**SECOND HARMONIC GENERATION IN NONLINEAR
OPTICAL FILMS**

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IMA Preprint Series # 1097

January 1993

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Abstract

Second harmonic generation, an important phenomenon in nonlinear optics, is modeled in this work. Our model is derived from a nonlinear system of Maxwell's equations, which overcomes the known shortcomings of some commonly used models in the literature. Existence and uniqueness of solutions are established by a combination of a variational approach and the contraction mapping principle. Some numerical results are also presented.

Key words: second harmonic generation, nonlinear optics, electromagnetic waves, nonlinear system of Maxwell's equations.

PACS numbers: 42.65, 02.60, 02.90

Running title: second harmonic generation

1 Introduction

This work is aimed at the mathematical and computational study of the phenomenon of optical second harmonic generation (SHG) in nonlinear films.

In principle, all optical media are nonlinear. However, nonlinear electromagnetic phenomena in the optical region normally are observed only with high intensity incident beams, generally requiring the application of a laser. Numerous nonlinear optical phenomena that have been discovered since the birth of the field of nonlinear optics in 1961 have created a revolution in optical technology. For example, the phenomenon of second harmonic generation (SHG) provides a way to generate frequency doubled laser light. A remarkable application of SHG is the generation of coherent radiation at frequencies shorter than that of available lasers. The underlying physics is simple: an intense incident (pump) beam induces a response through a nonlinear polarization in a given medium; the medium then reacts to modify the field in a nonlinear fashion. The former and latter processes are governed by the constitutive equations and a nonlinear system of Maxwell's equations respectively.

Our goal is to model SHG in nonlinear optical films mathematically. More precisely, we are interested in a model that may be used to accurately predict the field intensity distribution of both reflected and transmitted waves when an intense pump beam is incident on the surface of a nonlinear film. We derive a general nonlinear model based on Maxwell's equations, together with boundary conditions obtained from jump conditions. The wellposedness of this model is obtained by means of establishing existence and uniqueness results. We show that except for a discrete number of frequencies, the problem has a unique solution,

provided that the nonlinearities of the medium (measured by the magnitude of the nonlinear susceptibility tensors) are not too large. It is well known that most existing nonlinear optical materials have very small nonlinear susceptibilities, which justifies the utility of our results. The model problem is solved by a simple method of finite elements coupled with a fixed point iteration. Our numerical experiments exhibit several interesting nonlinear optical phenomena.

Most existing models have relied on either the undepleted pump approximation (UPA) or slowly varying approximation (SVA). The UPA assumes that the depletion of energy from the pump field can be neglected. Recently, a French group has obtained a number of results by using the UPA; see Refs. 1 and 2, and references cited there. Because of the UPA, the problem can be solved by the well established linear theory, for example Ref. 3. The model is actually a coupled linear system. Unfortunately, the theory of nonlinear optics (e.g. Ref. 4) asserts that the UPA is invalid when the output energy becomes significant compared to the pump beam. In other words, the approximation fails for the most interesting cases where high conversion efficiency is desired.

The SVA is useful in the case where the field varies slowly in some spatial direction, say the “ z ” direction. The SVA assumes that $|E''(z)| \ll |kE'(z)|$, where k is the wave number; in this case the original second order equation can be approximated by a first order equation. The SVA is valid if the energy transfer among waves is significant only after the waves travel over a distance much longer than their wavelengths^{4,5}. The drawback of the SVA is that it breaks down when the medium is sufficiently thin, as is the case with nonlinear coatings or nonlinear optical films.

Our model is derived from the full nonlinear system of equations. In addition to its generality, our model overcomes the difficulties mentioned above. The comparison of our model and the UPA is demonstrated through some numerical results presented in this paper. An interesting future project would be to compare the results of our model with those given by the SVA.

When the model is linear with periodic structure in one direction, results on existence and uniqueness were obtained by Chen and Friedman⁶ assuming the dielectric coefficient ϵ is a piecewise constant function. They showed that for all but a discrete number of ϵ 's, there exists a unique solution to the Maxwell equation by an integral equation approach, while standard jump conditions allowed them to reduce the system to a coupled pair of Fredholm equations. Similar results were obtained for Maxwell's equations in biperiodic structures in Dobson and Friedman⁷. However, little is known concerning the questions of existence and uniqueness for the nonlinear Maxwell equations that govern SHG.

