74.

STIMULATED HYPER-RAMAN EMISSION FROM SODIUM VAPOUR

D. COTTER, D.C. HANNA, W.H.W. TUTTLEBEE and M.A. YURATICH
Department of Electronics, The University, Southampton SO9 5NH, UK

Received 13 May 1977

The first observation of tunable stimulated hyper-Raman scattering is reported. A dye laser (ω_p) focussed into sodium vapour produced stimulated hyper-Raman emission ($2\omega_p - \omega_{3s\text{-}4p}$) at ~ 2.3 μm when $2\omega_p$ was tuned near the 3s-4d two-photon resonance. Photon conversion efficiencies up to 2% were obtained. The hyper-Raman emission was tuned over a ~ 160 cm^{-1} range, and could be observed with pump powers as low as ~ 2kW.

A number of two-photon and three-photon absorption processes in alkali vapours have been extensively studied in recent years. There is interest both in multiphoton ionisation and absorption to bound states. Another two-photon process, stimulated electronic Raman scattering, has also been the subject of recent investigations and has been shown to offer a simple and efficient means of converting tunable dye laser radiation into widely tunable infrared Stokes radiation [1–5]. In this letter we report observation of the three-photon analogue of stimulated Raman scattering, namely stimulated hyper-Raman scattering (SHRS), in which two pump photons (ω_p) are annihilated and a Stokes photon (ω_s) is created. Hyper-Raman scattering was first observed as a weak spontaneous process [6] and the first observation of stimulated hyper-Raman scattering appears to have been that of Yatsiv et al. [7], who briefly mention it as one of a number of nonlinear processes which they produced in K vapour. Recently SHRS has been investigated in some detail by Vrehen and Hikspoors [8,9]. For their pump source they used a 200 MW Nd:YAG laser, whose wavelength is fortuitously close to resonance with a two-photon transition in caesium. In this letter we show that by using a tunable dye laser as the pump source, SHRS can occur with high efficiency and that the frequency of the resulting Stokes emission can be tuned over a significant range. The results of a calculation of the expected SHRS threshold, based on the usual perturbation theory approach, is given for the two cases of circularly polarised pump radiation. Our experimental observations confirm that circular polarisation gives a significantly lower threshold. The presence of strong linebroadening mechanisms is indicated by the observation that the linewidth of the Stokes radiation is 20–30 times greater than both the linewidth of the pump laser and the Doppler width of the hyper-Raman transition.

We have studied the 3s → $4p_{3/2}$ hyper-Raman transition in sodium (fig. 1), the process being resonantly enhanced by tuning the dye laser (Rhodamine 6G) in the region of the 3s-4d two-photon resonance. Further resonant enhancement results from the proximity of

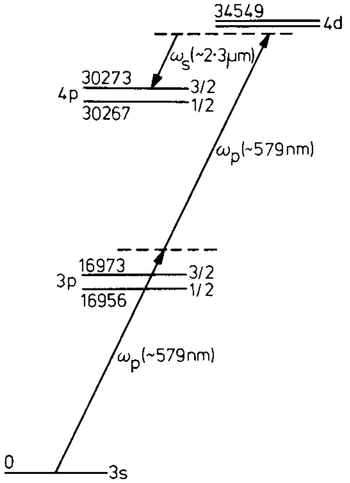

Fig. 1. Partial energy level diagram for neutral sodium showing the 3s → $4p_{3/2}$ hyper-Raman transition.

the dye laser frequency to the single-photon 3s-3p transition. Similar level schemes in other alkalis are also candidates for SHRS, but require the use of less convenient near infrared dye lasers.

There is an important difference between the level system we have studied and that used by Vrehen and Hikspoors [8,9]. In their experiment the intermediate level with which the pump frequency was most strongly resonant also acted as the final level, and therefore the wavelengths of the pump and scattered radiation were not widely different. In our scheme, by scattering to a higher lying final level, it is possible to generate Stokes radiation with a considerably longer wavelength than the pump. We note here that a related scattering process, in which radiation at frequency $2\omega - \Omega$ is produced when a pump laser is tuned close to an atomic transition Ω, has been described by Carlsten and Szoke [10] and Grynberg et al. [11]. They refer to the process as "three-photon scattering". If this process is considered within the framework of perturbation theory it is seen to be described by a susceptibility of the same form as that for hyper-Raman scattering and from this point of view the processes may be said to have a common identity. The important distinction we draw between hyper-Raman scattering and "three-photon scattering" is as follows. In "three-photon scattering" the intermediate levels which make the dominant contribution to the process are the final and initial atomic levels themselves. In hyper-Raman scattering intermediate levels other than the final and initial levels are dominant.

