[7] R. Burnham and N. Djeu, "Ultraviolet-preionized discharge-pumped lasers in XeF, KrF and ArF," *Appl. Phys. Lett.*, vol. 29, pp. 707-709, Dec. 1, 1976.

[8] A. J. Andrews, A. J. Kearsley, C. E. Webb, and S. C. Haydon, "A KrF fast discharge laser in mixtures containing NF_3, N_2F_4 or SF_6," *Opt. Commun.*, vol. 20, pp. 265-268, Feb. 1977.

[9] W. J. Sarjeant, A. J. Alcock, and K. E. Leopold, "A scalable multiatmosphere high-power XeF laser," *Appl. Phys. Lett.*, vol. 30, pp. 635-637, June 15, 1977.

[10] —, "Constant E/N pumping of high-power rare-gas halide lasers," *IEEE J. Quantum Electron.*, vol. QE-13, pp. 104D-106D, Sept. 1977.

[11] Type E-7 epoxy: Techkits Inc., Demarest, NJ.

[12] J. C. Martin, "Multichannel gaps," AWRE Switching Notes, no. 10, Mar. 5, 1970.

[13] J. C. Martin, "Duration of the resistive phase and inductance of spark channels," AWRE Switching Notes, no. 9, Dec. 1965.

[14] K. E. Leopold and A. J. Alcock, "Simple, fully isolated, high voltage trigger generator," *Rev. Sci. Instr.*, vol. 47, pp. 254-255, Feb. 1976.

[15] L. P. Bradley, private communication.

[16] R. Sze, T. Loree, and R. Scott, "Performance characteristics of discharge excited excimer lasers," Laser Induced Chemistry, *2nd Wint. Conf.*, Park City, UT, Feb. 13-16, 1977; C. Brau, private communication.

[17] G. N. Glasoe and J. V. Lebacqz, *Pulse Generators.* New York: Dover, 1965.

[18] W. J. Sarjeant, A. J. Alcock, and K. E. Leopold, unpublished.

[19] C. A. Brau, A. E. Greene, and S. D. Rockwood, "Theoretical model for discharge excited rare gas halide lasers," presented at *2nd Wint. Coll. Laser Induced Chem.*, Park City, UT, Feb. 13-16, 1977; C. Brau, "Excimer lasers," presented at *3rd Int. Cong. Int. Trade Fair*, June 20-24, 1977, Munich, West Germany.

[20] W. H. Long, Jr., "Electron kinetics in the KrF laser," *Appl. Phys. Lett.*, vol. 31, pp. 391-394, Sept. 15, 1977.

[21] J. Hsia, "A model for uv preionization in electric-discharge-pumped XeF and KrF lasers," *Appl. Phys. Lett.*, vol. 30, pp. 101-103, Jan. 15, 1977.

[22] J. C. Martin, "Pressure dependency of the pulse breakdown of gases," AWRE Dielectric Strength Notes, no. 15, Sept. 26, 1967; —, "Pulsed surface tracking in air and various gases," AWRE SSWA/JCM/745/735, May 1974.

[23] A. V. Grigor'ev, P. N. Dushuk, S. N. Markov, V. L. Shutov, and M. D. Yurysheva, "Low-inductance megampere-current commutator based on sliding discharge," *Instrum. Exp. Tech.*, (USSR), vol. 19, pt. 2, pp. 1104-1105, July-Aug. 1976.

[24] C. P. Christensen, "High-repetition-rate XeF laser with gas recycling," *Appl. Phys. Lett.*, vol. 30, pp. 483-484, May 1, 1977.

[25] W. J. Sarjeant and A. J. Alcock, "High-voltage probe system with subnanosecond rise time," *Rev. Sci. Instr.*, vol. 47, pp. 64-68, Oct. 1976.

[26] J. B. Laudenslager, T. J. Pacala, and C. Wittig, "Electric-discharge-pumped nitrogen ion laser," *Appl. Phys. Lett.*, vol. 29, pp. 580-582, Nov. 1, 1976.

[27] J. M. Hoffman, A. K. Hays, and G. C. Tisone, "High-power uv noble-gas-halide lasers," *Appl. Phys. Lett.*, vol. 28, pp. 538-539, May 1, 1976.

Saturation and Tuning Behavior of Stimulated Electronic Raman Scattering

DAVID COTTER AND DAVID C. HANNA

Abstract—The saturation behavior of stimulated electronic Raman scattering (SERS) in atomic vapors is studied theoretically and experimentally. Limits to the SERS output energy are imposed by both pump depletion and saturation of the Raman transition. The effects of these on the performance of tunable infrared lasers based on SERS are examined.

I. INTRODUCTION

STIMULATED electronic Raman scattering (SERS) produced by pulsed dye laser beams in atomic vapors provides a simple method of generating tunable IR radiation [1]-[8]. Previously in [9], we considered the threshold behavior of such a process, and in particular it was shown that the Stokes wave diffraction loss can be large and this has the effect of substantially increasing the threshold pump power. In this paper the region of large-signal growth above threshold is considered in detail. The mechanisms which limit the output from SERS and the influence which these have on the behavior of tunable IR lasers based on SERS are examined.

The most straightforward SERS arrangement is one in which the focused dye laser beam passes through the atomic vapor cell once only. It has been shown that this single-pass configuration is sufficient to provide wide IR tuning ranges with peak photon conversion efficiencies of up to 50 percent [2]-[4], [6]. The obvious advantage of such an arrangement is its simplicity, since it involves no critical alignment. Moreover, we have so far found that multipassing or resonating either or both the pump and Stokes waves (by deliberately aligning the beams perpendicular to the vapor cell windows or by using external mirrors) have produced only a marginal reduc-

---

Manuscript received August 24, 1977; revised October 17, 1977. This work was supported by a research grant from the Science Research Council, United Kingdom.