To facilitate our study, we make the following general assumptions:

- the fields are transverse,
- the nonlinear medium is stratified,
- the surface is flat, with normally incident pump beam.

It seems that this is perhaps the only situation where complete mathematical results may be obtained. Study on more general situations is in progress and will be presented elsewhere.

The plan of this paper is as follows: In Section 2, we derive the model, consisting of the nonlinear differential equations and boundary conditions derived from the nonlinear Maxwell

equations. We then proceed in Section 3 to establish existence and uniqueness of solutions to the model problem. Some numerical experiments are presented in Section 4. We conclude the paper in Section 5 with remarks and comments on interesting related research problems.

We refer the reader to the classic books of Bloembergen⁴ and Shen⁵ for the underlying physics of nonlinear optics. Many other interesting mathematical developments in nonlinear optics may be found in Newell and Moloney⁸ and references cited therein.

2 Formulation of the model

We assume that the media are nonmagnetic with constant magnetic permittivity everywhere, that no external charge or current is present in the field, and that the electric and magnetic fields are time harmonic, *i.e.*,

$$E = E(r)e^{-i\omega t}, \quad H = H(r)e^{-i\omega t}$$

where $r = (x_1, x_2, x_3) \in \mathbb{R}^3$.

We are interested in studying optical wave interaction in nonlinear media, or in particular SHG, which deals with the phenomenon of two wave mixing. Suppose that there is a pumping wave with some frequency $\omega_1 = \omega$. Consider the two wave fields $E(r, \omega_1)$ and $E(r, \omega_2)$, where $\omega_2 = 2\omega_1$. To simplify our notation, we denote $E(\omega_i) = E(r, \omega_i)$. The Maxwell equations yield the following coupled system:

$$\left[\nabla \times (\nabla \times) - \frac{\omega_1^2 \epsilon_1}{c^2} \right] E(\omega_1) = \frac{4\pi\omega_1^2}{c^2} \chi^{(2)}(\omega_1 = -\omega_2 + \omega_1) : E^*(\omega_1)E(\omega_2), \quad (2.1)$$

$$\left[\nabla \times (\nabla \times) - \frac{\omega_2^2 \epsilon_2}{c^2} \right] E(\omega_2) = \frac{4\pi\omega_2^2}{c^2} \chi^{(2)}(\omega_2 = \omega_1 + \omega_1) : E(\omega_1)E(\omega_1), \quad (2.2)$$

where E^* denotes the complex conjugate, and ϵ_1, ϵ_2 are the (linear) dielectric coefficients at frequencies ω_1 and ω_2 , respectively. Through the nonlinear coupling, energy can be transferred back and forth between fields at each frequency. The nonlinear nature of the medium is described by $\chi^{(2)}$, the second-order nonlinear susceptibility tensor. More discussions on nonlinear susceptibility tensors as well as the derivation of the above system from the Maxwell equations may be found in Shen⁵. Note that the presence of new frequency components is the most striking difference between nonlinear and linear optics.

Further, the usual jump conditions are valid⁴: the tangential components of E and H must be continuous at the boundary defining the interface between two homogeneous materials, and the normal components of D and H must also be continuous at the boundary for all frequencies. These jump conditions may be used to derive the necessary boundary conditions.

The formulation of the model can be simplified because of the general assumptions made in the Introduction. Since the medium is stratified, the fields vary only in one direction. The transversality assumption allows us to reduce the Maxwell system to a system of Helmholtz equations. For instance, by choosing the polarization properly, one may assume that $E(r, \omega) = E_1(z)\vec{y}$ and $E(r, 2\omega) = E_2(z)\vec{x}$.

Let us specify the geometry of the model. Assume that a slab of stratified nonlinear material (say, composed of many layers of different nonlinear media) is placed between two linear materials with dielectric constants ϵ_1 and ϵ_2 respectively. See Figure 1.

