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ABSTRACT

This note deals with the power and mode-matching requirements for generating wavelengths in the 16 μm region by stimulated Raman scattering in liquid nitrogen. Specific examples are given for pumping both resonant i.e. Raman oscillator, and non-resonant configurations with a Nd:YAG driven OPO, together with some comments on alternative sources.

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For efficient Raman conversion to the Stokes' shifted wavelength, the overall gain required to generate a strong signal from noise is $\sim e^{23}$, where the exponent is the intensity-dependent gain-length product $g_R I_P L$; g_R is the Raman gain coefficient, I_P the pump intensity and L the interaction length. For a liquid column situated at the focus of a gaussian pump beam, the axial intensity I_P at the beam-waist is related to the pump power P_P and the beam-waist radius w_o by $I_P = \frac{P_P}{\pi w_o^2}$, and an effective interaction length can be defined as the distance over which this intensity remains within a factor 2 of the peak value, ie $L = 2 z_o$ (the confocal parameter) where $z_o = \frac{\pi w_o^2}{\lambda}$. Combining these factors, we find a requirement on the pump power given by

$$P_P^{(1)} \simeq \frac{23 \lambda}{2K g_R} \qquad \ldots (1)$$

where K is an integration constant ($\pi/4 \leqslant K \leqslant \pi/2$ when $2 z_o \leqslant L \leqslant \infty$). This is a minimum power requirement for strong pump depletion and large Raman conversion in a single pass, and is obviously an upper limit on the pump power in the sense that no feedback is assumed. It can, however, be reduced by increasing the interaction length L by successive refocusing of the pump beam[(1)], or by multi-pass operation.

In the case of multi-pass gain, which would occur in a Raman oscillator, or double-pass gain provided by a mirror reflecting the pump radiation at the far end of the Raman cell, the pump power requirement decreases proportionally. However, for a Nd: YAG OPO pump source, the short pulse duration effectively limits the number of transits in the cavity to $N = L_p/L_c$, where the pulse length $L_p \approx c\tau_p$, τ_p is the pulsewidth and L_c the cavity length. The maximum interaction length is now L_p, and the required pump power becomes

$$P_P^{(2)} \simeq \frac{23\pi w_o^2}{K g_R L_p} \qquad \ldots (2)$$

with the matching requirement

$$L_p \sim \frac{L_c}{1 - \sqrt{R_1 R_2}} \qquad \ldots (3)$$

where R_1, R_2 are the mirror reflectivities for the Stokes' wavelength (this ensures that the mean residence time of a Raman scattered photon in the cavity matches the duration of the pump pulse). Clearly, the pumping power can be reduced if $P_p^{(2)} < P_p^{(1)}$, ie if

$$\frac{2\pi w_o^2}{\lambda L_p} < 1 \qquad \ldots (4)$$

which shows that tight focusing in a short cavity can reduce the input power requirement by a considerable factor, because we can make $L_c << L_p$ and choose high reflectivity mirrors to satisfy (3). In this way the seemingly unphysical condition (4) is readily achieved!

For a Stokes' shifted wavelength λ_s around 16μm - far from the pump wavelength λ_p = 3.4μm in liquid nitrogen - the question of mode matching the pump and Raman beams for maximum conversion arises. The larger the beam-waist, the higher the pump power required to achieve a given intensity. The choice of λ_s = 16 μm for determining the spot size at the focus of the pump beam is arguably not the optimum here; some compromise between minimising the pump power through sharp focusing of the input beam and maximising the coupling efficiency by mode matching to the Stokes' radiation is required. However, in the case of the Raman oscillator, it would probably be necessary to match the pump beam to the fundamental transverse cavity mode for the Stokes' wavelength, to reduce diffraction losses to a suitably low value.

Taking the published value of the Raman gain coefficient (2) in liquid N_2 at λ_p = 0.7 μm as g_R = 1.6 10^{-2} cm(MW)$^{-1}$, and assuming $g_R \propto \lambda_s^{-1}$, the estimated gain for λ_s = 16 μm is $g_R = 7.10^{-4}$ cm(MW)$^{-1}$. With this value for g_R and the geometric mean wavelength $\lambda = \sqrt{\lambda_p \lambda_s} \simeq 7.4$μm (as $P_p^{(1)} \propto \lambda_p \lambda_s$) we find the pump power requirement from equation (1): $P_p^{(1)} \simeq 24$ MW. Comparing this with the experimental value[1] of 16 MW for <u>three consecutive foci</u> of the 3.4 μm pump wavelength, for which we would estimate a pump power ∿ 8 MW, we see the agreement is satisfactory, particularly when diffraction losses resulting from the mode mis-match are taken into account.

