The inverse problem for second harmonic generation with amplitude-modulated pulses

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Abstract

For second harmonic generation, when restricted to one space dimension and pure amplitude modulation, we establish a direct relation between the incident pulse-shape of the fundamental wave and the asymptotic pulse-shape of the harmonic wave. In particular, we show that the inverse problem — i.e., to determine the input pulse-shape in order to achieve a required output pulse-shape — is much easier to solve than the forward problem. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

Second harmonic generation (SHG) has become a standard tool in experimental optics, such that there is practical interest to achieve optimal conditions for this process. For cw laser radiation, SHG is a well understood problem, its first experimental observation dating from almost four decades ago [1]. However, nowadays, in several situations one uses short-pulsed incident laser radiation [2]. Within this context, the theoretical description of this phenomenon is complicated by focussing conditions and by the walk-off of the pulses [3,4], and it requires the solution of partial differential equations for any specified conditions. Under the idealized conditions of only one space dimension and pure amplitude modulation SHG is a *solved problem* in the sense that the general solution is given explicitly by the Liouville equation [5,6]. In a previous paper, we used this solution and have reduced SHG with an arbitrary incident pulse at the fundamental frequency to the problem of solving an ordinary differential equation of Schrödinger type with the incident pulse shape playing the role of the Schrödinger *potential* [7]. In particular, we have shown that the pulse shapes of both waves change during the propagation in the medium, such that an asymmetrical incident wave may originate a symmetrical second harmonic pulse, and vice-versa.

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In the present letter we will establish a direct relation — a mathematical mapping — between the incident fundamental pulse-shape and the asymptotic harmonic pulse-shape which is approached in a sufficiently long medium. We address the following question: What a pulse shape must one take for the incident fundamental wave in order to get an asymptotic harmonic wave with a required pulse shape? Surprisingly this *inverse problem* is much easier to solve than the forward problem. Mathematically only one differentiation is required for the inverse problem while the forward problem requires to solve a Riccati equation (see Eq. (12) below).

This paper is outlined as follows: In Section 2 we discuss the general solution for SHG with amplitude-modulated pulses, following [7] with slight changes in notation due to our present purpose. In Section 3 we derive the relation between the incident pulse and the asymptotic second harmonic. In the following sections, we apply our formalism to some concrete pulse shapes (Section 4), and state our conclusions (Section 5).

2. Basic equations and a conservation law

We will use here slowly varying complex optical field amplitudes $B_j(x, t)$ defined in such a way that $|B_j|^2$ are the corresponding energy flux densities of the fundamental wave $(j = 1)$ and the harmonic wave $(j = 2)$. The connection to the electric field amplitudes A_j is given by $B_j = n_j \sqrt{\epsilon_0 c} A_j$, with n_j denoting the refraction numbers, ϵ_0 the vacuum dielectricity constant, and c the vacuum velocity of light. Then, in a second-order nonlinear medium, the interaction between two quasi-monochromatic plane electromagnetic waves with angular frequencies ω_1 and $\omega_2 = 2\omega_1$, is described by the two differential equations

$$\left(\partial_x + \frac{1}{v_1}\partial_t\right)B_1 \equiv \partial_1 B_1 = -\kappa B_2^* B_1,$$
$$\left(\partial_x + \frac{1}{v_2}\partial_t\right)B_2 \equiv \partial_2 B_2 = \kappa B_1^2, \tag{1}$$

where one-dimensionality in space has been assumed, namely the transverse structure can be neglected. We also assumed that the wave-numbers of both carrier waves fulfill the phase-matching condition

$k_2 = 2k_1$. The star denotes complex conjugation. x, t are laboratory space and time coordinates. Here we do not use characteristic coordinates. The symbols $\partial_j \equiv \partial_x + v_j^{-1}\partial_t$ are introduced as shorthand notations. The coupling constant κ is expressed as

$$\kappa = \frac{\pi \chi_{\text{eff}}^{(2)}}{n_1^2 \lambda_1 \sqrt{\epsilon_0 c}}. \tag{2}$$

