OPTI 571L Lab 11: Second-Harmonic Generation

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It is now more than 50 years since Franken, Hill, Peters, and Weinreich [1] initiated the field of experimental nonlinear optics with the generation of optical harmonics in a quartz crystal. (As an aside Prof. Peter A. Franken was the Director of the Optical Sciences Center from 1973-1983.) In that experiment a fundamental field from a ruby laser at 694.3 nm produced light at the second harmonic (SH) at 347.2 nm via the nonlinearity of the quartz crystal. Since those seminal experiments the field of harmonic generation has advanced greatly. For example, second-harmonic generation (SHG) is technologically important for new frequency generation that allows access to wavelengths where no lasers are currently available as well as devices such as green laser pointers, and high-harmonic generation is at the forefront of attosecond science and provides a source of radiation extending from the vacuum ultraviolet to soft x-rays starting from optical fields.

The goal of this Lab is to have you numerically explore some aspects of harmonic generation, in particular SHG with focussed Gaussian beams. This problem was solved in detail already in 1968 by Boyd and Kleinman [2]. However, it can be difficult to extract information from this forty three page paper and the idea is to lead you through the main results in steps. The lessons learned are valuable and of great utility if you become involved in experimental harmonic generation.

Keywords: Equations for SHG, phase-matching, plane-wave solution, undepleted pump beam approximation, Gaussian beam theory and optimization.

I. BASICS OF SHG

This Lab is not intended to be self-contained, you need to have or develop some background in nonlinear optics to reap the benefit of the simulations. The equations and notation for the Lab are contained in Chaps. 2.7 and 2.10 of the Nonlinear Optics text by Boyd [3]. The starting point is the equations for SHG including transverse diffraction of the fundamental and SH fields. This is followed by an examination of the case of plane-wave SHG in various limits as well as linear Gaussian beam propagation as stepping stones to discussing SHG with focused beams.

A. Nonlinear equations for SHG

First a brief overview of harmonic generation. In particular the optical polarization $P(t)$ of a medium in response to an applied electric field $E(t)$ is written as the power series

$$P(t) = \epsilon_0\chi^{(1)}E(t) + \epsilon_0\chi^{(2)}E^2(t) + \epsilon_0\chi^{(3)}E^3(t)\dots \quad (1)$$

$$= \sum_{q=1}^{\infty} P^{(q)}(t). \quad (2)$$

This power series represents the anharmonicity of the bound atomic electron motion in response to the applied electric field. In particular, $P^{(1)}(t) = \epsilon_0\chi^{(1)}E(t)$ is the familiar linear optical polarization with $\chi^{(1)}$ the linear susceptibility, whereas for $q > 1$ the nonlinear polarization is $P^{(q)}(t)$ with $\chi^{(q)}$ the corresponding q^{th} order nonlinear susceptibility. For simplicity in notation the tensor nature of these quantities has been neglected (Boyd Chap. 1). For this perturbative expansion to be valid it is required that all photon energies $\hbar\omega$ present in the applied electric field are well below the resonant transition frequencies of the material, and the magnitude of the electric field $|E(t)|$ is much less than the Coulombic electric field $E_{at} >> |E(t)|$ binding the electrons to their parent ions. Typically $E_{at} \simeq 5\times10^{11}$ V/m, MKS units being used throughout.

Based on the above discussion, for a monochromatic fundamental field of frequency ω_1

$$E(t) = \frac{1}{2}\left[A_1 e^{-i\omega_1 t} + c.c.\right], \quad (3)$$

with A_1 the complex amplitude of the fundamental field, the q^{th} order nonlinear polarization $P^{(q)}(t)$ will contain many terms including the one shown

$$P^{(q)}(t) = \frac{\epsilon_0\chi^{(q)}}{2^q}\left[A_1^q e^{-i(q\omega_1)t} + \dots\right]. \quad (4)$$

The significance of the term shown is that this nonlinear polarization can act as a source for a field of complex amplitude $A_q \propto A_1^q$ at the q^{th} harmonic with frequency $\omega_q = q\omega_1$. This is the physical underpinning of the process of harmonic generation.

