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**PARAMETRIC STUDY OF A RAMAN p-H₂ LASER
WITH OVER 30 MW EMISSION AT 16 μ m**

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Riassunto — In questo lavoro sono stati studiati i principali parametri che definiscono la soglia, la forma temporale e la stabilità di un laser Raman a para-idrogeno (16 μm). I nostri risultati mostrano un'alta efficienza di conversione di potenza (130%) per ciascuna riga del laser a CO₂ permettendo una buona tunabilità nel range dei 16 μm. Questo è fondamentale per la separazione isotopica di UF₆.

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Summary — In this work the main parameters which define the threshold, the temporal shape and stability of p-H₂ Raman Laser have been investigated. Our results show a very high power conversion efficiency (130%) and for each pump CO₂ line we can dispose of a correspondent line (pulse $\geq 1J/50 ns$) in the 16 μm range. This is fundamental as far as UF₆ isotopic separation is concerned.



COMITATO NAZIONALE PER LA RICERCA E PER LO SVILUPPO
DELL'ENERGIA NUCLEARE E DELLE ENERGIE ALTERNATIVE

M. BERNARDINI, A. FERRARIO, A. GIORGI, S. MARCHETTI, A. PALUCCI

**PARAMETRIC STUDY OF A RAMAN $p\text{-H}_2$
LASER WITH OVER 30 MW EMISSION AT $16 \mu\text{m}$**

RT/TIB/84/30

Testo pervenuto nel dicembre 1984

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1. - INTRODUCTION

Recently the multiple-photon processes have suggested the development of new laser sources covering the whole IR region from FAR to NEAR IR. In particular there has been a great deal of interest of 16 μm laser due to the proposed laser isotope separation scheme for UF_6 .

Since primary efficient sources are not available (discharge, solid state lasers, etc.....), different frequency conversion technics were studied to obtain a high-power and tunable source in the 16 μm range.

By using only an efficient laser, like the CO_2 one, we can obtain a suitable source with two different technics as optical pumping and S.R.S. (Stimulated Raman Scattering).

As proposed in ¹ we have chosen the S.R.S. because it shows a high and complete frequency conversion of pumping laser characteristics as coherence, bandwidth and tunability. However the very low Raman cross section depends on power and bandwidth of the laser pump so that particular care has to be devoted to construction of a suitable laser chain.

Moreover the S.R.S. gain in focused beam geometries with gaseous medium requires a very high pump power so that a very carefully designed Multiple Pass Cell (MPC) has been built to enhance the Raman gain and achieve consistent powerful radiation.

The $p\text{-H}_2$ gaseous active medium has a rotational transition $\text{So}(0) = 354.4 \text{ cm}^{-1}$, (Fig. 1) which provides a down-conversion from 10 μm range to 16 μm emission (Stokes) and up-conversion to 7 μm (Antistokes).

A first experiment, described in reference ², pointed out that a negative effect as optical breakdown in the gas or at the windows presented quite a low threshold to compromise an appreciable Raman effect unless particular care were taken in the experiment execution.

2. - EXPERIMENTAL

The experimental apparatus used to produce Stokes pulses appears in Fig. 2 and has been realized in collaboration with CISE (Contratto CNEN-CISE of November 1981). A hybrid oscillator with a low pressure cell can produce an optional single mode pulse which is amplified by means of two TEA Lumonics mod. 103. The laser chain can deliver 4J/110 ns, Fig. 2, ³ and a Fresnel rhomb provides to change the polarization from linear to circular to maximize the Raman gain ⁴.

In this case the effect is very important, because at the energy

necessary to observe Raman Scattering with linear polarization we generate breakdown in gas. The CO₂ laser pulses are conveniently mode-matched and injected into the multiple pass H₂ Raman cell where conversion to Stokes and Antistokes pulses occurs. In our laboratory we have designed a suitable MPC (25 passes) in which the principal elements are two home-made spherical copper mirrors of ~ 20 cm diameter, separated by $d = 430$ cm with confocal parameter $b = 106$ cm, where the p-H₂ is maintained at liquid N₂ temperature, at 500 Torr pressure.

The transmission of the CO₂ pump through the MPC indicated that the cell mirrors have a reflectivity of 98.3% at 10 μm .