D. Cotter was with the Department of Electronics, University of Southampton, Southampton, England. He is now with Abteilung Laserphysik, Max-Planck-Institute for Biophysical Chemistry, Göttingen, West Germany.

D. C. Hanna is with the Department of Electronics, University of Southampton, Southampton, England.

0018-9197/78/0300-0184$00.75 © 1978 IEEE

tion of threshold and increase of tuning range. This is in marked contrast with stimulated vibrational Raman scattering in gases, where typically it is found that the threshold can be considerably reduced by feedback from the cell windows [10]. The small effect of feedback in our SERS work is a consequence of the large IR diffraction losses prevailing under the conditions of our experiments [9]. Therefore, throughout the major part of this paper the treatment and discussion will be confined to single-pass generation.

II. Saturation Mechanisms

Previously in a SERS experiment using barium vapor, Carlsten and Dunn [2] clearly identified two different saturation processes. In the first case, the Stokes output is limited when the number of photons generated becomes comparable to the number of incident pump photons (*pump depletion*). The second limitation results from the excitation of the medium from the initial state to the final Raman level. As will be discussed later, in the various schemes which have been studied involving vapors of alkalis and alkaline earths, only an insignificant number of excited atoms relax back to the initial state within the pump pulse duration (typically in the range 5-30 ns). Therefore, the Stokes output is limited by the available number of initial state atoms (and we will refer to this as *atom depletion*). In the latter case Carlsten and Dunn [2] were able to demonstrate that the maximum Stokes energy generated in their experiment fell as the vapor pressure was reduced below a critical value at which the number of atoms present in the pump beam path was comparable to the number of incident pump photons.

It is the consequences of these two saturation mechanisms which are considered in detail here. In addition there are a number of other processes which will ultimately limit the generated output energy, but they are neglected in the analysis. Some of these limiting effects, including optical Stark shifts and gas breakdown, have been discussed elsewhere in connection with the behavior of other two-photon resonant nonlinear processes [11]-[14]. Also, the various competing parametric conversion processes are neglected in the analysis since the efficiency of these is generally much lower than that of SERS [15].

An important consequence of the atom depletion process is that it makes the Stokes output energy dependent on the small-signal Raman gain. If the small-signal gain is reduced (for example, by tuning the pump laser further from the intermediate single-photon resonances) the length of vapor column required for the small-signal exponential growth of the Stokes radiation is thereby increased. This effectively reduces the remaining length of the vapor column over which large-signal growth (involving atom depletion) can occur. Thus the number of atoms which are able to contribute to the output is reduced, and so the observed Stokes output energy is lowered. (Note that in most experiments it is the total *energy* of the Stokes output which is observed since the response time of the detector is usually longer than the pulse duration.)

These points concerning saturation of the Raman transition can be illustrated by means of the following simplified model for the SERS process. (A more detailed and quantitative analysis is described in Section IV.) Consider the one-dimensional problem of a pulse of the pump radiation which at time $t = 0$ enters the vapor column occupying the region $0 \leqslant z \leqslant L$. The fractional populations for the initial and final Raman levels are $\rho_g(z, t)$ and $\rho_f(z, t)$, respectively, and it is assumed that all the atoms in the vapor occupy the initial Raman level before the arrival of the pump pulse,

$$\rho_g(z, t) = 1 \text{ for } ct - z \leqslant 0. \tag{1}$$

The number density of Stokes photons in the forward traveling generated wave, given by $\mathfrak{N}_{sf}(z, t)$, is described by the photon transport equation

$$\left(\frac{\partial}{\partial z} + \frac{1}{c}\frac{\partial}{\partial t}\right)\mathfrak{N}_{sf} = gI_p(\rho_g - \rho_f)\mathfrak{N}_{sf} \tag{2}$$

where I_p is the pump intensity, and g is the SERS small-signal exponential gain coefficient calculated assuming that all the atoms are in the initial state. The rate of removal of atoms from the initial Raman level is the same as the rate of creation of Stokes photons

$$\frac{\partial \rho_g}{\partial t} = -\frac{c}{N}\left(\frac{\partial}{\partial z} + \frac{1}{c}\frac{\partial}{\partial t}\right)\mathfrak{N}_{sf} \tag{3}$$

where N is the total atomic number density, and for the present, a two-level atom approximation will be used

$$\rho_g + \rho_f = 1. \tag{4}$$

For mathematical convenience we will consider the case of a rectangular incident pump pulse of width τ

$$\begin{aligned} I_p(0, t) &= I_{po} \quad \text{for } 0 \leqslant t \leqslant \tau \\ &= 0 \text{ otherwise.} \end{aligned} \tag{5}$$

The analysis is further simplified by neglecting pump depletion

$$I_p(z, t) = I_p(0, t - z/c) \tag{6}$$

and also by neglecting backward wave Stokes generation. (All of these restrictions will be lifted in Section IV.)

This treatment of SERS has been formulated in such a way that it is very similar to the problem of pulse propagation and saturation in a laser amplifier, which has been analyzed by Frantz and Nodvik [16]. The steps which are followed in solving equations (1)-(6) are essentially the same as those in [16], and so only the following final solution will be given here

$$\mathfrak{N}_{sf}(z, t) = \frac{\mathfrak{N}_{so}}{1 - [1 - \exp(-gI_{po}z)] \exp[-2gI_{po}\mathfrak{N}_{so}(ct - z)/N]}. \tag{7}$$

The initial boundary condition

$$\begin{aligned} \mathfrak{N}_{sf}(0, t) &= \mathfrak{N}_{so} \quad \text{for } 0 \leqslant t \leqslant \tau \\ &= 0 \text{ otherwise} \end{aligned} \tag{8}$$

has been assumed, and thus $\mathfrak{N}_{so}$ represents the effective initial photon density produced by spontaneous scattering.

