

Efficient Frequency Conversion by Stimulated Raman Scattering

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Frequency conversion by parametric processes in stimulated Raman scattering is analyzed in the limit where the Raman shift is much smaller than the laser frequency. An exact solution is obtained which shows that the conversion efficiency to the higher orders may be large. The conditions for obtaining high efficiency are discussed.

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Recently there has been some renewed interest in stimulated Raman scattering (SRS) as a technique of frequency conversion.¹⁻⁷ Several groups have observed significant power in the higher-order anti-Stokes lines of hydrogen to the extent that the technique promises new and useful bright ultraviolet sources with the tunability and beam quality of visible or near infrared lasers. In a typical experiment a monochromatic laser is focused into a cell containing, for example, hydrogen gas, and emerges, perhaps after several passes through the cell, accompanied by several sidebands displaced from the incident frequency by multiples of the Raman shift. The measured conversion efficiency to the higher-order anti-Stokes lines is typically $\sim (0.1)^n$ for the n th-order wave, and significant power has been seen in the sixth-order waves, and beyond. In this Letter it is shown that in this process all the orders are mutually coupled together in a *multiwave parametric process* consisting of multiple four-wave interactions, in which each wave is coupled to all the other waves. The equations describing this multiwave process are solved exactly in the case where the total bandwidth is small in relation to the laser frequency. In this limit, the maximum conversion efficiency to the n th-order wave is approximately $1/n$. Thus very high conversion efficiencies appear to be possible with this technique.

Consider the following experiment. A set of overlapping parallel monochromatic plane waves propagates through a dispersionless Raman medium. Each wave is separated from its neighbor in frequency by the Raman shift ν_R . The waves are labeled j ; the wave with $j=0$ is the pump, and the anti-Stokes orders have $j \geq 1$. The frequency of the j th wave is

$$\nu_j = \nu_0 + j\nu_R \quad (1)$$

and its wave vector is

$$\vec{k}_j = (2\pi\nu_j/c)\vec{z}, \quad (2)$$

where \vec{z} is a unit vector along the z direction. As

the waves propagate, the medium responds by developing a polarizability which oscillates at ν_R . This modulation causes energy to be transferred among the waves and also generates new sidebands. The process is exactly analogous to ultrasonic wave modulation,⁸ except that in SRS the amplitude of the modulation depends on both the strength of the electric fields and the dynamics of the molecule in the presence of these fields.

The Maxwell-Schrödinger equations for this system may be reduced to the standard Maxwell-Bloch (MB) form. In general, the fields may be expressed as

$$\mathcal{E}_j = \frac{1}{2}[E_j \exp i\omega_j(t - z/c) + \text{c.c.}], \quad (3)$$

where E_j is slowly varying. Then the MB equations are as follows:

$$\partial E_j / \partial z = (\nu_j / \nu_0)(\beta^* E_{j+1} - \beta E_{j-1}), \quad (4)$$

$$\partial \vec{r} / \partial \xi = \vec{\gamma} \times \vec{r} - \Gamma(\vec{r} - \vec{r}_0), \quad (5)$$

where $\xi = t - z/c$ is the running time of the pump and \vec{r} is the Bloch vector in the rotating frame.⁹ We also define a Rabi frequency, and phase mismatch as follows:

$$\Omega e^{i\theta} = \sum_r 2\alpha_{12} E_r E_{r-1}^* / \hbar, \quad (6)$$

where α_{12} is the transition polarizability. Then the remaining parameters are

$$\vec{\gamma} = (\Omega, 0, \Delta + i\dot{\theta}), \quad (7)$$

$$\vec{r}_0 = (0, 0, -1), \quad (8)$$

$$\Gamma = \text{diag}(0, 1/T_2, 1/T_1), \quad (9)$$

and

$$\beta = [k_0 N \alpha / 2\epsilon_0] (r_1 - ir_2) e^{i\theta}. \quad (10)$$

Here N is the molecular density, Δ is the detuning (including the Stark shifts), and $T_{1,2}$ are relaxation times.¹⁰ It is interesting to compare the Rabi frequency, Ω , in Eq. (6) with the Rabi frequency of the standard MB equations, $\Omega = \mu E$, for a two-level atom making transitions via a single-

photon process. The two-photon, multiple-pair nature of the atomic transition is clearly evident in Eq. (6).

