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Novel Analytic Solutions to General Four-Wave-Mixing Problems in a Raman Medium

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A new approach to the problem of steady-state plane-wave propagation in a Raman-active medium has lead to general analytical solutions. Any arbitrary number of fields can interact via both Raman and four-wave processes, but all fields must satisfy the phase-matching condition. The solutions will be illustrated with a problem of current interest; multiple first Stokes generation from a multiple-mode pump field.

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In 1962 Armstrong *et al.*[1] presented the equations governing plane-wave steady-state propagation in a nonlinear medium. They derived explicit analytic solutions for second- and third-harmonic generation when the fields are assumed to satisfy the phase-matching condition, and indicated the procedure to be followed for a higher-order harmonic generation. In 1964 Platonenko and Khokhlov[2] 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 have been found which can describe pump depletion and/or saturation. For example, Butylkin *et al.*[3] in 1976 described the generation of the first anti-Stokes field for which it is assumed that there is 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 philosophy 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.[4] In some cases where solutions describing actual experimental conditions are desired a computer may be the only path to a solution. However, in many cases the existence of analytic solutions would allow a valuable qualitative understanding of the physics of a process even if the physics is only absolutely valid for some more simplied model.

In this Letter, I would like to show that a new approach to the original equations derived in 1962 for four-field interactions can lead to simple physically transparent 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 match exactly 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. I will also make some standard approximations and assumptions:

(1) The two-photon Rabi frequency[5] is always sufficiently small such that there is no molecular

dynamics. (The 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

$$dA_m/dz = \beta\omega_m\{(|A_{m-1}|^2 - |A_{m+1}|^2)A_m + \sum_l [A_l^* A_{l+1} A_{m-1}\exp(i\Delta k_{m-1,l}z) - A_l A_{l+1}^* A_{m+1}\exp(-i\Delta k_{m,l}z)]\}, \quad (1)$$

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 $\Delta k_{m,l}$ is defined as

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

The lth electric field is given by

$$E_l(z,t) = \mathrm{Re}\{A_l \exp[i(k_l z - \omega t)]\}, \quad (3)$$

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) indices 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 by using two subscripts, where the first one identifies the chain and the second identifies the field. This multiple indexing will be necessary in the example. 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 to 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.]

The equations of motion assuming all fields satisfy the phase-matching condition, $\Delta k_{m,l} = 0$, are simply

$$dA_m/dz = \beta\omega_m[(\sum_l A_l^* A_{l+1})A_{m-1} - (\sum_l A_l A_{l+1}^*)A_{m+1}]. \quad (4)$$

Since the quantity $\sum_l A_l A_{l+1}^*$ is now no longer directly coupled via the phase mismatch, I can create its equation of motion from (4). One can easily discover that the phase, ψ, of this complex variable is a constant of the motion where

$$\sum_l A_l A_{l+1}^* \equiv |\sum_l A_l A_{l+1}^*| e^{i\psi}. \quad (5)$$

After defining a new variable θ where

$$d\theta/dz \equiv |\sum_l A_l A_{l+1}^*|, \quad (6)$$

One can substitute (5) and (6) into (4) obtaining

$$dA_m/dz = \beta\omega_m(d\theta/dz)(A_{m-1}e^{i\psi} - A_{m+1}e^{i\psi}). \quad (7)$$

By assuming all the fields depend on z only through θ and by making the transformation

$$A_m = B_m e^{-im\psi}, \quad (8)$$

I find

$$dB_m/d\theta = \beta\omega_m(B_{m-1} - B_{m+1}) \quad (9a)$$

$$d\theta/dz = \sum_l B_l B_{l+1}^*. \quad (9b)$$

Since $d\theta/dz$ is real, the constant ψ is specified by the initial conditions such that at $z=0$ the imaginary part of

$$\sum_l B_l B_{l+1}^* = \sum_l A_l A_{l+1}^* e^{-i\psi}$$

is identically zero. The solutions will then evolve such that this imaginary part is always zero. Since I can arbitrarily choose $\theta(0)=0$, I find that θ remains a real variable.

Equations (9a) are simply first-order linear coupled differential equations which can be easily solved by standard techniques for the fields B_m as a function of θ.[6] The solutions for the B_i's are simply sine and cosine functions making the qualitative dynamics transparent. However, the quantitative dynamics is complicated by the nonlinear dependence of θ on z. Each chain of higher Stokes and anti-Stokes fields generated from a single pump field forms a complete set of equations (9a) which are not coupled to other chains through equations (9a). Therefore, energy initially in a particular pump field always remains in its chain of higher Stokes and anti-Stokes fields, and is never transferred between chains. Equation (9b), however, takes the information in each chain and combines this information to determine the overall dynamics of all chains through the variable θ. For example, the simplest Raman process can have its dynamics advanced more rapidly toward conversion simply by the addition of other pump fields. In particular, I will show that the gain for each pump-Stokes conversion is now equal to the sum of the gains for each individual conversion process.[7] If we recall the original derivation of (1),[1] we can recognize that the bilinear product of fields in (9b) is essentially the two-photon Rabi frequency for the transition, and it is for this reason that the dynamics for *all* fields evolve through the quantity θ. After obtaining the solutions to (9a) we substitute them into (9b) which becomes a first-order nonlinear differential equation for θ. Since the equation is first order, it can be easily solved on the computer if necessary. However, I will illustrate our general results with a problem for which θ can be evaluated analytically.

