J. Phys. B: At. Mol. Opt. Phys. 22 (1989) L231-L233. Printed in the UK

LETTER TO THE EDITOR

Sideband generation by coherent anti-Raman scattering in quartz of a two-frequency high-power laser beam

A Dyson, A E Dangor and A K L Dymoke-Bradshaw
Blackett Laboratory, Imperial College, London SW7, UK

Received 6 December 1988

Abstract. Coherent anti-Raman scattering in quartz has been observed using two copropagating high-power laser beams at 1.064 μm and 1.053 μm from a neodymium glass laser. This is due to the beat frequency at 98.2 cm^{-1} being within the broad-band Raman gain of quartz which extends from about 20 to 550 cm^{-1}. The beams were each of intensity $3\times10^9\ \mathrm{W\,cm}^{-2}$ and had an interaction length of ≃5 cm. The measured gain is $9.5\times10^{-10}\ \mathrm{cm\,W}^{-1}$ in agreement with the established value.

When two intense laser beams are sent into a Raman active medium, strong molecular or lattice vibrations with a high degree of temporal and spatial coherence can be generated. These vibrations modulate the incoming light generating sidebands by coherent anti-Raman scattering (CARS). The Stokes and anti-Stokes sidebands have the frequency separation of the pump laser beams (Maier 1976). This has applications in the generation of coherent radiation and spectroscopy (Reintjes 1985, Shen 1984). When the Raman active medium is a plasma the vibrations are replaced by plasma resonances. The excitation of a relativistic plasma wave by the beating of two intense laser beams has aroused considerable interest because of its application to particle acceleration (Tajima and Dawson 1979).

In this letter we report on the observation of sideband generation by two intense laser beams over a short interaction length of quartz. These results were obtained during an experiment designed to generate a plasma wave by the beat wave process.

The experiment was performed using the Vulcan neodymium glass laser at the Rutherford Appleton laboratory. Two oscillators produced the wavelengths 1.064 μm (YAG) and 1.053 μm (YLF) which were amplified in separate rod chains to give beams of ≃50 J in 200 ps. The beam diameter was ≃10 cm giving a peak intensity of $3\times10^9\ \mathrm{W\,cm}^{-2}$ in each beam. The laser beams followed separate air paths and were orthogonally polarised before being recombined under vacuum using the arrangement shown in figure 1. This arrangement was used to avoid Raman scattering in atmospheric

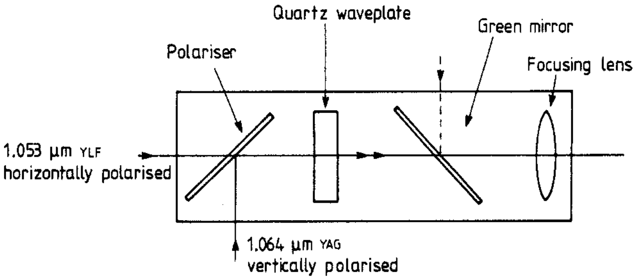

Figure 1. Arrangement for recombining the pump beams.

0953-4075/89/090231+03$02.50 © 1989 IOP Publishing Ltd

nitrogen (Dangor *et al* 1986). The beams interacted in the quartz waveplate, which matched the polarisations, and in a mirror and a focusing lens. The transmitted beams were attenuated by two 99% reflecting mirrors before emerging at full diameter into the atmosphere. The estimated interaction length in quartz is ≃5 cm. The sidebands formed on the pump beams were monitored using a grating spectrometer coupled to a Hadland Imacon 675 streak camera fitted with an S1 photocathode and intensifier. To detect the low intensity sidebands the pump light was preferentially attenuated by a factor of 100 using an Nd strip at the entrance slit of the streak camera.

The observed sidebands are shown in figure 2. The Stokes and anti-Stokes lines are clearly evident. The sidebands were not observed when the pumps were desynchronised or when the pumps were orthogonally polarised by removing the waveplate.

A significant difference between the Raman spectra of quartz and other materials is the breadth of the spectrum. This extends from about 20 to 550 cm^{-1} (Stolen *et al* 1972). The beat frequency of the pump laser beams is at 98.2 cm^{-1} which lies within this broad band. The sideband intensities are weak, being only about 1% of the pump intensities. The pump intensities can be assumed to be unaffected. Thus the intensities of the Stokes and the anti-Stokes sidebands $I_{-1,2}$ are (Maier 1976)

$$I_{-1,2}(z) = \tfrac{1}{2}(\omega_{-1,2}/\omega_0)^2(\gamma I_{0,1} z)^2 I_{1,0} \qquad (1)$$

where the mode numbers 0, 1 refer to the 1.064 μm (YAG) and the 1.053 μm (YLF) pumps, ω_m is the frequency and γ is the Raman gain.