From (1), all of the atoms encountered by the leading edge of the pump pulse as it travels through the vapor are in the initial state. Thus at the leading edge ($ct - z = 0$), (7) shows that the Stokes photon density grows according to the familiar exponential law

$$\mathcal{N}_{sf}(z, z/c) = \mathcal{N}_{so} \exp (gI_{po}z). \tag{9}$$

However, in the tail of the pump pulse the initial state population has already been reduced by the leading part of the pulse, and so the Stokes radiation grows at a slower rate.

The parameter which is of practical importance is the total Stokes *energy* gain G_E calculated by integrating (7)

$$G_E(z) = \frac{\int_{-\infty}^{+\infty} \mathcal{N}_{sf}(z, t)\, dt}{\mathcal{N}_{so}\tau} = \frac{1}{2gI_{po}\eta} \cdot \ln \{1 + \exp (gI_{po}z) [\exp (2gI_{po}\eta) - 1]\} \tag{10}$$

where $\eta = \mathcal{N}_{so}c\tau/N$. Two limiting regimes of (10) are of interest. In the small-signal growth regime near the beginning of the vapor column (i.e., for small z), there is a negligible reduction of the atomic initial state population and the exponential growth behavior of the Stokes photon density remains valid throughout the pump pulse. Thus, from (7) it can be inferred that the trailing edge will also be amplified exponentially ($\mathcal{N}_{sf}(z, \tau + z/c) \sim \mathcal{N}_{so} \exp (gI_{po}z)$) only if

$$\exp (gI_{po}z) \ll (2gI_{po}\eta)^{-1}. \tag{11}$$

By inserting the inequality (11) into (10), an expression for the exponential growth of the Stokes *energy* in the small-signal limit is recovered

$$G_E \sim \exp (gI_{po}z) \tag{12}$$

which, as expected, has the same form as the small-signal growth of *power*. However in the large-signal regime at distances further into the vapor (in the limit $gI_{po}z \gg 1$), (10) leads to

$$G_E \sim \frac{z}{2\eta}. \tag{13}$$

The large-signal growth of energy is thus *linear* with distance z, and (13) implies that the Stokes photon density has grown to the point at which it is sufficient to deplete (via SERS) the ground state atomic population in the latter part of the vapor column. This depletion reaches the point at which the Raman gain falls to zero when the populations in the initial and final Raman levels have become equal, and this accounts for the factor 2 in (13). As discussed in Section IV, another case of experimental interest [cf. (4)] is given by

$$\rho_f \approx 0 \quad \text{for all } z, t \tag{14}$$

corresponding to the situation where atoms decay rapidly from the final Raman level. The analysis can be repeated in this case with (4) replaced by (14) to give identical solutions (7)-(13) except that all factors of 2 are omitted.

A consequence of the atom depletion is that the output energy may be much lower than that determined solely by pump depletion. Atom depletion effects rapidly become noticeable in SERS experiments in alkali vapors at pressures of less than a few torr, and an example of this behavior is shown in Fig. 2.

III. EXPERIMENTAL OBSERVATIONS OF SATURATION

All of the experimental results in this paper refer to a tunable IR laser system based on SERS in caesium vapor. The practical details of this have been described elsewhere [6]. Briefly, it consists of a nitrogen laser pumped dye laser which produces an output of 140-180 μJ in 6-7-ns pulses (20-25 kW), 0.1-cm^{-1} bandwidth and diffraction-limited beam, tunable in the region 448-467 nm. The dye laser output pumps the 6-7 s electronic Raman transition in caesium (Raman shift 18536 cm^{-1}), as shown in Fig. 1. The caesium vapor is contained in a heat pipe oven. The resulting IR output is tunable in the range 2.7-3.5 μm with a peak output power of up to 1.5 kW. For the work described here, the energy of the IR beam emerging from the heat pipe was measured using a calibrated pyroelectric detector in combination with optical filters or a monochromator. Care was taken to ensure that the measurements were made under conditions in which absorption of the dye laser beam by Cs_2 dimers was insignificant [6].

The upper curve in Fig. 2 was obtained using a caesium vapor pressure of 10 torr and with the dye laser beam focused confocally over the ~20-cm vapor column (i.e., focused to a waist at the center of the vapor with a confocal parameter equal to the vapor column length). In [6] it is shown that these are the optimum operating conditions for this laser system. The dye laser was tuned ~38 cm^{-1} below the $6s$-$7p_{1/2}$ resonance, this being in the region of maximum IR output energy for these conditions. A dye laser energy greater than ~5 μJ is necessary to obtain a detectable IR output (>0.8 nJ). Above this threshold the IR output increases rapidly as the dye laser energy is increased, until dye laser energies of greater than ~40 μJ produce IR outputs which are limited by pump depletion. The IR energy then increases almost linearly with pump energy up to the maximum available, and this represents a nearly constant photon conversion efficiency of 40-50 percent.

The lower curve in Fig. 2 was obtained using the same degree of focusing but with the caesium pressure reduced to ~0.015 torr. (This was achieved by reducing the vapor cell temperature to ~164°C. The argon buffer gas pressure was kept at 10 torr, and the cell was therefore no longer operating in the "heat-pipe" mode. The vapor column length was then ~10 cms.) In this case the dye laser was tuned ~4 cm^{-1} below the $6s$-$7p_{1/2}$ resonance. The fact that the pump was tuned very much nearer to the intermediate resonances explains why the SERS pump threshold was comparable despite the lower vapor pressure. However the IR output was no longer limited by pump depletion, but by the reduced number of atoms in the pump beam path. With the full dye laser input to the cell (~140 μJ), the number of Stokes photons detected was ~6.5 × 10^{12}. This is equal to the number of atoms in a cylindrical vapor column of radius ~$2W_p$ where W_p is the 1/e field radius of the dye laser beam (~140 μm). The onset of saturation due to atom depletion as shown in Fig. 2 is less abrupt than in the observations by Carlsten and Dunn [2]. An explanation for this is that in their experiment, Carlsten and

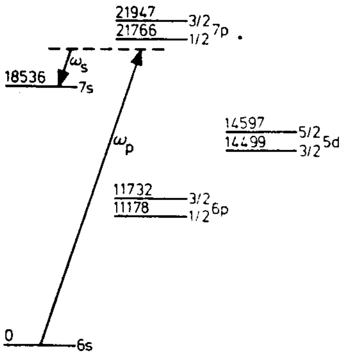

Fig. 1. Caesium energy-level diagram showing the 6s-7s SERS transition (energies in cm^{-1}).

