A Possible Room Temperature Terrahertz Wave Source Via Harmonic Generation in a Microwave Pumped Ferroelectric Crystal

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***Abstract*— This paper explores theoretically a model for harmonic generation in ferroelectric crystals which, being resonant in the far infrared will also respond to microwave frequencies. The essential idea is that frequency doubling or tripling of microwave radiation incident on the ferroelectric brought about by second or third harmonic generation will produce waves in the terahertz region. The strong response and nonlinearities of ferroelectric crystals in their interaction with microwaves of frequencies of in the range 200 to 300 GHz makes this a viable proposal.**

Landau-Devonshire theory has been used to calculate nonlinear susceptibility coefficients. Here susceptibility coefficients for second and third harmonic generation are given and it is shown how the slowly varying amplitude (SVA) approximation together with a susceptibility coefficient to solve the wave equation that describes the propagation of the generated waves in the crystal. Conditions on the length through which the waves travel that optimize the phase matching are a useful result of this analysis.

***Index Terms*— microwaves, terahertz wave source, ferroelectrics, Landau theory, nonlinear suseptibilities**

I. INTRODUCTION

The Landau-Devonshire expansion of the free energy F in terms of the electric polarization vector $\mathbf{P}$ may be used to give a phenomenological description of the properties of ferroelectric materials. From this theory formulae for the static dielectric constant and the nonlinear dielectric response have been obtained [1], [2]. Using the Landau-Khalatnikov (LK) dynamical equations Ishibashi and Orihara [3] have extended the theory to give expressions for the nonlinear dynamic dielectric response, that is, the nonlinear susceptibility coefficients, for third order nonlinearities in the paraelectric phase above the Curie temperature T_c. Subsequently calculations of the susceptibilities have been extended to the ferroelectric phase for a variety of cases [4], [5], [6]

In this paper the we give an outline of the basic formalism required to calculate nonlinear susceptibility coefficients and then give specific forms of the coefficients for second and third harmonic generation. We stick to a relatively simple symmetry which gives useable results without too much complication since our main aim is to demonstrate the principle that terahertz waves can be produced from a microwave input via the harmonic generation. Since there is much interest now in terahertz waves this is believed to be a timely contribution to the field.

Finally through the example of third harmonic generation, we show how, having obtained harmonic generation susceptibility coefficients, it is possible to study the wave propagation of the generated waves by solving the Maxwell wave equation under the SVA approximation [7], [8] in which the amplitude of the waves varies slowly relative to their wavelengths. It will be shown that a phase mismatch factor occurs that has minima at certain values of the propagation length L. This information is very useful for designing the correct length for a device such as a Fabry-Perot etalon that might be used to generate the harmonics since the smaller the phase mismatch the greater is the generated wave intensity.

II. FORMALISM

We consider a bulk ferroelectric for which the free energy per unit volume F is written as an expansion of the elastic Gibbs function in terms of polarization $\mathbf{P} = (P_x, P_y, P_z)$

$$F = \frac{1}{2}\frac{A}{\epsilon_0}P^2 + \frac{1}{4}\frac{B}{\epsilon_0^2}P^4 + \frac{1}{6}\frac{C}{\epsilon_0^3}P^6 - \mathbf{P}\cdot\mathbf{E}, \qquad (1)$$

where $P^2 = P_x^2 + P_y^2 P_z^2$, $A = a(T - T_0)$ and $\mathbf{E}$ is the electric field due to incident microwave radiation. For first order transitions the constant coefficients satisfy $B < 0$ and $C > 0$ and the transition from the paraelectric to ferroelectric phase is a discontinuous jump occurring at a temperature $T_0 \leqslant T_c$ [2]. A second order transition, in contrast, occurs continuously and can described with $B > 0$ and $C = 0$. The factors $\epsilon_0$are included so that A, B and C have mechanical dimensions, with a the inverse of the Curie constant. The spontaneous polarization that exists in the ferroelectric state in the absence of an external field $\mathbf{P}_0$ can be found from the free energy since it is the minimum of F with respect to P (with $E = 0$) which can easily be found from $\partial F/\partial P = 0$; in doing this we assume for simplicity that P_0 is aligned along the positive z direction so that $\mathbf{P}_0 = (0, 0, P_0)$.

