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# A multilens Raman cell as a tool to obtain high optical quality and efficient 1st Stokes backward conversion

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## 1. Introduction

Raman conversion is a simple and efficient tool for frequency down-conversion but, as well reported in many previous works, when the pump power is driven at a level much larger than the threshold to obtain an efficient power conversion (>50%), a large cascade both in the antiStokes and Stokes branches is produced, reducing the effective 1st Stokes conversion [1,2]. This fact is particularly evident when the pump frequency is much larger than the Raman frequency. To avoid this effect it has been suggested and experimentally demonstrated that the collimation of the pump beam inhibites the four wave mixing process that is the dominant origin of the multiStokes emissions [1]. However unfocused pumping needs very large power while strong focusing is necessary to reach the Raman scattering threshold with the power of common pulsed lasers. This problem can be solved in guides of small section [3], that require low powers, although in that case there are upper limits for the injectable power.

It has been shown in some works that a strong focusing of a narrow band pump laser induces a dominant backward 1st Stokes (BS) scattering [4]. In fact for a bandwidth pump  $\Delta_p$  smaller than the Raman linewidth  $\Delta_r$ , the forward and backward gains  $g_f$  and  $g_b$ are comparable being from [5]:

$$g_b \approx g_f \frac{\Delta_r}{\Delta_p + \Delta_r} \tag{1}$$

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### ABSTRACT

A multilens high pressure  $H_2$  cell has been used to demonstrate that efficient, high optical quality and low threshold down-conversion to 1st Stokes can be obtained also with a poor quality broadband pump, with just the condition that the pump pulsewidth is larger than the cell transit time. A backward 1st Stokes with 0.7 overall quantum efficiency conversion has been obtained from a broadband Nd:YAG duplicated laser.

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and the backward dominance can be induced by the destructive interference of forward Stokes–antiStokes process (FS and FAS respectively). This fact has been both theoretically [6] and experimentally observed [4,7–9] when there is an exact phase matching for the four wave mixing (FWM) process generating the FAS and this phase matching is ensured when strong focusing is used [4,8]. On the contrary this interference is not present for the BS emission because no phase matching is possible between the BS wave and the forward ones [4]. This fact also ensures that FWM generation of a backward Stokes is impossible and as a backward Stokes cascade needs very large powers, a very efficient downconversion can be obtained in BS scattering.

However these results are limited to narrow band pump lasers, while normally high power lasers are broadband especially many tunable pulsed dye lasers. In this particular case many works have demonstrated that the mixing induces a Stokes cascade that reduces the efficient down-conversion on a single Raman frequency [2,10,11]. However with a suitable Raman design it is possible to obtain a high quantum conversion in the backward scattering also with multimodal broadband pumping. We demonstrate this possibility in a simple single cell set-up, otherwise a more complex dual-cell system can be used to obtain a good efficiency [12].

The problem of the multimodal structure of the beams has been analysed in [13]. By using that approach it is easy to obtain the total gain for forward and backward 1st Stokes scattering:

$$\mathbf{g}_f = \mathbf{g}\Gamma_f = \mathbf{g}\frac{2\Delta_p \Delta_f}{\Delta_p^2 + \Delta_f^2},\tag{2}$$

$$g_{b} = g\Gamma_{b} = g\frac{2\Delta_{b}\Delta_{p}}{\Delta_{b}^{2} + \Delta_{p}^{2}} \exp\left[-\frac{1}{c^{2}}\frac{\Delta_{b}^{2}\Delta_{p}^{2}}{\Delta_{b}^{2} + \Delta_{p}^{2}}z^{2}\right],$$
(3)



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where  $\Delta_p$ ,  $\Delta_f$ ,  $\Delta_b$  are the bandwidths of the pump, FS and BS radiations respectively and g is the Stokes gain for monochromatic radiation. While Eq. (2) shows the well known result that the maximum forward Stokes gain  $g_f = g$  is obtained for  $\Delta_f = \Delta_p$ , Eq. (3) shows again the relationship (1) if  $\Delta_b \ll \Delta_p$ , but we also see that for  $\Delta_b = \Delta_p$  a  $g_b \cong g$  gain can be obtained for a coherence distance  $Z_c \cong \sqrt{\frac{2c}{\Delta_p}}$ . So, if we can produce a backward BS mode with a confocal parameter  $b \cong Z_c$  at the first focal spot, we have a dominant  $g_b$  gain at the entrance and a high quantum efficiency BS conversion can be observed also with a broadband pump. This can be achieved using a system of short focal lenses.