In Eq. (2), $\chi_{\text{eff}}^{(2)}$ is the effective second-order susceptibility of the medium, and λ_1 the vacuum wavelength of the fundamental mode. For later use we introduce the group velocity mismatch

$$\frac{1}{v} = \frac{1}{v_2} - \frac{1}{v_1}. \tag{3}$$

Under the restriction to purely amplitude-modulated waves we may take the amplitudes B_j as real. With this assumption from the SHG equations (1) it is easy to derive the conservation law

$$\partial_2 \left(\partial_1 B_2 + \kappa B_2^2\right) = 0. \tag{4}$$

Consequently we get

$$\partial_1 B_2 + \kappa B_2^2 = f(t - x/v_2). \tag{5}$$

3. The input–output relation

Let us consider typical initial conditions for second harmonic generation, namely

$$B_1(0, t) = b_1(t), \qquad B_2(0, t) = 0, \tag{6}$$

equivalent to the *restricted Cauchy problem* as treated in [7]. From the evolution equations (1) it is clear that for an infinitely long medium the ground wave B_1 goes to zero and a freely propagating harmonic wave B_2 is generated,

$$\lim_{x \to \infty} B_2(x, t + x/v_2) = b_2(t). \tag{7}$$

Now we will see that the conservation law established in the preceding section leads to a simple relation connecting the input pulse $b_1(t)$ with the output pulse $b_2(t)$.

From the definition of ∂_1, ∂_2 together with (3) we have

$$\partial_t = \nu(\partial_2 - \partial_1). \tag{8}$$

Thus the combination of (5) with the second of Eqs. (1) leads to

$$\partial_t B_2 - \kappa \nu \left(B_1^2 + B_2^2 \right) = -\nu f(t - x/v_2). \tag{9}$$

Specification to $x = 0$ then gives

$$f(t) = \kappa b_1^2(t) \tag{10}$$

while the limit $x \to \infty$ leads to

$$\nu f(t) = -\partial_t b_2 + \kappa \nu b_2^2. \tag{11}$$

Summarizing we find

$$b_1^2(t) = b_2^2(t) - \frac{1}{\kappa \nu} \partial_t b_2(t). \tag{12}$$

This remarkably simple relation is connecting input and output. For given input b_1, $\int_{-\infty}^{\infty} b_1^2(t) < \infty$, we have to solve the Riccati equation with the limiting condition $b_2 \to 0$ for $t \to -\infty$. (The Riccati equation is equivalent to a Schrödinger-type equation, so that the connection to the former paper [7] can be seen.) Surprisingly the inverse problem is much simpler to be solved: When the output b_2 is given only one differentiation is required in order to find the input b_1. Note that the mapping $b_2(t) \to b_1(t)$ is a local one. That is, in order to get $b_1(t_0)$ at some fixed time point t_0 we only need to know the value $b_2(t_0)$ and its derivative at the corresponding time. From the initial condition (6) together with the second of the differential equations (1) it is seen that B_2 — and hence b_2 — cannot be negative. For any positive definite, continuous and piecewise differentiable output function $b_2(t)$ fulfilling

$$b_2^2(t) - (\kappa \nu)^{-1} \partial_t b_2(t) \geqslant 0, \tag{13}$$

the inverse problem is solved by (12). As an important consequence of the condition (13) $b_2(t)$ cannot vanish faster than $1/|t|$ for $t \to -\infty$ ($+\infty$) if $\nu > 0$ (< 0).

For a numerical specification we may consider SHG in LiIO$_3$. With values taken from [4], $\chi_{\text{eff}}^{(2)} = 2d_{\text{eff}} = 9.2 \times 10^{-12}$ m/V, $\nu = 1.2 \times 10^9$ m/s, $\lambda_1 = 780$ nm, $n_1 = 1.8$ we find

$$\kappa = 2.2 \times 10^{-5} \text{ W}^{-1/2},$$
$$\kappa \nu = 2.7 \times 10^4 \text{ W}^{-1/2} \text{ m/s}. \tag{14}$$

4. Examples of pulse shapes

By use of (12) — having in mind restriction (13) — one would be able to establish a catalogue of analytical input and related output pulse shapes. Instead, we will provide here not more than three examples, for which only $b_2(t)$ will be explicitly written. $b_1^2(t)$ then is easily available, but some of the related expressions are considerably messier. Thus it might be more instructive for the reader to look at the three figures.

