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**EFFICIENT RAMAN CONVERSION FROM LIQUID NITROGEN  
CONTAINED IN AN ORDINARY DEWAR**

by

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ABSTRACT

Efficient backward Raman conversion is obtained by focusing laser radiation near the surface of liquid nitrogen contained in an ordinary laboratory Dewar. Shifting of both the fundamental frequency (1.064  $\mu\text{m}$ ) and the second harmonic (532 nm) of a Q-switched Nd:YAG is demonstrated.

RÉSUMÉ

Une bonne efficacité de conversion Raman a été observée en focalisant un faisceau laser à la surface d'un Dewar ordinaire rempli d'azote liquide. Cette technique de conversion est démontrée pour la radiation fondamentale (1.064  $\mu\text{m}$ ) d'un laser Nd:YAG déclenché ainsi que pour sa deuxième harmonique (532 nm).



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FIGURES 1 AND 2





## 1.0 INTRODUCTION

Raman shifting is a well known technique for extending the wavelength coverage of the few efficient and high-power solid-state laser sources. In practice only a handful of Raman media are being widely used. Pressurised gases like methane, H<sub>2</sub> and D<sub>2</sub> are the most commonly used because they offer good efficiency (40-70%) and because simple high-pressure Raman cells are easily implemented. Liquid nitrogen (LN<sub>2</sub>) has been proven [1] to be one of the best media as it offers a large Raman cross section, a narrow linewidth and a much higher molecular density than the mentioned gases at their normal operating pressure. It has also a very broad transmission range in the UV, visible and infrared. Photon conversion efficiencies higher than 95 percent have been demonstrated with LN<sub>2</sub> [2-3]. Despite these impressive results, LN<sub>2</sub> has not been extensively used. This is mostly due to the need of using sophisticated cryogenic cells in which each window is composed of cold and warm windows separated by a vacuum gap. This method is necessary to prevent any condensation of water vapor on the window in contact with the 77°K LN<sub>2</sub>. In this thechnical note, we propose a simple setup that makes use of the backward-Raman process instead of the more usual forward process. With this approach an ordinary laboratory dewar can be used as the LN<sub>2</sub> container. This work was carried out at DREV between November 1995 and June 1996 under Thrust 2d "Land Force Tactical Surveillance and Counter-Surveillance" and more specifically under project 2da28 "New Solid-State Laser Sources".

## 2.0 BACKGROUND

The backward to forward Raman gain ratio is affected by many parameters related to the Raman medium, the pump laser linewidth and the interaction geometry. In the case of narrow-band pumping and in liquids, having an homogeneous broadening, it is usually

very close to one. In the case of a pump laser having a linewidth broader than the Raman resonance bandwidth ( $0.067 \text{ cm}^{-1}$  for  $\text{LN}_2$  [1]), the Raman interaction is characterised by significantly different coherence lengths for the forward and the backward processes [3-4]. It turns out that a low threshold, comparable to the one observed for a narrow-pump linewidth, is achieved only if the interaction length in the medium is smaller than the respective coherence length. For a visible or near infrared pump laser having a bandwidth of the order of  $0.5 \text{ cm}^{-1}$  this restricts the interaction length for the backward scattering to values in the 0.5-1 cm range whereas it is between 200-500 cm for the forward process. To obtain similar thresholds for both processes, a convenient method is to restrict the interaction length by focusing the pump beam with a short focal length lens.

### 3.0 EXPERIMENTAL SET-UP

The experimental set-up used to demonstrate backward conversion in  $\text{LN}_2$  is schematically shown in Fig. 1. The pump laser is a commercial multi-transverse and multi-longitudinal modes  $1.06\text{-}\mu\text{m}$  Nd:YAG (JK Laser Mini-Q). The laser linewidth as measured with a scanning Fabry-Perot interferometer is of the order of  $0.45 \text{ cm}^{-1}$ . The laser is mounted horizontally and a dichroic beamsplitter at  $45^\circ$  redirects the beam vertically toward the  $\text{LN}_2$  Dewar through a focusing lens. The backward Raman beam generated in the focal volume is collected and collimated by the same lens and redirected horizontally by a  $45^\circ$ -mirror after passage through the dichroic beamsplitter. Alternatively a 2X demagnifying telescope, a  $3\times 3\times 5 \text{ mm}^3$  KTP crystal cut for type II doubling and a  $1.06 \mu\text{m}$  blocking filter can be put in front of the laser in order to work with the second harmonic at  $0.532 \mu\text{m}$ . The typical conversion efficiency to  $0.532 \mu\text{m}$  is 40 percent. The energy of both the pump and the Raman beam was measured with a pyroelectric joulemeter (Gentec ED-200). Unfiltered  $\text{LN}_2$  taken from a 20-liters Dewar was used. This is in contrast with most previous experiments [1-3] where great care was

