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Observation of the suppression of the stimulated hyper-Raman scattering ($3S_{1/2}$–$4P_{3/2}$) near the sodium 4D two-photon resonance

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Abstract. The stimulated hyper-Raman scattering (SHRS) emissions associated with the excitation of the $4P_{3/2}$ state in sodium vapour have been observed in both the forward and backward directions as the pumping laser was tuned near the 3S–4D two-photon resonance. The results show that though the forward SHRS process is greatly reduced by a destructive interference between it and the four-wave mixing (FWM) process induced by itself, the forward SHRS emission has a positive gain and can still be observed under a certain condition.

1. Introduction

The stimulated hyper-Raman scattering (SHRS) associated with the excitation of the 4P states in sodium vapour was first observed by Cotter *et al* (1977) by using a circularly polarized laser tuned near the 3S–4D two-photon resonance. Recently, a destructive interference between the SHRS process and the four-wave mixing (FWM) process induced by the SHRS itself was discussed by Moore *et al* (1988), Wunderlich *et al* (1990) and Garrett *et al* (1992). They also observed the SHRS in sodium vapour, using a linearly polarized pumping laser tuned near the 3S–3D or 3S–4D two-photon resonance. According to their observations and analysis, they generally pointed out that the SHRS emission associated with the excitation of a state which is also dipole-coupled to the ground state is almost totally suppressed by the destructive interference with the self-induced FWM if the pumping laser is linearly polarized and the following condition in SI units (Garrett *et al* 1992) is satisfied,

$$\frac{e^2 N_0 f_{0j} z}{8\pi \varepsilon_0 m c^2} \gg \Gamma \tag{1}$$

where Γ is the larger of the laser bandwidth or the pressure-broadened width of the $|0\rangle \rightarrow |j\rangle$ transition, z is the pathlength in the medium, N_0 is the number density of the resonant species, f_{0j} is the oscillator strength of the $0 \rightarrow j$ transition, ε_0 and c are the permittivity and the light velocity in vacuum respectively, and e and m are the charge and mass of an electron, respectively. Obviously this condition is independent of the laser intensity. Since FWM emission only occurs in the forward direction due to phase-matching considerations, only the forward SHRS emission is completely suppressed, but

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the backward SHRS emission is not influenced. It means that the interference effect only yields backward-propagating SHRS emission, as demonstrated in their Na vapour experiments. This destructive interference effect was also studied theoretically by Malakyan (1989). According to her analysis, the SHRS emission in the forward direction will still have a positive gain if the following condition is satisfied,

$$R=\omega_2\mu_{23}^2\Omega_2^2/\omega_3\mu_{31}^2\Delta^2\approx f_{32}\Omega_2^2/f_{13}\Delta^2>1 \tag{2}$$

where ω_2 and ω_3 are the frequencies of SHRS and the self-induced field in a FWM process, respectively; μ_{ij} and f_{ij} are the dipole matrix element and the oscillator strength of the $i\rightarrow j$ transition, respectively; $\Delta=2\omega_p-\omega_{21}$ denotes the detuning from the two-photon

