LA-UR-81-1228

TITLE: STIMULATED RAMAN SCATTERING AND FOUR-WAVE MIXING: STEADY-STATE PLANE-WAVE ANALYTIC SOLUTIONS

MASTER

AUTHOR(S): Jay R. Ackerhalt

SUBMITTED TO: Conference on Optics, 81

DISCLAIMER

By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes.

The Los Alamos Scientific Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy

University of California

LOS ALAMOS SCIENTIFIC LABORATORY

Post Office Box 1663 Los Alamos, New Mexico 87545

An Affirmative Action/Equal Opportunity Employer

Form No. 836 R3
St. No. 2629
12/78

UNITED STATES
DEPARTMENT OF ENERGY
CONTRACT W-7405-ENG-36

A311

Stimulated Raman scattering and four-wave mixing: steady-state plane-wave analytic solutions

Jay R. Ackerhalt
Theoretical Division, Los Alamos National Laboratory
Los Alamos, New Mexico 87545

Abstract

The equations for plane-wave steady-state propagation of n fields interacting via four wave processes in a Raman medium have been known since at least 1962. Complete analytic solutions have only been found for the process of harmonic generation. However, solutions have been obtained in the gain regime assuming zero pump depletion. Assuming all fields satisfy the phase matching condition we have found general analytic solutions to these equations. The solutions are complete in that they describe both pump deletion and saturation. Some example problems will be studied.

Introduction

In 1962 Armstrong, Bloembergen, Ducuing and Pershan[1] presented the equations governing plane-wave steady-state propagation in a non-linear medium. They derived explicit analytic solutions for second and third harmonic generation assuming the fields satisfied the phase matching condition, and indicated the procedure to be followed for higher order harmonic generation. In 1964 Platonenko and Khokhlov presented analytic solutions to the simplest problem which can exist in a Raman medium pump conversion to first Stokes. Both these analytic solutions contained the dynamics of the entire process allowing complete depletion of the initial pump fields and saturation of the generated final product fields. Since that time no new analytic solutions to these equations have been found which can describe pump depletion and/or saturation. For example Butvikin, et. al[2] in 1976 described the generation of the first anti-Stokes field assuming no back reaction of the anti-Stokes field on the pump-Stokes conversion process. Even with this approximation these solutions are very complicated Gauss hypergeometric functions and indicate the prevailing [illegible] that analytic solutions are sufficiently difficult or impossible to obtain and too complex to understand, necessitating the direct use of a computer to generate solutions.[3]

In this letter we would like to show that a new [illegible] to the original equations derived in 1962 for four field interactions can lead to [illegible] physically [illegible] analytic solutions for a very large group of problems when applied to a Raman active medium. Any arbitrary number of fields can be considered as long as all fields are phase matched and as long as each field has at least one other corresponding field with a frequency chosen to exactly [illegible] the two-photon Raman transition i.e., for example a problem with a pump, first-Stokes, second-Stokes, etc., or several different pump fields and their corresponding Stokes fields. We will also make some standard approximations and assumptions:

(1) The two-photon Rabi frequency is always sufficiently small such that there is no molecular dynamics. (Our equations will be written down explicitly for the case where all molecules are in their ground state.

(2) The plane wave monochromatic fields all propagate on a common z axis and are linearly polarized along the same x axis.

(3) All the fields' frequencies are far off resonance with intermediate state energy levels such that all the frequency denominators in the third order susceptibility $\chi^{(3)}$, which describes the medium's response for steady-state propagation, are approximately independent of the fields' frequencies.

The equations of motion generalized to include any number of fields interacting via Raman and four-wave interactions are

$$\frac{dA_m}{dz} = \gamma_m \left(A_{m-1}^2 - A_{m+1}^2 \right) A_m \tag{1}$$

$$+ \sum_i \left(A_i^* A_{i+1} A_{m-1} e^{i\Delta k_{m-1,i} z} - A_i A_{i+1}^* A_{m+1} e^{i\Delta k_{m,i} z} \right)$$

A311

where the terms independent of Δk are the usual Raman terms and the remaining terms are the four-wave mixing terms.[1] The frequency ω_m of the mth field is related to the wave vector k_m of the mth field in the standard way. The m, ℓ field pair phase mismatch $\Delta k_{m,\ell}$ is defined as