taken to ensure a high-purity since much longer interaction lengths were used. It was nevertheless found that best results were obtained when the Dewar was well cleaned before filling ( because this results in a quiet medium with little bubbling). The pump laser energy was varied by inserting neutral density filters and glass plates before the 45°-dichroic beamsplitter.

#### 4.0 RESULTS

Backward Raman generation was easily observed with this setup at both pump wavelengths. The best results were obtained when the lens was positioned so that the focal spot was just slightly (5-10 mm) below the surface of the LN<sub>2</sub>. This is consistent with the need to keep the interaction length to a small value. The results presented below were obtained with a lens having a focal length of 7.5 cm. However it was found to be not a critical parameter.

Fig. 2 gives the input-output results for both pump wavelengths, taking into account the losses suffered by the pump and the Raman beam in the dichroic beamsplitter. It can be observed that the conversion efficiency has a linear behaviour and that the threshold is fairly low. Slope efficiencies of 0.33 and 0.39 were obtained when pumping at 1.064  $\mu\text{m}$  and 0.532  $\mu\text{m}$  respectively. This nearly corresponds to equal quantum efficiencies of 0.44 and 0.45 respectively. These data were taken at a repetition rate of 0.3 pps to ensure that all the bubbles generated in the LN<sub>2</sub> by the thermalisation of the vibrational energy deposited in the medium have been removed before the next shot. In practice, satisfactory operation at the highest available energy were obtained at up to 3 pps. Ample conversion was also obtained at 10 pps but the pulse to pulse stability was then very poor. In all cases, only a single line at the first Stokes (either 1.414  $\mu\text{m}$  or 0.607  $\mu\text{m}$ ) was observed in the backward beam. The divergence of the backward beam was just slightly larger than that of the pump beam and the pointing stability was

extremely good. This might appear somewhat surprising as even at 0.3 pps the LN<sub>2</sub> does not have time to settle completely after the brief boiling episode that follows each pump pulse. In fact, the backward beam has the interesting property to be very nearly the conjugate of the pump pulse and as such does not depend in first order to the quality of the air-LN<sub>2</sub> interface. This is no more true if a bubble is still present in the focal volume of the pump beam as it sometimes happens at high-pump energy and at repetition rates higher than approximately 1 pps. In this condition, conversion still occurs but the backward beam divergence is considerably affected.

In this experiment, forward Raman conversion must also occur since at best the backward to forward threshold ratio is one. This was verified by using a 600- $\mu\text{m}$  optical fiber to couple the light coming from the interior of the dewar to a spectrograph fitted with a CCD linear array. Signals at the 1<sup>st</sup> to the 4<sup>th</sup> antistokes lines (0.853, 0.712, 0.611 and 0.535  $\mu\text{m}$ ) were observed when pumping at 1.06  $\mu\text{m}$ . We could not verify if higher order Stokes lines were generated because no suitable detector was available. Similarly light at the first to the third Stokes (0.607, 0.707 and 0.846  $\mu\text{m}$ ) as well as at the first and second antistokes (0.473 and 0.426  $\mu\text{m}$ ) was observed when pumping at 0.532  $\mu\text{m}$ . The first antistokes line comes mainly from a four-wave mixing process driven by the pump and the first Stokes beams [5]. Order  $n$  antistokes are mainly driven by pump, first Stokes and the  $(n-1)$  antistokes component. In the forward process, generation of the high-order Stokes as well as antistokes serves as a limiting factor. This in turn may help to prevent the forward process to dominate the interaction and allows the good efficiency observed in the backward conversion.

The ordinary Dewar that was used has a good quality silver coating to prevent thermal radiation from the surrounding to enter the Dewar. This coating is highly reflective in the visible and near infrared and this might couple some energy from the forward Raman conversion to the backward process. This might help an early onset of the backward conversion and a concomitant increase in conversion. This cannot be ruled out

but it was observed that a decent although more erratic conversion was still obtained with LN<sub>2</sub> contained in a black paper lined styrofoam coffee cup, despite a constant boiling of the LN<sub>2</sub> . Thus, it can be concluded that the coupling if it exists is not essential to the backward Raman generation.

