Recentl y, we examined a Raman oscillator containing an intracavity second-harmonic generator that freq uency doubles the resonant Stokes field [1]. We found that there is an optimum ratio between the nonlinear coupling of the stimulated Raman scattering (SRS) and the nonlinear coupling of the intracavity second-harmonic generation (SHG) for maximizing the second harmonic of the Stokes frequency. We called the system the SRS-SHG device. Since the SHG interaction is performed inside the resonant cavity, it takes advantage of the high circulating power of the resonant Stokes field. An alternative intracavity frequency upconversion process is sum-frequency generation (SFG) between the pump and resonant Stokes radiation. We refer to the device that combines SFG and SRS in a single resonator as the SRS-SFG device. For the same SRS material, the SRS-SFG device yields a higher energy photon and results in less heat deposited per high-frequency photon in the SRS material than the SRS-SHG device. It is also possible to reduce the power loading of the SRS material when the SFG interaction precedes the SRS material in the SRS-SFG device. This allowed them to produce radiation suitably shifted from the second harmonic of the pump for performing differential absorption measurements. In this paper, we use both plane-wave analysis and three-dimensional (3-D) simulation to examine design considerations of a Raman oscillator containing an intracavity SFG interaction between the pump and circulating Stokes field.

In designing an oscillator with an intracavity sum-frequency generator, there is typically an optimum interaction coupling that maximizes the frequency-upconverted output of the device. If the SFG interaction is much weaker than the SRS interaction, the SFG interaction does not effectively convert the Stokes and pump fields. If the SFG interaction is much stronger than the SRS interaction, the circulating Stokes field never builds up to achieve significant conversion. The aim of this paper is to determine the optimum ratio between coupling in the SRS interaction and coupling in the SFG interaction for a Raman oscillator with an intracavity sum-frequency generator. Since the pump field interacts with both materials, the ordering of the interactions is significant. We refer to the device in which the pump is first incident on the SRS material and then the SFG material as the SRS → SFG device and the opposite case as the SFG → SRS device.

Our analysis of the SRS → SFG device shows that states with high conversion efficiency that completely deplete the pump are unstable. This can be understood from the fact that the SRS process depletes the pump so rapidly that it is difficult to maintain pump and Stokes waves at significant levels to efficiently generate the sum frequency. The SFG → SRS device, by contrast, appears to operate stably and with high efficiency over a large dynamic range of incident pump intensities. Although we have studied the dynamics for both orderings of the nonlinear interactions, in this paper we present mainly the analysis of the more useful SFG → SRS configuration.

In previous work [3], [4], we discussed how to choose the relative couplings for parametric oscillation and intracavity SFG interactions. The approach we use here is similar, but since the dynamics of the SRS process are so different from three-wave mixing, the results are very different. In particular, the plane-wave equations describing three-wave mixing indicate that the pump field, under appropriate conditions, can be completely depleted. In the plane-wave equations describing SRS the pump field can approach zero exponentially but never actually reaches zero. Therefore, there are no states of operation that completely deplete the pump in the SFG → SRS device, where the SRS process is the last interaction of the pump field. While the SRS → SFG device has states of

**Abstract**—We use a plane-wave analysis to examine a Raman oscillator containing an intracavity sum-frequency interaction that frequency sums the circulating first-order Stokes radiation with the pump radiation. We find that there is an optimum ratio between the nonlinear coupling in the Raman medium and the nonlinear coupling in the sum-frequency generator. We also find that higher order Stokes radiation should be suppressed with the optimum choice of nonlinear coupling in the sum-frequency interaction. Numerical integration of the equations containing transverse effects predicts a time-averaged power-conversion efficiency of 61.4% for conversion of 532- to 273.5-nm radiation using a CW mode-locked frequency-doubled Nd:YAG laser with Ba(NO₃)₃ for the Raman material and CsLiB₃O₁₀ (CLBO) for the sum-frequency material.

**Index Terms**—Nonlinear optics, optical frequency conversion, optical mixers, optical mixing, optical propagation in nonlinear media, Raman lasers.