$$\Delta k_{m,\ell} = (k_m - k_{m+1}) - (k_\ell - k_{\ell+1}) \quad . \tag{2}$$

The ℓth electric field is given by

$$E_\ell(z,t) = \mathrm{Re}(A_\ell e^{i(k_\ell z - \omega_\ell t)}) \tag{3}$$

where A_ℓ is the complex amplitude of the field assumed here to be independent of time. The parameter β which is assumed here independent of the fields' frequencies is proportional to $\chi^{(3)}$. The indexing in (1) is chosen such that if A_m is a pump field, then A_{m+1} (A_{m-1}) is its corresponding first Stokes (first anti-Stokes) field. Successive higher (lower) indicies refer to successively higher Stokes (anti-Stokes) fields. If additional independent chains of fields are of interest in the problem, then the indexing can be easily generalized using two subscripts where the first one identifies the chain and the second identifies the field. The summation in (1) refers to a sum over all other pairs of fields whose frequency difference is resonant with the Raman transition. It is important to realize that this sum includes pairs of fields which may not appear in a common chain of higher Stokes or anti-Stokes fields due a single pump field, but exist due to the possible initial presence of additional pump fields. As we will see, energy is only transferred within individual chains, not between chains, but the overall propagation dynamics is determined by all fields. (We should notice here that the Raman terms in (1) can be naturally included in the four wave mixing terms by simply allowing the sum to include all field pairs. The exponential $\Delta k_{m,m}$ terms in this case are identically unity.)

Raman conversion (Example 1)

Instead of illustrating the general method of solution with equations (1) let us consider a simple exactly solvable example problem, pump conversion to first Stokes. Since there are only two fields in this problem let us refer to the pump and Stokes fields as P and S, respectively. The equations of motion describing this system are

$$\frac{dP}{dz} = i\omega_p |S|^2 P \tag{4a}$$

$$\frac{dS}{dz} = -i\omega_s |P|^2 S \quad . \tag{4b}$$

The standard approach to solving these equations is to use the condition

$$\frac{|P|^2}{\omega_p} + \frac{|S|^2}{\omega_s} = \text{constant} \tag{5}$$

which when substituted into (4a) for $|S|^2$ or (4b) for $|P|^2$ gives a single differential equation for P or S, respectively. These new equations are easily solved by direct integration.

Instead of using this standard method which is not suitable for solving the general problem let us rewrite (4) as

$$\frac{dP}{dz} = -\omega_p (PS^*)S \tag{6a}$$

$$\frac{dS}{dz} = \omega_s (P^*S)P \quad . \tag{6b}$$

Using (6) we can construct the equation of motion for the pair product PS^*

A311

$$\frac{d}{dz}(PS^*) = \beta(\omega_s|P|^2-\omega_p|S|^2)(PS^*) \tag{7}$$

which can subsequently be rewritten as

$$\frac{d}{dz}\ln(PS^*) = \beta(\omega_s|P|^2-\omega_p|S|^2) \quad . \tag{8}$$

If we now define some new variables

$$PS^* = \frac{d\theta}{dz}e^{i\phi} \tag{9}$$

where $\frac{d\theta}{dz}$ is real and equal to the magnitude of PS^*, then substitution of (9) into (8) gives

$$\frac{d\theta}{dz}\left(\ln\frac{d\theta}{dz} + i\frac{d\phi}{dz}\right) = \beta(\omega_s|P|^2-\omega_p|S|^2) \quad . \tag{10}$$

Equating real and imaginary parts shows that ϕ is a constant in z. The value of ϕ is chosen from (9) using the initial values for P and S and requiring that $\frac{d\theta}{dz}$ is real.

Substituting (9) into (6) gives

$$\frac{dP}{dz} = -\omega_p\frac{d\theta}{dz}T \tag{11a}$$

$$\frac{dT}{dz} = \omega_s\frac{d\theta}{dz}P \tag{11b}$$

where we have removed the phase ϕ by a redefinition of the Stokes field,

$$T = Se^{i\phi} \quad . \tag{12}$$

If we assume the fields depend on z only through the new variable θ, then (11) becomes

$$\frac{dP}{d\theta} = -\omega_p T \tag{13a}$$

$$\frac{dT}{d\theta} = \omega_s P \tag{13b}$$

which are two first-order linear coupled differential equations with constant coefficients. The solution of (13) can be obtained by standard methods,

$$P(\theta) = P_0\cos\theta - \frac{\omega_p T_0}{\Omega}\sin\theta \tag{14a}$$

$$T(\theta) = T_0\cos\theta + \frac{\omega_s P_0}{\Omega}\sin\theta \tag{14b}$$

where

$$\Omega = (\omega_p\omega_s)^{1/2} \tag{15}$$

and P_0 and T_0 without an explicit dependence on z refer to $z = 0$ ($\theta = 0$), our choice of initial conditions for z. The solution of the entire problem is now reduced to finding