In the regions D^+ and D^- , where the material is linear, the electric fields may be expressed easily following the theory of linear optics, for example Born and Wolf⁹. Specifically, in the

region D^+ ,

$$E_1(z) = E_I e^{ik_{11}z} + E_{1R} e^{-ik_{11}z} \quad (2.3)$$

$$E_2(z) = E_{2R} e^{-ik_{21}z} \quad (2.4)$$

and in the region D^- ,

$$E_1(z) = E_{1T} e^{ik_{12}z} \quad (2.5)$$

$$E_2(z) = E_{2T} e^{ik_{22}z} \quad (2.6)$$

where the incident electric field is $(0, E_I e^{ik_{11}z}, 0)$, E_{1R} and E_{2R} are the reflectivity constants, E_{1T} and E_{2T} are the transmittance constants corresponding to frequencies ω_1 and ω_2 respectively. In these expressions, the constants k_{ij} are defined by

$$k_{i1} = \frac{\omega_i \sqrt{\epsilon_1}}{c}$$

$$k_{i2} = \frac{\omega_i \sqrt{\epsilon_2}}{c},$$

for $i = 1, 2$. In the nonlinear medium, the Maxwell system in the previous section reduces to

$$\left[\frac{d^2}{dz^2} + k_1^2 \right] E_1 = \chi_1 E_2 E_1^* \quad (2.7)$$

$$\left[\frac{d^2}{dz^2} + k_2^2 \right] E_2 = \chi_2 E_1 E_1 \quad (2.8)$$

where for $i = 1, 2$, $k_i^2 = \omega_i^2 \epsilon(\omega_i)/c^2$, $\epsilon(\omega_i)$ are the linear dielectric constants, and $\chi_1 = -4\pi(\omega_1^2/c^2)\chi_{2,2,1}^{(2)}(\omega_1)$, $\chi_2 = -4\pi(\omega_2^2/c^2)\chi_{1,2,2}^{(2)}(\omega_2)$. Note that k_j and χ_j , $j = 1, 2$ are not necessarily constant; we have taken them to be functions of z .

The boundary conditions can be derived from the standard jump conditions. Indeed, since the tangential components of E_i and H_i are continuous across the boundary Γ_i for $i = 1, 2$, we get on Γ_1

$$[[E_1]]_T = 0 \Rightarrow E_I e^{ik_{11}l} + E_{1R} e^{-ik_{11}l} = E_1(l), \quad (2.9)$$

$$[[E_2]]_T = 0 \Rightarrow E_{2R} e^{-ik_{21}l} = E_2(l), \quad (2.10)$$

$$[[H_1]]_T = 0 \Rightarrow E'_1(l) = ik_{11} E_I e^{ik_{11}l} - ik_{11} E_{1R} e^{-ik_{11}l}, \quad (2.11)$$

$$[[H_2]]_T = 0 \Rightarrow E'_2(l) = -ik_{21} E_{2R} e^{-ik_{21}l}, \quad (2.12)$$

and on Γ_2

$$[[E_1]]_T = 0 \Rightarrow E_1(0) = E_{1T}, \quad (2.13)$$

$$[[E_2]]_T = 0 \Rightarrow E_2(0) = E_{2T}, \quad (2.14)$$

$$[[H_1]]_T = 0 \Rightarrow E'_1(0) = ik_{12} E_{1T}, \quad (2.15)$$

$$[[H_2]]_T = 0 \Rightarrow E'_2(0) = ik_{22} E_{2T}, \quad (2.16)$$

where E_I is given as the incoming electric field intensity, and E_{1R} , E_{2R} , E_{1T} , and E_{2T} are constants that may be found as follows

$$E_{1R} = (E_1(l) - E_I e^{ik_{11}l}) e^{ik_{11}l}, \quad (2.17)$$

$$E_{2R} = E_2(l) e^{ik_{21}l}, \quad (2.18)$$

$$E_{1T} = E_1(0), \quad (2.19)$$

$$E_{2T} = E_2(0). \quad (2.20)$$

Define the domain $\Omega = (0, l)$. We have derived the following two-point boundary value

problem

$$\left(\frac{d^2}{dz^2} + k_1^2\right)E_1 = \chi_1 E_1^* E_2 \quad \text{in } \Omega, \quad (2.21)$$

$$\left(\frac{d^2}{dz^2} + k_2^2\right)E_2 = \chi_2 E_1^2 \quad \text{in } \Omega, \quad (2.22)$$