With regard to the polarization vector F is written formally as an expansion of the invariants of the paraelectric phase and consequently the crystal symmetry of ferroelectric crystal (in its paraelectric phase) is reflected in the symmetry of the free energy with respect to the polarization. The expression in (1) belongs to the isotropic point group symmetry ∞, and as such does not correspond to an actual crystal. This is often used however as an approximation to the actual crystal and in fact for the simplest transition of a typical perovskite ferroelectric such as $PbTiO_4$ or $BaTiO_4$ from its cubic paraelectric phase to a tetragonal ferroelectric phase (1) has appropriate symmetry

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as is brought out by Strukov and Lenanyuk [9]. A more comprehensive description of such perovskites would include extra terms because $P_x^4+P_y^4+P_z^4$ and $P_x^2P_y^2+P_y^2P_z^2+P_z^2P_x^2$ are separately invariant for cubic crystals. For second order transitions susceptibility coefficients using a cubic free energy expression have been derived by Murgan et al. [6]. However for the purposes of this paper we stick with the simpler case described by (1) as it is enough to demonstrate the basic principles of harmonic generation discussed here.

The dynamic response of the ferroelectric to the incident radiation is modelled using the LK equations of motion

$$\hat{O}P_i = -\partial F/\partial P_i, \quad i = x, y, z, \tag{2}$$

where

$$\hat{O} = m\frac{\partial^2}{\partial t^2} + \gamma\frac{\partial}{\partial t} \tag{3}$$

for oscillatory dynamics and relaxational dynamics can be handled by setting the inertial parameter m to zero leaving only the damping parameter γ. The susceptibility coefficients are derived by inverting (3) to find a response function that is linear in $\mathbf{P}$ and nonlinear in $\mathbf{E}$. This, as is described in detail elsewhere [5], [6], is done by expanding $\mathbf{P}$ as a Taylor series in powers of $\mathbf{E}$ with the susceptibility coefficients, which are tensors, being the coefficients. This may be compactly written in component form for the first three orders as

$$\begin{aligned} P_i &= P_i^{(1)} + P_i^{(2)} + P_i^{(3)} \\ &= \epsilon_0\chi_{il}^{(1)}E_l + \epsilon_0\chi_{ilm}^{(2)}E_lE_m + \epsilon_0\chi_{ilmn}^{(3)}E_lE_mE_n, \end{aligned} \tag{4}$$

where P_i^n is the nth-order polarization induced by the incident field $E_i(t)$. Here we consider only a single frequency sinusoidal incident field but a more complicated field which may be represented by a Fourier sum [5], [7] can readily be dealt with within the formalism. The single frequency field is written in terms of complex amplitudes following the convention in Butcher and Cotter [7] as follows

$$E_i(t) = \frac{1}{2}\left[E_{oi}\exp(-i\omega t) + E_{oi}^*\exp(-\omega t)\right]. \tag{5}$$

The corresponding response function, again following Butcher and Cotter [7], is defined by

$$\left(P_i^{(n)}\right)_{\omega_\sigma} = \epsilon_0 K(-\omega_\sigma;\omega_1,\ldots,\omega_n) \tag{6}$$

$$\times\, \chi_{i\alpha_1\alpha_2\cdots\alpha_n}^{(n)}(-\omega_\sigma;\omega_1,\ldots,\omega_n) \tag{7}$$

$$\times \left(E_{\sigma_1}\right)_{\omega_1}\left(E_{\sigma_2}\right)_{\omega_2}\cdots\left(E_{\sigma_n}\right)_{\omega_n}, \tag{8}$$

where $\omega_\sigma = \omega_1 + \omega_2 + \cdots + \omega_n$ [5], [7]. The factor K is a product of combinatorial factors and is defined in Ref. [7]. For the purposes of this paper it is sufficient to state that $K = 1/2$ and $1/4$ for second and third harmonic susceptibilities respectively.