The beam enters through two BaF₂ windows ($T = 0.92$ to 10 μm) and a 3 cm hole in the mirrors allows the incoming beam to be directed into the MCP. The output windows are a ZnSe ($T < 0.95$ to 10 μm , $T = 0.7$ to 16 μm and 7 μm) and a KCl ($T \cong 0.9$).

The exit beam contains the 16 μm Stokes radiation, the residual 10 μm pump and a very little component of 7 μm Antistokes as well. A LiF window, which reflects the Stokes, provides the separation for the measurements or the use of single radiation. Two Rofin photon-drags are used for the temporal analysis while two Scientech Joulemeters are used for measuring the energy.

To simultaneously analyze all three frequencies the LiF plate was replaced with a grating which reflected at different angles the three radiations, whose detection was performed by means of the previous named photon-drags.

RESULTS

In typical running conditions, considered satisfactory, we obtained 16 μm energy pulses of 1.2J/about 40 ns, taking into account the windows absorption, starting with 10R20 pump pulse of 3.3J/110 ns. This result gives an energetic conversion efficiency of 40% and a power conversion efficiency $\eta = 130\%$.

To compare this experimental value with the theoretically expected one we report the conversion equation after n -passes in a MCP as calculated by R.T.V. Kung⁵ according to a generally accepted theory where also the pump depletion is considered:

$$(1) \ln \frac{P_{s,n}}{P_{so}} - \ln \frac{P_{p,n}}{P_{po}} = \frac{1-R^n}{1-R} \frac{4(\alpha P_{po} + \alpha P_{so})}{\lambda_s + \lambda_p} \text{tg}^{-1} (d/b)$$

where P's are the total powers of Stokes and pump beams after n passes (0 means initial beams), R mirror reflectivity, α 's plane wave gain coefficients (cm/MW), λ 's wavelengths, d mirror distance and b confocal parameter of the cell. P_{so} is the spontaneous Stokes noise power, reasonably estimated $P_{so} \cong 1.3 \cdot 10^{-12}$ W. At the saturation limit we have, according to Ref. 5,

$$(2) \eta_{\text{limit}} = \frac{w_s}{w_p} R^{n-1}$$

which, in our case, assumes the value $\eta = 42\%$. Concerning the power we have a conversion greater than the theoretical value because of the MPC crossings^{5,6}.

From Eq. 1 and by using the results of Fig. 3 we find at the pump pulse of 2.3J/110 ns an α value of $\alpha_p = (6 \pm 1)10^{-5}$ cm MW⁻¹ when no crossing effect is present. However another approach can be used when the conversion is very low. In this case the Eq. 1 can be written

$$(3) \ln P_s \sim A + \frac{1-R^n}{1-R} \frac{4\alpha P_{po} \text{tg}^{-1} d/b}{\lambda_p + \lambda_s} = A + BP_{po}$$

with a linear semilog dependence in P_{po} 's values. Obviously in this case the crossing effects are uncontrolled. From Fig. 4 we estimate $\alpha = 6 \pm 1 \cdot 10^{-5}$ cm MW⁻¹ as previously found.

By varying the cw gain we can lengthen the pumps pulse duration (T_p) till 170 nsec. Also in this case we observed Raman emission and although the overall efficiency was reduced the estimated α value reaches $1.5 \cdot 10^{-4}$ cm MW⁻¹ as evidenced in Fig. 5 where the onset of Raman emission in two extremal cases are reported: 5a) pump energy 1.9J/120 nsec and 5b) 2.8J/170 nsec with a long tail.

This very surprising effect is due to the collision dephasing time (T_c) whose value is about 10 nsec. From Ref. 7 we have that Eq. 1 is valid for $T_p \gg T_c$ while when $T_p \sim T_c$ the Raman gain is fluence dependent. Probably in our case we are in an intermediate state. From Fig. 4

we can observe that the Raman gain with a multimode pump is just a little larger but it has to be remarked that also the breakdown threshold is lower with severe drawbacks.

The conclusion is that our multipass system is a very good device to obtain powerful Stokes radiation, which is our goal, but allows only a rough estimation of the small signal plane wave gain. Nevertheless the found values are completely in agreement with those deduced in similar experiments⁸. We have previously pointed out that some important effects are due to the beam crossings in the MPC.

