Raman conversion using crossed broadband pump beams and bisecting Stokes

D. Korff and E. Mazur*

North East Research Associates, Inc., Woburn, Massachusetts 01801

C. Duzy and A. Flusberg

Avco Research Laboratory/Textron, Everett, Massachusetts 02149

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The effects of stimulated Raman conversion under the conditions of crossed pump beams and a bisecting injected Stokes are examined. It is found that the beam quality of the emerging Stokes wave and the effective gain can be characterized by the following two parameters: the temporal coherence of the crossed pump beams and the angle between the pumps.

INTRODUCTION

The problem of Raman "beam cleanup" has recently attracted considerable attention.[1,2] Technically a misnomer, this term refers to stimulated Raman scattering of a high-beam-quality (near-diffraction-limited) Stokes wave by an aberrated pump wave. If the Stokes wave amplifies at the expense of the pump wave without aberrating, it is said to have undergone Raman cleanup. In the case of a monochromatic pump, Raman beam cleanup is indicated by the right-hand side (rhs) of the Stokes equation in the slowly varying amplitude approximation[3]:

$$\partial_z \mathcal{E}_s + (2ik_s)^{-1}\nabla_\perp{}^2\mathcal{E}_s = g|\mathcal{E}_p|^2\mathcal{E}_s/2. \tag{1}$$

In Eq. (1), k_s is the wave vector of the Stokes, $|\mathcal{E}_p|^2$ represents the pump intensity, $\mathcal{E}_s$ is the (complex) Stokes amplitude, and g is the gain (in centimeters per watt). $\mathcal{E}_s$ and $\mathcal{E}_p$ are assumed to be collinear. We note that the phase of the pump does not enter the rhs of the equation. In what follows, we assume that the pumps are broadband and the Raman band width is less than the laser mode separation. We likewise assume near-uniform intensity variation of the pump. If the pump is not uniform, effects such as anomalous Raman dispersion[4] and transient refractivity[5] can become important. In the case of a broadband pump (many longitudinal modes), the Stokes equation is generalized to[6]

$$\partial_z \mathcal{E}_{sj} + (2ik_s)^{-1}\nabla_\perp{}^2\mathcal{E}_{sj} = (g/2)\mathcal{E}_{Pj}\sum_k \mathcal{E}^*{}_{pk}\mathcal{E}_{sk}\exp(i\alpha_{jk}z), \tag{2}$$

where $\mathcal{E}_{sj}$ is the amplitude of the jth mode and

$$\alpha_{jk} = (k_{pj} - k_{pk})\left(1 - \frac{n_s}{n_p}\right), \tag{3}$$

where n_s and n_p are the refractive indices at the Stokes and the pump wavelengths, respectively. If we restrict ourselves to the lowest laser transverse mode and if all longitudinal modes have the same transverse wave front,[7] then one suspects that there may again be no transfer of phase information from the pump laser to the Stokes. This argument is ultimately a ray-optics argument since it ignores the effects of the diffraction. Diffraction can convert pump phase to pump intensity, which can have profound effects on the Stokes wave. The ray-optics domain of validity can be characterized by saying that the variation of the pump phase over a Fresnel zone (as viewed from the end of the Raman cell) is small compared with a radian. We shall henceforth assume this to be the case.[8]

Furthermore, for most cases of interest, the dispersion term on the rhs of Eq. (2) can be neglected. We may then write Eq. (2) in the form (now neglecting diffraction)

$$\partial_z|S\rangle = |P\rangle\langle P|S\rangle, \tag{4}$$

where we have adopted the bra and ket notation from quantum mechanics.[9] If N is the total number of modes,

$$|P\rangle = \begin{pmatrix} P_1 \\ \vdots \\ P_N \end{pmatrix}, \qquad \langle P| = (P^*{}_1 \ldots P^*{}_N), \tag{5}$$

where similar definitions apply to the Stokes and

$$P_i = (\omega_s\chi''{}_R/n_s c)^{1/2}\mathcal{E}_{pi} = (g/2)^{1/2}\mathcal{E}_{pi}, \tag{6a}$$

$$S_i = (\omega_p\chi''{}_R/n_p c)^{1/2}\mathcal{E}_{si}. \tag{6b}$$

In Eqs. (6a) and (6b) ω_s and ω_p are the angular frequencies of the Stokes and pump lasers at line center, c is the speed of light in vacuum, and $\chi''{}_R$ is the real part of the third-order polarizability. Equation (4) can be augmented by a corresponding pump equation and the combined system solved,[10] but we shall treat only the pump-nondepletion limit here. A formal solution of Eq. (4) can be written in the form ($|S_0\rangle$ is the Stokes at $z = 0$ and $P^2 \equiv \langle P|P\rangle = \sum |P_i|^2$):