III. Nonlinear Susceptibility Coefficients for Second and Third Harmonic Generation

On the basis of the theory outlined above expressions for second and third order susceptibility coefficients have been derived [5] and are given next for the ferroelectric phase.

The second harmonic susceptibility tensors $\chi^{(2)ilm}(-2\omega;\omega,\omega)$ are symmetric on interchange of l and m and $K = 1/2$. Explicitly the nonvanishing ones are:

$$\left.\begin{aligned} \chi_{xxz} &= \chi_{yyz} = \chi_{xzx} = \chi_{yzy} \\ &= (1/2)g_x\sigma(2\omega)\sigma(\omega)s(\omega) \\ \chi_{zxx} &= \chi_{zyy} = (1/2)g_x s(2\omega)\sigma^2(\omega) \\ \chi_{zzz} &= (1/2)g_z s(2\omega)s^2(\omega) \end{aligned}\right\} \begin{array}{c}\text{second-harmonic}\\ \text{generation}\end{array} \tag{9}$$

The third-harmonic susceptibilities $\chi^{(3)ilmn}(-3\omega;\omega,\omega,\omega)$ are symmetric on permutation of (lmn) and have $K = 1/4$. The nonvanishing ones are:

$$\left.\begin{aligned} \chi_{xxxx} &= \chi_{yyyy} \\ &= (1/2)\sigma(3\omega)\sigma^3(\omega) \\ &\quad\times\left[g_x^2 s(2\omega) + q\right] \\ \chi_{xxyy} &= \chi_{yyxx} = (1/3)\chi_{xxxx} \\ \chi_{xxzz} &= \chi_{yyzz} \\ &= (1/3)\sigma(3\omega)\sigma(\omega)s^2(\omega) \\ &\quad\times\left[(1/2)g_xg_zs(2\omega) + (1/2)p\right] \\ \chi_{zzxx} &= \chi_{zzyy} \\ &= (1/3)s(3\omega)s(\omega) \\ &\quad\times\left[g_x^2\sigma(2\omega) \right.\\ &\qquad + (1/2)g_xg_zs(2\omega) \\ &\qquad \left. + (1/2)p\right] \\ \chi_{zzzz} &= (1/2)s(3\omega)s^3(\omega)\left[g_z^2 s(2\omega) + p\right] \end{aligned}\right\} \begin{array}{c}\text{third-harmonic}\\ \text{generation}\end{array} \tag{10}$$

The functions used in the above expressions are:

$$\sigma(\omega) = -\frac{1}{\epsilon_0(m\omega^2 + i\omega\gamma)} \tag{11}$$

$$s(\omega) = -\frac{1}{-\epsilon_0(m\omega^2 + i\omega\gamma + f_z)} \tag{12}$$

$$f_z = -2AP_0^2\left(\frac{B}{\epsilon_0^2} + \frac{2C}{\epsilon_0^3}P_0^2\right) \tag{13}$$

$$g_x = -2P_0\left(B + \frac{2C}{\epsilon_0}P_0^2\right) \tag{14}$$

$$g_z = -6P_0B - 20\frac{2C}{\epsilon_0}P_0^3 \tag{15}$$

$$p = -2\left(\epsilon_0 B + 10CP_0^2\right) \tag{16}$$

$$q = -3\left(\epsilon_0 B + 2CP_0^2\right). \tag{17}$$

For relaxational dynamics ($m = 0$) ϵ_0 must be replaced by $-\epsilon_0$ in (12).

As an illustration the real part of the χ_{zzz} susceptibility coefficient for second harmonic generation has been plotted in Fig. 1.