$$E_1'(l) + ik_{11}E_1(l) = 2ik_{11}e^{ik_{11}l}E_l, \quad (2.23)$$

$$E_2'(l) + ik_{21}E_2(l) = 0, \quad (2.24)$$

$$E_1'(0) - ik_{12}E_1(0) = 0, \quad (2.25)$$

$$E_2'(0) - ik_{22}E_2(0) = 0. \quad (2.26)$$

Next, we derive the weak form. Let ϕ be a test function. Multiplying both sides of equations (2.21) and (2.22) by ϕ , we get after some simple integration by parts and making use of the boundary conditions (2.23-2.26),

$$\int_{\Omega} E_1' \bar{\phi}' - \int_{\Omega} k_1^2 E_1 \bar{\phi} + ik_{11}E_1(l)\bar{\phi}(l) + ik_{12}E_1(0)\bar{\phi}(0) = - \int_{\Omega} \chi_1 \bar{E}_1 E_2 \bar{\phi} + 2ik_{11}e^{ik_{11}l}E_l \bar{\phi}(l) \quad (2.27)$$

and

$$\int_{\Omega} E_2' \bar{\phi}' - \int_{\Omega} k_2^2 E_2 \bar{\phi} + ik_{21}E_2(l)\bar{\phi}(l) + ik_{22}E_2(0)\bar{\phi}(0) = - \int_{\Omega} \chi_2 \bar{E}_1^2 \bar{\phi}. \quad (2.28)$$

We now present a result on the conservation of energy. As usual the incoming energy is denoted by $e_{in} = k_{11}|E_I|^2$. Similarly, one can define the energy corresponding to the frequency components ω and 2ω by

$$e_1 = k_{11}|E_{1R}|^2 + k_{21}|E_{2R}|^2, \quad (2.29)$$

$$e_2 = k_{12}|E_{1T}|^2 + k_{22}|E_{2T}|^2, \quad (2.30)$$

respectively.

Theorem 2.1 *The energy is conserved if $\chi_1(z)$ and $\chi_2(z)$ are identical real valued functions.*

Proof. We wish to show that

$$e_{in} = e_1 + e_2 .$$

Choosing $\phi = E_1$ in the weak form (2.27) and $\phi = E_2$ in (2.28) respectively, we obtain, by taking the imaginary parts on both sides,

$$\begin{aligned} k_{11}|E_1(l)|^2 + k_{12}|E_1(0)|^2 &= -Im \int \chi_1 E_2 \bar{E}_1 \bar{E}_1 + 2Re\{k_{11}e^{ik_{11}l} \bar{E}_1(l)E_I\} \\ k_{21}|E_2(l)|^2 + k_{22}|E_2(0)|^2 &= -Im \int \chi_2 E_1^2 \bar{E}_2 . \end{aligned}$$

Substituting the expressions of E_{iR} and E_{iT} for $i = 1, 2$ into those of e_1 and e_2 , we have

$$\begin{aligned} e_1 + e_2 &= k_{11}|E_1(l)e^{ik_{11}l} - e^{2ik_{11}l}E_I|^2 + k_{12}|E_1(0)|^2 + k_{21}|E_2(l)e^{ik_{21}l}|^2 + k_{22}|E_2(0)|^2 \\ &= k_{11}|E_1(l)|^2 + k_{12}|E_1(0)|^2 + k_{21}|E_2(l)|^2 + k_{22}|E_2(0)|^2 \\ &\quad + k_{11}(-2Re\{\bar{E}_1(l)e^{ik_{11}l}E_I\}) + e_{in} \\ &= -Im \int (\chi_1 E_2 \bar{E}_1^2 + \chi_2 E_1^2 \bar{E}_2) + e_{in} \end{aligned}$$

and the conclusion follows from the assumption that $\chi_1 = \chi_2$ is a real-valued function. $\square$

3 Existence and uniqueness

Recall that $\Omega = (0, l)$. From now on, we assume that ϵ_1 , ϵ_2 , χ_1 , and χ_2 are fixed L^∞ functions in Ω . Let $A_j : H^1(\Omega) \rightarrow H^{-1}(\Omega)$ be the linear operator defined by $B_j(u_1, u_2) = \langle A_j u_1, u_2 \rangle$, where

$$B_j(u_1, u_2) = \int_{\Omega} u_1' \bar{u}_2' - \int_{\Omega} k_j^2 u_1 \bar{u}_2 + ik_{j1} u_1(l) \bar{u}_2(l) + ik_{j2} u_1(0) \bar{u}_2(0)$$

Recall that $k = \frac{\omega\sqrt{\epsilon}}{c}$, where ω is the frequency.