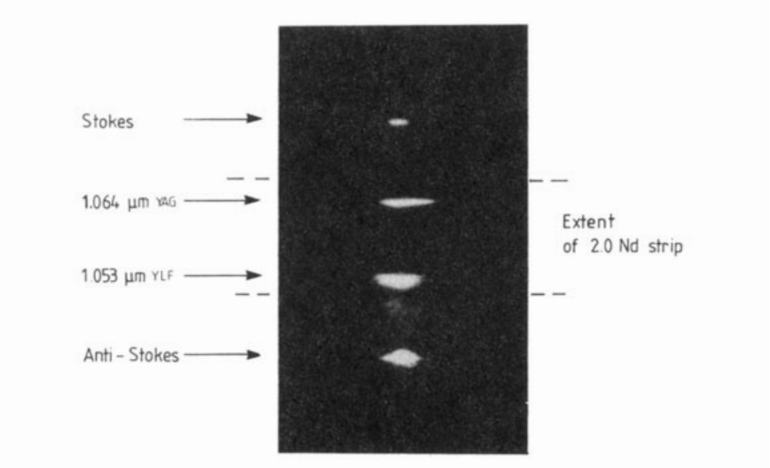

Figure 2. Streak record showing the pumps and sidebands.

Microdensitometer traces of the transmitted intensities obtained for a shot with equal pump beams are shown in figure 3. Using equation (1) and the assumed interaction length of 5 cm the observed intensity ratios give a gain of $\gamma = 9.5 \times 10^{-11}$ cm W^{-1}. According to Johnston *et al* (1968) the Raman gain scales as the cube of the wavelength. Thus our result at 1.075 μm implies a gain of 1.2×10^{-11} cm W^{-1} at 5330 Å. This is in good agreement with the value 1.3×10^{-11} cm W^{-1} measured at this wavelength (Stolen *et al* 1972). It should be noted that this result was deduced from spontaneous Raman scattering data obtained with a single low-power CW laser.

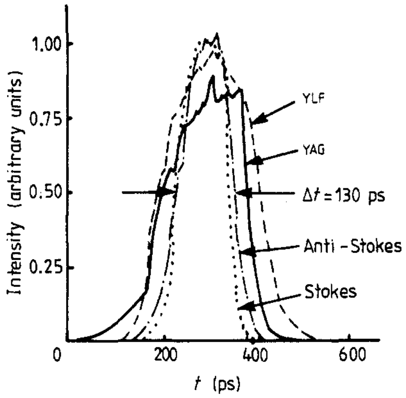

Figure 3. Microdensitometer traces of the pumps and sidebands. The pump intensities are reduced by a factor of 100.

The dependence of sideband intensity on the pumps in equation (1) implies that for Gaussian pumps of equal intensity the pulse length of the sidebands scales as $(\frac{1}{3})^{1/2}$ that of the pumps. For the pumps which are of pulse length ≃200 ps this gives ≃120 ps for the sidebands in agreement with the measured value of 130 ps.

In this experiment which was designed to measure the plasma wave excited by the beat wave process the expected sidebands were orders of magnitude smaller than those due to the CARS effect in quartz. Thus if measurements are to be made of the plasma wave the pumps must not have a common path in quartz.

The authors are grateful to Dr Mike Damzen for extremely useful discussions and for providing some of the references. Thanks are also to due to Colin Danson, Taraj Ashfar-Rad, Adrian Cole and Tony Damerell of the Rutherford Appleton Laboratory for their contribution to the experiment.

References

Dangor A E, Dymoke-Bradshaw A K L, Dyson A, Garvey T, Mitchell I, Cole A J, Danson C, Edwards C B and Evans R G 1986 *Rutherford Appleton Laboratory Annual Report* RAL-86-046 A1.1
Johnston W D, Kaminow I P and Bergman J G 1968 *Appl. Phys. Lett.* **13** 190
Maier M 1976 *Appl. Phys.* **11** 209
Reintjes J F 1985 *Laser Handbook* vol 5 (Amsterdam: North-Holland)
Shen Y R 1984 *The Principles of Nonlinear Optics* (New York: Wiley)
Stolen R H, Ippen E P and Tynes A R 1972 *Appl. Phys. Lett.* **20** 62
Tajima T and Dawson J M 1979 *Phys. Rev. Lett.* **43** 267