Now the MB equations form an infinite variable problem. However, the problem splits into two parts: the molecular dynamics and the propagation or conversion dynamics. From Eqs. (4) and (6), we find

$$(\partial/\partial z)(\Omega e^{i\theta}) = 2i(\nu_R/\nu_0)\beta(\Omega_0^2 + \Omega^2)^{1/2}, \quad (11)$$

where Ω_0 is a function of ξ only. Now the molecular dynamics is formulated only in terms of \vec{r} , Ω , and θ ; the full complexity of the infinite-field problem has been removed. Solving the dynamical equations for \vec{r} gives β and then the solution of (4) gives the conversion efficiencies. In this Letter, we will carry out this procedure for a simple molecular system, in order to demonstrate the potential for high conversion efficiency in this process. This simple model appears to describe the process in H_2 gas quite well.

The simplest molecular model assumes that there is no molecular excitation, $r_3 \simeq -1$, and that the bandwidth is small, $\nu_R/\nu_0 \sim 0$. From (11), it follows that Ω and θ are independent of z . The solution for β is

$$\beta = \frac{1}{4}i\gamma_R \epsilon_0 c \int_{-\infty}^{\xi} d\xi' T_2 \exp[-(\xi - \xi')/T_2] \times \sum_r E_r^{in}(\xi') E_{r-1}^{in}(\xi')^*, \quad (12)$$

where γ_R is the steady-state exponential gain coefficient (units of centimeters per watt) and the fields are evaluated at the input to the medium, $z=0$. Then the solution of Eq. (4) for the fields is easily obtained:

$$E_n(z, \xi) = \sum_p e^{i(n-p)\varphi} E_p^{in}(\xi) J_{p-n}(z|\beta|), \quad (13)$$

where φ is the phase of β and J_n is a Bessel function. The parameter $z|\beta|$ (the parametric gain) determines the output spectrum, which consists of a number of waves, the bandwidth being increased from the input bandwidth by approximately $(2z|\beta|)\nu_R$. Each wave in the output has the same phase angle φ between itself and its lower neighbor. In the interesting case where the input contains just two waves, a pump and a weak Stokes or anti-Stokes line, the intensity in the n th order (Stokes or anti-Stokes) is

$$I_n(\xi) = I_0(\xi) J_n^2(z|\beta|). \quad (14)$$

Figure 1 plots the conversion efficiency $\eta = J_n^2(z|\beta|)$ as a function of its argument. Clearly high conversion efficiencies are predicted by Eq.

(14). However, for some combinations of input waves β may vanish; conversion is not guaranteed in all cases.

To apply this analysis to the case where just one wave is injected into the cell, a model for the parametric gain, $z|\beta|$, is required. In the single-wave input case, the fields in the focus are approximated by plane waves, and the Stokes wave grows initially from quantum noise and has a large solid angle. That part of the Stokes wave which is phase matched with the pump with respect to parametric generation of anti-Stokes or second Stokes radiation eventually grows strong enough to initiate the parametric process. A reasonable evaluation of the appropriate input intensities for the present analysis might be to take that value of the Stokes intensity at the output corresponding to the threshold for the parametric process. Although the Stokes input is amplified noise, its phase is strongly correlated with the pump, and has no memory of the initial random source.^{11,12} Only that part of the initial noise which reproduces the pump phase is amplified. Thus the appropriate input for the parametric analysis is