It is convenient to put $\kappa \nu = 1$ corresponding to an appropriate scaling. That is, one may choose a proper unit of intensity and then measure the time in units of $t_{\text{unit}} = 1/(\kappa \nu \sqrt{P_{\text{unit}}})$. One may also introduce a characteristic length $L_{\text{char}} = 1/(\kappa \nu \sqrt{P_{\text{unit}}})$ with the meaning that for an energy flux density $P = P_{\text{unit}}$ the crystal length must be large compared to L_{char} in order to reach the asymptotic region. When, e.g., we choose $P_{\text{unit}} = 10$ GW/cm^2 then for our specified LiIO$_3$ example we would arrive at $t_{\text{unit}} = 3.8$ ps and $L_{\text{char}} = 4.5$ mm.

As the first example, we take a "flat-top" second harmonic

$$b_2(t) = \begin{cases} 1/\sqrt{1 + (t - 10)^2}, & t < 10, \\ \frac{1}{2}[1 - \tanh[2(t - 20)]], & t > 10, \end{cases} \tag{15}$$

which is approximately constant for $10 < t < 19$. This yields a ground wave which also shares this property, as can be seen in Fig. 1. Remarkable differences between input and output do appear only in the leading front and in the trailing edge. An important feature is the sharp peak at the trailing edge of the fundamental wave, which is responsible for the "flat-top" shape of $b_2(t)$. This characteristic is not present, for instance, in the asymptotic second harmonic originated by a square input pulse, for which the sharp peak is absent [7].

For a second harmonic

$$b_2(t) = \begin{cases} \frac{2(t-10)\tanh[4(t-10)]}{4 - (t-10)^3} - \frac{0.8(t-10)}{4 + (t-6)^2}, & t < 10, \\ 0, & t > 10, \end{cases} \tag{16}$$

however, the fundamental and harmonic waves may have completely different shapes. For instance, in Fig. 2 we show how a single-peaked second harmonic is related to a double-peaked fundamental. Similar double-peaked structures appear very frequently in the temporal [8] or spectral [8,9] profile of a pulse, so that
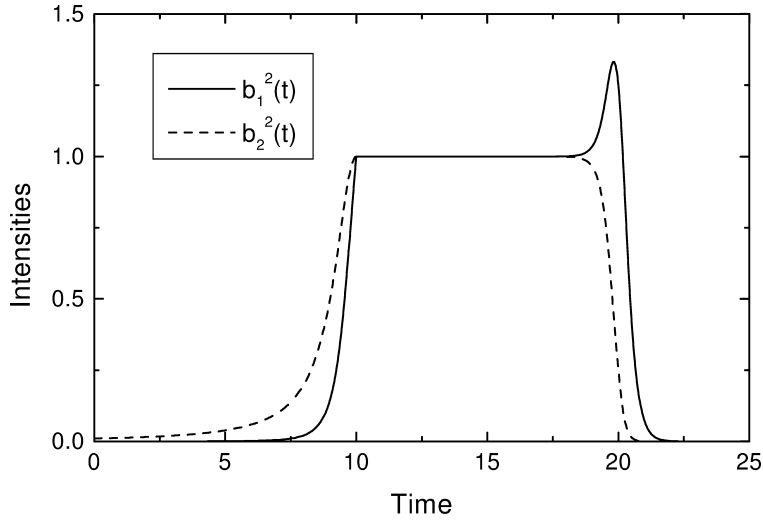
Fig. 1. "Flat-top" asymptotic second harmonic, given by Eq. (15) (dashed line) and corresponding initial ground wave (solid line). The energy flux densities b_j^2 and time t are measured in adjusted units P_{unit} and t_{unit}, see the text.
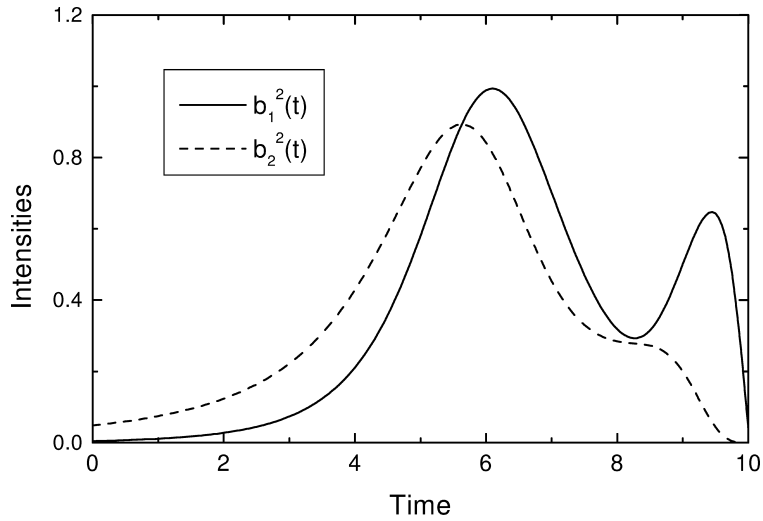