I. INTRODUCTION

**R**ECENTLY, we examined a Raman oscillator containing an intracavity second-harmonic generator that frequency doubles the resonant Stokes field [1]. We found that there is an optimum ratio between the nonlinear coupling of the stimulated Raman scattering (SRS) and the nonlinear coupling of the intracavity second-harmonic generation (SHG) for maximizing the second harmonic of the Stokes frequency. We called the system the SRS-SHG device. Since the SHG interaction is performed inside the resonant cavity, it takes advantage of the high circulating power of the resonant Stokes field. An alternative intracavity frequency upconversion process is sum-frequency generation (SFG) between the pump and resonant Stokes radiation. We refer to the device that combines SFG and SRS in a single resonator as the SRS-SFG device. For the same SRS material, the SRS-SFG device yields a higher energy photon and results in less heat deposited per high-frequency photon in the SRS material than the SRS-SHG device. It is also possible to reduce the power loading of the SRS material when the SFG interaction precedes the SRS material in the SRS-SFG device. This allowed them to produce radiation suitably shifted from the second harmonic of the pump for performing differential absorption measurements. In this paper, we use both plane-wave analysis and three-dimensional (3-D) simulation to examine design considerations of a Raman oscillator containing an intracavity SFG interaction between the pump and circulating Stokes field.

In designing an oscillator with an intracavity sum-frequency generator, there is typically an optimum interaction coupling that maximizes the frequency-upconverted output of the device. If the SFG interaction is much weaker than the SRS interaction, the SFG interaction does not effectively convert the Stokes and pump fields. If the SFG interaction is much stronger than the SRS interaction, the circulating Stokes field never builds up to achieve significant conversion. The aim of this paper is to determine the optimum ratio between coupling in the SRS interaction and coupling in the SFG interaction for a Raman oscillator with an intracavity sum-frequency generator. Since the pump field interacts with both materials, the ordering of the interactions is significant. We refer to the device in which the pump is first incident on the SRS material and then the SFG material as the SRS → SFG device and the opposite case as the SFG → SRS device. Our analysis of the SRS → SFG device shows that states with high conversion efficiency that completely deplete the pump are unstable. This can be understood from the fact that the SRS process depletes the pump so rapidly that it is difficult to maintain pump and Stokes waves at significant enough levels to efficiently generate the sum frequency. The SFG → SRS device, by contrast, appears to operate stably and with high efficiency over a large dynamic range of incident pump intensities. Although we have studied the dynamics for both orderings of the nonlinear interactions, in this paper we present mainly the analysis of the more useful SFG → SRS configuration.

In previous work [3], [4], we discussed how to choose the relative couplings for parametric oscillation and intracavity SFG interactions. The approach we use here is similar, but since the dynamics of the SRS process are so different from three-wave mixing, the results are very different. In particular, the plane-wave equations describing three-wave mixing indicate that the pump field, under appropriate conditions, can be completely depleted. In the plane-wave equations describing SRS the pump field can approach zero exponentially but never actually reaches zero. Therefore, there are no states of operation that completely deplete the pump in the SFG → SRS device, where the SRS process is the last interaction of the pump field. While the SRS → SFG device has states of...
Fig. 1. Schematic diagram of the SFG→SRS device. The incident pump field is first frequency-summed with the circulating Stokes field to produce the desired frequency \( \omega_d = \omega_s + \omega_p \). The unconverted pump then drives the SRS medium to produce gain at the Stokes frequency \( \omega_s \). The Stokes field is circulated in a ring cavity with intensity reflectivity \( R \).

complete pump depletion, we find this device is less efficient and dynamically less well behaved than the SFG→SRS device.

We first model the SRS and SFG interactions using equations describing continuous plane-wave interactions. We begin by calculating the efficiency for converting pump photons to sum-frequency photons in a parameter space where system operation takes place along a straight line through the origin. The slope of the straight line is a system design parameter describing the relative nonlinear couplings of the two interactions. As an additional design parameter, we include the possibility of phase mismatch in the SFG interaction.

A schematic diagram of the SFG→SFG device is shown in Fig. 1. The field propagates around the ring cavity along the \( z \) axis. The entrance to the SFG medium is taken to be \( z_1 \), the entrance to the SRS medium is \( z_2 \), and the exit of the SRS medium is \( z_3 \). The length of the SRS interaction is taken to be \( L_s \) and the length of the SFG interaction is \( L_d \). We assume the round-trip losses can be modeled as a single output coupler with intensity reflectivity \( R \) at the Stokes frequency and placed after the second interaction. If \( R \) is close to unity, the placement of the output coupler does not significantly affect the dynamics.