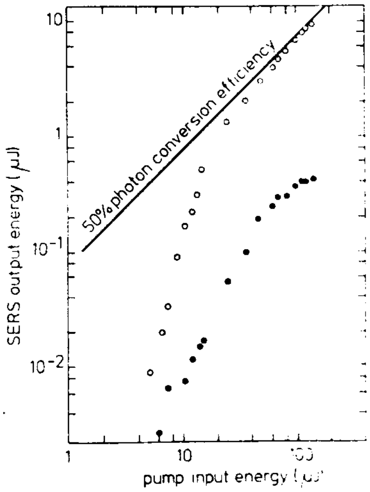

Fig. 2. SERS input-output characteristics at Cs vapor pressures of 10 (o) and 0.015 torr (•). The dye laser frequency was not the same for the two pressures (see text).

Dunn [2] had available a high-energy (250 mJ) dye laser beam and so could use a large beam area (larger than the cross-sectional area of the vapor region). They therefore had a uniform beam intensity over a well-defined area of vapor column. However, the dye laser energy available for our measurements shown in Fig. 2 was much lower, and in this case it was necessary to tightly focus the beam in the vapor. (As well as achieving a good Raman gain, it is necessary to focus the dye laser beam in the vapor in order to saturate the absorption by Cs_2 dimers, and thus keep this absorption to an insignificant level [6]). The beam had a nearly Gaussian intensity distribution, and this would mean that close to threshold only those atoms which are near to the beam axis are able to contribute to the output, whereas well above threshold, the useful beam diameter is effectively increased and thus a greater number of atoms can contribute.

For the results shown in Fig. 3 the vapor pressure was held fixed at 10 torr, and the output versus input curves were obtained with the dye laser tuned progressively further from the intermediate 6s-7p resonances. The pump energy input to the vapor cell was varied by means of calibrated attenuating filters. As the dye laser is tuned further from resonance the SERS threshold is seen to increase. This is to be expected because of the reduced small-signal gain. Also it is seen from Fig. 3 that the IR output energy for a given dye laser energy decreases as the dye laser is tuned off resonance. Clearly the IR output energy is not then limited by pump depletion. The fact that the IR output energy is then found to be *consistently* lower than the pump depletion limit does however indicate the presence of a saturation process, and this behavior is readily understood in terms of the simple notion of atom depletion described above. It should be noted that to amplify the spontaneous Raman emission to a level at which the pump is depleted in our experimental arrangement (and in the SERS experiments reported by others) requires a small-signal power gain of the order of exp 36. If it is argued that the reduction of IR output with detuning is simply because the gain is reduced (to exp 34 say), then small fluctuations of pump intensity would produce very large changes in the value of this exponential and hence of IR output energy. This is not observed, although it is observed that the IR output energy fluctuations do increase steadily with detuning. This is also explained (in Section IV) in terms of the atom depletion model.

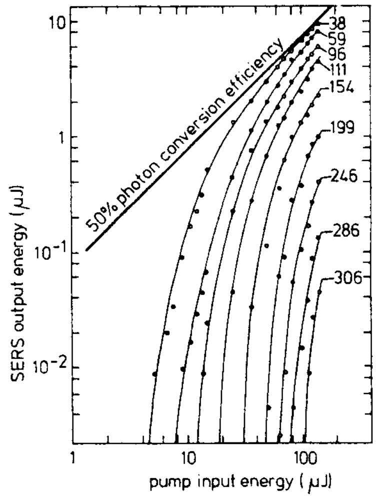

Fig. 3. SERS input-output characteristics at 10-torr Cs vapor pressure and for various amounts of pump detuning $\Omega_{6s-7p_{1/2}} - \omega_p$ (in cm^{-1}).

Recently Kung and Itzkan [7], [8] have published curves similar to those in our Fig. 3 and have argued that by taking the gradient (using a semilogarithmic plot of SERS output versus pump input) one can arrive at a value of the Raman gain coefficient gl_p. The values which they obtained are however as much as an order of magnitude smaller than the required $gl_p \sim 30$-40. We claim that their results (and ours) cannot be interpreted in this way and that the true explanation lies in the atom depletion behavior described above. Meaningful gain measurements can be made by observing the amplification of an injected pulse of radiation at the Stokes frequency, but it would be necessary to carry out this measurement under carefully controlled conditions of low gain if the influence of both pump depletion and initial state depletion is to be avoided (as for example in [17]). In a separate experiment we have used the SERS process to amplify injected radiation from a He-Ne laser at 3.39 μm (the dye laser having been tuned to provide Raman gain at that wavelength). The results of this experiment are again consistent with the notion of atom depletion. Thus, *without* the injected 3.39 μm radiation and with the dye laser power at its maximum, the spontaneous Raman emission at 3.39 μm was amplified to a level which gave an IR output of ~30-W peak power, being limited by atom depletion. The dye laser power was then reduced until there was no detectable 3.39-μm signal. When the He-Ne radiation (~90 μW CW) was then injected, the pulsed IR signal returned, and finally with the dye laser restored to full power the IR output was then ~120-W peak power, again limited by atom depletion. The observed signal was larger because a smaller length of vapor column was needed for small-signal gain, thus leaving a greater length of vapor over which atom depletion could occur. Clearly also the small-signal gain was greatly in excess of the values inferred by Kung and Itzkan [7], although we have not attempted to calculate this gain because of the uncertain magnitude of diffraction loss and effect of the narrow linewidth of the He-Ne signal, and also because the IR output is still limited by atom depletion.