$$E_S^{in}(\xi) = bE_P^{in}(\xi). \quad (15)$$

For a flat pump pulse, the parametric gain is

$$z|\beta| = \gamma_R I_0 z b [1 - \exp(-\xi/T_2)] \quad (16)$$

and may be significantly less than the nonparametric gain, $\gamma_R I_0 z$. The output of the single input experiment consists of a non-phase-matched component, which cascades down the Stokes ladder,¹³ and a phase-matched component, which is described by the Bessel function solution. Because the parameter b is not easily evaluated, there is some ambiguity in comparing this analysis with the single-input experiments. However, if b is small enough so that the parametric gain is $z|\beta| \sim 0.1$, then the energy in each order varies as

$$\eta_n = I_n/I_0 = (z|\beta|)^{2n}/(2n!)^2 \quad (17)$$

in rough agreement with the experimental data.¹⁻⁷

The multiwave theory given here is related to the standard theory of SRS through the phase-matching conditions. In a nondispersive medium, all the waves are phase matched in the forward direction. On the other hand, dispersion causes the phase-matched waves to develop in cones, reducing the gain length for the higher orders. However, for the two-wave process, where the pump couples only to the Stokes wave, and so on

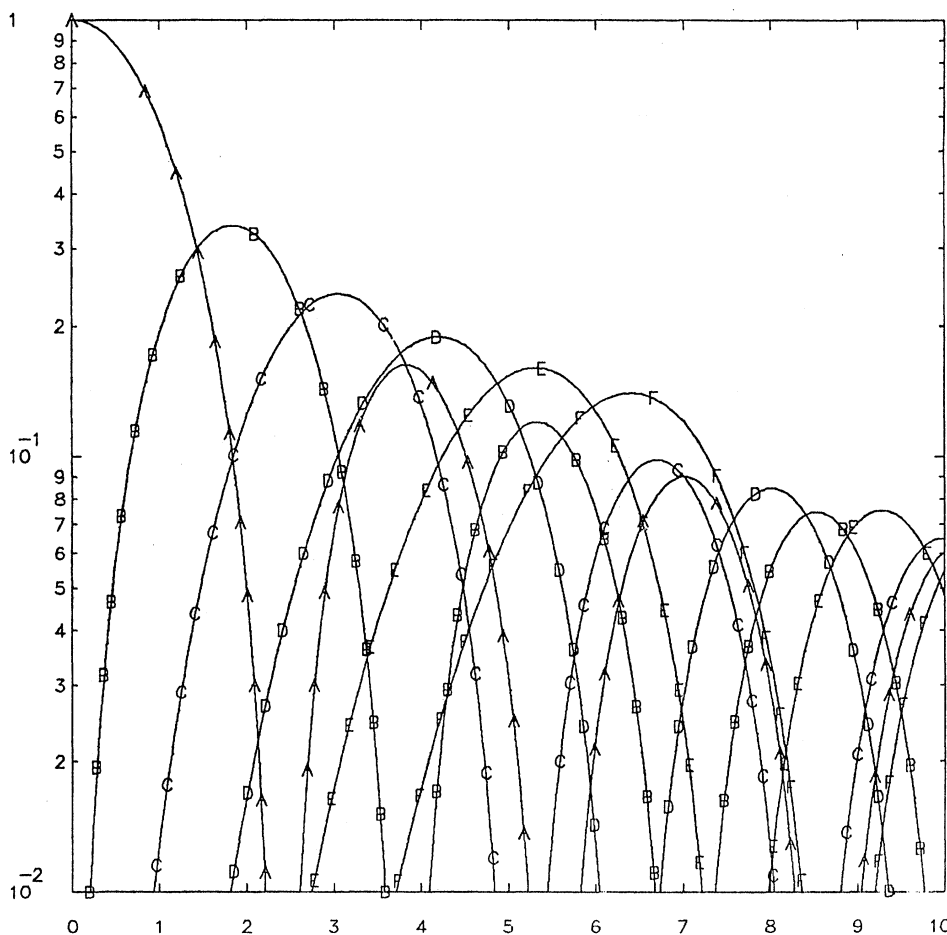


FIG. 1. Conversion efficiency η_n [see Eq. (14)] as a function of the parametric gain, $z|\beta|$. The pump is labeled "A," the first Stokes and anti-Stokes waves "B," the second sidebands "C," etc. Note that the peak value of $\eta_n \sim 1/n$.