SHRS is characterised by an exponential growth of Stokes intensity with distance through the scattering medium. The gain coefficient G can be calculated as for stimulated Raman scattering [e.g. 12] except that the hyper-Raman process is described by a fifth order nonlinear susceptibility, $\chi_{\mathrm{HR}}^{(5)}(-\omega_s;\omega_p,\omega_p,-\omega_p,-\omega_p,\omega_s)$. In the general case, in which pump and Stokes waves can have any polarisation and spin-orbit splitting of the intermediate levels cannot be neglected, the calculation of $\chi_{\mathrm{HR}}^{(5)}$ is rather lengthy. It can be greatly shortened by using spherical tensor techniques (as in ref. [12]) and this general calculation has been performed for the alkali atoms. Detailed numerical calculations have been made for the above transition in Na, and the analogous transition (6s → 7p) in Cs [13]. For the Cs calculation spin-orbit splitting of the 6p and 6d doublets cannot be neglected. However for the Na scheme the fine structure can be ignored since the $3p_{3/2}-3p_{1/2}$ splitting (17 cm^{-1}) is small compared to the detuning $\Delta\omega_1 = \omega_{3s3p} - \omega_p$ (typically ~ 300 cm^{-1}), and the $4d_{5/2}-4d_{3/2}$ splitting (0.03 cm^{-1}) is small compared to the dye laser linewidth (0.1 cm^{-1}). Thus we present here the results of an analysis in which fine structure of all but the final 4p level is ignored and, in the summation over intermediate states, only the 3p and 4d levels, which make the dominant contribution, are taken into account. The power gain coefficient G is (in SI units)

$$G = \frac{N I_p^2 \omega_s e^6}{30 \hbar^5 (5\epsilon_0 c)^3 \Gamma} \Phi \Theta \tag{1}$$

where

$$\Phi = \frac{400}{27}\left\{\frac{\langle 4p|r|4d\rangle\langle 4d|r|3p\rangle\langle 3p|r|3s\rangle}{\Delta\omega_1 \Delta\omega_2}\right\}^2 \tag{2}$$

if the final level is $4p_{3/2}$. When the final level is $4p_{1/2}$, Φ takes exactly half this value. Here $\Delta\omega_2 = \omega_{3s4d} - 2\omega_p$, Γ is the HWHM linewidth (rad s^{-1}) of the spontaneous hyper-Raman emission, N is the atomic number density and I_p is the pump intensity. The angular dependence [12,13] is contained in the factor Θ, which is $(3\cos^2\beta + 1)/4$ in the case of linearly polarised pump and Stokes waves, where β is the angle between the two polarisations. For pump and Stokes waves propagating collinearly in the same direction and circularly polarised in the same sense, Θ takes the value 9/4. For circular polarisation in opposite senses Θ is zero. In generating a Stokes output by amplification of the spontaneous noise, the Stokes wave will take the polarisation for which the hyper-Raman gain is greatest, and therefore the gain coefficient is effectively 9/4 times greater for a pump wave which is circularly rather than linearly polarised.

The experiment used a Rhodamine 6G dye laser similar to that described elsewhere [4,14], giving an output of up to 8 mJ in a 15 ns pulse (500 kW peak power), with a 0.1 cm^{-1} linewidth and nearly diffraction-limited beam. The linearly polarised beam was passed through a Fresnel rhomb to produce circular polarisation and then focussed so as to be confocal over a ~ 25 cm column of sodium vapour at 10 torr pressure contained in a heat pipe oven. The infrared radiation was detected after a 30 cm grating monochromator. As the dye laser was tuned across the 3s-4d two-photon transition a strong infrared emission, collinear with the dye laser beam, was observed. Its energy was greater than 0.2 nJ over a dye laser tuning range of ~ 80 cm^{-1} and the measured wavelengths agreed to within experimental error (± 2 cm^{-1}) with those calculated from $\omega_s = 2\omega_p - \omega_{3s4p_{3/2}}$.

Bloom et al. [15] observed a 4-wave parametric process in Na when they pumped near the 3s-3d two-photon resonance. If an analogous process were responsible for the IR we observed then a tunable UV output near 30273 cm^{-1} (3s-4p) with about eight times the Ir energy (the ratio of UV and IR photon energies) should have accompanied the IR output. Such an emission was not found although two much weaker UV emissions were seen, one of fixed frequency, 30366 cm^{-1}, with peak energy ~ 2 μJ and one which tuned as $2\omega_p - 4426\ \mathrm{cm}^{-1}$ with peak energy ~ 60 nJ. The latter signal has its origin in a 4-wave parametric process and the fourth wave, at 4426 cm^{-1} with an energy of ~ 5 nJ was just detectable. The origin of the fixed frequency UV signal is not clear at present.

The IR emission at $2\omega_p - \omega_{3s4p_{3/2}}$ was also seen in the backward direction (although over a much smaller tuning range). This gives further confirmation that this IR emission was not parametric in origin since the coherence length for the backward wave parametric process is extremely small.