Lemma 3.1 *For all but a discrete set of the frequencies ω , there exist constants $C_j(\omega, l)$ such that*

$$\|A_j^{-1}\| \leq C_j, \quad j = 1, 2.$$

Proof. Since the argument is the same for both operators A_j , $j = 1, 2$, we drop the subscript j for the rest of the proof. Write $B = B^1 + \omega^2 B^2$ where

$$\begin{aligned} B^1(u_1, u_2) &= \int_{\Omega} u'_1 \bar{u}'_2 + ik_1 u_1(l) \bar{u}_2(l) + ik_2 u_1(0) \bar{u}_2(0), \\ B^2(u_1, u_2) &= -\frac{1}{c^2} \int_{\Omega} \epsilon u_1 \bar{u}_2. \end{aligned}$$

Splitting B^1 into real and imaginary parts, and using the fact that k_1 is real and positive, we see that

$$|B^1(u_1, u_1)| \geq c \left(\int_{\Omega} |u'_1|^2 + k_1 |u_1(l)|^2 \right).$$

It then follows easily from Poincaré's inequality that B^1 is coercive, that is,

$$|B^1(u_1, u_1)| \geq c(k_1, l) \|u_1\|_{H^1(\Omega)}^2,$$

and thus the Lax-Milgram theorem implies that the operator A^1 defined by $\langle A^1 u_1, u_2 \rangle = B^1(u_1, u_2)$ has a bounded inverse. To emphasize that A^1 depends on k_1 , and hence on the frequency ω_1 (recall that $k_1 = \frac{\omega_1 \sqrt{\epsilon_1}}{c}$), we write $A^1(\omega_1)$. Notice that the operator $A^2 : H^1(\Omega) \rightarrow H^{-1}(\Omega)$ defined by $\langle A^2 u_1, u_2 \rangle = B^2(u_1, u_2)$ is compact.

Holding ω_1 fixed, consider the operator $A(\omega_1, \omega) = A^1(\omega_1) + \omega^2 A^2$. We see that $A(\omega_1, \omega)^{-1}$ exists by Fredholm theory for all $\omega \notin \mathcal{E}(\omega_1)$, where $\mathcal{E}(\omega_1)$ is some discrete set. It is clear that

$$\|A^1(\omega) - A^1(\omega_1)\| \rightarrow 0, \quad \text{as } \omega \rightarrow \omega_1.$$

Thus, since $\|A(\omega, \omega) - A(\omega_1, \omega)\| = \|A_1(\omega) - A_1(\omega_1)\|$ is small for $|\omega - \omega_1|$ sufficiently small, it follows from the stability of bounded invertibility (see e. g. Ref. 10, Theorem IV-1.16) that $A(\omega, \omega)^{-1}$ exists and is bounded for $|\omega - \omega_1|$ sufficiently small, $\omega \notin \mathcal{E}(\omega_1)$. Since $\omega_1 > 0$ can be taken to be any real number, we have shown that $A(\omega, \omega)^{-1}$ exists for all but a discrete set of points. $\square$

We wish to find u and $v \in H^1(\Omega)$ such that for $f \in H^{-1}(\Omega)$, and $\chi_1, \chi_2 \in L^\infty(\Omega)$, there holds

$$A_1 u = \chi_1 v u^* + f, \quad (3.1)$$

$$A_2 v = \chi_2 u^2. \quad (3.2)$$

Consider the following fixed-point iteration. Set $u_0 = 0$ and for $k = 0, 1, \dots$, define

$$\begin{aligned} v_k &= A_2^{-1}(\chi_2 u_k^2) \\ u_{k+1} &= A_1^{-1}(\chi_1 u_k^* v_k + f). \end{aligned}$$

Further

$$u_{k+1} = F(u_k)$$

where

$$F(u) = A_1^{-1}\{\chi_1 u^* A_2^{-1}(\chi_2 u^2) + f\}.$$

It is easy to see that F maps $H^1(\Omega)$ to $H^1(\Omega)$.