in a cascade, there is no phase-matching condition. Thus in a sufficiently dispersive medium the parametric process is dominated by the two-wave (non-phase-matched) process. This is the standard situation. In hydrogen gas, both processes may occur. In the other experiment, where a comb of frequencies is injected into the cell, dispersion gives rise to a finite coherence length, limiting the gain, and tending to suppress the sidebands. Now, efficient sideband generation requires that the cascade process be dominated by the parametric process. Consequently the medium must be shorter than the coherence length and the cascade gain must be small enough to prevent build up of the non-phase-matched components from noise. The cascade gain exponent calculated from (14) is approximately

$$G_c = (I_0/I_{-1})^{1/2} \ln(z\beta) \quad (18)$$

and the condition that this be small enough to suppress the cascade process, namely $G_c \lesssim 30$, is easily met. Consequently, the parametric process is capable of dominating the cascade process for two-wave input, but not for single-wave input. The interpretation of the single-input experiments is complicated by the cascade process for the Stokes orders. However, the anti-Stokes orders are generated purely by the parametric process and consequently the present analysis applies to them. In the single-input experiments the cascade process causes strong deviations from the symmetry between Stokes and anti-Stokes waves implied by Eq. (14). Note also that the cascade process generates output with a large angular divergence, whereas the parametric process generates tightly focusable output (from focusable input) through the phase-matching condition.

In conclusion, we have shown that frequency

conversion using SRS is potentially a very efficient process. The generation of useful radiation in the sidebands requires suppression of the cascade process, and the conversion efficiency to the n th order is limited only by the shift ratio (ω_R/ω_0) and the input spectrum. For example, if this limit ($\nu_R/\nu_0 \sim 0$) is not taken, then one can show that the parametric gain is bounded¹⁴:

$$z|\beta| < (\nu_0/\nu_R) [\sum_r E_r E_{r-1}^* / \sum_r |E_r|^2]. \quad (19)$$

Thus for large bandwidth (e.g., vibrational SRS in hydrogen) the parametric gain and hence the conversion efficiency to high orders is limited. A fuller account of the process and further exact solutions for more general molecular dynamics and propagation where ω_R/ω_0 is not small will be published elsewhere.¹⁴

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Possibility of Achieving Ignition in a High-Field Ohmically Heated Tokamak

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There exists a regime in parameter space where a small high-field Ohmically heated tokamak may be capable of reaching thermonuclear ignition. Results of numerical simulations of the minimum ignition conditions are presented, including empirical, sawtooth, and magnetic-field ripple diffusion and the effects of impurities. An ignition condition is derived and compared with the results of the numerical simulations.

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It has been suggested that thermonuclear ignition may be achievable in a small high-field tokamak¹ with or without auxiliary heating provided by compression, rf, and/or neutral-beam auxiliary heating.² Here, with full transport-model numerical simulations, I confirm the accuracy of these analytic estimates and point out that recent engineering calculations³ and experimental and theoretical results suggest the feasibility of achieving ignition with Ohmic heating alone. This would have vast advantages in terms of the simplicity of

a proof-of-principle experiment and in terms of the economic viability of reactor concepts emerging from this approach.³

The severe engineering constraints of materials stress and heat removal in the toroidal field and Ohmic heating magnets dictate a window in size for the major radius $0.6 \lesssim R \lesssim 0.9$ m. Within these dimensions, toroidal magnetic fields of 16 to 20 T appear feasible.³ Accordingly, I have investigated numerically the minimum ignition conditions at radii lying in this "window."