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$$|S\rangle = |S_0\rangle + [\exp(z|P|^2) - 1] \frac{|P\rangle \langle P|S_0\rangle}{\langle P|P\rangle}$$

$$= \exp(z|P|^2)|P\rangle \frac{\langle P|S_0\rangle}{\langle P|P\rangle} + \left(1 - \frac{|P\rangle \langle P|}{\langle P|P\rangle}\right)|S_0\rangle, \quad (7)$$

showing the well-known result that the gain is the broadband Raman gain. Equation (7) also shows that it is only the component of the Stokes along the pump (in the Hilbert-space[11] sense) that amplifies; the orthogonal component does not grow.

MISALIGNED SINGLE-PUMP-STOKES SYSTEM

If the pump and Stokes beams have an angular offset, new complications arise. For sufficiently large mismatch angles, amplified spontaneous Raman conversion will occur. This arises since, as we shall see, the broadband gain decreases with increased mismatch angle. Eventually a point is reached at which the amplified wave at the higher gain (along the pump direction) dominates the injected Stokes at the lower gain (along the injected-Stokes direction). We restrict ourselves to the angular region where Raman-amplified spontaneous emission is not significant.

If the pump and Stokes are misaligned, then the rhs of Eq. (2) must be modified. The rhs of Eq. (2) becomes (without dispersion)

$$\text{rhs} = (g/2)\mathcal{E}_{Pj} \sum_k \mathcal{E}^*_{pk}\mathcal{E}_{sk} \exp\{i(k_{pj} - k_{pk}) \times [(\cos\psi - 1)z + \sin\psi x]\}, \quad (8)$$

where the propagation vectors of the pump and the Stokes lie in the xz plane, with the Stokes taken along the z axis, and ψ is the angle mismatch. The effect of the z exponentials is to reduce the gain—the effect of the x exponentials is to reduce the beam quality. Conditions for high gain with good beam quality can be found by simply requiring the exponential terms to be negligible. (We use similar techniques with crossed pump beams as well.) From the form of Eq. (8), we have (we assume that $\psi \ll 1$)

$$(\Delta k\psi^2 L_c)/2 \ll 1 \quad (9)$$

(vanishing of the z exponential) and

$$(\Delta k\psi D) \ll 1 \quad (10)$$

(vanishing of the x exponential). In inequalities (9) and (10), Δk is the bandwidth of the pump laser, L_c is the conversion length ($\sim\langle|P|^2\rangle^{-1}$), and D is the beam diameter.

CROSSED PUMP BEAMS WITH BISECTING STOKES

The geometry that we consider is outlined in Figs. 1 and 2. The angle between the pump beams is 2θ, and the incident Stokes is assumed to bisect the pump beams. We choose our coordinate system such that the z axis makes an angle Ω with the incident Stokes. We may represent the pumps as

$$P_1 = \sum_n P_{1n} \exp\{ik_{pn}[z\cos(\Omega + \theta) - x\sin(\Omega + \theta)]\}, \quad (11)$$

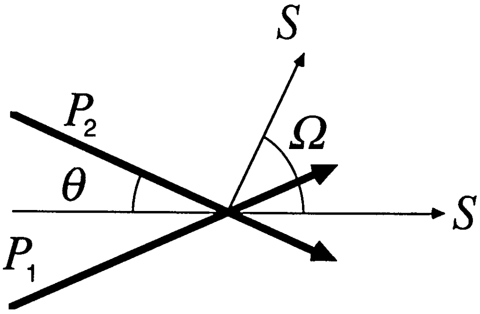

Fig. 1. Geometry for stimulated Raman scattering with two pump beams P_1 and P_2 and an incident Stokes beam S^{inc} bisecting the pump beams. S represents radiation scattered at an angle Ω with respect to the incident Stokes beam.