II. PLANE-WAVE EQUATIONS AND THEIR SOLUTIONS

In this section, we introduce our notation for the plane-wave equations describing SRS and SFG and their solutions. Throughout the paper, we refer to the sum frequency of the Stokes and pump frequencies as the desired frequency component. We assume that none of the sum frequency is circulated in the cavity, so that no sum frequency exists at the entrance to the SFG interaction.

A. Sum-Frequency Generation

The equations describing SFG in the plane-wave approximation are

\[
\frac{d}{dz} E_s = i \left( \frac{d \omega_s}{cn_s} \right) E_p E_p^* \exp[iq_b z] \tag{1}
\]

\[
\frac{d}{dz} E_p = i \left( \frac{d \omega_p}{cn_p} \right) E_s E_d^* \exp[iq_b z] \tag{2}
\]

\[
\frac{d}{dz} E_d = i \left( \frac{d \omega_d}{cn_d} \right) E_s E_p \exp[-i q_b z] \tag{3}
\]

where \( E_s, E_p, \) and \( E_d \) are the Stokes, pump, and sum-frequency (desired) electric-field amplitudes, respectively, \( n_s, n_p, \) and \( n_d \) are the indices of refraction at the Stokes, pump, and desired frequencies \( \omega_s, \omega_p, \) and \( \omega_d \) respectively, \( c \) is the speed of light, \( d_b \) is the effective nonlinear coefficient, and the wavevector mismatch \( q_b = k_d - k_s - k_p \), where \( k_s, k_p, \) and \( k_d \) are the Stokes, pump, and desired wavevectors, respectively. For \( j = s, p, d \), we substitute \( E_j = \sqrt{\omega_j / n_j \mu_j} \exp[i \phi_j] \), where \( \rho_j \) and \( \phi_j \) are real quantities, into (1)–(3) to obtain the real equations

\[
\frac{d}{dz} \rho_s = -i k_b \rho_p \sin(\theta) \tag{4}
\]

\[
\frac{d}{dz} \rho_p = -i k_b \rho_s \sin(\theta) \tag{5}
\]

\[
\frac{d}{dz} \rho_d = i k_b \rho_p \sin(\theta) \tag{6}
\]

\[
\frac{d}{dz} \theta = q_b + \left( \frac{\rho_s \rho_p}{\rho_d} - \frac{\rho_p \rho_d}{\rho_s} - \frac{\rho_s \rho_d}{\rho_p} \right) \nu_b \cos(\theta) \tag{7}
\]

where \( \theta = \phi_s + \phi_d - \phi_s - \phi_p \) and

\[
\nu_b = \frac{d_b}{c} \sqrt{\frac{\omega_b \omega_s \omega_p}{n_s n_p n_d}}. \tag{8}
\]

The variables \( \rho_j^2 \) are proportional to photon flux densities.

The solutions to (4)–(7) use the three constants of the motion [5], [6]

\[
D_s = \rho_s^2 + \rho_d^2 \tag{9}
\]

\[
D_p = \rho_p^2 + \rho_d^2 \tag{10}
\]

which are the Manley–Rowe relations, and

\[
\nu_b \rho_s \rho_p \cos(\theta) + \frac{1}{2} q_b \rho_d = 0 \tag{11}
\]

where we have made use of the assumption that \( \rho_d^2(z_1) = 0 \). For the sake of continuity, we define the initial phase \( \theta(z_1) = \pi/2 \).

We further define \( D_s < D_p < D_d \) as the smaller and larger roots of the equation

\[
(D_s - D)(D_p - D) - \left( \frac{Q_b}{\nu_b L_b} \right)^2 D = 0 \tag{12}
\]

where \( Q_b = q_b L_b / 2 \) is a dimensionless detuning parameter. We also define \( m = D_s / D_d \). The solutions can then be written as

\[
\rho_s^2(z) = D_s \sin^2(Z_b \mid m_b) \tag{13}
\]

\[
\rho_p^2(z) = D_p - D_s \sin^2(Z_b \mid m_b) \tag{14}
\]

\[
\rho_d^2(z) = D_p - D_s \sin^2(Z_b \mid m_b) \tag{15}
\]

\[
\sin(\theta) = \sqrt{D_s D_p / \rho_s(z) \rho_p(z)} \tag{16}
\]

where \( Z_b = \sqrt{D_s / \nu_b L_b} \) and \( \sin, \cos, \) and \( \tan \) are Jacobi elliptic functions [7]. At the end of the crystal, we replace \( Z_b \) with \( \Delta \mu = \sqrt{D_s / \nu_b L_b} \).

