Creating a Terahertz Wave Source Through Nonlinear Difference Frequency Generation in Ferroelectric Crystals

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Abstract— **This paper proposes that terahertz waves may be generated from higher frequency sources by exploiting the difference-frequency nonlinear effect in ferroelectric crystals. The nonlinear response of ferroelectric crystals together with the fact that they are resonant in the terahertz region is the main reason for proposing their use.**

The proposal is explored theoretically using Landau-Devonshire theory to calculate nonlinear susceptibility coefficients for difference frequency generation. Also it is shown how the slowly varying amplitude approximation can be used to solve the wave equation, thus describing 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 part of the analysis.

An example of why such an approach is useful is to consider that quantum cascade lasers can produce frequencies above the terahertz range at room temperature, but need to be cooled to low temperatures in order to operate in the terahertz range. However two room-temperature produced frequencies can be used to generate a terahertz-range output by arranging for the difference of the frequencies to fall into this range.

I. Introduction

The terahertz region of the electromagnetic spectrum covers the range of frequencies from 0.3 THz to 3 THz or the wavelength range from 0.1 mm to 1 mm. This region has immense applications in imaging (medical, manufacturing and security), spectroscopy (scientific such as in condensed matter physics, environmental studies and security), and even in free space communication at high altitudes. Until recently there were few sources of such radiation. Efforts were made from the microwave end of the spectrum to extend the operation of sources in the terahertz region, either directly generating terahertz radiation or indirectly through the use of multipliers[1]. It is found that output power reduces as the square of the frequency with increasing frequency. On the other hand, efforts were also made from the optical end of the spectrum using lasers. As with the microwave end, two methods have been of interest: direct methods, such as free electron lasers and quantum cascade lasers; and indirect methods using nonlinear optical properties of materials. Free electron lasers are bulky, and extremely expensive. On the other hand, although great advances have been made with quantum cascade lasers, they are not very tunable and do not operate at room temperature for terahertz generation[2]. Among indirect methods, generation of terahertz as a difference frequency of two optical waves is of interest. By tuning one or both the optical frequencies, such a scheme allows tunability. In one technique called photo mixing, the difference frequency is generated in an ultra-fast photoconductive material , low temperature grown GaAs with two optical inputs of wavelengths around 780 nm [3]. The mixing is due to the modulation of conductivity. An applied electric field therefore produces a current at terahertz frequency which is then radiated out by an antenna. Another type of difference frequency generation is called optical rectification[4] which is the inverse of the electro-optic effect. This occurs in media, such as ZnTe, with large second order susceptibility. The common procedure is to illuminate the medium with ultra-short laser pulses (typically 1035nm 300fs). The pulse has a large number of frequency components and terahertz radiation is obtained from the difference of these frequencies. Apart from ZnTe, $LiNbO_3$ has also been used. The analysis of optical rectification [4] was carried out using a generalized Beer absorption law taking into account multiple photon absorption.

In this paper an analysis is carried out for difference frequency generation by a ferroelectric crystal (such as barium titanate) on which two continuous wave sources are incident. The advantages of ferroelectrics is that they are resonant in the terahertz region when subject to incident electromagnetic radiation and have a nonlinear response (many other nonlinear crystals may not be resonant in the terahertz region and would therefore not respond as strongly). The difference frequency component of the nonlinear response can be exploited. As an example of possible inputs consider a quantum cascade laser operating at room temperature. At this temperature the output is above the terahertz range but it is possible to arrange for two output frequencies whose difference is in this range. So the analysis to be carried out here will take these two frequencies as inputs and then show how the difference frequency can be generated in the ferroelectric crystal.

The theoretical basis is as follows. A Landau-Devonshire expansion of the free energy F in terms of powers of the components of the electric polarization vector $\mathbf{P}$ may be used to give a phenomenological description of the properties of ferroelectric materials. From this, formulae for the static dielectric constant and the nonlinear dielectric response have been obtained [5], [6]. Using the Landau-Khalatnikov (LK) dynamical equations Ishibashi and Orihara [7] have extended the theory to give expressions for the nonlinear dynamic dielectric response, that is, the nonlinear susceptibility coeffi-

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cients, 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 [8], [9], [10]

In the work here a relatively simple symmetry will be employed which gives usable results without too much complication, since our main aim is to demonstrate the principle that terahertz waves can be produced via difference-frequency generation. We go on to show how, having obtained difference-frequency susceptibility coefficients, it is possible to study the wave propagation of the generated waves by solving the Maxwell wave equation under the slowly varying amplitude approximation [11], [12] 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 difference-frequency output since the smaller the phase mismatch the greater 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 $\mathbf{P} = (P_x, P_y, P_z)$