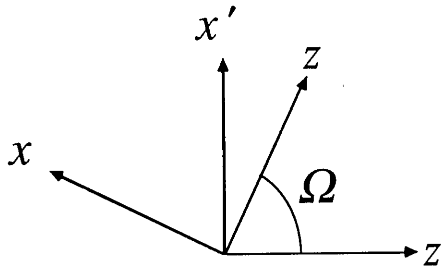

Fig. 2. The coordinate system for the problem. The z axis is in the direction of the scattered Stokes. The z' axis is in the direction of the incident Stokes. The unprimed coordinate system is used throughout.

$$P_2 = \sum_m P_{2m} \exp\{ik_{pm}[z\cos(\Omega - \theta) - x\sin(\Omega - \theta)]\}. \quad (12)$$

The scattered Stokes at the angle Ω can be written as

$$S = \sum_l S_l \exp(ik_{sl}z) \quad (13)$$

and the Stokes in the incident direction as

$$S^{\text{inc}} = \sum_q S_q^{\text{inc}} \exp[ik_{sq}(z\cos\Omega - x\sin\Omega)]. \quad (14)$$

Generalizing Eqs. (2) and (8) to include crossed Raman beams now yields

$$\partial_z S_n = \sum_{r=1}^{4} B_{rn}, \quad (15)$$

where

$$B_{1n} = P_{1n} \sum_m P^*_{1m} S_m^{\text{inc}} \times \exp(i\{k_{pn}[z\cos(\Omega + \theta) - x\sin(\Omega + \theta)] - k_{pm}[z\cos(\Omega + \theta) - x\sin(\Omega + \theta)] + k_{sm}(z\cos\Omega - x\sin\Omega) - k_{sn}z\}), \quad (16)$$

$$B_{2n} = P_{2n} \sum_m P^*_{2m} S_m^{\text{inc}} \times \exp(i\{k_{pn}[z\cos(\Omega - \theta) - x\sin(\Omega - \theta)] - k_{pm}[z\cos(\Omega - \theta) - x\sin(\Omega - \theta)] + k_{sm}(z\cos\Omega - x\sin\Omega) - k_{sn}z\}), \quad (17)$$

$$B_{3n} = P_{1n} \sum_m P^*_{2m} S_m^{\text{inc}} \times \exp(i\{k_{pn}[z\cos(\Omega+\theta) - x\sin(\Omega+\theta)] - k_{pm}[z\cos(\Omega-\theta) - x\sin(\Omega-\theta)] + k_{sm}(z\cos\Omega - x\sin\Omega) - k_{sn}z\}), \quad (18)$$

$$B_{4n} = P_{2n} \sum_m P^*_{1m} S_m^{\text{inc}} \times \exp(i\{k_{pn}[z\cos(\Omega-\theta) - x\sin(\Omega-\theta)] - k_{pm}[z\cos(\Omega+\theta) - x\sin(\Omega+\theta)] + k_{sm}(z\cos\Omega - x\sin\Omega) - k_{sn}z\}), \quad (19)$$

We will find that the B_1 and B_2 terms are inherently different from B_3 and B_4. Consider just the x exponential in B_3. This vanishes when

$$-k_{pn}(\theta+\Omega) + k_{pm}(\Omega+\theta) - k_{sm}\Omega = 0. \quad (20)$$

Ignoring intrapump and intra-Stokes wave-vector differences, this leads to the condition

$$\Omega = \Omega_- \equiv -\frac{2k_p\theta}{k_s}. \quad (21)$$

Similarly, the x exponential in B_4 vanishes when

$$\Omega = \Omega_+ \equiv \frac{2k_p\theta}{k_s}. \quad (22)$$

There is a simple interpretation of these angles. The crossed pumps form a grating with period λ_g,[12] where (λ_p is the pump wavelength)

$$\lambda_g = \frac{\lambda_p}{2\theta}. \quad (23)$$

The Stokes then scatters off this grating at an angle $\pm\Omega_s$, where

$$\Omega_s = \frac{\lambda_s}{\lambda_g} = \frac{2k_p\theta}{k_s}. \quad (24)$$

To obtain significant wide-angle scattering the z exponential must be small; otherwise integration along the scattering direction will average out to negligible values. This requirement yields (L is the cell length)

$$\frac{2k_p^2}{k_s} L\theta^2 \ll 1 \quad (25)$$

or

$$\theta < \theta_1 \equiv (k_s/2k_p^2L)^{1/2} \approx (2k_pL)^{-1/2}. \quad (26)$$

This corresponds to $\lambda_pL \ll \lambda_g^2$, that is, an optically thin grating.

As the wide-scattered radiation grows, S^{inc} on the rhs of Eqs. (15)–(18) must be replaced by $S^{\text{inc}} + S$. However, we shall not pursue amplified wide-scattered radiation further.