The focus here is on the process of SHG for which $q = 2$. The *nonlinear* equations governing propagation of the fundamental and SH fields along the z-axis are

$$\frac{\partial A_1}{\partial z} = \frac{i}{2k_1}\nabla_T^2 A_1 + \frac{i\omega_1^2\chi^{(2)}}{k_1c^2}A_2A_1^* e^{-i\Delta kz}, \quad (5)$$

$$\frac{\partial A_2}{\partial z} = \frac{i}{2k_2}\nabla_T^2 A_2 + \frac{i\omega_2^2\chi^{(2)}}{2k_2c^2}A_1^2 e^{i\Delta kz}, \qquad (6)$$

where A_j with $j = 1, 2$ are the complex amplitudes for the fundamental and SH fields respectively, $k_j = n_j\omega_j/c$ are the wavenumbers for the fields along the z-axis, with n_j the corresponding refractive-indices in the *birefringent* crystal. In these equations one may approximate $n_2 = n_1$ and $k_2 = 2k_1$ everywhere except in the wavevector mismatch

$$\Delta k = (2k_1 - k_2) = \frac{2\omega_1}{c}(n_1 - n_2) = \frac{2\omega_1}{c}\Delta n, \qquad (7)$$

with n_1 being the refractive-index experienced by the fundamental and n_2 that for the SH. These equations coincide with Eqs. (2.7.10) and (2.7.11) of Boyd generalized to include transverse diffraction via the transverse Laplacian terms, and $d_{eff} \equiv \frac{\chi^{(2)}}{2}$ being the effective nonlinear coefficient which depends on the material (see Boyd Chap. 1).

In the propagation Eqs. (5,6) the complex amplitudes $A_j(r, z)$ depend not only on the propagation coordinate z but also the transverse radial displacement r in cylindrical coordinates (r, θ, z): Cylindrical symmetry is assumed to be maintained so the fields are independent of the azimuthal angle θ. The goal is to investigate solutions of Eqs. (5,6) for an initial fundamental field $A_1(r, z_0)$ and zero incident SH $A_2(r, z_0) = 0$ at $z = z_0$.

(a) The parameters $d_{eff} = 10^{-12}$ m/V, $\lambda_1 = 10^{-6}$m, and $n_1 = 1.6$ are used unless otherwise stated. Show that Eqs. (5,6) reduce to the form

$$\frac{\partial A_1}{\partial z} = \frac{i}{2k_1}\nabla_T^2 A_1 + i\eta A_2 A_1^* e^{-i\Delta kz}, \qquad (8)$$

$$\frac{\partial A_2}{\partial z} = \frac{i}{4k_1}\nabla_T^2 A_2 + i\eta A_1^2 e^{i\Delta kz}, \qquad (9)$$

and find the value for η in MKS units.

(b) The beam intensity for either the fundamental or SH is given by $I_j(r, z) = 2\epsilon_0 n_j c|A_j(r, z)|^2$, and the corresponding beam power by

$$P_j(z) = \int_0^\infty 2\pi r dr\ I_j(r, z).$$

For a fundamental Gaussian beam of the form $A_1(r, 0) = \mathcal{A}_1 e^{-r^2/w_0^2}$ with spot size $w_0 = 10\ \mu$m and peak intensity of $I_1(0, 0) = 10^{13}$ W/m^2 find the peak field strength $\mathcal{A}_1$ and beam power $P_1(0)$. These numbers give an idea of the scale of the field parameters used in the simulations.

The nonlinear partial differential equations (PDEs) in Eqs. (8,9) are the basis for the remainder of the Lab. Note that in the absence of the nonlinearity $\eta = 0$ these equations are simply the paraxial wave equations describing linear diffraction of the fundamental and SH fields.

B. Plane-wave theory

As a first step consider the plane-wave limit of Eqs. (8,9) in which transverse diffraction is neglected and $A_1(z)$ and $A_2(z)$ depend only on the propagation coordinate z

$$\frac{dA_1}{dz} = i\eta A_2 A_1^* e^{-i\Delta kz}, \quad \frac{dA_2}{dz} = i\eta A_1^2 e^{i\Delta kz}. \qquad (10)$$

These nonlinear ordinary differential equations (ODEs) are solved subject to the boundary conditions $A_1(z_0) = \mathcal{A}_1, A_2(z_0) = 0$, and for a medium of length $L = 10^{-2}$ m.

(a) Write a MATLAB code to solve the above ODEs subject to the boundary conditions with $z_0 = 0$. You can write own code based on, for example, the Euler or Runge-Kutta method, or use a MATLAB supplied routine such as `ode45`. For the *phase-matched case* $\Delta k = 0$, and an incident fundamental intensity $I_1(0) = 10^{13}$ W/m^2, plot $I_{1,2}(z)$ versus $z = [0, 10^{-2}]$ m.

(b) Your numerical simulation from part (a) should show close to 100% *depletion* of the fundamental and conversion to the SH, and an analytic solution is (Boyd Chap. 2.7)

$$A_1(z) = \mathcal{A}_1 sech\left(\frac{z}{\ell}\right),$$

$$A_2(z) = \mathcal{A}_1 tanh\left(\frac{z}{\ell}\right),$$

$$\ell = \frac{\sqrt{2n_1^2 n_2\epsilon_0 c^3}}{2\omega_1 d_{eff}\sqrt{I_1}}.$$

Validate your code from part (a) by comparing your numerical simulation with this solution. Near 100% conversion can be realized in waveguides [4].