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

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 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 $T_0 \leqslant T_c$ [6]. A second order transition, in contrast, occurs continuously and can be described with $B > 0$ and $C = 0$. The factors involving ϵ_0 are not strictly necessary, but 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 satisfies $\partial F/\partial P = 0$; 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, as is brought out by Strukov and Lenanyuk [13]. A more comprehensive description of such perovskites would include extra terms because $P_x^4 + P_y^4 + P_z^4$ and $P_x^2P_y^2 + P_y^2P_z^2 + P_z^2P_x^2$, for example, 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. [10]. However for the purposes of this paper we will deal with the simpler case described by (1) as this is sufficient to demonstrate the basic principles of difference-frequency generation of interest.

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 the oscillatory dynamics of interest here (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 [9], [10], 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, since we are interested in difference-frequency generation, we only need to consider an incident field made up of two single-frequency sinusoidal components, but a more complicated field which may be represented by any Fourier sum (or integral) [9], [11] can readily be incorporated into the formalism. We express the field in terms of complex amplitudes following the convention in Butcher and Cotter [11] as follows

$$\begin{aligned} E_i(t) = \frac{1}{2}\Big[&E_{\omega_1 i}\exp(-i\omega_1 t) + E^*_{\omega_1 i}\exp(i\omega_1 t) \\ &+E_{\omega_2 i}\exp(-i\omega_2 t) + E^*_{\omega_2 i}\exp(i\omega_2 t)\Big]. \end{aligned} \tag{5}$$

The corresponding response function, again following Butcher and Cotter [11], is defined by

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

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

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

where $\omega_\sigma = \omega_1 + \omega_2 + \cdots + \omega_n$ [9], [11]. The factor K is a product of combinatorial factors defined in Ref. [11]. For the purposes of this paper it is sufficient to state that $K = 1$ for the difference-frequency generation susceptibilities.

III. Nonlinear Susceptibility Coefficients for Difference Frequency Generation

On the basis of the theory outlined above expressions for difference frequency generation susceptibility coefficients have been derived [9] and are given below for the ferroelectric phase. These susceptibilities are for two input frequencies ω_1 and ω_2, which correspond to a second order nonlinearity. Note that second order susceptibilities vanish in the paraelectric phase ($T > T_0$) since the symmetry becomes non-polar [9]. Thus the ferroelectric state is essential for difference-frequency generation.

The non-vanishing susceptibility tensors $\chi^{(2)}_{ilm}(-(\omega_1 - \omega_2); \omega_1, -\omega_2)$, are:

$$
\begin{aligned}
\chi_{xxz} &= \chi_{yyz} = (1/2) g_x \sigma(\omega_1 - \omega_2)\sigma(\omega_1)s^*(\omega) \\
\chi_{xzx} &= \chi_{yzy} = (1/2) g_x \sigma(\omega_1 - \omega_2)s(\omega_1)\sigma^*(\omega_2) \\
\chi_{zxx} &= \chi_{zyy} = (1/2) g_x s(\omega_1 - \omega_2)\sigma(\omega_1)\sigma^*(\omega_2) \\
\chi_{zzz} &= (1/2) g_z s(\omega_1 - \omega_2)s(\omega_1)s^*(\omega_2)
\end{aligned}
\tag{9}
$$

where

$$\sigma(\omega) = -\frac{1}{\epsilon_0(m\omega^2 + i\omega\gamma)} \tag{10}$$

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

$$g_x = -2P_0\left(B + \frac{2C}{\epsilon_0}P_0^2\right) \tag{12}$$

$$g_z = -6P_0B - 20\frac{2C}{\epsilon_0}P_0^3 \tag{13}$$

$$f_z = \frac{P_0 g_x}{\epsilon_0^2} \tag{14}$$

For relaxational dynamics ($m = 0$) ϵ_0 must be replaced by $-\epsilon_0$ in (11).

As an illustration the real and imaginary parts of the χ_{zzz} susceptibility coefficient for difference-frequency generation have been plotted in Fig. 1 for two different temperatures, one close to the transition temperature at $T = 0.8T_0$, and the other close to room temperature. Strong resonances along the lines $\bar{\omega}_{1,2} = (1 - T/T_0)^{1/2}$ and $\bar{\omega}_1 - \bar{\omega}_2 = (1 - T/T_0)^{1/2}$ can be seen (the dimensionless frequencies are defined by $\bar{\omega} = \omega/\omega_0$, with $\omega_0^2 = 2aT_0/(\epsilon_0 m)$).