Finally, we have shown that the atom depletion model provides an explanation for the observed tuning profiles in SERS. This is examined in detail in the following section.

IV. TUNING BEHAVIOR

Fig. 4 shows the measured IR output energy as a function of pump frequency when the dye laser energy input to the vapor was ~110 μJ. (Here, as before, the dye laser beam was focused confocally in the vapor which was at a pressure of 10 torr.) Fig. 5 uses a logarithmic scale to show the region below the $6s$-$7p_{1/2}$ resonance in greater detail. By comparing Figs. 3 and 5 it can be seen that the peak output is limited by pump depletion whereas at frequencies further from the $6s$-$7p_{1/2}$ resonance the output is limited by atom depletion.

In order to investigate this tuning behavior, a quantitative analysis of the saturation processes already described has been carried out. This analysis is more general than the illustrative calculations in Section II and includes the competition between forward and backward traveling Stokes waves and the effects of pump depletion. The coupled photon transport equations that are used are

$$\left(\frac{\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}+\alpha\right)\mathfrak{N}_{sf}=gch\omega_p\mathfrak{N}_p\{(\rho_g-\rho_f)\mathfrak{N}_{sf}+\rho_g\mathfrak{N}_{so}\} \tag{15a}$$

$$\left(\frac{-\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}+\alpha\right)\mathfrak{N}_{sb}=gch\omega_p\mathfrak{N}_p\{(\rho_g-\rho_f)\mathfrak{N}_{sb}+\rho_g\mathfrak{N}_{so}\} \tag{15b}$$

$$\left(\frac{\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}\right)\mathfrak{N}_p=-gch\omega_p\mathfrak{N}_p\{(\rho_g-\rho_f)(\mathfrak{N}_{sf}+\mathfrak{N}_{sb})\}. \tag{15c}$$

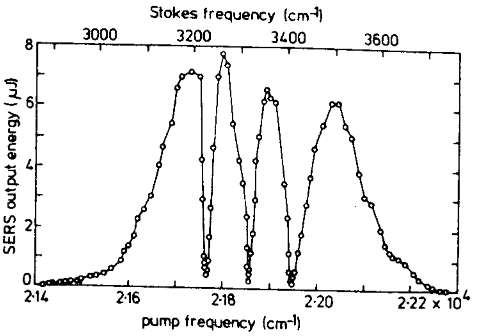

Fig. 4. SERS output tuning profile.

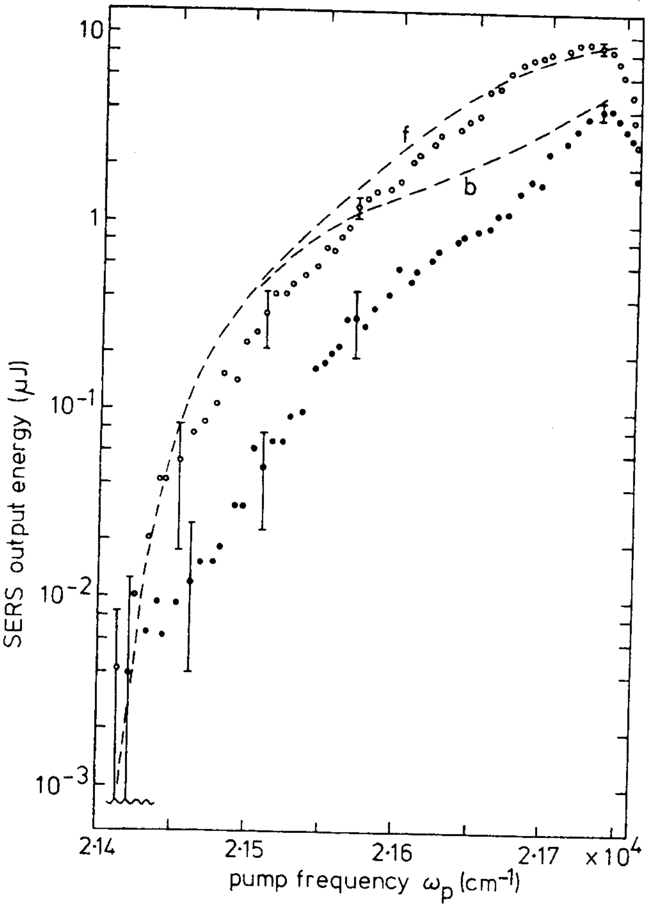

Fig. 5. SERS output tuning profile in the region below the $6s$–$7p_{1/2}$ resonance, showing forward (○) and backward (●) outputs. Detector noise limit is indicated by the wavy line. Broken lines show the calculated values for the forward (f) and backward (b) outputs.

The subscripts f and b refer to the Stokes photon number densities in the forward ($+z$) and backward ($-z$) traveling waves, respectively, and ω_p is the pump frequency. The term α has been introduced to represent the various losses suffered by the Stokes radiation. Since the IR absorption in alkali and alkaline earth atomic vapors is small, the major contribution to α is the diffraction loss, as discussed in [9]. Thus, although the treatment here is strictly one dimensional, the effects of diffraction can be included phenomenologically as a refinement.