Theorem 3.1 *Given $f \in H^{-1}(\Omega)$ there exists a constant $\epsilon > 0$ depending only on $\|f\|_{H^{-1}}$ and Ω , such that if $\|\chi_1\|_{L^\infty}\|\chi_2\|_{L^\infty} \leq \epsilon$ then the model problem, (3.1), (3.2), has a unique solution $u, v \in H^1(\Omega)$.*

The proof of Theorem 3.1 can be given by using the contraction principle, see for example Hutson and Pym¹¹, which follows immediately from the following Lemmas 3.2 and 3.3.

Lemma 3.2 *For the product of $\|\chi_1\|_{L^\infty}$ and $\|\chi_2\|_{L^\infty}$ sufficiently small, there exists $R > 0$ such that $\|u\|_{H^1} \leq R$ implies $\|F(u)\|_{H^1} \leq R$.*

Proof. Let $g = A_1^{-1}f$, then $g \in H^1$. Write

$$F(u) - g = A_1^{-1}\{\chi_1(u - g)^* A_2^{-1}(\chi_2 u^2)\} + A_1^{-1}\{\chi_1 g^* A_2^{-1}(\chi_2 u^2)\}.$$

Then using Schauder's lemma, we can estimate

$$\begin{aligned} \|F(u) - g\|_{H^1} &\leq C\|\chi_1(u - g)^* A_2^{-1}(\chi_2 u^2)\|_{L^2} + C\|\chi_1 g^* A_2^{-1}(\chi_2 u^2)\|_{L^2} \\ &\leq C\|\chi_1\|_{L^\infty}(\|(u - g)^* A_2^{-1}(\chi_2 u^2)\|_{L^2} + \|g\|_{H^1}\|A_2^{-1}(\chi_2 u^2)\|_{H^1}) \\ &\leq \|\chi_1\|_{L^\infty}\|\chi_2\|_{L^\infty}(C_1\|u\|_{H^1}^2\|u - g\|_{H^1} + C_2\|u\|_{H^1}^2). \end{aligned}$$

Choosing $R = 2\|g\|_{H^1}$, since $\|\chi_1\|_{L^\infty}\|\chi_2\|_{L^\infty}$ is sufficiently small we then have

$$\|F(u)\|_{H^1} \leq \|F(u) - g\|_{H^1} + \|g\|_{H^1} \leq R.$$

□

Lemma 3.3 *Given a radius R , assume that $\|\chi_1\|_{L^\infty}\|\chi_2\|_{L^\infty}$ is sufficiently small. Then F is a contraction on*

$$B_R = \{u \in H^1 : \|u\|_{H^1} \leq R\}$$

i.e.,

$$\|F(u_1) - F(u_2)\|_{H^1} \leq C_0\|u_1 - u_2\|_{H^1}$$

for all $u_1, u_2 \in B_R$ where $C_0 < 1$.

Proof.

$$\begin{aligned}
\|F(u_1) - F(u_2)\|_{H^1} &= \|A_1^{-1}\{\chi_1 u_1^* A_2^{-1}(\chi_2 u_1^2) - \chi_1 u_2^* A_2^{-1}(\chi_2 u_2^2)\}\|_{H^1} \\
&\leq C\|\chi_1\{u_1^* A_2^{-1}(\chi_2 u_1^2) - u_2^* A_2^{-1}(\chi_2 u_2^2)\}\|_{L^2} \\
&= C\|\chi_1\{(u_1 - u_2)^* A_2^{-1}(\chi_2 u_1^2) - u_2^* A_2^{-1}(\chi_2(u_2^2 - u_1^2))\}\|_{L^2} \\
&\leq C\|\chi_1\|_{L^\infty}\{\|u_1 - u_2\|_{H^1} \cdot \|A_2^{-1}(\chi_2 u_1^2)\|_{H^1} \\
&\quad + \|u_2\|_{H^1} \cdot \|A_2^{-1}(\chi_2(u_2^2 - u_1^2))\|_{H^1}\} \\
&\leq C\|\chi_1\|_{L^\infty}\|\chi_2\|_{L^\infty}(\|u_1\|_{H^1}^2 + \|u_2\|_{H^1}^2)\|u_1 - u_2\|_{H^1} .
\end{aligned}$$

□

4 Numerical experiments

We solve the model problem by a combination of the method of finite elements and fixed-point iterations. The convergence is fast for all conventional materials. Our numerical experiments also show that the iteration scheme breaks down when the product of $\|\chi_1\|_{L^\infty}$ and $\|\chi_2\|_{L^\infty}$ becomes very large, as predicted by Theorem 3.1. Note that the scheme works in the case that the value of either χ_1 or χ_2 is large as long as the product is not too large.