A311

the solution of a sin[illegible] linear equation in θ,

$$\frac{d\theta}{dz} \doteq \frac{I}{2} \sin\left[2(\pi\Omega\theta+\xi)\right] \tag{16}$$

where we have assumed that the intial pump field is much greater than the initial Stokes field, $|P| \gg |S|$, and

$$I \equiv \frac{\Omega}{\omega_p} |P|^2 \tag{17}$$

$$\xi \equiv \frac{1}{2} \frac{(PT^*+P^*T)}{(\Omega/\omega_p)|P|^2} \ll 1 \tag{18}$$

Since the gain for the growth of the Stokes field as defined in the region $z \approx 0$ ($\theta \approx 0$), we can rewrite (16) as

$$\frac{d\theta}{dz} \approx I\pi\theta + I\xi \tag{19}$$

where the solution of (19) is

$$\theta \approx (e^{2\pi\omega_s |P|^2 z} - 1)(\xi/\pi\Omega) \quad . \tag{20}$$

For small θ (14b) becomes

$$T(z) \approx \frac{\pi\omega_s^2 P}{\omega_p} \theta + T \tag{21}$$

which after substituting of (20) reduces to

$$T(z) = Te^{2\pi\omega_s |P|^2 z} \tag{22}$$

where the gain for the Stokes amplitude is defined as

$$g = \pi\omega_s |P|^2 \quad . \tag{23}$$

Equation (16) can be integrated exactly

$$\tan \xi = \xi e^{gz} \tag{24}$$

where

$$F = \pi\Omega\theta + \xi \quad . \tag{25}$$

From (24) and (25) we observe that ξ is initially [illegible] at $\theta = 0$ ($z = 0$) and grows until [illegible] ≈ 2 ($z \approx$). Making the same approximations in (14) as was done to arrive at (16) allows (14) to to be written in the form

$$|P(z)|^2 = |P|^2 \cos^2 \xi \tag{26a}$$

$$|S(z)|^2 = \frac{\pi\omega_s}{\omega_p} |P|^2 \sin^2 \xi \tag{26b}$$

A311

where we have written (26) for the field intensities. Using standard trigonometric identities for $\tan^2\phi$ in terms of $\cos^2\phi$ or $\sin^2\phi$ allows (26) to be written in a more familiar form.

$$|P(z)|^2 = |P|^2 / \left(1 + \frac{\omega_p |S|^2}{\omega_s |P|^2} e^{2gz}\right) \tag{27a}$$

$$|S(z)|^2 = |S|^2 e^{2gz} / \left(1 + \frac{\omega_p |S|^2}{\omega_s |P|^2} e^{2gz}\right) \tag{27b}$$

where we have also used (12), (14) and (18). Equation (24) now allows us to easily define saturation, the point where the pump intensity is depleted to half its initial value, $\phi = \pi/4$,

$$gz_{SAT} = \ln\left(\frac{1}{\xi}\right) \tag{28}$$

While this method of solution for this simple problem seems very complicated, we should point out that this method is completely general in that the n field problem can be solved by this same approach. It is also interesting to notice that the exponential solutions (27) are expressed in terms of sine and cosine functions of a nonlinear variable in z. It is this concept of simple functions of a nonlinear variable which allows all these types of problems to exhibit simple solutions.

"Multipass cell" (Example 2)

Let us now consider a problem recently studied by Trutna, et. al[1] where k modes of a single pump field have a one-to-one correspondence with k modes of a first-Stokes field. Since this problem deals with multiple chains of field pairs, we will generalize our previous notation such that the ith chain's pump and Stokes fields are labeled P_i and S_i, respectively. After specifying the initial fields, γ is chosen such that the imaginary part of $\sum_i P_i S_i^* e^{-\gamma z}$ is zero. Using (12) the initial conditions for the fields S_i can be transferred into initial conditions for the constructed fields T_i. In this case (13) generalizes to a set of k pairs of differential equations with the solutions

$$P_i(\phi) = P_i \cos \phi_i \phi - \frac{\omega_i T_i}{\omega_i} \sin \phi_i \phi \tag{29a}$$

$$T_i(\phi) = T_i \cos \phi_i \phi + \frac{\omega_i P_i}{\omega_i} \sin \phi_i \phi \tag{29b}$$

where $P_i(T_i)$ is the initial value of the ith pump (Stokes) field with frequency ω_i (ω_i) such that

$$\omega_i = (\omega_i \omega_i)^{1/2} \quad . \tag{30}$$

At this point we observe from (29) that the pump-Stokes conversion proceeds in order of decreasing ω_i where the field pair with the largest frequency product goes first. By making the approximations that the initial pump field is much greater than the initial Stokes field ($P_i \gg S_i$) and that the modes are spaced sufficiently close ($1.1 \omega_i \omega_i$ .9 for all i,j)[2], Eq. (16) can be expressed as