The Figure 6 shows a typical multipass output (Raman effect) pulse where the residual pump is strongly depleted in favor of the onset of Stokes radiation. Observing the Stokes pulse, we can see that all energy is concentrated in a peak of 40 ~ 50 ns FWHM while the original pump peak is depleted for a time equivalent to the transit time of MPC. This pulse compression permits to generate a power conversion greater than one. It is the time interval between two adjacent crossings which finally determines the Stokes pulse width. This effect has been already observed at Exxon Laboratories whose authors developed also a theoretical interpretation⁶. In case of our cell we made the proper calculations and the presence and the distance between the dips in the residual pump tail (Fig. 6) confirm quite well the theory. If the experimental condition of gain is very high it is possible to observe the onset of 16 μm emission also at different crossing times non-synchronous with the main one Fig. 7. In some cases with a very large pump tail another weak peak at $(n + 1)d/c$ delay is observed also due to the reamplified window reflection radiation (Fig. 8).

Conversion efficiency measurements have been performed at different operating pressure of $p\text{-H}_2$. Among 350 and 800 Torr we found no appreciable effect as we would expect if the collisional broadening were the main responsible of Raman linewidth. In fact we have from Ref. 9 that gain $\rho \propto n/\Delta\nu_R$, for quite large pressures, goes to $N/\gamma N = 1/\gamma$.

As a matter of fact the homogeneous line broadening is $\Delta\nu_{\text{coll}} \sim \sim 80 \text{ MHz}$ ⁹ already at 350 Torr and 77° K sensibly larger than the Doppler broadening $\Delta\nu_D \sim 22 \text{ MHz}$.

Concerning the first Antistokes radiation we measured its energy amount transmitted through the LiF plate. We estimated 20 ~ 50 mJ when the experimental conditions were to be considered satisfactory for the

Stokes production. Nevertheless, the operating parameters of the multi-pass cell do not influence the AS radiation whose optimization can be performed by varying the pump pulse characteristics. The results are the object of another paper¹⁰.

CONCLUSIONS

In this work the main parameters which define the threshold, the temporal shape and stability of p-H₂ Raman laser have been investigated. The spurious effects which accompany the use of MPC have been evidentiated as well.

Our results show a dependence on the pump pulse duration which do not agree perfectly with a simplified theory. Besides we have never observed an appreciable Stokes emission with a linear polarization of the pump pulse not even when the energy achieved a value of $1.7 \div 1.8$ times greater than the one which with circular polarization was able to generate a reasonable Stokes emission.

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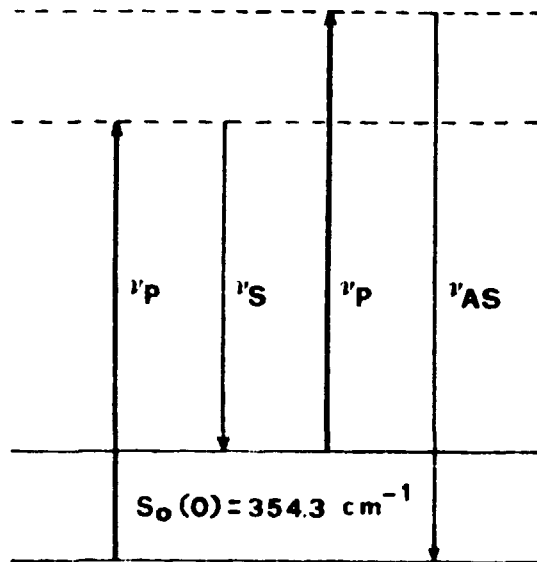


Fig. 1

Emission scheme of p-H₂ gaseous active medium: ν_p (10 μm), ν_s (16 μm), ν_{AS} (7 μm), $\Delta\nu = 354.4 \text{ cm}^{-1}$.