To simplify the mathematics, we define

\[
x = \frac{1}{2} \log \left( \frac{D_p}{D_s} \right). \tag{17}
\]
Now consider that the product of the roots of (12) is equal to $D_s D_p$. Using this and the definition of $m_b$, it follows that $D_s = \sqrt{D_s D_p/m_b}$. Using this and (12), we can obtain a quadratic equation for $\sqrt{m_b}$:

$$m_b - 2 \cosh(x) \sqrt{m_b} + 1 - \left( \frac{Q_b}{2\alpha_b} \right)^2 = 0. \tag{18}$$

Since $0 \leq m_b \leq 1$, the solution to (18) can be written as

$$\sqrt{m_b} = \cosh(x) - \sqrt{\sinh^2(x) + \left( \frac{Q_b}{2\alpha_b} \right)^2}. \tag{19}$$

### B. Stimulated Raman Scattering

For the SRS interaction, we ignore higher order Stokes and anti-Stokes processes. We show later in the paper that this approximation is consistent with operation in the optimal configuration. The equations describing the SRS interaction are conveniently written in terms of the photon flux densities

$$\frac{d}{dz} \rho_s^2 = G_{sr} \rho_s^2 \rho_p^2 \tag{20}$$

$$\frac{d}{dz} \rho_p^2 = -G_{sr} \rho_s^2 \rho_p^2 \tag{21}$$

where $\rho_s^2$ and $\rho_p^2$ are the Stokes and pump photon flux densities, respectively, and $G_{sr}$ is the Raman photon-flux gain coefficient. The sum of the two equations yields a constant of the motion, the total number of Stokes and pump photons in the SRS medium

$$C = \rho_s^2 + \rho_p^2. \tag{22}$$

This allows the equations to be integrated to give solutions

$$\rho_s^2(z) = \frac{G_{sr} \rho_s^2(z_0)}{\rho_s^2(z_0) + \rho_p^2(z_0) \exp\left[-CG_{sr}(z - z_0)\right]} \tag{23}$$

$$\rho_p^2(z) = \frac{G_{sr} \rho_s^2(z_0)}{\rho_s^2(z_0) + \rho_p^2(z_0) \exp\left[-CG_{sr}(z - z_0)\right]} \tag{24}$$

### III. Steady-State Operation in the $(g_a, g_b)$-Plane

In this section, we derive the steady-state operation of the SFG→SRS device. It is convenient to describe the two interactions by their nonlinear coupling parameters. We define

$$g_a^2 \equiv G_{sr} L_a \rho_s^2(2z) \tag{25}$$

and

$$g_b^2 \equiv G_{sr} L_b \rho_p^2(2z). \tag{26}$$

Note that these definitions show how the nonlinear coupling of the SRS process grows linearly with increasing length, while the SFG coupling grows quadratically with increasing length. Both $g_a^2$ and $g_b^2$ are proportional to the incident pump photon flux density, so their ratio is independent of this density.

### A. Small-Signal Gain

The threshold for the SRS-SFG device is determined from the product of the small-signal gains of the SRS and SFG interactions. In the small-signal regime, the two orderings of the interactions are equivalent and the net small-signal gain per pass is of the form $G = R G_a G_b$. The small-signal gain of the SRS interaction $G_a$ is

$$G_a = \exp\left[\frac{G_{sr} L_a \rho_s^2(2z)}{g_a^2}\right] = \exp\left[\frac{g_a^2}{g_a^2}\right] \tag{27}$$

where we used $\rho_s^2(2z) = \rho_p^2(2z)$ in the small-signal regime. The small-signal gain of the SFG interaction is [4]

$$G_b = 1 - \frac{g_b^2}{g_b^2 + \Delta} \sinh^2\left(\sqrt{g_b^2 + \Delta}\right). \tag{28}$$

It is useful to calculate the small-signal gain of the second-order Stokes frequency $G_{a(2)}$ to estimate the threshold for second-order Stokes oscillation. We assume the first-order Stokes is undepleted and has the $z$ dependence given by (23) within the SRS medium. We then integrate the equation describing the $z$ dependence of the second-order Stokes field to obtain

$$G_{a(2)} = \left[\left(\rho_p^2(2z) \exp\left(CG_{sr} L_a\right) + \rho_p^2(2z)\right) / C \right]^{C_{sr}(2)} \tag{29}$$

where $G_{sr}(2)$ is the Raman photon-flux gain at the second-order Stokes frequency. This small-signal gain represents an upper bound of the actual small-signal gain for the second-order Stokes, since SFG between the second-order Stokes and the pump, as well as SFG between the second-order Stokes and the first-order Stokes, could significantly reduce the small-signal gain of the second-order Stokes. The significance of the SFG contribution to the small-signal gain depends on the spectral acceptance bandwidth and other phase-matching characteristics of the sum-frequency interaction.