Fig. 2. A double-peaked second harmonic pulse generates a single-peaked second harmonic peak with a "shoulder" as it is given by Eq. (16).

controlling such features is of practical interest. In our third example, as shown in Fig. 3, both input and output pulses are periodically modulated. We take

$$b_2(t) = 1 + 0.375 \sin t. \tag{17}$$

As a further test, one can easily verify that, for the examples discussed in [7], the initial pulse shapes are recovered applying the input–output relation for the asymptotic second harmonic.

5. Conclusions

We provide an analytical relation for second-harmonic generation with short pulses connecting the as-
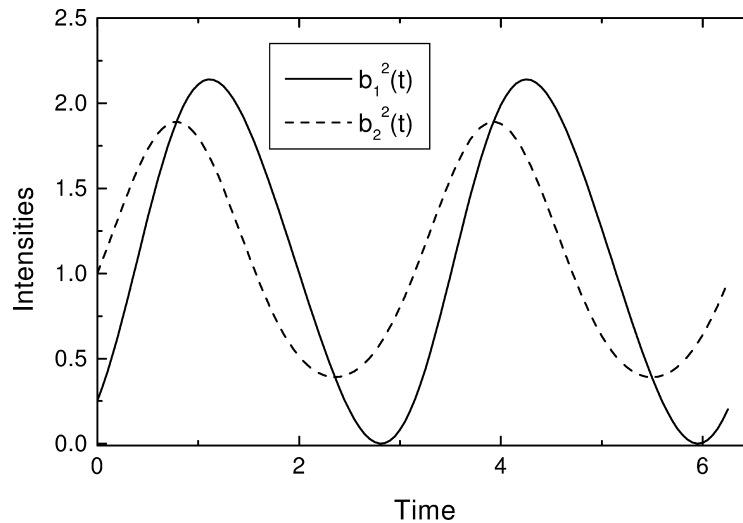
Fig. 3. Periodically modulated input and asymptotic output waves, where the harmonic output is given by Eq. (17). Note that the intensity of the fundamental wave reaches zero but that of the harmonic does not.

ymptotic second harmonic and the initial pulse at the fundamental frequency. This relation is valid for amplitude-modulated pulses of arbitrary shapes, and gives the corresponding input wave for a given asymptotic second harmonic. This relation has been obtained in form of a local transformation: in order to compute the fundamental wave at a given time, only the second harmonic at the same (then retarded) time together with its derivative are necessary. Thus the *inverse problem* — i.e., to find the input fundamental pulse shape from the required harmonic output pulse shape — considerably simpler than the direct problem, solved in a previous paper [7].

Acknowledgements

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