Re}(\chi_{zzz})$. Thinner line: $\text{Im}(\chi_{zzz})$).

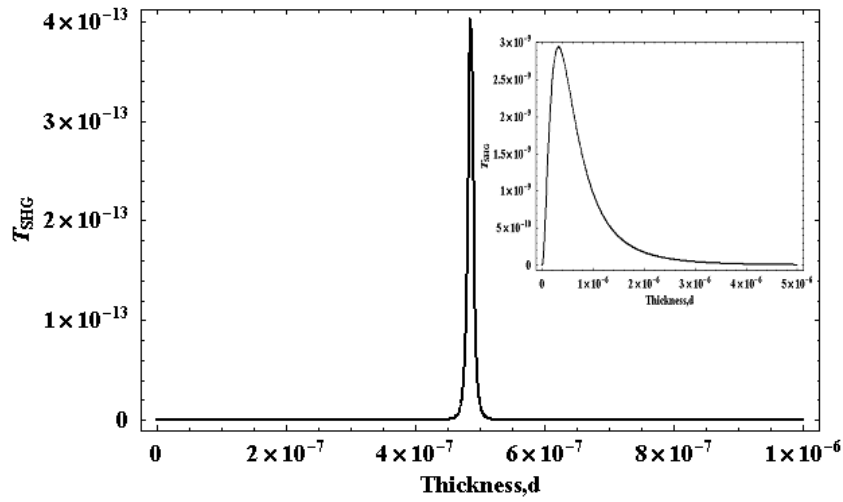


Figure 5: Generated SHG transmission signal, $T_{SHG}=|t_{SHG}|^2$, versus film thickness for FE resonator system with 95% mirror reflectivity. Inset: The T_{SHG} curve for a film without mirror.

Here we consider an input frequency $\omega_1=1.5 \times 10^{13}$ Hz where as shown by Figure 2, it is away from resonance, and consider the generation of SHG signal at frequency

$2\omega_1=3.0\times 10^{13}$ Hz. Figure 4 shows the transmission curve of the generated SHG versus film thickness for film without mirror (inset) and with 95% mirror reflectivity. There exists a high peak demonstrating appreciable SHG transmittance at a particular film thickness for film with mirror. As expected the transmissivity is much lower in magnitude for the case with mirror since here most of the input waves are reflected back into medium 1. The film thickness appears to be in the submicron range. At this film thickness, it is expected that there exists an almost complete constructive interference between the input waves propagating in the film at frequency $\omega_1=1.5\times 10^{13}$ with the generated SHG waves propagating at frequency $2\omega_1$ [20]. The value of T_{SHG} generated is small as expected since the formalism is based on weak nonlinearity (undepletion of the input waves) and phase mismatch condition. However, with the current availability of FIR sources and the use of photon multiplier, it should be detectable.

CONCLUSION

We have presented the formalism to derive the linear and nonlinear susceptibility coefficients for bulk FE material with tetragonal symmetry and have analyzed SHG wave transmission through a FE film, based on a standard non-linear optics approach. We have given a full account on the behavior of SHG wave transmission for a film with and without partial mirror. In this work, we have used our derived expressions of $\chi_{zz}^{(1)}$ and χ_{zzz}^{SHG} , and have concentrated only on z polarized input waves, and have specifically used BaTiO₃ parameters in the numerical calculation. The work can be further extended to include the study of generated SHG signals using other nonvanishing SHG susceptibility tensor components, other symmetry like rhombohedral [14], and other type of FE material for example PbTiO₃. We have shown that even though FE material has high damping but it is capable of generating an appreciable SHG signal and it can be generated at a submicron film thickness.

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