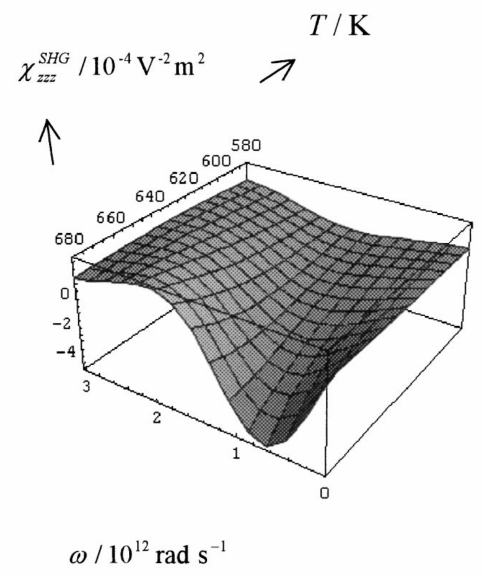

Fig. 1. Frequency and temperature dependence of the real part of the second harmonic generation susceptibility coefficient $\chi^{(2)}_{zzz}$ for second order phase transitions. The following values have been taken from published data [10], [11] for $PbTiO_4$: $a = 2.47 \times 10^{-6}$ K^{-1}, $B = 1.27 \times 10^{-13}$ J^{-1}m^3, $T_c = 765$ K, $m = 5.15 \times 10^{-18}$ JmA^{-2} and $\gamma = 2.43 \times 10^{-5}$ WmA^{-2}.

IV. Harmonic Generation of Terahertz Waves

Consider a single frequency microwave source of about 200 GHz frequency doubling or through second harmonic generation or tripling from third harmonic generation will push the generated frequency into the terahertz wave range. This section shows how the wave equation can be solved to elucidate the propagation of generated waves in ferroelectric crystal.

The example of third harmonic generation (THG) will be taken with no depletion of the pump beam (the microwave incident radiation). Suppose the incident beam is propagating with a a single frequency ω' in the y direction with the spontaneous polarization aligned along the z axis, consistent with the formalism above. With these assumptions the driving field is in general is in the x-z plane. The relevant wave equation to be solved for the THG field is [7], [8]

$$\frac{\partial E_i^{3\omega'}}{\partial y^2} = \mu_0 \frac{\partial^2}{\partial t^2}\left[\epsilon_0 E_i^{3\omega'} + P_i^L + P_i^{NL}\right], \quad i = x, z, \quad (18)$$

in which P^L and P^{NL} are the linear and nonlinear contributions, respectively, to polarization in the bulk ferroelectric, and $E_i^{3\omega'}$ is the ith-component-generated field propagating at frequency $3\omega'$. For the z component, for example, this field is

$$E_z^{3\omega'} = \frac{1}{2}\left[\mathrm{E}_i^{3\omega'}(y)\exp(ik_{3z}y)\exp(-i3\omega' t) + \text{c.c.}\right], \quad (19)$$

where $k_{2z} = \epsilon_{3z}(3\omega')^2/c^2$ is the wave number of THG beam with ϵ_{3z} at frequency $3\omega'$ evaluated using a similar method to that in Ref. [6], and c.c. stands for complex conjugate. From this together with the nonvanishing coefficients in (10) and the linear coefficients $\chi^{(1)}_{xx}$ and $\chi^{(1)}_{zz}$ (which are straightforward to calculate as is shown in Ref. [5]), we have

$$\mathbf{P}^L = \epsilon_0\left[\chi^{(1)}_{xx}(-3\omega;\omega)E_x^{3\omega'}\hat{\mathbf{x}} + \chi^{(1)}_{zz}(-3\omega;\omega)E_z^{3\omega'}\hat{\mathbf{z}}\right], \quad (20)$$

for linear polarization and

$$\begin{aligned}\mathbf{P}^{NL} = \epsilon_0\Big\{ & \big[\chi^{(3)}_{xxxx}(-3\omega;\omega,\omega,\omega)E_x^{\omega'}E_x^{\omega'}E_x^{\omega'} \\ & + \chi^{(3)}_{xxzz}(-3\omega;\omega,\omega,\omega)E_x^{\omega'}E_z^{\omega'}E_z^{\omega'} \\ & + \chi^{(3)}_{xzxz}(-3\omega;\omega,\omega,\omega)E_z^{\omega'}E_x^{\omega'}E_z^{\omega'} \\ & + \chi^{(3)}_{xzzx}(-3\omega;\omega,\omega,\omega)E_z^{\omega'}E_z^{\omega'}E_x^{\omega'}\big]\hat{\mathbf{x}} \\ & + \big[\chi^{(3)}_{zxxz}(-3\omega;\omega,\omega,\omega)E_x^{\omega'}E_x^{\omega'}E_z^{\omega'} \\ & + \chi^{(3)}_{zxzx}(-3\omega;\omega,\omega,\omega)E_x^{\omega'}E_z^{\omega'}E_x^{\omega'} \\ & + \chi^{(3)}_{zzxx}(-3\omega;\omega,\omega,\omega)E_z^{\omega'}E_x^{\omega'}E_x^{\omega'} \\ & + \chi^{(3)}_{zzzz}(-3\omega;\omega,\omega,\omega)E_z^{\omega'}E_z^{\omega'}E_z^{\omega'}\big]\hat{\mathbf{z}}.\Big\}\end{aligned} \quad (21)$$