$$\frac{d\phi}{dz} = \frac{1}{2} \sin [2(\phi + \phi)] \tag{31}$$

where

$$I_i = (\omega_i \omega_i) P_i^2 \tag{32}$$

A311

$$\xi_i = \frac{1}{2}\,\frac{(P_i T_i^* + P_i^* T_i)}{(\Omega_i/\omega_i)|P_i|^2} \ll 1 \tag{33}$$

$$I = \sum_i^k I_i \tag{34}$$

$$\langle\Omega\rangle = \sum_i^k I_i \Omega_i / I \tag{35}$$

$$\langle\xi\rangle = \sum_i^k \xi_i I_i / I \quad . \tag{36}$$

The gain for the field amplitudes, $\bar{g}$ can be defined in the region $\theta \sim 0$ ($z \sim 0$). By following the same procedure we used to go from (19)-(23) we find

$$\bar{g} = I\beta\langle\Omega\rangle = \beta\sum_i^k \alpha_i |P_i|^2 \quad . \tag{37}$$

The gain for each pump-Stokes pair is the same and equal to the sum of the gains for each individual pump-Stokes pair as if all the other fields were absent.[5] Therefore, a single pump-Stokes conversion can be substantially enhanced by the simple presence of other pump modes.

Equations (24), (25) and (28) now become

$$\tan\psi = \langle\xi\rangle e^{\bar{g}z} \tag{38}$$

$$\psi = \beta\langle\Omega\rangle\theta + \langle\xi\rangle \tag{39}$$

$$\bar{g}z_{SAT} \sim \ln\left(\frac{1}{\langle\xi\rangle}\right) \tag{40}$$

The dynamics for this problem are now complete. We know the order that the field mode pairs saturate from (29), and in conjunction with (40) we know the location of saturation for each pair.

If we now allow this process to occur in a multipass cell[1] whose cavity is defined by plane parallel mirrors, then we can account for the reflectivity of these mirrors[11] within the present framework of our equations (assuming all fields have the same mirror reflectivity). Since each field is reduced by the reflectivity R each time it is reflected at a mirror, we can observe from (31) that θ remains a continuous function of z. Therefore, we integrate (31) in single-pass steps. The result is to multiply the right side of (29) by $R^{n/2}$, i.e. $P_i(\theta) \to R^{n/2} P_i(\theta)$ and $T_i(\theta) \to R^{n/2} T_i(\theta)$, and to replace z in (38) and z_{SAT} in (40) by $((1-R^n)/(1-R))L$ where n is the number of passes in the cavity of length L. The overall effect of mirror losses is to simply extend the distance over which the dynamics occurs.

The general method of solving these types of problems has been published elsewhere.[11] We have used this forum to illustrate the method and give solutions to some example problems of current interest.

The author would like to thank Norman Kurnit and John Carlsten for suggesting the problem and for many valuable discussions.

References

1. J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev. 127, 1918 (1962).

2. V. T. Platonenko and R. V. Khokhlov, Zh. Eksp. Teor. Fiz. 46 555 (1964) [Translation: Sov. Phys. JETP 19, 378 (1964)].

A311

3. V. S. Butylkin, V. G. Venkin, V. P. Protasov, P. S. Fisher, Yu, G. Khroncpulo, and M. F. Shalyaev, Zh. Eksp. Teor. Fiz. 70, 829 (1976), [Translation: Sov. Phys. JETP 43, 430 (1976)].

4. D. von der Linde, M. Maier, and W. Kaiser, Phys. Rev. 178, 11 (1969), and Y. R. Shen and N. Bloembergen, Phys. Rev. 137, A 1787 (1965).

5. M. G. Raymer, J. Mostowski and J. L. Carlsten, Phys. Rev. A 19, 2304 (1979).

6. W. R. Truntna, Jr., Y. K. Park and R. L. Byer, IEEE JQE 15, 648 (1979).

7. This form of approximate solution was suggested to me by R. T Pack, private communication.

8. J. A. Giordmaine and W. Kaiser, Phys. Rev. 144, 676 (1966), see also ref. 8.

9. W. R. Trutna and R. L. Byer, Appl. Opt. 19 301 (1980); P. Rabinowitz, A. Stein, R. Brickman and A. Kaldor, Appl. Phys. Lett 35, 739 (1979); J. L. Carlstein and N. A Kurnit, Paper Q8, XI International Quantum Electronics Conference June 23-26, 1980 - Boston, Massachusetts.

10. We are neglecting several effects which cannot be dealt with within the framework of our equations: (1) beam overlap which leads to pulse shaping and gain effects and (2) focusing effects due to the cavity mirrors which are in reality convex.

11. Jay R. Ackerhalt, to be published in Phys. Rev. Lett. (April 6 issue 1981).