Let us consider a problem recently studied by Trutna, Park, and Byer[8] 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 A_{i0} and A_{i1}, respectively. After specifying the initial fields, A_{ij}, ψ is chosen such that the imaginary part of $\sum_i A_{i0}A_{i1}{}^* e^{-i\psi}$ is zero. By using (8) the initial conditions for the fields A_{ij} can be transferred into initial conditions for the constructed fields B_{ij}. Since each chain only contains two fields, we will for convenience and clarity simply refer to the ith chain's pump and Stokes fields, B_{i0} and B_{i1}, by P_i and S_i, respectively, eliminating the need for dual indexing. In this case (9) is a set of k pairs of differential equations with the solutions

$$P_i(\theta)=P_i\cos(\beta\Omega_i\theta)-\frac{\omega_i S_i}{\Omega_i}\sin(\beta\Omega_i\theta) \tag{10a}$$

$$S_i(\theta)=S_i\cos(\beta\Omega_i\theta)+\frac{\Omega_i P_i}{\omega_i}\sin(\beta\Omega_i\theta), \tag{10b}$$

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

$$\Omega_i \equiv (\omega_i\alpha_i)^{1/2}. \tag{11}$$

At this point we observe from (10) 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 ($1.1 \gtrsim \Omega_i/\Omega_j \gtrsim 0.9$ for all i,j),[9] Eq. (9b) can be expressed as

$$d\theta/dz \sim (I/2)\sin[2(\beta\langle\Omega\rangle\theta+\langle\xi\rangle)], \tag{12}$$

where

$$I_i=(\Omega_i/\omega_i)|P_i|^2, \tag{13}$$

$$\xi_i=\tfrac{1}{2}(P_iS_i{}^*+P_i{}^*S_i)/[(\Omega_i/\omega_i)|P_i|^2]\ll 1, \tag{14}$$

$$I=\sum_i^k I_i, \tag{15}$$

$$\langle\Omega\rangle=\sum_i^k I_i\Omega_i/I, \tag{16}$$

$$\langle\xi\rangle=\sum_i^k \xi_i I_i/I. \tag{17}$$

Since the gain for the field amplitudes, g, can be defined in the region $\theta\sim 0$ ($z\sim 0$) one can expand (12) holding only the lowest term proportional to θ and identify the gain as the coefficient of this term as g is the rate of exponential growth for θ, and therefore the fields (10):

$$g=I\beta\langle\Omega\rangle=\beta\sum_i^k\alpha_i|P_i|^2. \tag{18}$$

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.[7] Therefore, a single pump-Stokes conversion can be substantially enhanced by the simple presence of other pump modes.

Equation (12) can be integrated in closed form:

$$\tan\Phi=\langle\xi\rangle e^{gz}, \tag{19}$$

where

$$\Phi=\beta\langle\Omega\rangle\theta+\langle\xi\rangle. \tag{20}$$

From Eqs. (19) and (20) we see that Φ is initially $\langle\xi\rangle$ at $\theta=0$ and grows until $\Phi=\pi/2$. If I define saturation as the point (z_{sat}) where the pump modes on the average are depleted to half their initial intensity, then $\tan\Phi\sim 1$ giving

$$gz_{sat}=\ln(1/\langle\xi\rangle). \tag{21}$$

The dynamics for this problem are now complete. We know the order that the field mode pairs saturate from (10), and in conjunction with (21), we know the location of saturation for each pair. Because of the approximation made in going from (9b) to (12), I have simplified the solution to this complex multimode problem such that it appears identical in form to the simple Raman problem.[2] If I restrict these multimode solutions to represent a single pump-Stokes pair, then these solutions are exact, and are in agreement with Ref. 2 after using some trigonometric identities.

I have shown that steady-state plane-wave propagation of phase-matched fields in a Raman medium can be completely described by two equations. The first, a set of first-order linear coupled equations, are easily solved by standard methods; and the second, a first-order nonlinear equation, may be analytically integrated or easily solved by using a computer. These equations account for both pump depletion and saturation. In a longer paper, I will further illustrate the method by studying two additional problems: higher Stokes generation and first anti-Stokes generation. In addition, I will show that both a linear absorption and/or a mirror reflectivity (as one has in a multiple-pass cell) can be included analytically in the solutions with a very simple extension of the method.

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