for the nonlinear polarization, where $E_i^{\omega'}$ is the ith component of the driving field and for the x component it is

$$E_i^{\omega'} = \frac{1}{2}\left[\mathrm{E}_i^{\omega'}\exp(ik_{1x}y)\exp(-i\omega' t) + \text{c.c}\right], \quad (22)$$

where $\mathrm{E}_i^{\omega'}$ is a constant. (20) and (21) show the possible driving field polarization configurations. For simplicity, we concentrate on the case where the driving field is purely x polarized. This leads to the generation of $\mathbf{P}^{NL}$ and thus the THG beam in the x direction as depicted by (21). Based on the slowly varying amplitude approximation [7], [8] in which the envelope $\mathrm{E}_\mathrm{i}^{3\omega'}$ is assumed to have slow spatial variation compared with the exponential in (19) , (18) reduces to

$$\frac{d\mathrm{E}_x^{3\omega'}}{dy} = \frac{9i\omega'^2}{8k_{3x}c^2}\chi^{(3)}_{xxxx}(-3\omega;\omega,\omega,\omega)(\mathrm{E}_x^{\omega'})^2\exp(i\Delta k_x y), \quad (23)$$

in which $\Delta k_x = 3k_{1x} - k_{3x}$. The THG terahertz field generated over a distance L can be determined by integrating (23) over y to give

$$\left|\mathrm{E}_x^{3\omega'}(L)\right|^2 = \frac{81\omega'^4}{64\,|k_{3z}|^2 c^4}(\mathrm{E}_x^{\omega'})^4\left|\chi^{(3)}_{xxxx}\right|^2\frac{\sin(\Delta k_x L/2)}{\Delta k_x L/2}. \quad (24)$$

From this it is seen that the condition for a maximum in the THG (optimum phase matching) is $\Delta k_x L = n\pi$, $n = 1, 3, 5, \ldots$. The coherence length is defined as $l_c = \pi/\Delta k_x$.

V. Conclusion

Using Landau-Devonshire theory it is possible to calculate nonlinear susceptibility coefficients for second and third harmonic generation as has been illustrated. Terahertz waves can be generated from these from a microwave input. Calculations to substantiate this based on the wave equation have been made leading to conditions on the propagation length that give maximum values for the generated wave amplitude. Therefore, even if the generated wave had a tendency to be weak, working with an optimum length would enhance it. This is important in designing devices based on ferroelectric crystals (for example

a Fabry-Perot etalon) that would enable the terahertz wave generation discussed in this paper to be put in to practice. Further support for the feasibility of the method discussed here is apparent from a recent paper by Inoue et al. [12] in which, among other things, it is demonstrated that terahertz waves can propagate through a ferroelectric crystal. Also microwave transmission has been observed in ferroelectric liquid crystals [13] and propagation of microwaves in bulk and thin-film ferroelectrics has been discussed by Tagantsev et al. [14]. Of particular relevance to the work here is that Tagenstev et al. [14] show that single crystal samples of the ferroelectric $SrTiO_3$ exhibit a low loss factor (below 10^{-3}) at microwave frequencies and go on to show that ferroelectric $KaTaO_3$ crystals have very similar properties.

ACKNOWLEDGEMENT

The author wishes to thank Swinburne University of Technology Sarawak Campus for financial support and Dr Manas Kumar Haldar for useful discussions.

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