IV. Difference-Frequency Generation of Terahertz Waves

As discussed in the introduction it is possible to find two sources at ω_1 and ω_2 whose difference $\omega_1 - \omega_2$ is in the terahertz range. This section shows how the wave equation can be solved in order to find the generated waves in the ferroelectric crystal, and how resonance enhancement can strengthen these waves which otherwise might be rather weak compared to the input wave since they are produced from second-order nonlinearities.

In what follows it is assumed that the depletion of the incident waves as they travel through the crystal is negligible when calculating the generated wave. Suppose that the incident beam is travelling in the y direction with the spontaneous polarization aligned along the z axis, consistent with the formalism above. With these assumptions the driving field in general is in the x-z plane. The relevant wave equation to be solved for the difference-frequency field is [11], [12]

$$\frac{\partial^2 E_i^{\omega_1-\omega_2}}{\partial y^2} = \mu_0 \frac{\partial^2}{\partial t^2}\left[\epsilon_0 E_i^{\omega_1-\omega_2} + P_i^L + P_i^{NL}\right], \quad i = x, z, \tag{15}$$

in which P^L and P^{NL} are, respectively, the linear and nonlinear contributions to polarization in the bulk ferroelectric, and $E_i^{\omega_1-\omega_2}$ is the ith-component of the generated field propagating at frequency $\omega_1 - \omega_2$. For the z component, for example, this field is

$$E_z^{\omega_1-\omega_2} = \frac{1}{2}\Big[\mathrm{E}_z^{\omega_1-\omega_2}(y)\exp(ik_{3z}y)\exp(-i(\omega_1-\omega_2)t) + \text{c.c.}\Big], \tag{16}$$

where $k_{3z} = \epsilon_{3z}(\omega_1 - \omega_2)^2/c^2$ is the wave number of the field with ϵ_{3z} at frequency $\omega_1 - \omega_2$ evaluated using a similar method to that in Ref. [10], and c.c. stands for complex conjugate. From this together with the non-vanishing coefficients in (9) and the linear coefficients $\chi^{(1)}_{xx}(-(\omega_1 - \omega_2); \omega_1, \omega_2)$ and $\chi^{(1)}_{zz}(-(\omega_1 - \omega_2); \omega_1, \omega_2)$ (which are straightforward to calculate as is shown in Ref. [9]), we have

$$\mathbf{P}^L = \epsilon_0\Big[\chi^{(1)}_{xx}(-(\omega_1-\omega_2);\omega_1,\omega_2)\hat{\mathbf{x}} + \chi^{(1)}_{xx}(-(\omega_1-\omega_2);\omega_1,\omega_2)\hat{\mathbf{z}}\Big], \tag{17}$$

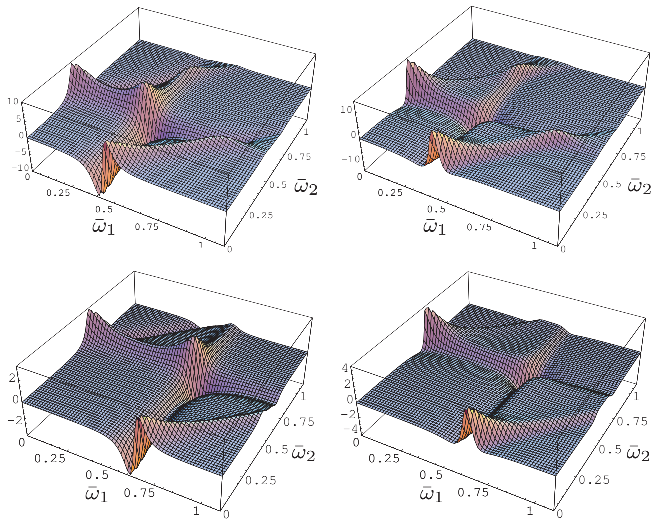

Fig. 1. Frequency and temperature dependence of the difference-frequency generation susceptibility coefficient $\chi^{(2)}_{zzz}(-(\omega_1 - \omega_2); \omega_1, \omega_2)$ in the ferroelectric phase. Horizontal axes: in multiples of $\omega_0 = (2aT_0/\epsilon_0 m)^{1/2}$, so that the dimensionless frequencies are $\bar{\omega}_{1,2} = \omega_{1,2}/\omega_0$; Vertical scale: in 10^{-5} mV^{-1}. Left: Imaginary part; right: real part; top: $T/T_0 = 0.8$ (close to the transition temperature); bottom: $T/T_0 = 0.58$ (close to room temperature); $T_0 = 473$ K, the value for barium titanate. For all plots the damping is set by $\gamma/(m\omega_0) = 0.1$. Other parameter values for the plot, chosen to by typical for barium titanate, are the same as in Ref. [14].