In the case of SFG→SRS, it might be convenient, if the sum frequency can be transmitted by the SRS medium, to pass the sum frequency through the SRS material before it is coupled out of the cavity. In such a case, it is necessary to consider the possibility of the sum frequency interacting with the SRS material to produce Raman-shifted Stokes components. The small-signal gain of the process $G_{a(2)}$ is similar to (27)

$$G_{a(2)} = \exp\left[G_{sr(2)}^{C_{sr}(2)} L_a \rho_s^2(2z)\right] \tag{30}$$

where $G_{sr(2)}$ is the Raman photon-flux gain coefficient for the sum-frequency-pumped SRS material. The Raman photon-flux gain coefficient scales as $\omega^2$, and, since the sum frequency will be less than $2\omega_p$, $G_{sr(2)} < G_{sr(2)}$. We define the quantum efficiency for SFG as $\eta_{SFG} = \rho_s^2(2z) / \rho_p^2(2z)$, which has an upper bound of 0.5 in steady state. This allows us to put an upper bound on the small-signal gain for sum-frequency-pumped SRS

$$G_{a(2)} < \exp\left[4 G_{sr} L_a \rho_s^2(2z) \eta_{SFG}\right] = \exp\left(4 g_a^2 \eta_{SFG}\right). \tag{31}$$

We calculate the quantum efficiency $\eta_{SFG}$ in the $(g_a, g_b)$-plane for producing the sum frequency of the Stokes and pump frequencies. The $(g_a, g_b)$ plane is a convenient parameter space.
to analyze the system performance, since operation takes place along a straight line through the origin. Movement up and down this straight line occurs as the pump intensity varies. This variation could be from temporal variations in the pulse envelope or from transverse variations in the pump profile. While the continuous plane-wave theory does not directly account for these effects, the analysis gives us insight into the problem. The intuition gained from the solutions is expected to be good whenever diffraction effects and spatial walkoff or group-velocity mismatch and group-velocity dispersion are not too large.

Since the expressions for the efficiency $\eta_\text{kl}$ and the second-order Stokes small-signal gain $G_a^{(2)}$ are not easily obtained in terms of $g_a$ and $g_b$, we parameterize the steady-state solutions and $g_a$ and $g_b$ in terms of $\Lambda_b$ and $\kappa$. We use the Manley–Rowe relations for the SFG and SRS interactions, (9), (10), and (22), together with the steady-state cavity condition

$$\rho_a^2(z_3) = R \rho_a^2(z_3)$$

(32)
to obtain relationships for all the variables in terms of these two parameters.

B. Steady-State Solutions

To calculate the steady state of the SFG$\rightarrow$SRS device, we can use the Manley–Rowe relations (9), (10), and (22) and the steady-state cavity condition (32) to obtain expressions for the Stokes, pump, and desired fields

$$\rho_a^2(z_1) = e^{-2\kappa} \rho_a^2(z_1)$$
$$\rho_b^2(z_2) = (e^{-2\kappa} - \eta_b) \rho_b^2(z_1)$$
$$\rho_s^2(z_3) = \frac{e^{-2\varphi}}{R} \rho_s^2(z_1),$$
$$\rho_p^2(z_2) = (1 - \eta_b) \rho_p^2(z_1)$$
$$\rho_s^2(z_3) = \left[1 - 2\kappa + e^{-2\varphi} \left(1 - \frac{R}{\rho_s^2(z_1)} \right) \right] \rho_p^2(z_1)$$
$$\rho_a^2(z_2) = 0$$
$$\rho_a^2(z_2) = \eta_b \rho_a^2(z_2).$$

From (23), we can solve for $\rho_a^2$ to obtain

$$\rho_a^2 = \frac{1}{1 - 2\kappa + e^{-2\varphi}} \times \log \left\{ \frac{e^{-2\kappa}(1 - \eta_b)}{R(1 - 2\kappa + e^{-2\varphi}) + e^{-2\varphi}} \right\},$$