Our basic idea has been to use a multilens Raman cell with lenses of very small focal lengths at a variable distance to maximize the backscattering process. In this case not only Raman scattering is possible for small pump power but also a very efficient down-conversion on the backward 1st Stokes emission is obtained with an improved peak power and higher optical quality of the beam. The generation of a beam with these characteristics, i.e. high power and good optical quality, can be useful for several applications, like for LIDAR experiments.

### 2. Experiment and results

The experimental set-up with the multilens cell is sketched in Fig. 1. In our experiment three lenses are used but in principle more lenses can be used, because the lens problems affecting the forward scattering (reflection losses, defocusing due to the confocal parameter compression [10], phase matching interference [6]) do not influence the backward emission that is always produced at the focus of the first lens. In our case to avoid even the loss of the input window the first lens  $(f_1)$  is positioned directly as the input window. The alignment of a multilens cell is not a problem if each lens is mechanically referred to the cell structure. This result is obtained by gluing the lens to iron rings exactly matching the cell bore. Being the cell in steel, the rings can be easily translated and fixed along the cell by means of external magnets to obtain the maximum backward scattering. The typical working H<sub>2</sub> pressure is 30 bar and the cell is 70 cm long while the focal length is 10 cm for the first two lenses and  $f^3 = 5$  cm for the 3rd lens. By applying the geometrical optics to Fig. 1 three foci are produced at F1 = f1, F2 = f1 + 4f2 and F3 = f1 + 4f2 + 4f3, respectively, corresponding to a matched cell length f1 + 4f2 + 6f3 = 80 cm and the radiation is well transmitted without dispersion from the cell wall. Our cell is not well matched (L < f1 + 4f2 + 6f3) but adapting the f2 and f3 lens positions we have again three foci although now the beams are weakly collimated by the waveguide diameter producing a diffuse radiation. This fact induces negligible losses on the FS scattering but it has a great positive effect on the BS one. So the lens positions can be optimised to maximize alternatively the FS or the BS emission. In our configuration the lens positions are



**Fig. 1.** Sketch of apparatus: LS = laser source, BS, P, FS, FAS = laser beams, phd = photodiode, d = dichroic mirror, f = filter, S = screen, l = lens, m = magnet, W = window, L = cell length, x, y = photodiode distances from the cell sides (with x = y).



**Fig. 2.** Emitted pulses at the onset of the Raman process with 1 mJ dye laser pulse. P = pump, FS = forward 1st Stokes (dotted line), BS = backward 1st Stokes;  $\lambda_P = 600$  nm;  $\lambda_{1S} = 799$  nm.

not highly critical, meaning that a lens displacement of a few mm does not change dramatically the results.

Spatial and temporal analysis of the emitted (transmitted) beams is performed at the same distance (25 cm) from the cell windows in the two directions by fast photodiodes and a CCD camera.

The initial measurements have been performed with a dye laser having 1 mJ pulse energy and 15 GHz estimated bandwidth, about 10 times larger than the Raman linewidth. This laser has been used in a previous work [10], where two lenses were used and no backward Stokes emission was observed even at the maximum pulse energy (4 mJ). Although the lens positions are optimised to produce FS, the use of a third lens does not increase the forward conversion. However the Raman threshold is lowered and an evident backward emission is now observed. For example in Fig. 2 three pulses are detected at the onset of process when the pump is not depleted. We see that the FS is dominant as expected and, in agreement with [14], it induces at a delay nearly equal to  $\frac{L}{2}$  a BS emission. That means we are in the "pseudo cavity regime" (PCR) that is largely influencing the dynamics of process [15]. The PCR influences the timing of the Raman process when we have  $e^{G} * R_{w} \gg 1$ , where G is the total gain of a single transit and  $R_w$  is the very weak window reflectivity inside the mode of the Raman beam. Therefore the use of a third lens produces a backward gain large enough to amplify the fraction back reflected by the exit window although  $g_b$ should be 10 times smaller than  $g_f(Eq. (1))$ . The PCR can stimulate a weak BS emission also with a single lens if the gain is large enough [11], but in this case a forward prevalence is always observed. This prevalence is reduced in [11] by increasing the total cell gain for a "cinematic" effect, when the incoming pump meets an intense BS wave near the entrance lens when the BS field is maximum and the FS minimum. Fig. 2 also shows that after the onset of the Raman scattering, the large reflections of the Stokes beams due to the optical components inside the cell ensure the pseudo cavity stationary conditions for the scattering also in pump tail. The pulse modulations in Fig. 2 are due to the dye mode structure that is more evident in the Stokes pulses due to the nonlinear nature of the Raman scattering. In this case it is noteworthy that the Fourier transforms of the Stokes pulses don't show the dye bandwidth but just the 600 MHz detector bandwidth nearly corresponding to a 0.5 ns response time.

Fig. 2 also shows that an initial FS emission is necessary and to observe a BS dominance a larger gain (more lenses or more power)

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