The initial conditions of the medium are assumed as previously in (1), and subsequently each scattering event alters the atomic populations

$$\frac{\partial\rho_g}{\partial t}=+\frac{c}{N}\left(\frac{\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}\right)\mathfrak{N}_p. \tag{16}$$

Equation (16) differs from (3) by including the rate of creation of backward traveling Stokes photons.

The fractional atomic populations have been modeled in two different ways according to the type of SERS system being examined. Firstly, there is the type of SERS transition in which the final Raman level is a metastable state (lifetime much longer than the pump pulse duration). This is typified by the experiment of Carlsten and Dunn in barium vapor [2], in which they observed SERS from the singlet $^1S_0$ ground state to the lowest triplet $^3D_1$ and $^3D_2$ states. Then, neglecting absorption by the intermediate states, the simple two-level approximation (4) is used, and in this case saturation of the Raman gain at a point in the vapor occurs when the populations in the initial and final levels become equal.

Secondly, there is the type of transition, typified by SERS in alkali metal vapors, in which the lifetime of the final level is short compared to the pump pulse duration. For example, SERS transitions from the ground state of alkali metals involve $^2S_{1/2}$ states as final levels and the measured lifetimes of these final levels are typically a few tens or hundreds of nanoseconds. But the SERS process tends to produce a population inversion between the final Raman level and lower lying ^2P levels, and this inversion rapidly reaches a level at which strong amplified spontaneous emissions (ASE) can occur. To give a specific example, in the case of the SERS source using the $6s$-$7s$ transition in caesium (see Fig. 1), the tunable Stokes output is accompanied by strong fixed frequency emissions at 1.35 and 1.47 μm, which correspond to ASE on the transitions $7s$-$6p_{1/2,3/2}$. A simple calculation (assuming Doppler-broadened transitions) shows that population inversions on these transitions of as little as 10^{17} m^{-3} (cf. total number density of $\sim 10^{20}$-10^{23} m^{-3} at 0.01-10 torr) will be sufficient to produce stimulated emission gain coefficients of $\sim 10^3$ m^{-1}. Thus, the population produced in the $7s$ level by SERS will be rapidly transferred to the $6p$ levels (the lowest lying ^2P levels) and the effective lifetime of the $7s$ level may be reduced to the subnanosecond region. In the case of SERS transitions to higher excited $^2S_{1/2}$ states in caesium [4], ASE outputs with wavelengths corresponding to the cascading ladder of transitions from the final Raman level to the lowest ^2P levels are observed. The growing populations in these lowest ^2P levels are effectively radiation trapped there owing to the excess ground state population. This radiation trapping can result in effective lifetimes for these ^2P levels of hundreds of nanoseconds or more [18]. Atoms so trapped are prevented from returning to the ground state and being "recirculated" by SERS. The modeling of this highly complex situation has therefore been simplified by assuming that the atomic population in the final level is always practically zero, as in (14), and (16) still allows for depletion of the population in the initial level.

Computer solutions for the coupled equations (15), (16), and (4) or (14) have been obtained for a variety of practical situations, and the results obtained using the two different models are qualitatively similar. The results shown here were obtained using the second model [with (14)] to simulate the tuning behavior of the caesium SERS source described earlier. The computer was used to integrate the coupled equations over a complete input pump pulse in order to calculate the total Stokes output energy for various dye laser wavelengths. The nonlinear atomic susceptibility, and hence the gain coefficient g, were calculated as a function of pump frequency as described in [5]. It was assumed that the dye laser input to the vapor occurs in a sinc^2 pulse (truncated at the first zero) of FWHM duration 6 ns and total energy 100 μJ, and the other parameters were also given values appropriate to the practical SERS source.

The dashed curves in Fig. 5 show the calculated SERS output energy in the forward and backward directions. The fit between theory and experiment for the forward wave output is very good, and is indeed better than might be expected in view of the many simplifications in the theory. This fit was obtained by scaling only *one* variable parameter, the spontaneous Raman linewidth Γ which enters into the expression for the nonlinear susceptibility [5]. The experimentally measured IR output linewidth is in the range 0.25–0.44 cm^{-1} (FWHM), and within this range the linewidth is observed to increase as the dye laser is tuned closer to the $7p_{1/2}$ resonance [6]. The theoretical curves in Fig. 5 which gave a best fit to the experimental results were calculated by using at each point in the tuning range a value for Γ (FWHM) which is twice the measured output linewidth.

The numerical calculations also predict that the peak *backward* Stokes output energy should be a factor 2-2.5 times smaller than that in the forward direction, and this is in good agreement with experiment (Fig. 5). (The explanation for this lies in the dynamic competition between the forward and backward Stokes waves in which the forward wave benefits from traveling together with the pump input.) However there are certain observations which are not adequately explained by the theory. One of these is that over most of the tuning range the backward wave output is much lower than predicted. In this respect the discrepancy between theory and experiment is as much as an order of magnitude at some parts of the tuning range, as shown in Fig. 5.

A further unexplained observation is the slight asymmetry of the tuning profile shown in Fig. 4, the IR output peaks in the region of the $6s$-$7p_{3/2}$ resonance being somewhat lower than those around the $6s$-$7p_{1/2}$ resonances. Kung and Itzkan [7] have recently offered an explanation of an asymmetry in their observations in terms of gain focusing. However, the mechanism which they invoke is that of dispersion focusing which depends on whether $\omega_p - \omega_s$ is greater or less than the *two-photon* Raman transition frequency. The asymmetric behavior in Stokes output which they observed was with respect to the pump frequency tuned above or below the *single-photon* intermediate resonance. These two effects are entirely unrelated.

The theoretical model as described does not account for the behavior in the regions close to the intermediate resonances where sharp dips in the IR output energy are observed (Fig. 4). This is because the theory neglects depletion of the pump or excitation of atoms resulting from the various processes acting in competition with SERS. In addition to single-photon absorption of the pump by the intermediate levels (which are strongly resonance broadened), there are a number of nonlinear processes such as multiphoton ionization [19] which become significant under these highly resonant conditions.