(c) In the *phase-mismatched case*, $\Delta k \neq 0$, total depletion is no longer possible. Rather incomplete oscillations between the fundamental and SH, called *Maker fringes*, occur with a period given by twice the coherence length $L_{coh} = \frac{\pi}{|\Delta k|}$. For an incident fundamental intensity $I_1(0) = 0.1 \times 10^{13}$ W/m^2, plot $\frac{I_2(z)}{I_1(0)}$ versus $z = [0, 10^{-2}]$ m, for $\Delta n = 10^{-4}, 3\times10^{-4}$. Demonstrate that the Maker fringes have the predicted period, and estimate the percentage depletion of the fundamental in each case.

(d) The *undepleted pump beam approximation* arises in the phase-mismatched case when there is little depletion of the fundamental and $A_1(z) \approx A_1(0)$. In this approximation the SH field may be expressed as the integral (you should prove this for yourself)

$$A_2(z) = i\eta\int_{z_0}^{z} dz' A_1^2(0)e^{i\Delta kz'}. \qquad (11)$$

Write a MATLAB code to evaluate $\frac{I_2(z)}{I_1(0)}$ versus z using this integral for $\Delta n = 3 \times 10^{-4}$. You may write your own routine using, for example, the trapezoidal rule, or use a MATLAB supplied routine such as `integral`. Validate the integral approach by comparing with your numerical results from part (c). The integral solution will be generalized to form the basis of the treatment of SHG with focused beams.

Further exploration: Here you explored the exact solution for the phase-matched case but Boyd (Chap. 2.7) gives solutions also for the phase-mismatched case. You may want to challenge yourself and your code by comparing against these more general exact solutions. The experiment in Ref. [4] is a spectacular validation of the use of the plane-wave-like theory in the context of SHG in waveguides. In that paper a periodically poled lithium niobate waveguide was used. This can be used to produce quasi-phase-matching (QPM) in which an intrinsic phase-mismatch is cancelled by a periodic variation of $d_{eff}(z)$ imposed by the poling. It is left as a challenge to build this QPM into your code.

C. Gaussian beam propagation

In the absence of nonlinearity ($\eta = 0$) the paraxial wave equation for the fundamental field in Eqs. (8,9) has the Gaussian beam solution (Boyd Chap. 2.10.2)

$$A_1(r,z) = \frac{\mathcal{A}_1}{(1+2iz/b)} e^{-\frac{r^2}{w_0^2(1+2iz/b)}} \tag{12}$$

$$= \mathcal{A}_1 \left(\frac{w_0}{w(z)}\right) e^{i\Phi(z)} e^{-\frac{r^2}{w^2(z)}} e^{\frac{ik_1 r^2}{2R(z)}}, \tag{13}$$

with w_0 the focused Gaussian spot size at $z = 0$, $\mathcal{A}_1$ the peak on-axis field at focus, $b = k_1 w_0^2$ the confocal parameter, $w(z)$ and $R(z)$ are the z-dependent spot size and radius of curvature, and $\Phi(z)$ is the Gouy phase-shift.

(a) By comparing the two forms of the Gaussian solution obtain expressions for spot size $w(z)$ and Gouy phase $\Phi(z)$ in terms of w_0 and b.

(b) For a beam with $w_0 = 10 \times 10^{-6}$ m plot $\frac{w(z)}{w_0}$ and $\frac{\Phi(z)}{\pi}$ for $z = [-3b, 3b]$. Using your results illustrate that the beam remains almost focused over a confocal parameter around the origin, and there is close to a π phase shift over the same range.

II. SHG WITH A FOCUSED GAUSSIAN BEAM

The treatment is within the undepleted pump beam approximation. A full solution requires solving the paraxial wave equation for the SH field in Eqs. (8,9) with the fundamental field given by Eq. (12). This is done in Boyd Chap. 2.10.3 and here a summary is given: The general discussion in Sec. I.A indicates that $A_2 \propto A_1^2$ suggesting a SH field of the form

$$A_2(r,z) = \frac{\mathcal{A}_2(z)}{(1+2iz/b)} e^{-\frac{2r^2}{w_0^2(1+2iz/b)}}. \tag{14}$$

The final step extends Eq. (11) to allow for the on-axis variation of the fundamental field

$$\mathcal{A}_2(z) = i\eta\mathcal{A}_1^2 \underbrace{\int_{z_0}^{z} dz' \frac{e^{i\Delta k z'}}{(1+2iz'/b)}}_{J_2(\Delta k, z_0, z)}. \tag{15}$$

The underbraced term is what Boyd calls $J_q(\Delta k, z_0, z)$ in his Eq. (2.10.11b), with $q = 2$ for SHG. Even with $\Delta k = 0$ the Gouy phase shift precludes high SH conversion.