for linear polarization and

$$
\begin{aligned}
\mathbf{P}^{NL} =&\epsilon_0\Big\{\Big[\chi^{(2)}_{xxz}(-(\omega_1-\omega_2);\omega_1,\omega_2)E_x^{\omega_1}E_z^{\omega_2} \\
&+\chi^{(2)}_{xzx}(-(\omega_1-\omega_2);\omega_1,\omega_2)E_z^{\omega_1}E_x^{\omega_2}\Big]\hat{\mathbf{x}} \\
&+\Big[\chi^{(2)}_{zxx}(-(\omega_1-\omega_2);\omega_1,\omega_2)E_x^{\omega_1}E_x^{\omega_2} \\
&+\chi^{(2)}_{zzz}(-(\omega_1-\omega_2);\omega_1,\omega_2)E_z^{\omega_1}E_z^{\omega_2}\Big]\hat{\mathbf{z}}.\Big\}
\end{aligned} \tag{18}
$$

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

$$
\begin{aligned}
E_x^{\omega_1-\omega_2} = \frac{1}{2}\Big[&\mathrm{E}_x^{\omega_1}\exp(ik_{1x}y)\exp(-i\omega_1 t) \\
&\times \mathrm{E}_x^{\omega_2}\exp(ik_{2x}y)\exp(i\omega_2 t)+\mathrm{c.c}\Big],
\end{aligned} \tag{19}
$$

where $\mathrm{E}_x^{\omega_1}$ and $\mathrm{E}_x^{\omega_2}$ are constant amplitudes. (17) and (18) suggest that there are three possible driving field polarization configurations: x-z plane polarized, purely x polarized, and purely z polarized. 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 difference-frequency beam in the x direction as described by (18). Based on the slowly varying amplitude approximation [11], [12] in which the envelope $\mathrm{E}_z^{\omega_1-\omega_2}$ is assumed to have slow spatial variation compared with the exponential in (16), (15) reduces to

$$
\begin{aligned}
\frac{d\mathrm{E}_z^{\omega_1-\omega_2}}{dy} = &\frac{i(\omega_1-\omega_2)^2}{2k_{3z}c^2}\chi^{(2)}_{zxx}(-(\omega_1-\omega_2);\omega_1,\omega_2) \\
&\times \mathrm{E}_x^{\omega_1}\mathrm{E}_x^{\omega_2}\exp(i\Delta k_x y),
\end{aligned} \tag{20}
$$

in which $\Delta k_x = k_{1x} - k_{2x} - k_{3z}$. The difference-frequency terahertz field generated over a distance L can be determined by integrating (20) over y to give

$$
\begin{aligned}
\left|\mathrm{E}_z^{\omega_1-\omega_2}(L)\right|^2 = &\frac{\omega'^4}{4\left|k_{3z}\right|^2 c^4}(\mathrm{E}_x^{\omega_1}\mathrm{E}_x^{\omega_2})^2 \\
&\times \left|\chi^{(2)}_{zxx}(-(\omega_1-\omega_2);\omega_1,\omega_2)\right|^2 \frac{\sin(\Delta k_x L/2)}{\Delta k_x L/2}.
\end{aligned} \tag{21}
$$

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

We can now see that the key to generating an appreciable difference-frequency field is to achieve good phase matching. In this way, even though the second order conversion is not as strong as the linear fields, a usable difference-frequency field in the terahertz range can be generated.

V. Conclusion

Using Landau-Devonshire theory it is possible to calculate nonlinear susceptibility coefficients for difference-frequency generation, as has been illustrated. Terahertz waves can be generated from inputs outside this range, such as higher frequencies from a quantum cascade laser. Calculations substantiating this based on the wave equation have been made leading to phase-matching conditions on the propagation length that give maximum values for the generated wave amplitude. Therefore, even if the generated wave has a tendency to be weak, working with phase-matched lengths 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. [15] in which, among other things, it is demonstrated that terahertz waves can propagate through a ferroelectric crystal.

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