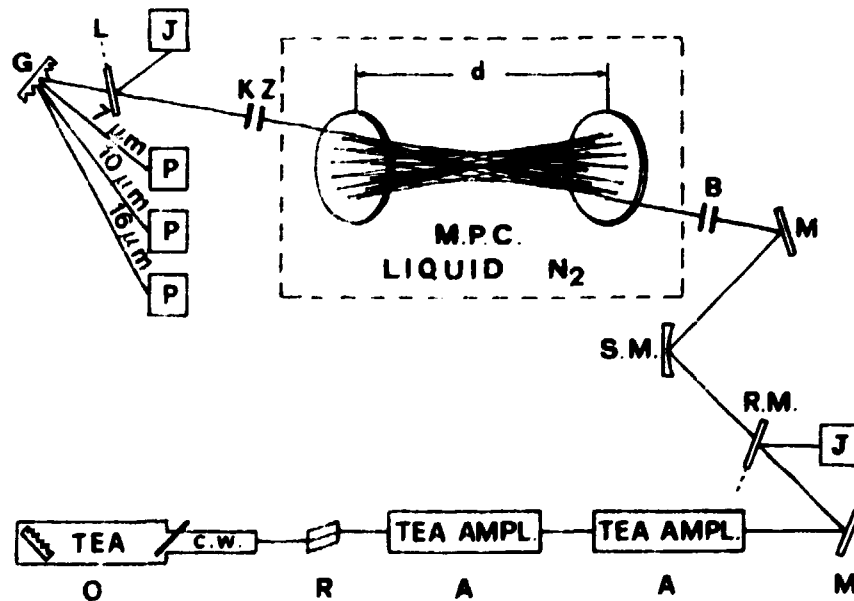


Fig. 2

Experimental apparatus: O - Hybrid oscillator; R - Fresnel rhomb; A - TEA Lumonics mod 103; J - Joulemeter, B - Two BaF₂ windows; MPC - Multipass cell (25 passes, $d = 430 \text{ cm}$, $b = 106 \text{ cm}$); Z - ZnSe window; K - KCl window; L - Removable LiF plate; G - Grating; P - Photon-drag.