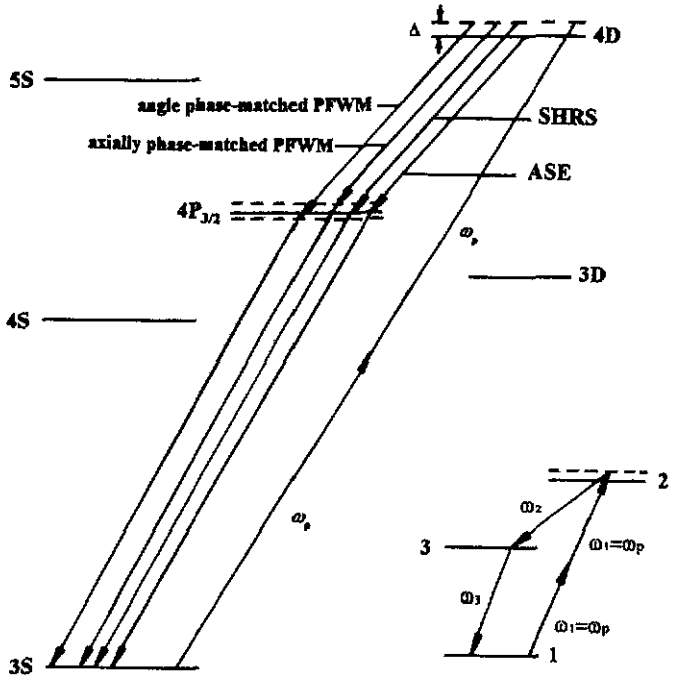

Figure 1. Partial diagram of the sodium atomic energy levels. Here the processes expected to be observed are illustrated. A sketch of the SHR process is shown in the lower right part.

resonance ω_{21} (see figure 1) and Ω_2 is the two-photon Rabi frequency defined in SI units by

$$\Omega_2=\frac{1}{2nc\varepsilon_0\hbar^2}\left|\sum_m\frac{\mu_{2m}\mu_{m1}}{\omega_{m1}-\omega_p}\right|I_p\ (\text{rad s}^{-1}). \tag{3}$$

Here I_p and ω_p are the laser intensity and frequency respectively, $h=2\pi\hbar$ is the Planck constant, and ω_{ij} is the transition frequency from the i state to the j state, n is the refractive index. The result means that the SHRS in the forward direction could be observed in the experiment where the condition given by equation (2) is met. From equation (3) it is found that the condition given by equation (2) is dependent on the laser intensity. Obviously this prediction contradicts that of Moore *et al* (1988), Wunderlich *et al* (1990) and Garrett *et al* (1992).

Most recently we observed the SHRS emissions associated with the 2S–2P and 2S–3P transitions in lithium vapour, pumped with a linearly polarized laser which was

tuned near the 2S–3S and 2S–4S two-photon resonances (Lu and Liu 1993). It was found that as the laser was tuned near the 2S–4S two-photon resonance, the forward SHRS emission associated with the 2S–3P transition was observed. In this case, a calculation shows that the condition given by equation (2) was nearly satisfied. However, the forward SHRS emission associated with the 2S–2P transition was not observed as the laser was tuned near the 2S–3S or 2S–4S two-photon resonance. The calculations show that in these cases the R in equation (2) is much smaller than one, i.e. the condition given by equation (2) can not be met.

It is interesting for us to observe the forward SHRS emission associated with the 3S–4P transition in sodium vapour, pumped with a linearly polarized laser which is tuned near the 3S–4D two-photon resonance, under circumstances where both the conditions given by equations (1) and (2) are satisfied. All the results presented here verify that the condition given by equation (2) is correct.

2. The experiment

The experimental set-up is shown in figure 2. A pulsed R590 (Exciton) dye laser (Quanta-Ray, PDL-1) was used as a pumping source. The operating parameters of the dye laser were: 20 ns pulse duration, 2–8 mJ energy per pulse, 0.25 cm^{-1} linewidth and 10 Hz repetition rate. In this experiment a linearly polarized pumping light was used.

A conventional heat pipe with a 30 cm heated zone was operated with sodium vapour. Argon at 1.5 mbar was used as a buffer gas. The laser beam was directed along the axis of the heat pipe and focused by an 80 cm focal-length lens to a spot with a diameter of 0.25 mm at the centre of the heat pipe. A 4 mm aperture was placed in the forward output beam from the heat pipe to discriminate against the conical emission produced by angle phase-matched FWM. Instead of this aperture a 5 mm beam stop was used to block the axial part of emission in order to observe the angle phase-matched FWM emission. For the forward measurement a colour filter (RG780) was used against the strong pumping laser. The output emissions were detected in both the forward and backward directions by a photomultiplier (Hamamasu R928) in the visible region and by a Si/PbS two-colour IR detector (Spex 1429C) in the infrared region after 1 m monochromator (Spex 1704). In this experiment the forward and backward SHRS emissions were not measured on the same scale.