## 5.0 CONCLUSION

In this technical note, we show that efficient backward Raman scattering in LN<sub>2</sub> can be obtained even with a multi-longitudinal modes pump source. This allows the use of a common laboratory Dewar to contain the LN<sub>2</sub> instead of a more sophisticated cryogenic optical cell. Moreover, no filtration of the LN<sub>2</sub> was necessary. The backward generated beam is free of other Stokes and antistokes components and has good pointing stability and spatial quality.

## 6.0 ACKNOWLEDGEMENTS

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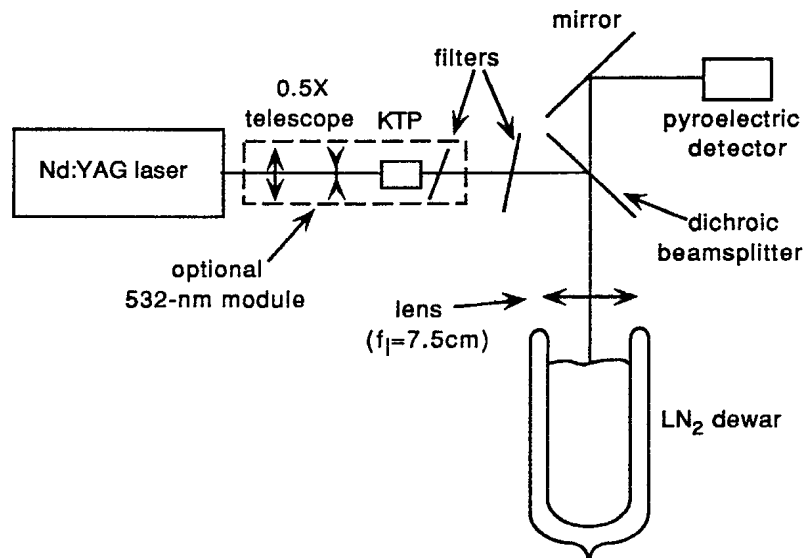


FIGURE 1 - Experimental setup. The elements shown in the dotted box are used only when experimenting with the second harmonic.

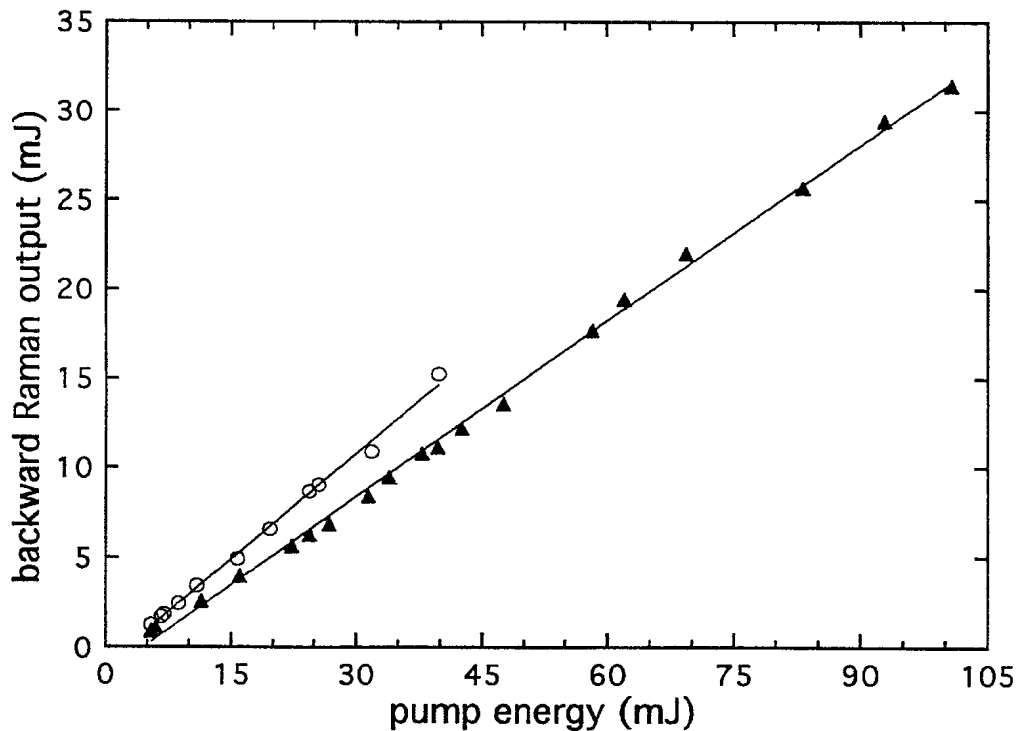


FIGURE 2 - Measured conversion efficiency at 532 nm (open circles) and 1.064 μm (triangles). The lines are best fits to the shown data