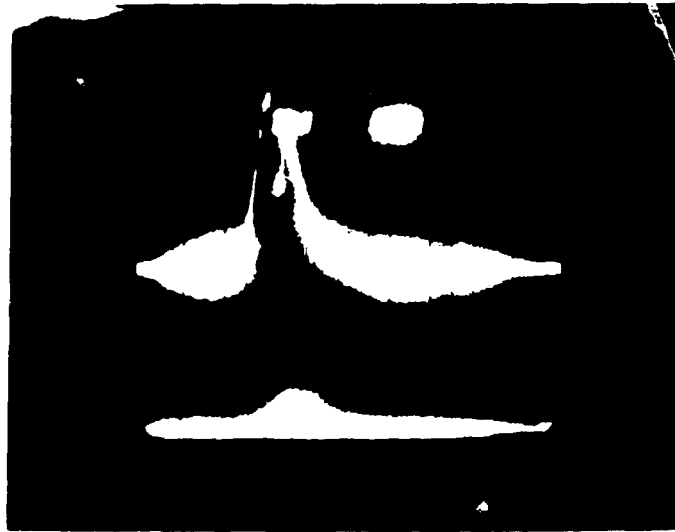


Fig. 3

Onset of Raman emission. a) Depleted pump pulse, b) Stokes emission

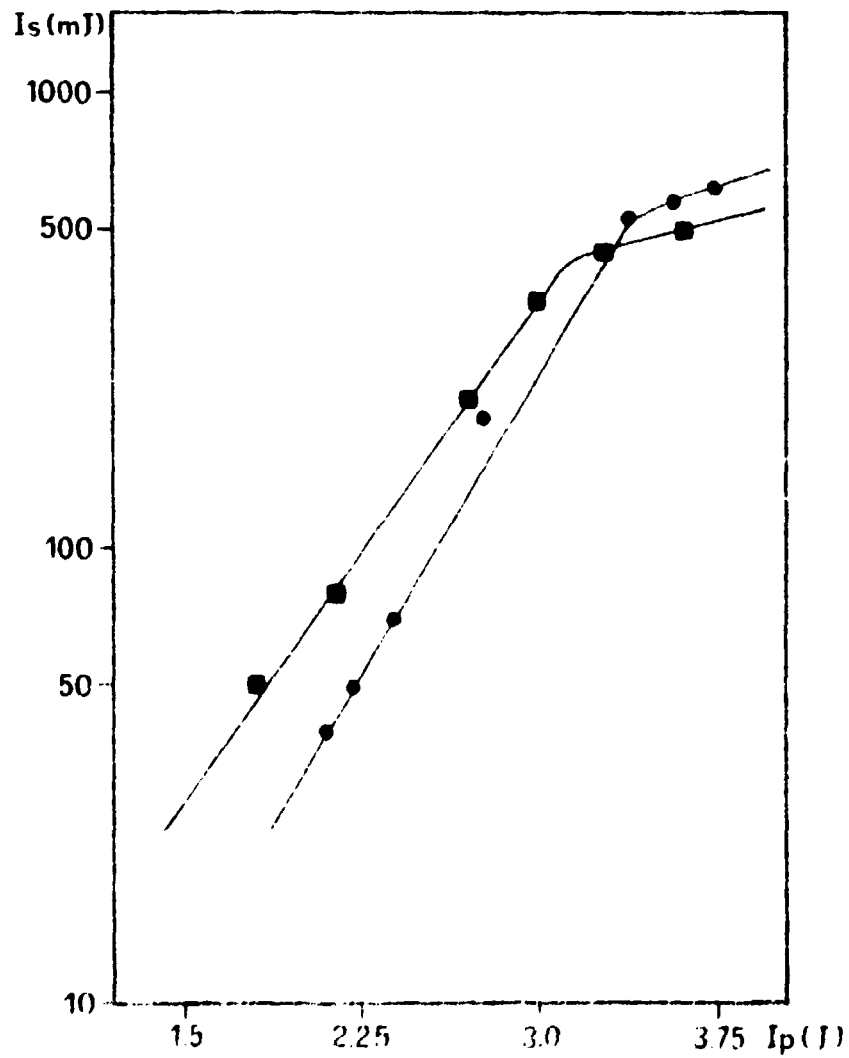


Fig 4

Stokes power emission in function of pump power pulse. a) Multimode, b) Single mode pump pulse.

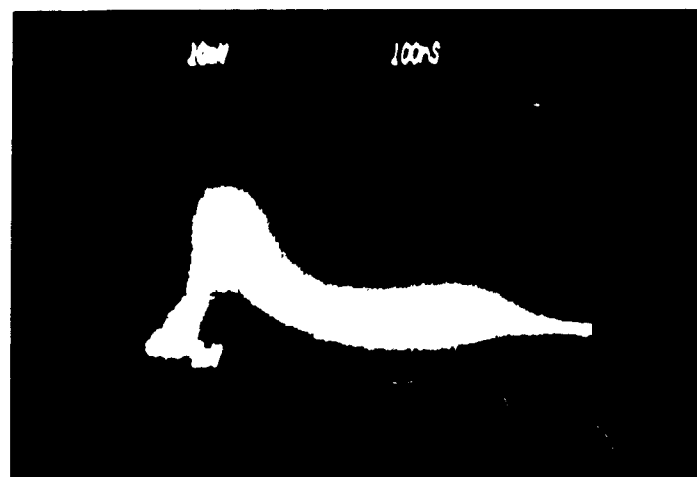
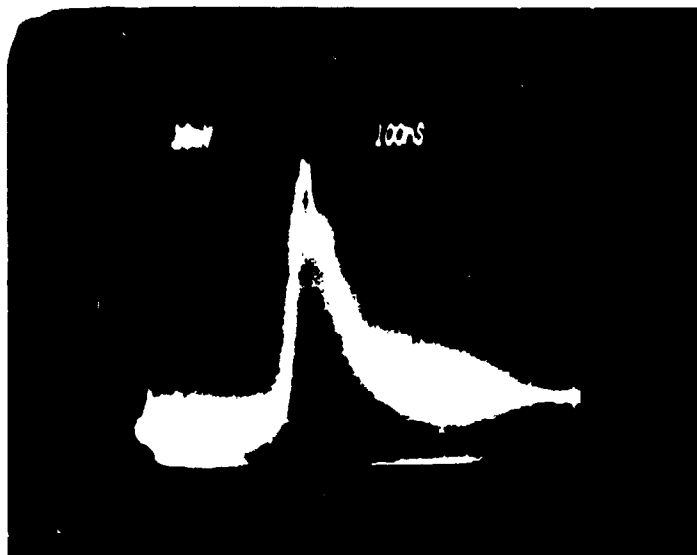


Fig. 5

Pump waveform: a) 1.9 J/120 ns; b) 2.8 J/170 ns.