The influence of these competing effects on SERS is complex, and a detailed treatment is beyond the scope of the present work. Similarly, the dip in the IR output for pump frequencies nearly midway between the resonance doublet is not accounted for in the theory. As explained in [5], this dip is *not* caused by a cancellation of Raman susceptibility contributions from the $7p_{3/2}$ and $7p_{1/2}$ levels. This minimum is not accompanied by any observable increase in attenuation of the dye laser beam, and we believe that in this region a large proportion of the generated Stokes radiation is subsequently absorbed by the considerable population of excited $6p_{1/2}$ atoms. This level is populated by ASE from the $7s$ and $5d$ levels. At the pump frequencies corresponding to this minimum IR output (21855 ± 2 cm^{-1}) the corresponding Stokes frequency is then in close resonance with the $6p_{1/2}$ - $5d_{3/2}$ transition [5]. The $6s$-$7s$ SERS transition in caesium appears to be unique in having this close coincidence of transition frequencies.

A further aspect of the saturation and tuning behavior of SERS sources is their shot-to-shot amplitude stability. The vertical bars on the experimental points in Fig. 5 indicate the ranges of energies which are measured during a sequence of pulses. At the peaks of the tuning profile, where the output is limited by pump depletion, the output amplitude stability is the same as that of the dye laser (about ± 5 percent with the nitrogen laser pumped dye lasers in use in our laboratory). However, towards the edges of the tuning range, where the output is limited by atom depletion, a small variation in the pump energy can produce a large change in the IR output, and shot-to-shot variations can be as great as an order of magnitude.

Finally, a brief description is given of experimental results obtained using a double-pass SERS configuration. It is apparent from the theory that by reflecting a proportion of the generated SERS radiation back into the vapor cell it should be possible to obtain a higher output energy since a greater number of atoms would then be capable of contributing to the large-signal growth. The arrangement shown in Fig. 6 was used. Allowing for the losses in the "Infrasil" heat pipe windows, Ge filter and KBr lens, the proportion of the forward IR energy fed back was certainly no greater than 10 percent. Nevertheless, as shown in Fig. 7, the backward output energy was increased by a factor of 2.5-4 over most of the tuning range. (The long distance (~1 m) between the lens $L1$ and IR detector at the opposite end of the heat pipe ensured that the observed increase in output energy was not simply due to the reflected portion of the forward wave being *added* to the backward wave output.) A detailed quantitative analysis of this experiment has not been attempted here. However, it is clear that by using a dye laser pulse of longer duration than the 6-7 ns available for this experiment, it should be possible to construct a multipass SERS resonator. Whilst it is possible that the IR diffraction loss in such a resonator may result in only a marginal reduction in SERS threshold and increase of tuning range [9], nevertheless a useful improvement in output energy could be expected. (However many passes are required for the small-signal growth, in a resonator *all* of the pumped atoms would finally be able to contribute to the IR output.) A SERS resonator could then exhibit the saturation behavior characteristic of a laser oscillator, with the output increasing rapidly above threshold and limited mainly by pump depletion over most of the tuning range. To test these suggestions will, however, require a higher power and longer pulse than is available from our dye laser. The higher power will allow the use of a larger area beam and hence a reduction in the SERS diffraction loss.

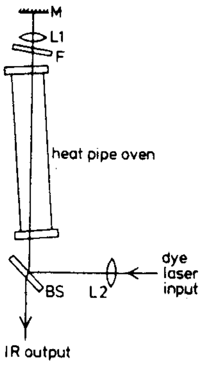

Fig. 6. Double-pass arrangement for SERS generation: BS–dichroic beam splitter (reflecting the dye laser beam and transmitting the generated IR), M–plain aluminum mirror, $L1$–5-cm KBr lens, $L2$–1-m glass lens (producing confocal focusing of dye laser beam in vapor), F–polished wedged Ge slice (prevents damage to M by dye laser beam).

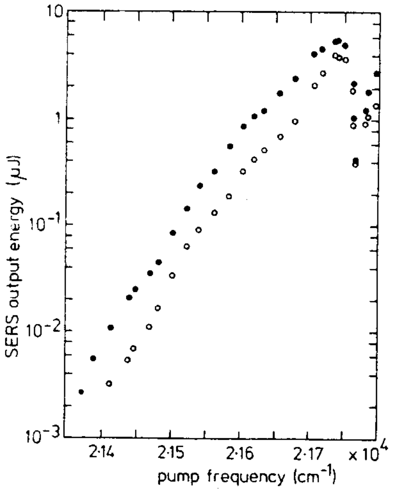

Fig. 7. Tuning profiles comparing the backward SERS output energies obtained using a single-pass arrangement without feedback (○) and using the double-pass arrangement shown in Fig. 6 (●).

V. CONCLUSIONS

In this paper the saturation behavior of tunable IR lasers based on SERS in atomic vapors has been described. Limits to the IR output energy are imposed by the onset of pump depletion and the limited number of atoms in the beam path. It has been demonstrated and explained that even when using vapor pressures at which the number of atoms in the beam exceeds the number of incident pump photons, the IR output may still be considerably less than the limit imposed by pump depletion alone.

The theory provides an explanation for a number of features of the observed profile of IR output energy as a function of pump frequency. The use of multipass SERS configurations should result in a substantial increase in output energy over a large part of the tuning range, although a better understanding of the backward wave generation is still needed. An understanding of the saturation behavior of SERS is also important for a quantitative analysis of the various hybrid nonlinear schemes which use parametric mixing processes in which one of the interacting waves is provided by SERS [1], [15].