Inequality (25) gives an upper bound on θ at which wide-angle scattering can occur. A lower limit can be seen from Eq. (23). When the spacing of the grating planes exceeds the diameter of the beam, then the pump beams can be considered collinear. Alternatively, the wide-scattered radiation is within the diffraction angle. We shall return to the collinear case below. In any event, we note that wide-scattered radiation can be observed only in the range

$$\lambda_s/D \equiv \theta_{\text{diff}} < \theta < \theta_1 = (2k_pL)^{-1/2}. \quad (27)$$

Let us return now to the B_1 and B_2 terms. We will show that these produce forward scattering. Outside the wide-scattering regime given by Eq. (27) these are the only terms that play a role in Raman conversion. The x-dependent terms in B_1 are of the form

$$[(k_{pm} - k_{pn})(\theta+\Omega) - k_{sn}\Omega]x. \quad (28)$$

These terms can be neglected only for $\Omega = 0$, for then this expression becomes

$$(k_{pm} - k_{pn})x\theta (\lesssim \Delta kD\theta), \quad (29)$$

which we assume to be small, as in inequality (10) above (or else beam quality is not preserved). The vanishing of the z exponential yields the same result as inequality (9) (with θ substituted for ψ). Likewise, the conditions under which the B_2 terms lead to good beam quality with high gain reproduce inequalities (9) and (10). Let us now assume that

$$\Delta kD\theta \ll 1, \qquad \theta \ll \theta_2 \equiv (D\Delta k)^{-1}, \quad (30)$$

$$\Delta k\theta^2L/2 \ll 1, \qquad \theta \ll \theta_3 \equiv (2/\Delta kL_c)^{1/2}. \quad (31)$$

We now have for forward Raman scattering

$$\partial_z S_n = B_{1n} + B_{2n}. \quad (32)$$

Using the Hilbert-space notation, we may recast Eq. (32) in the form

$$\partial_z|S\rangle = [|P_1\rangle\langle P_1| + |P_2\rangle\langle P_2|]|S\rangle. \quad (33)$$

This equation can be formally solved as

$$|S\rangle = \exp[z(|P_1\rangle\langle P_1| + |P_2\rangle\langle P_2|)]|S(z=0)\rangle. \quad (34)$$

Expanding term by term, we find that

$$|S\rangle = [1 + z(|P_1\rangle\langle P_1| + |P_2\rangle\langle P_2|) + \ldots]|S(z=0)\rangle. \quad (35)$$

We consider now the following two limits:

(1) The two pumps are temporally correlated with the same spectral distribution:

$$|P_2\rangle = \beta|P_1\rangle, \quad (36)$$

where β is a constant. From Eq. (7) we now have [let $|S_0\rangle = |S^{\text{inc}}(z=0)\rangle$]

$$|S\rangle = |S_0\rangle + \{\exp[(1+\beta^2)\langle P_1|P_1\rangle z] - 1\}|\hat{P}_1\rangle\langle\hat{P}_1|S_0\rangle, \quad (37)$$

where $|\hat{P}_1\rangle$ is a unit vector in the (Hilbert-space) direction of $|P_1\rangle$. In this case we say that the Raman amplification proceeds through *multibeam* gain.

(2) The two pumps are temporally uncorrelated:

$$\langle P_1|P_2\rangle = [(\langle P_1|P_1\rangle\langle P_2|P_2\rangle)/N]^{1/2}\exp(i\chi), \quad (38)$$

with χ random and N equal to the number of modes.[11] In this case

$$|S\rangle = |S_0\rangle + [\exp(\langle P_1|P_1\rangle z) - 1]|\hat{P}_1\rangle\langle\hat{P}_1|S_0\rangle + [\exp(\langle P_2|P_2\rangle z) - 1]|\hat{P}_2\rangle\langle\hat{P}_2|S_0\rangle. \quad (39)$$

What emerges is now a Stokes beam, with one piece temporally correlated with the first pump (with a gain proportional to the first pump's intensity) and the other temporally correlated to the second pump (with a gain proportional to the *second* pump's intensity).

The conditions for the temporal coherence of the multimode crossed beams are the following:

(1) The lasers originate in the same master oscillator,

(2) $\delta L \Delta k \ll 1,$ (40)

(3) $D\theta\Delta k \ll 1,$ (41)

where δL is the path mismatch and Δk is the pump's bandwidth.

Temporal coherence also obtains in the case of single-longitudinal-mode lasers. To obtain multibeam gain in a beam-combining experiment with two different lasers, rather than two laser beams originating from the same master oscillator, very-narrow-band lasers must be used, since for typical excimer lasers the mode spacings are of the order of 100 MHz.