(40)

In addition, from the definition of $\Lambda_b$, and the expression for $D_s$, we can write $\rho_b^2$ in terms of $\Lambda_b$ as

$$\rho_b^2 = \sqrt{\eta_b \Lambda_b^2 e^{\varphi}}.$$  

(41)

Finally, using (13) and (33), we can write the efficiency for the SFG$\rightarrow$SRS device as

$$\eta_\text{kl} = e^{-\varphi} \sqrt{\eta_b \rho_a^2(\Lambda_b \mid \eta_b)},$$

(42)

C. Stability of Steady-State Operation

We now consider the stability of steady-state operation of the SFG-SRS device to a small perturbation in the circulating Stokes field. We begin by considering small perturbations to the steady-state values $\rho_s \rightarrow \rho_s + \rho_s \delta_s$, $\rho_p \rightarrow \rho_p + \rho_p \delta_p$, $\rho_a \rightarrow \rho_a + \rho_a \delta_a$ of the variables induced by a small perturbation to the input Stokes wave $\delta_s(z_1)$. It is convenient to define $\epsilon_s = \rho_s \delta_s$. The perturbation at the end of the SRS interaction of the circulating Stokes field $\delta_s(z_3)$ results in a perturbation $\sqrt{R} \delta_s(z_3)$ to the circulating Stokes field at the input of the SFG interaction on the next pass through the resonator. In order for the steady state to be stable, it is necessary and sufficient that $|S| < 1$, where $S = \sqrt{R} \delta_s(z_3)$.

The analysis of the SFG interaction is identical to that performed in [4]. The main results of that analysis are

$$\epsilon_s(z_3) = \left(1 - \Delta \right) \epsilon_s(z_1)$$
$$\epsilon_p(z_2) = -\Delta \epsilon_s(z_1)$$

(43)
(44)

where $\Delta$ is given by [4, eq. (48)].

For the stability analysis of the SRS component of the device, we begin by examining the variation of $C$ [see (22)]

$$\delta C = 2[\epsilon_s(z_2) + \epsilon_p(z_2)],$$

(45)

We use this in the variation of (23) to obtain

$$\epsilon_s(z_3) = \epsilon_s(z_2) \Gamma_1 + \epsilon_p(z_2) \Gamma_2$$

(46)

where

$$\Gamma_1 = \frac{\left[C + \rho_s^2(z_2)\right]}{B} - \frac{C \rho_p^2(z_2)}{B^2} \left[1 - C \rho_p^2(z_2) \exp(-C \rho_p^2(z_2)) \right]$$

(47)
$$\Gamma_2 = \frac{\rho_a^2(z_2)}{B} - \frac{C \rho_p^2(z_2)}{B^2} \exp(-C \rho_p^2(z_2)) \left[1 - C \rho_s^2(z_2) \right]$$

(48)

and

$$B = \rho_s^2(z_2) + \rho_p^2(z_2) \exp(-C \rho_p^2(z_2)).$$

(49)

This leads to an expression for the stability parameter,

$$S = \frac{\Re_s(z_3)}{\epsilon_s(z_1)},$$

(50)

$$= R \left[ \Gamma_1 - \Delta (\Gamma_1 + \Gamma_2) \right].$$

(51)

We then calculate $S$ in the $(g_a, g_b)$ plane. We find that for the case of exact phase matching ($Q_b = 0$), no unstable steady states appear in the region plotted in Figs. 2 and 3.

In Fig. 2, we plot contours in the $(g_a, g_b)$ plane of the steady-state quantum efficiency $\eta_\text{kl}$ of the sum frequency between the first-order Stokes field and the pump field for the case of exact phase matching in the SFG interaction, $Q_b = 0$ for the SFG$\rightarrow$SRS device. The straight line through the origin represents an appropriate operating line for the device with a slope of $g_a/g_b = 0.4$. The dashed line represents the small-signal gain threshold for the SRS-SFG device for a cavity reflectivity of $R = 0.95$. 
In this section, we report the results of iterating the plane-wave solutions given above as the system moves along the operating line in Figs. 2 and 3. We use the solutions listed in (13)–(15), (23), and (24) to construct a mapping that represents the wave solutions given above as the system moves along the operating line in Figs. 2 and 3. We use the solutions listed in

\[ a = \frac{g_0}{g_n} \]

and that the SRS photon-flux gain coefficient \( g_n^{(2)} \) as a function of the normalized pump intensity \( g_n^2 \). For normalized incident pump intensities \( g_n^2 \) greater than 1, the quantum efficiency exceeds 40%. When the SFG interaction is detuned to \( Q_b = 2.0 \), the efficiency decreases by only a few percent. This indicates that high conversion can take place over a large dynamic range of pump intensities and should be very tolerant of phase mismatch in the SFG.