For a confocal cavity, the relation of the beam-waist radius w_o and the cavity length $L_c = 2z_o$ is

$$L_c = \frac{2\pi w_o^2}{\lambda} \qquad \ldots (5)$$

If the pump beam is refocused and reflected into the cavity for a second pass, the interaction length $L = 2L_p$ and the required pump power for achieving oscillation and efficient conversion to the Stokes' wavelength becomes

$$P_p^{(3)} \simeq \frac{\left[23 + (L_P/L_c)\ \ln\left(\frac{1}{R_1R_2}\right)\right] 4w_o^2}{g_R\ L_p} \qquad \ldots (6)$$

where the second term in the numerator is the cavity loss (threshold gain) summed over the total number of transits during the development of the Stokes' radiation. Writing $N = L_p/L_c$, the effective number of transits for a <u>single</u> pass of the pump beam, and assuming mode-matching of the pump light to the cavity mode at the Stokes' wavelength λ_s, we have finally

$$P_p^{(3)} \simeq \frac{\left[23 + N\ \ln\left(\frac{1}{R_1R_2}\right)\right] \lambda_s}{\pi\ N\ g_R} \qquad \ldots (7)$$

Comparison with the single pass saturation gain requirement of equation (1) shows a reduction in pump power by a factor $\lesssim$ 2N. For a double pass of an OPO pulse at $\lambda_p = 3.4$ µm in a Raman oscillator cavity with $\sqrt{R_1R_2} = 0.9$ ($N \simeq 10$), we find from equation (8)

$$P_p^{(3)} \simeq 1.8\ \text{MW} \qquad \ldots (8)$$

which represents a reduction of the estimated single-pass non-cavity pump power $P_P^{(1)}$ by more than an order of magnitude. For a 10 ns pulse, the corresponding pump energy is 18 mJ.

A further requirement relates to the linewidth of the pump radiation.[1] In addition to the mode-matching condition for minimising diffraction loss at the generated wavelength λ_s, the linewidth should be comparable with the width for spontaneous Raman scattering in liquid nitrogen, if the actual pump power is to agree with the values calculated above for matching the gain requirement. The measured spontaneous width reported by Clements and Stoicheff[3] is $\Delta\bar{\nu} = 0.067$ cm^{-1}, which means that the linewidth at the pump wavelength λ_p should be

$$\Delta\bar{\nu}_p \lesssim 0.1 \text{ cm}^{-1} \quad (\Delta\nu_p \lesssim 3 \text{ GHz}) \qquad \ldots (9)$$

This condition is easily met for low pressure gas lasers, but a parametric oscillator requires intra-cavity etalons to achieve the necessary reduction in linewidth.

As for alternatives to the parametric oscillator, there are no molecular gas lasers at the appropriate pump wavelength of 3.4 μm, and no powerful tunable sources other than the OPO in this region. One possibility is frequency tripling of CO_2 laser radiation in liquid CO[4], assisted by two-photon resonance with the fundamental of the Q-branch at $\bar{\nu}_v = 2138$ cm^{-1}. This is very close to coincidence with the second harmonic of the R(6) rotational line in CO_2 at $\bar{\nu}_6 = 1068.9$ cm^{-1} ($\lambda = 9.355$ μm), but it will give a Stokes' frequency $\bar{\nu}_s \simeq 880$ cm^{-1} after Raman conversion in LN_2 ($\bar{\nu}_R = 2326$ cm^{-1}). The R(34) line at $\bar{\nu} = 984.3$ cm^{-1} would be a better choice, except that the third harmonic conversion in CO would be non-resonant and presumably inefficient. Thus resonant frequency tripling of CO_2 radiation in liquid CO is not directly applicable to the problem, some other combination of frequencies being needed for generating 3.4 μm by mixing in a suitable non-linear medium.

The effect of a parametric instability in stimulated Raman scattering[5], or of oscillation with feedback by Rayleigh scattering[2], is to reduce the power requirement for the onset of gain saturation (and efficient conversion to the Stokes' radiation) by about a factor of two when compared with the simple estimate based on exponential gain in the beam of the pump laser. This result was observed for stimulated Raman scattering in liquid nitrogen with a ruby laser[2] and, because the critical intensity I_c for parametric instability scales inversely with the Raman gain[5], the ratio should be similar for $\lambda_s = 16$ μm, whereas feedback scattering would be ruled out by the λ_s^{-4} dependence of the Rayleigh scattering cross-section. In the Raman oscillator, the situation will depend on the intensity I_o at the beam-waist in the confocal cavity (whether $I_o >$ or $< I_c$[6]) so that at best the power of the pump laser at $\lambda_p = 3.4$ μm could be reduced to about half.

In summary, the estimated requirements for efficient 16 μm generation in liquid nitrogen are as follows: pump wavelength λ_p = 3.4 μm (tunable), pump power $P_p \simeq 2$ MW (Raman oscillator), linewidth $\Delta\bar{\nu}_p < 0.1$ cm^{-1}, pulse energy $E_p \simeq 20$ mJ for a pulse width $\tau_p \simeq 10$ ns, intensity profile - gaussian, matched to TEM_{00} mode of Raman oscillator cavity.

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6. $I_c \simeq$ 700 MW/cm^2 for λ_s = 16μm (cf. ref. (5)).