Finally, we address the problem of forward scattering when the angle between the beams is less than the diffraction angle. In this case the spacing between grating layers exceeds the beam width, and the beam may be taken to be collinear. Hence we must solve Eq. (15) by keeping all four terms. Taking θ and Ω to be equal to zero, the exponentials in Eq. (15) vanish, and we obtain

$$\partial_z|S\rangle = \left(\sum_{n=1}^{2}|P_n\rangle\right)\left(\sum_{m=1}^{2}\langle P_m|\right)|S\rangle. \tag{42}$$

Assuming now *lack* of temporal coherence, we obtain by formal integration

$$|S\rangle = |S_0\rangle + \left(\sum_{n,m=1}^{2}|P_n\rangle\langle P_m|S_0\rangle\right)\left(\sum_{j=1}^{2}\langle P_j|P_j\rangle\right)^{-1} \times\left[\exp\left(z\sum\langle P_j|P_j\rangle\right)-1\right], \tag{43}$$

so that in this case (collinear pumps) the multibeam gain is obtained in the *absence* of temporal coherence.

There is, however, a price to be paid in beam quality of the amplified beams if the pump beams have different transverse phase fronts. This can be seen by noting that since for the mth beam we may write

$$|P_m\rangle = |P_{m0}\rangle\exp[i\phi_m(\mathbf{x})], \tag{44}$$

where $\phi_m(\mathbf{x})$ represents the phase front of the mth beam, there is *no* cancellation of pump phases on the rhs of Eq. (42). Cancellation obtains only if the transverse phase fronts of the different pump beams are identical. This situation is to be contrasted with the finite-crossing-angle case (above the grating angle) in which the crossed beams could have different phase fronts and still preserve the Stokes beam quality.

SUMMARY AND CONCLUSIONS

We may most conveniently summarize our results by angles of the crossed beams.

(1) *Collinear* ($\theta < \lambda/D$). In this case the multibeam gain is attained even if the pumps are incoherent. If they are coherent, the beams alternatingly reinforce and cancel along the propagation direction. The beam quality is not preserved unless the phase fronts of the interacting beams are the same.

(2) *The Grating Regime* ($\theta_1 > \theta > \lambda/D$). Wide-angle scattering occurs in this regime. (The amount of scattering depends on the temporal correlation among the pumps and on the temporal correlation between the Stokes and the pumps.) Forward scattering also occurs with single-pump gain if the pump beams are incoherent; otherwise it occurs with multipump gain.

(3) *The Forward-Scattering Regime* (θ_3, $\theta_2 > \theta > \theta_1$). There is no wide-angle scattering. Multibeam gain obtains if the pumps are temporally coherent; otherwise one has single-pump gain. Beam quality is preserved even if the pumps have different phase fronts.

(4) *Single-Beam Dropoff* ($\theta > \theta_2$ *and/or* $\theta > \theta_3$). If $\theta > \theta_2$ there is a dropoff in beam quality. If $\theta > \theta_3$ there is a dropoff in gain. Both of these effects, however, also occur in the case of a single pump with a misaligned Stokes.

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* Permanent address, Department of Physics and Division of Applied Sciences, Harvard University, Cambridge, Massachusetts 02138.

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11. Note that $\langle P|S_0\rangle$ is an inner product in Hilbert space. The value of this inner product depends on the temporal correlation

between $|P\rangle$ and $|S_0\rangle$. In general, if $|A\rangle$ and $|B\rangle$ are two Hilbert-space vectors, then, if they are temporally correlated, the phases of the modes are the same and

$$\langle A|B\rangle = \sum_{i=1}^{N} A_i^x B_i = \sum_{i=1}^{N} |A_i||B_i|.$$

If the amplitudes $|A_i|$ and $|B_i|$ are independent of i, then

$$\langle A|B\rangle \approx N|A||B| = (N|A|^2 \cdot N|B|^2)^{1/2} = (\langle A|A\rangle \langle B|B\rangle)^{1/2}.$$

If $|A\rangle$ and $|B\rangle$ are temporally uncorrelated, then, by a random-walk argument,

$$\langle A|B\rangle = \sum_{i=1}^{N} |A_i||B_i| \exp[i(\phi_{A_i} - \phi_{B_i})]$$
$$\approx N^{1/2}|A||B| \exp(i\chi),$$

where χ is random. We shall use these results repeatedly in what follows.

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