Our numerical experiments exhibit an important phenomenon of nonlinear optics, that of *phase matching*, see Shen⁵. It is known that the maximum rate of energy transfer should occur when both energy and momentum matching conditions are satisfied. Since the energy matching condition $\omega_1 + \omega_2 = \omega_3$ is automatically satisfied in the steady state case, in order to achieve the maximum energy conversion rate, the momentum matching condition $k = k_1 + k_2$

should also be satisfied. In other words, $\Delta k = k_1 + k_2 - k_3$ should vanish, where Δk is the phase mismatch between the waves. Figure 2 shows a numerical experiment where it is clear that the energy conversion rate is the highest when the phase mismatch vanishes. The lack of symmetry with respect to Δk in Figure 2 should be expected: since the experiment involves a “thin” film, it can be seen that even in the linear case, the field intensity inside the film has no symmetry with respect to perturbations in k . The energy conversion rate is largely determined by the field intensity of the pump beam inside the film, hence one would not expect symmetry in the energy conversion rate with respect to perturbations in k either.

Figure 3 shows that the deeper the medium is, the more energy may be transferred to the frequency doubled wave. Note the easily observed “wavelength” of each field, with the second harmonic at exactly half the wavelength of the pump. Although the energy inside the film varies, the total *radiated* energy is conserved when the nonlinear susceptibilities are real and equal, as per Theorem 2.1.

We also compare our results to those obtained by applying UPA. It is worth pointing out that UPA is nothing but the first step of our fixed-point iteration. Our numerical experiments indicate when χ_1 and χ_2 are small, the UPA solution matched our solution closely. However, as shown in Figure 4, even with a relatively small increase in the magnitude of the nonlinear susceptibility tensors, the solutions given by our model differ greatly from those given by UPA. This indicates that UPA is not a good approximation when the nonlinearity of the medium is large. We also found that UPA becomes relatively less accurate as the film gets deeper.

A comparison of our model with SVA would be very worthwhile, with the goal of estab-

lishing conditions under which the predictions of the two models is substantially different. We believe that our model is primarily useful in the case of thin films (where the film depth is on the order of tens to hundreds of wavelengths). Since our goal here was only to introduce our model, such a comparison is outside the scope of the present paper.

5 Concluding remarks

The techniques described here may be extended to study some more general problems in nonlinear optics.

One interesting direction is to study second harmonic generation in diffraction gratings (periodic structures), with bare materials or with nonlinear films and coatings. Previous experiments and theory indicate that a great enhancement in conversion efficiency may be achieved by applying gratings over the flat surface of a nonlinear material. Presumably the enhancement is due to resonance phenomena in the diffraction grating. The techniques described here, both analytical and numerical, can be extended to study this situation and should be useful in examining different materials (with different $\chi^{(2)}$) versus grating parameters (period, groove depth, coated material, etc.), in an effort to optimize the nonlinear effects.

Our model may also be generalized to deal with nonlinear optical media that involve more complicated nonlinear susceptibility tensors. For example, many applications have indicated the usefulness of $\chi^{(3)}$, the third order nonlinear susceptibility tensor, in the modeling process. In fact, it is known⁵ that $\chi^{(2)}$ vanishes in media with inversion symmetry. In par-

ticular, all liquids, gases, and centrosymmetric solids have zero second order susceptibility tensors. Therefore, the nonlinear properties of these media are governed by the third order susceptibility tensor.

Acknowledgements

We wish to thank Dr. J. Allen Cox of Honeywell, Inc. Systems and Research Center for introducing us to this problem and some of its applications. The first author was partially supported by a grant from Honeywell, Inc. The second author was partially supported by an NSF Mathematical Sciences Postdoctoral Fellowship.

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Captions

Figure 1: The geometry of the problem.

Figure 2: Energy of the doubled frequency wave vs. phase mismatch Δk .

Figure 3: The intensity distributions of the fields $E(\omega)$ (Figure 3a) and $E(2\omega)$ (Figure 3b).

Film depth in Figure 3b is double that of Figure 3a.

Figure 4: Comparison of intensity distributions of the electric fields obtained by using our model and those given by UPA. a) $E(\omega)$ fields. b) $E(2\omega)$ fields.

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