A pyroelectric detector was used to measure the energy of the SHRS signal in the forward direction as a function of dye laser tuning, as shown in fig. 2. The output energy was greater than 0.1 μJ (~ 15 W) over the 45 cm^{-1} range of pump frequencies (90 cm^{-1} Stokes tuning range) as shown. By using a more sensitive InAs detector, infrared energies greater than 0.2 nJ (30 mW) were observed throughout a total infrared tuning range of ~ 160 cm^{-1}, from 4240 – 4400 cm^{-1}. The infrared pulse duration was 7 ns, and so the maximum observed energy of ~ 37 μJ represents a peak power of ~ 5 kW; this also corresponds to a value of ~ 2% for the conversion efficiency from pump photons to Stokes photons. A simple calculation shows that under these conditions the number of generated Stokes photons was comparable to the number of atoms in the region of the focussed dye laser beam, and the output was therefore limited by depletion of the atomic ground state population. It also follows that for atoms at the distant end of the vapour column, the SHRS rate exceeds the three-photon ionisation rate. In fig. 2 the maximum detected energy is approximately the same for either linear or circular pump polarisation. This is to be expected because of atomic saturation, as described above. However as the pump is tuned further from the two-photon resonance, the SHRS gain falls. The smaller the gain, the greater is the fraction of the vapour column length needed for the Stokes intensity to grow to a level at which the ground state atomic population is significantly depleted. Thus the volume of vapour from which Stokes energy can be extracted is reduced. Numerical calculation shows that this adequately accounts for the general shape of the low frequency side of the tuning profile, and the difference between the observed profiles for circular and linear polarisations. When the dye laser was exactly resonant with the 3s-4d two-photon transition, a dip in hyper-Raman output occurred as shown in fig. 2. We believe this to be due to pump depletion via two-photon absorption and three-photon ionisation. However, at present we are unable to explain the asymmetry of the tuning profile shown in fig. 2, the output energy and tuning range being less than expected for pump frequencies tuned above the two-photon resonance. A similar (and unexplained) asymmetry has previously been observed on stimulated electronic Raman transitions [3].

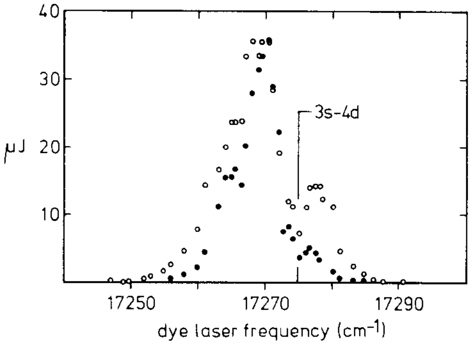

Fig. 2. Observed energy of stimulated hyper-Raman emission as a function of tuning for linear (●) and circular (○) pump polarisation.

No SHRS corresponding to the lower gain 3s → $4p_{1/2}$ transition could be observed; this can be explained by the atomic saturation produced by the stronger 3s → $4p_{3/2}$ transition. Attempts to observe SHRS with the dye laser tuned close to the $3s\text{-}3p_{1/2,3/2}$ single-photon resonances were unsuccessful, probably as a result of competition from the very strong single-photon absorption of the pump light. By using two dye pumps (ω_{p1} and ω_{p2}) tuned so that $\omega_{p1} + \omega_{p2}$ is close to the two-photon resonance, and one pump tuned to take better advantage of the 3s-3p single-photon resonance [16], a further reduction in threshold should be observed. Nevertheless with a single pump source ($\omega_{p1} = \omega_{p2} = \omega_p$), we have found that the two-photon resonance enhancement is sufficient to allow observation of SHRS with a pump power as low as 2kW.

The linewidth of the hyper-Raman emission was measured using an infrared grating monochromator (instrumental resolution 0.5 cm^{-1}) and typically found to be ~ 2–3 cm^{-1}. This is 20–30 times greater than both the pump laser linewidth (0.1 cm^{-1}) and the Doppler width of the 3s-4$\mathrm{p}_{3/2}$ transition (~ 0.13 cm^{-1}). A similar degree of linebroadening has been observed in stimulated electronic Raman scattering [4]. The origin of this has not been conclusively identified.