The variation of the SH conversion may be assessed in terms of the above solution. We consider the case of focusing at the center of the medium and set $z_0 = -L/2$ and $z = L/2$. Then defining the dimensionless parameters $\xi = L/b$, $\sigma = b\Delta k/2$, and $\zeta = 2z/b$, yields

$$J_2 = \frac{L}{2\xi} \int_{-\xi}^{\xi} \frac{e^{i\sigma\zeta} d\zeta}{(1+i\zeta)}. \tag{16}$$

The focusing parameter ξ is the ratio of the medium length to the confocal parameter: When $\xi < 1$ the beam is collimated over the medium. The phase-mismatch parameter σ is half the phase-shift difference between the fundamental and SH fields accumulated over a confocal parameter. The utility of this formalism is that the output SH power P_2 may be expressed in terms of the fundamental input power P_1 as [5]

$$P_2 = KLk_1 h(\sigma,\xi) P_1^2, \quad h(\sigma,\xi) = \frac{1}{4.27\xi} \left| \int_{-\xi}^{\xi} \frac{e^{i\sigma\zeta} d\zeta}{(1+i\zeta)} \right|^2, \tag{17}$$

where $K = \frac{2.14\omega_1^2 d_{eff}^2}{\epsilon_0 n_1^2 n_2 c^3 \pi}$, which depends on the material and field parameters, and the factor $h(\sigma,\xi)$ describes the dependence on geometry via the phase-mismatch parameter σ and focusing parameter ξ. The goal of the following is to explore how the geometrical factor $h(\sigma,\xi)$ can be optimized as opposed to evaluating the absolute conversion efficiency as was done in Sec. I.B for plane-waves. For example, for a fixed medium length L what focused spot size should be used for optimal conversion?

(a) Find the relation between the Gaussian spot sizes of the fundamental and SH fields, and show that they yield the same Rayleigh range (this is key to the above solution). It is important to note this difference in the spot sizes.

(b) Write a MATLAB code to calculate and plot the function $h(\sigma,\xi)$ in Eq. (17) versus $\sigma = [-3, 3]$ for $\xi = 2.84$. You may write your own routine using, for example, the trapezoidal rule, or use a

MATLAB supplied routine such as `integral`. You should find a maximum in the conversion for $\sigma > 0$, and that there is little conversion for $\sigma < 0$, so a first lesson for optimization is to have $n_1 > n_2$.

(c) Adapt your code from part (b) to plot the function $h(\sigma, \xi)$ versus $\xi = [1, 6]$ for $\sigma = 0.56$. You should find that there is a maximum in the conversion in this range, showing that optimization is desirable.

(d) The bigger question regards optimization in both parameters. To address this adapt your code to plot $h(\sigma, \xi)$ versus $\xi = [1, 4]$ and $\sigma = [0, 2]$ and display the results using `imagesc`. You should see a global maximum which corresponds to the optimized conditions. Compare the optimal conditions with those in parts (b) and (c).

(e) Consider a nonlinear medium of length $L = 3 \times 10^{-3}$ m. What are the required spot size of fundamental beam at the medium entrance and refractive-index difference $(n_1 - n_2)$ to obtain optimal SH conversion? Typically the refractive-index difference can be tuned by rotating the birefringent crystal or by controlling the medium temperature.

(f) Building on your answer from part (e) and using $d_{eff} = 15 \times 10^{-12}$ m/V, what is the value of E_{NL} appearing in the expression $P_2 = E_{NL} P_1^2$ [5], and the SH output power for a fundamental input power of 10 W? This will give you an idea of the conversion efficiency in the focused geometry.

Further exploration: Here we have tacitly assumed that the incident fundamental field and generated second-harmonic field propagate in the same direction, even though the two fields have different polarizations and the medium is birefringent: This is the limit of non-critical phase-matching (NCPM). More generally, medium birefringence causes the two fields to spatially separate, a phenomenon called walk-off, and critical phase-matching (CPM) must be used by eg. changing the angle of the crystal. The paper by Boyd and Kleinmann [2] treats the case including walk-off, and you can test your skills by investigating CPM. Finally, the analysis has also been extended to include elliptical instead of circular beams [6] as elliptical beams have been found to extend the efficiency of SHG in the presence of walk-off.

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