In Fig. 4, we plot the quantum efficiency of the sum frequency between the pump and circulating Stokes fields as a function of normalized incident pump intensity \( g_n^2 \) using the iterative technique. For a cavity reflectivity of \( R = 0.95 \), threshold for the device occurs around \( g_n^2 = 0.5 \). For normalized incident pump intensities \( g_n^2 \) greater than 1, the quantum efficiency exceeds 40%. When the SFG interaction is detuned to \( Q_b = 2.0 \), the efficiency decreases by only a few percent. This indicates that high conversion can take place over a large dynamic range of pump intensities and should be very tolerant of phase mismatch in the SFG.

In Fig. 5, we plot the small-signal gain of the second-order Stokes \( G_n^{(2)} \) and the small-signal gain of the Stokes frequency generated by the sum frequency in the Raman material \( G_n^{(2)} \) as a function of the normalized pump intensity \( g_n^2 \) along the operating line in Figs. 2 and 3. For these calculations, we assume the Raman photon-flux gain for the second-order Stokes is equal to that of the first-order Stokes process \( G_{r}^{(2)} = G_{r}^{(2)} \) and that the SRS photon-flux gain coefficient \( G_{r}^{(2)} \) for the Stokes frequency-pumped SRS interaction is twice as large as that for the SRS interaction pumped by the fundamental \( G_{r}^{(2)} = 2G_{r} \), which represent upper bounds on these gains.

The small-signal gain for the second-order Stokes \( G_n^{(2)} \) can be kept below 50 for the phase-matched SFG interaction, but increases to as high as 300 for the nonphase-matched SFG interaction over the range of pump intensities considered here. This indicates that it is relatively easy to suppress second-order Stokes generation in the device over this range.

\[ G_n^{(2)} = 10^4 \]

1 Jacobi elliptic functions were computed using programs in [8].
The small-signal gain for the sum-frequency-pumped Raman material is low over this range, but is increasing exponentially with increasing pump intensity. So, at higher pump intensities, it may be necessary to outcouple the sum frequency to avoid its interaction with the SRS material.

V. PHYSICAL EXAMPLE AND NUMERICAL INTEGRATION RESULTS

As a physical example, consider generation of ultraviolet radiation for differential absorption laser ranging and detection of ozone and other atmospheric gases. Using the second harmonic of a Nd:YAG laser at 532 nm to pump a Ba(NO$_3$)$_2$ Raman medium produces 563 nm. Summing the pump and Stokes frequencies produces 273.5 nm. The Raman intensity gain for Ba(NO$_3$)$_2$ is $G_r = 2G_s/(\epsilon_0\omega_p c) = 47 \text{ cm/GW}$ [9]. If we assume we have an 5 cm Ba(NO$_3$)$_2$ crystal, then a pump power of 1.6 kW into a 70-$\mu$m spot gives a peak nonlinear drive of $g_2 = 4.9$. We use CsLiB$_6$O$_{10}$ (CLBO) for the SFG interaction. It phase matches for type-I SFG at $\theta = 58.6^\circ$; the material nonlinear drive is $Z_b = n_b^2/\alpha_0\omega_p = 94/\text{GW}$ [12]. A 0.64-cm length of CLBO gives a ratio of nonlinear coupling parameters for plane waves $g_b/g_a = \sqrt{Z_bL_b^2/(\hat{G}_rL_a)} = 0.40$. The walk-off angle in CLBO is 2$^\circ$, warranting the employment of walkoff compensation. Let $x$ be the walkoff dimension and $y$ be the orthogonal transverse dimension. The walkoff in each CLBO crystal of length 0.32 cm is $\pm 111 \mu$m. Using the peak nonlinear drives in the plane-wave analysis gives power-conversion efficiencies on the order of 92%.