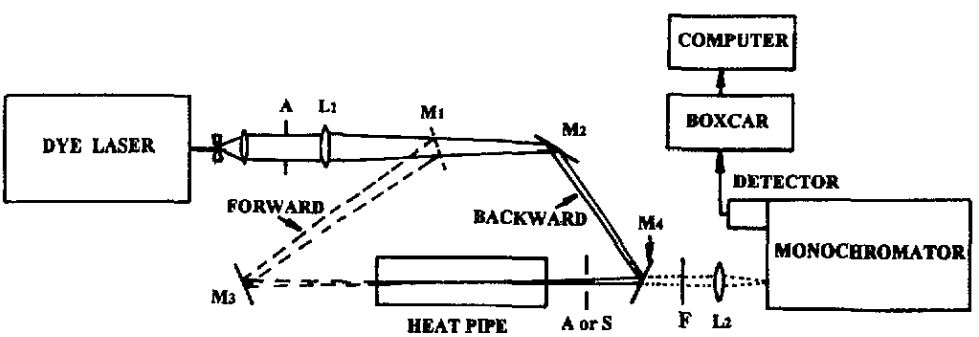

Figure 2. Experimental set-up. The mirrors M_1 and M_3 are used for the forward measurement, and the mirrors M_2 and M_4 for the backward measurement.

3. Results and discussion

As the pumping laser was tuned near the sodium 3S–4D two-photon resonance (578.89 nm), the emission spectra in both the forward and backward directions were measured in the 2.3 μm wavelength region. Here the fine structure of the 4D state is ignored since the splitting (0.03 cm^{-1}) of the $4D_{3/2}$ and $4D_{5/2}$ is much smaller than the laser linewidth 0.25 cm^{-1}. In the measured wavelength region, at small detuning from the two-photon resonance, four spectral lines related to the $4P_{3/2}$ state could be expected in the forward direction, as shown in figure 1, corresponding to four processes: ASE ($4D_{3/2,5/2}$–$4P_{3/2}$), SHRS ($3S_{1/2}$–$4P_{3/2}$), angle phase-matched and axially phase-matched FWM ($2\omega_p = \omega_1 + \omega_2$). However, only two spectral lines corresponding to the ASE and SHRS processes could occur in the backward direction due to the phase-matching consideration. Indeed, at 0.5 cm^{-1} detuning to the high energy side of the two-photon resonance we observed all the spectral lines mentioned above. The results are shown in figures 3(*a*)–(*d*). Figure 3(*a*) shows the emission spectrum measured in the forward direction without any beam stop or aperture. Figures 3(*b*) and (*c*) show the spectra measured with a 4 mm aperture and a 5 mm beam stop, respectively. Since most of the emission generated by the angle phase-matched FWM process can be eliminated by the aperture, from figure 3(*b*) it can be seen that there are three peaks which correspond to the SHRS ($3S_{1/2}$–$4P_{3/2}$), ASE ($4D_{3/2,5/2}$–$4P_{3/2}$) and axially phase-matched FWM enhanced by near resonance of the $4P_{3/2}$ state. However, the axial part of the forward emission can be blocked by the 5 mm beam stop, so figure 3(*c*) shows the spectrum which is only from the conical part of the forward emission, corresponding to the angle phase-matched FWM process. In this case, though the spectral lines corresponding to the angle phase-matched and axially phase-matched FWM processes are widely spreading, very strong and very close to the ASE and SHRS spectral lines, all the lines still can be resolved in evidence by using the aperture and beam stop. Figure 3(*d*) shows the backward emission spectrum produced by the SHRS and ASE processes only. Here the ASE signal is much stronger than the SHRS signal. In order to clarify the backward SHRS signal, we subtract the ASE signal from the backward emission spectrum by using a simple data processing, in which a symmetric lineshape for the ASE is assumed. Figure 3(*e*) shows the spectrum corresponding to the backward SHRS after the processing. Comparing figure 3(*e*) with figure 3(*b*), we find that the backward SHRS peak wavelength is exactly the same as the forward. For comparison, the observed and calculated IR wavelengths for the ASE, SHRS, axially phase-matched and angle phase-matched FWM are listed in table 1. The IR wavelength of the axially phase-matched FWM was calculated in the same way as we did for potassium vapour (Lu and Liu 1992). The refractive indices of sodium vapour at different wavelengths were given by the Sellmeier formula. The angle phase-matched FWM in sodium vapour has been well analysed by Krasinski *et al* (1985). In a similar way we calculated the IR wavelength of the angle phase-matched FWM with the Sellmeier formula and found that the phase matching could be satisfied in a wavelength range from 2.3382 to 2.3544 μm, corresponding to a cone half angle range from $\pi/2$ rad to zero. It can be expected that the spectrum of the angle phase-matched FWM could be broad band and the peak should occur near 2.3382 μm, where the phase-matching point is closer to the $3P_{3/2}$ level and therefore the process has a larger gain due to the resonant enhancement. Actually we observed the peak at 2.3380 μm. According to the calculation, the cone half angle at this wavelength is 0.05 rad (3°). In all the calculations, a pure sodium vapour was assumed. Usually the vapour is a mixture of sodium and argon. A sodium vapour mixed with some argon is less dispersive than pure sodium