A comparison between the predicted and observed SHRS threshold was made, using eqs. (1) and (2) to calculate the expected threshold pump intensity. The result of this calculation is strongly dependent on the value assumed for Γ which, in view of the linebroadening observed in the experiment, is rather ill-defined; this constitutes a weakness in the calculation. Threshold is reached when the gain exponent $GL = 30$, this being the minimum gain required to amplify the spontaneous hyper-Raman signal to a detectable level. Thus if L (length of vapour column), N and Γ are known, the threshold pump power can be calculated as a function of ω_p. A further refinement in the calculation is to include the effects of diffraction [17]. Fig. 3 shows the calculated threshold power for our experiment, taking $\Gamma = 15\ \mathrm{cm}^{-1}$. This choice of linewidth gives reasonable agreement both with the observed SHRS threshold and tuning range in the region of the 3s-4d two-photon resonance, and also with the measured linewidth of the generated infrared radiation, allowing for gain narrowing.

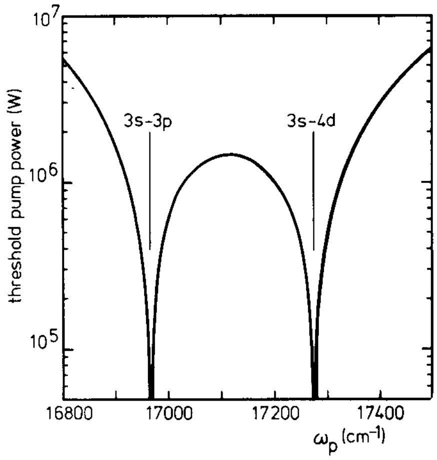

Fig. 3. Calculated threshold pump power for stimulated hyper-Raman emission on the 3s → 4$\mathrm{p}_{3/2}$ transition.

It is to be expected that similar results to those reported here would be observed on the 4s → 5p transition in K, 5s → 6p in Rb and 6s → 7p in Cs. These may be of some interest since the Stokes wavelengths would be progressively longer, reaching 16 μm in Cs. A significant reduction in threshold should result from the use of two dye laser pumps, to take full advantage of both the one-photon and two-photon resonances. However the considerable linebroadening we have observed both in stimulated Raman scattering and SHRS constitutes a serious drawback, both in raising the threshold and in producing a broad output linewidth. Proposed schemes for using transitions between Rydberg levels as sources of tunable infrared radiation [18] may be vitiated by this linebroadening.

This work has been supported by the Science Research Council. D. Cotter holds an 1851 Exhibition Research Fellowship.

References

[1] M. Rokni and S. Yatsiv, Phys. Letters 24A (1967) 277; M. Rokni and S. Yatsiv, IEEE J. Quant. Elect. QE-3 (1967) 329; P.P. Sorokin, N.S. Shiren, J.R. Lankard, E.C. Hammond and T.G. Kazyaka, Appl. Phys. Letters 10 (1967) 44.
[2] P.P. Sorokin, J.J. Wynne and J.R. Lankard, Appl. Phys. Letters 22 (1973) 342.
[3] J.L. Carlsten and P.C. Dunn, Opt. Commun. 14 (1975) 8.
[4] D. Cotter, D.C. Hanna, P.A. Kärkkäinen and R. Wyatt, Opt. Commun. 15 (1975) 143; D. Cotter, D.C. Hanna and R. Wyatt, Opt. Commun. 16 (1976) 256.
[5] D. Cotter and D.C. Hanna, J. Phys. B9 (1976) 2165.
[6] P.D. Maker, Physics of quantum electronics, Proc. Intern. Conf. San Juan, Puerto Rico, 1965 (McGraw-Hill, New York, 1966) p. 60.
[7] S. Yatsiv, M. Rokni and S. Barak, IEEE J. Quant. Elect. QE-4 (1968) 900.

[8] Q.H.F. Vrehen and H.M.J. Hikspoors, in: Proc. 9th Intern. Conf. on Quantum electronics, Amsterdam 1976; Opt. Commun. 18 (1976) 113.

[9] Q.H.F. Vrehen and H.M.J. Hikspoors, (1977) to be published.

[10] J.L. Carlsten and A. Szöke, Phys. Rev. Letters 36 (1976) 667.

[11] G. Grynberg, F. Biraben, M. Bassini and B. Cagnac, Phys. Rev. Letters 37 (1976) 283.

[12] M.A. Yuratich and D.C. Hanna, J. Phys. B9 (1976) 729.

[13] M.A. Yuratich, Ph. D. Thesis, University of Southampton (1977) unpublished.

[14] D.C. Hanna, P.A. Kärkkäinen and R. Wyatt, Opt. Quant. Elect. 7 (1975) 115.

[15] D.M. Bloom, J.T. Yardley, J.F. Young and S.E. Harris, Appl. Phys. Letters 24 (1974) 427.

[16] J.E. Bjorkholm and P.F. Liao, Phys. Rev. Letters 33 (1974) 128.

[17] D. Cotter, D.C. Hanna and R. Wyatt, Appl. Phys. 8 (1975) 333.

[18] A.M.F. Lau, W.K. Bischel, C.K. Rhodes and R.M. Hill, Appl. Phys. Letters 29 (1976) 245.