While the plane-wave analysis is encouraging, it cannot answer questions about the influence of walk-off, diffraction, and different spot sizes in the two nonlinear materials or the expected beam quality of the output. We have developed a numerical-integration code based on fast Fourier transforms and fourth-order Runge–Kutta integration to address such concerns. We use this code, described previously [1], [10], to model the SFG—SRS device described above. The calculations require about 200 passes of the Stokes field through the resonator to reach steady state. The Stokes field is attenuated in power by a factor $\hat{R} = 0.95$ after each pass through the crystals to account for undesirable, but inevitable, resonator losses. The parameter choices for the simulations described here work well but are not necessarily optimal.

We suppose that the device operates in a symmetric bowtie resonator with two curved mirrors with 1-m radius of curvature and two flat mirrors. The fold angle of the resonator is assumed to be small. The crystals are located in the leg of length 108.3 cm between the curved mirrors, such that the resonator waist is centered in the Raman crystal. The two CLBO crystals are next to each other before the Raman crystal and separated from it by a gap of length 2 cm. A gap is required to provide room for an output coupler for the sum frequency, which is not transmitted by barium nitrate. The leg between the flat mirrors has a length 87.3 cm, providing a round-trip cavity length appropriate for synchronous pumping at 76 MHz. The radius of the Stokes field at its waist in the Raman crystal is 125 $\mu$m. The pump is focused at the same position and is collinear with the Stokes field.

We assume that a peak pump power of 1.6 kW at 532 nm is available. This is typical of power levels we have obtained previously [11] by doubling 100-ps pulses from a 20-W average-power CW mode-locked Nd:YAG laser in tandem crystals of LBO. We investigated the effects of varying the pump radius. The steady-state power-conversion efficiency, shown in Fig. 6, is optimized for a pump radius of 70 $\mu$m, though the efficiency does not vary greatly over the range $w_p = 50$ to 125 $\mu$m. We note that using a pump beam which is narrow compared to the Stokes beam was also found to improve the efficiency in our numerical studies of the SRS-SHG device. For the present case, the steady-state power distribution with a 70-$\mu$m pump is 71.7% to the sum frequency, 8.5% to undepleted pump, 16.8% to cavity loss, and 3.0% to Raman heating. The beam quality $M^2$ [13] at the sum frequency for the $(x,y)$ dimensions is $(1.0021,1.0005)$ and the radius is $w_d = (78.9,626) \mu$m. We tried varying the nonlinear coefficient of the CLBO in the simulation and confirmed that the true value is near optimal. This suggests that
plane-wave theory is an appropriate way to choose $I_3^o/L_o$.

Since the SFG interaction in CLBO involves one e-wave and two o-waves, four inequivalent configurations of the two crystals are obtainable by flipping one of the crystals [14]–[16]. These correspond to choices as to whether the walk-off is compensated or not (WC or NWC) and whether there is a sign change in the nonlinear coefficient (SC or NSC). For an 80-μm pump radius, the quantum efficiencies were WC-NSC, 0.3681; NWC-NSC, 0.3458; NWC-SC, 0.2633; and WC-SC, 0.0004. Although the NWC-NSC configuration, which is equivalent to using a single crystal, provides good efficiency, the sum-frequency beam is spread out in the $x$ direction, $w_x = (128.7, 70.0)$ μm, and the beam quality $(1.058, 1.0001)$ in this dimension is adversely affected by the large walk-off. The NWC-SC configuration produces two noncollinear beams at the sum frequency. The WC-SC configuration produces almost no sum-frequency radiation, since conversion in the first CLBO crystal is canceled by backconversion in the second crystal.

We repeated the calculation with the 70-μm pump for a series of pump powers. The results are shown in Fig. 7, where we suppose that the pump pulse is a Gaussian in time: $P_3^o(t) = 1.6$ kW $\exp(-t^2)$. The sum-frequency power is fitted by $P_2(t) = 1.148$ kW $\exp(-1.2t^2 - 0.17t^4)$. Numerical integration of the fitted curve yields a time-integrated power-conversion efficiency from $\omega_p$ to $\omega_d$ of 61.4%. Further increases in efficiency should be possible using a pump with higher peak power or a resonator with lower losses.

VI. SUMMARY

We have used a CW plane-wave analysis of a Raman oscillator containing an intracavity sum-frequency generator to frequency sum the pump field with the circulating Stokes field. We find that there is an optimum ratio of sum-frequency nonlinear coupling to Raman nonlinear coupling which optimizes the sum-frequency output. The small-signal gain of the second-order Stokes in the SRS material is significantly reduced under optimal choice of the ratio. Three-dimensional simulations indicate high efficiency should be possible.

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