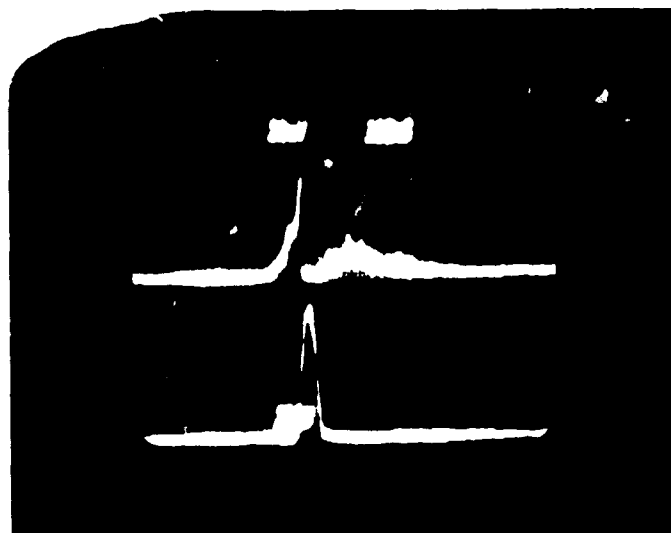


Fig. 6

Typical waveform of a Raman effect: a) Big depletion of pump pulse;
b) Stokes emission.

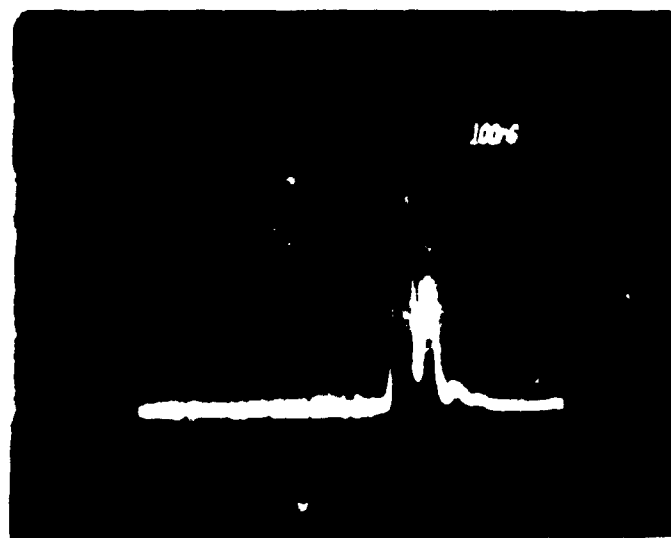
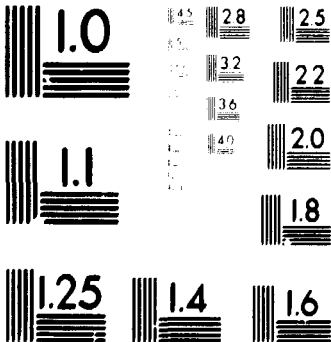


Fig. 7

Stokes emission wave form. The main peak is followed by other peaks
due to different crossings.



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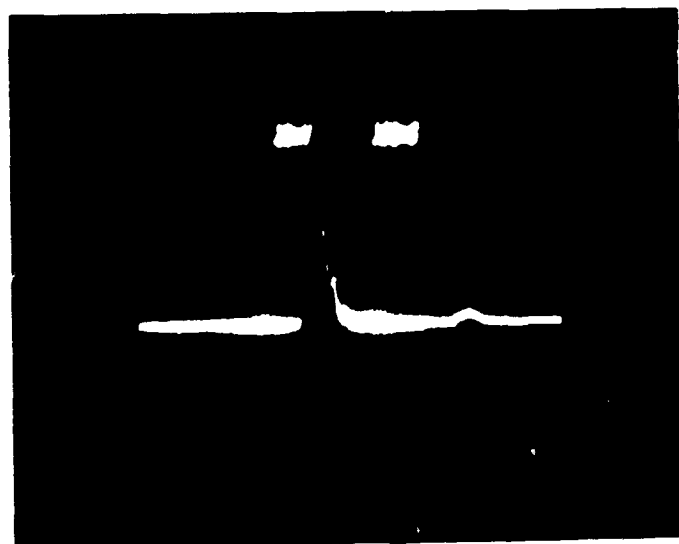


Fig. 8

Stokes emission waveform. The small peak 380 ns after the main one is due to the window reflection and to the long pump tail.

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