It is interesting to compare the saturation behavior of SERS with that of other types of IR generation by stimulated Raman scattering. In stimulated vibrational Raman scattering in gases (see for example [20]), the molecular number densities commonly used are three or more orders of magnitude greater than in the atomic vapors used for SERS. Consequently, it is usually possible to operate in such a way that the effects of depleting the molecular ground state are negligible, and the Stokes output is limited by pump depletion and by generation of higher order Stokes and anti-Stokes emissions [21]. The opposite is true of pulsed spin-flip Raman lasers in which the primary output limitation is saturation of the electron spin transition [22].

Finally, similar saturation and tuning behavior to that described here has recently been observed by us in stimulated electronic *hyper*-Raman scattering in sodium vapor [23].

ACKNOWLEDGMENT

The authors would like to thank Dr. M. A. Yuratich for useful discussions.

REFERENCES

[1] P. P. Sorokin, J. J. Wynne, and J. R. Lankard, "Tunable coherent IR source based upon four-wave parametric conversion in alkali metal vapours," *Appl. Phys. Lett.*, vol. 22, pp. 342-344, 1973.

[2] J. L. Carlsten and P. C. Dunn, "Stimulated Stokes emission with a dye laser: Intense tunable radiation in the infrared," *Opt. Commun.*, vol. 14, pp. 8-12, 1975.

[3] D. Cotter, D. C. Hanna, P. A. Kärkkäinen, and R. Wyatt, "Stimulated electronic Raman scattering as a tunable infrared source," *Opt. Commun.*, vol. 15, pp. 143-146, 1975.

[4] D. Cotter, D. C. Hanna, and R. Wyatt, "A high power, widely tunable infrared source based on stimulated electronic Raman scattering in caesium vapour," *Opt. Commun.*, vol. 16, pp. 256-258, 1976.

[5] D. Cotter and D. C. Hanna, "Effects of spin-orbit coupling on resonant enhancement of stimulated electronic Raman scattering," *J. Phys. B: Atom. Molec. Phys.*, vol. 9, pp. 2165-2171, 1976.

[6] —, "Stimulated electronic Raman scattering in Cs vapour: A simple tunable laser system for the 2.7 to 3.5μm region," *Opt. Quantum Electron.*, vol. 9, pp. 509-518, 1977.

[7] R. T. V. Kung and I. Itzkan, "Tunable infrared laser generated via stimulated Raman scattering in Cs vapor," *IEEE J. Quantum Electron.*, vol. QE-13, pp. 73-79, Mar. 1977.

[8] —, "16μ and 8.5μ generated via stimulated Raman process in potassium vapor," *Appl. Phys. Lett.*, vol. 29, pp. 780-783, 1976.

[9] D. Cotter, D. C. Hanna, and R. Wyatt, "Infrared stimulated Raman generation: Effects of gain focussing on threshold and tuning behaviour," *Appl. Phys.*, vol. 8, pp. 333-340, 1975.

[10] G. Haidacher and M. Maier, "Explanation of anomalies in the stimulated Raman scattering in H_2 gas," *IEEE J. Quantum Electron.*, vol. QE-10, pp. 786-787, Sept. 1974.

[11] R. B. Miles and S. E. Harris, "Optical third-harmonic generation in alkali metal vapors," *IEEE J. Quantum Electron.*, vol. QE-9, pp. 470-484, Apr. 1973.

[12] P. F. Liao and J. E. Bjorkholm, "Direct observation of atomic energy level shifts in two-photon absorption," *Phys. Rev. Lett.*, vol. 34, pp. 1-4, 1975.

[13] E. A. Stappaerts, "Harmonic generation at high field strengths: Frequency shifts and saturation phenomena," *Phys. Rev. A.*, vol. 11, pp. 1664-1667, 1975.

[14] J. N. Elgin and G. H. C. New, "Semi-classical theory of two-photon resonant third-harmonic generation," *Opt. Commun.*, vol. 16, pp. 242-246, 1976.

[15] P. A. Kärkkäinen, "Two-photon resonant parametric generation in alkali metal vapours," *Appl. Phys.*, vol. 13, pp. 159-163, 1977.

[16] L. M. Frantz and J. S. Nodvik, "Theory of pulse propagation in a laser amplifier," *J. Appl. Phys.*, vol. 34, pp. 2346-2349, 1963.

[17] J. J. Wynne and P. P. Sorokin, "Optical mixing in atomic vapors," in *Nonlinear Infrared Generation, Topics in Applied Physics*, vol. 16, Y.-R. Shen, Ed. Berlin, Heidelberg, New York: Springer-Verlag, 1977.

[18] T. Holstein, "Imprisonment of resonance radiation in gases," *Phys. Rev.*, vol. 72, p. 1212, 1947;
—, "Imprisonment of resonance radiation in gases. II," *Phys. Rev.*, vol. 83, pp. 1159-1168, 1951.

[19] E. H. A. Granneman and M. J. Van der Wiel, "Two-photon ionisation measurements on atomic caesium by means of an argon ion laser," *J. Phys. B: Atom. Molec. Phys.*, vol. 8, pp. 1617-1626, 1975.

[20] R. Frey and F. Pradere, "Tunable infrared generation by stimulated Raman scattering," *Infrared Phys.*, vol. 16, pp. 117-120, 1976.

[21] A. J. Glass and J. M McMahon, "Pulse shapes and spatial modes in Raman lasers," *IEEE J. Quantum Electron.*, vol. QE-5, pp. 1-6, Jan. 1969.

[22] W. J. Firth, "Power output of a pulsed Raman laser with saturable excitation," *IEEE J. Quantum Electron.*, vol. QE-8, pp. 869-872, Dec. 1972.

[23] D. Cotter, D. C. Hanna, W. H. W. Tuttlebee, and M. A Yuratich, "Stimulated hyper-Raman emission from sodium vapour," *Opt. Commun.*, vol. 22, pp. 190-194, 1977.