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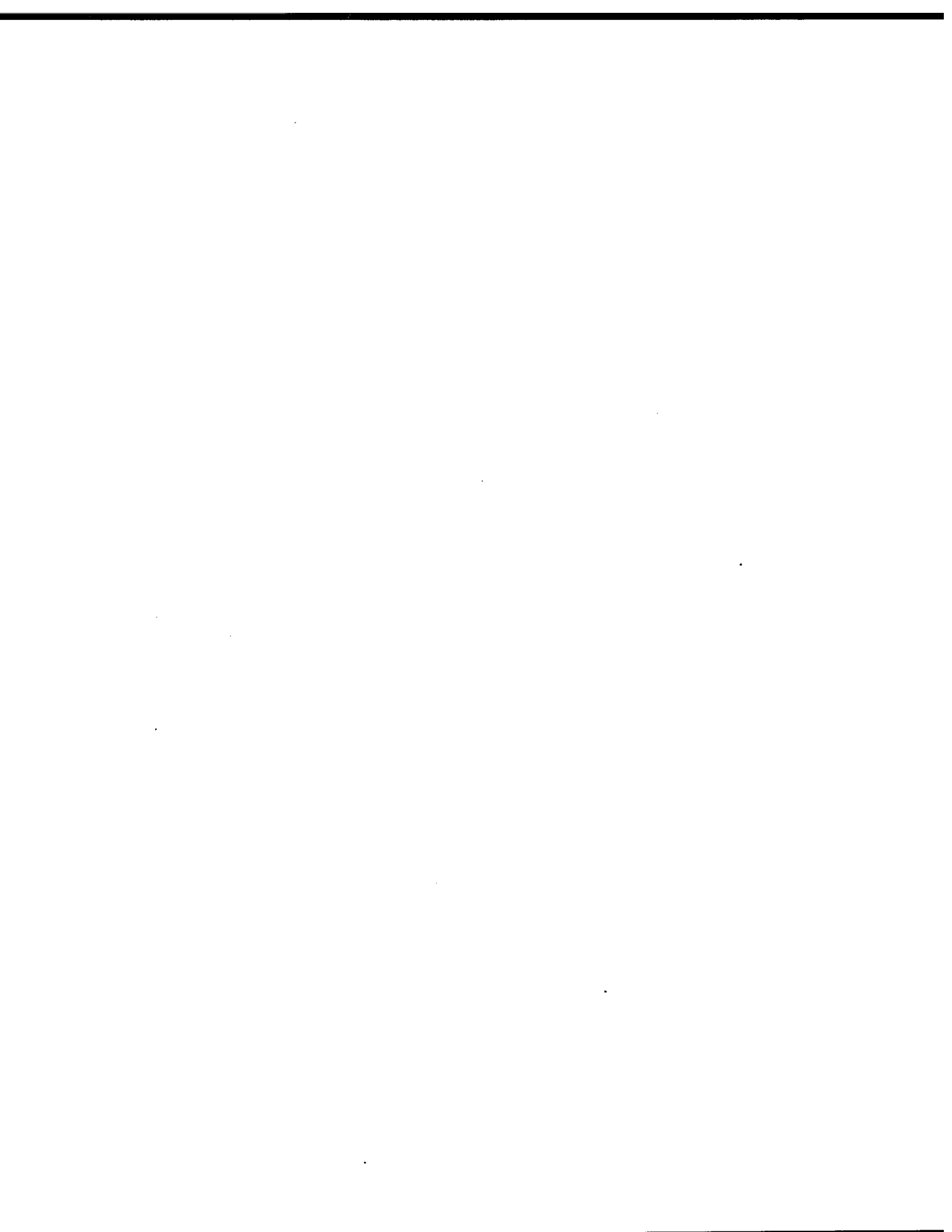
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