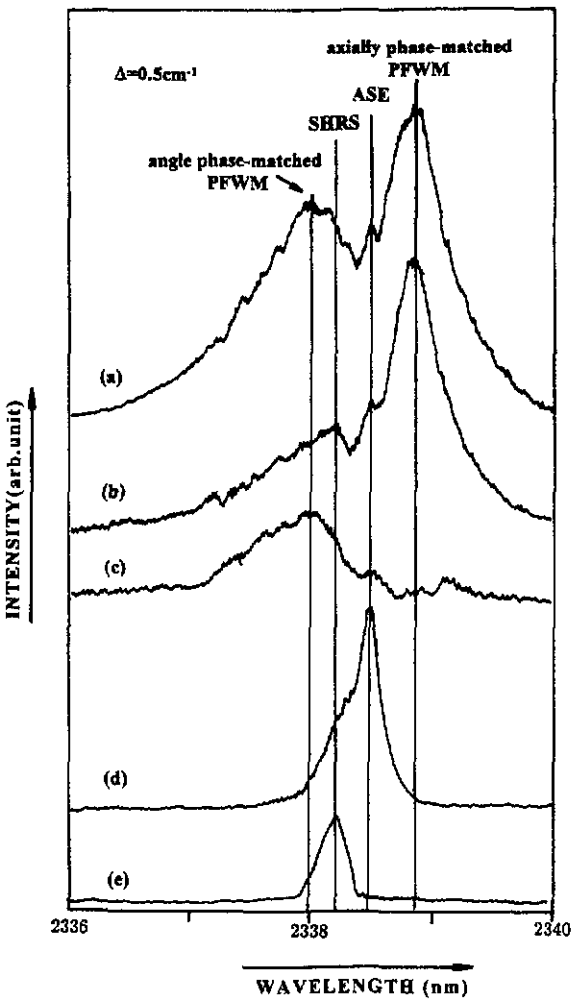

Figure 3. Emission spectra measured in the forward direction (*a*) without aperture or beam stop, (*b*) with a 4 mm aperture, and (*c*) with a 5 mm stop; and (*d*) measured in the backward direction. The spectrum (*e*) is the result from the spectrum (*d*) minus the ASE signal. Here the detuning from the 4D two-photon resonance is $\Delta = 0.5\ \text{cm}^{-1}$ and the vapour pressure is ~2.4 mbar.

Table 1. Calculated and observed IR wavelengths for various processes at the detuning $\Delta = 0.5\ \text{cm}^{-1}$ from the 3S–4D two-photon resonance.

Process	Calculated (μm)	Observed (μm)
ASE	2.3385	2.3385
SHRS	2.3382	2.3382
Axially phase-matched FWM	2.3391	2.3389
Angle phase-matched FWM	2.3382†	2.3380

† This wavelength is the limitation on the long wave side.

vapour at the wavelengths we observed in this experiment. So the phase-matching points calculated for pure sodium vapour will move to the $4P_{3/2}$ level if a mixed vapour is considered. Table 1 shows good agreement between the predicted and the observed wavelengths.

We also measured the forward and backward emission spectra in the wavelength range 2.333–2.345 μm at different detunings. At 2.4, 3.6 and 4.8 cm^{-1} detunings to the low energy side of the resonance the results measured without any aperture and beam

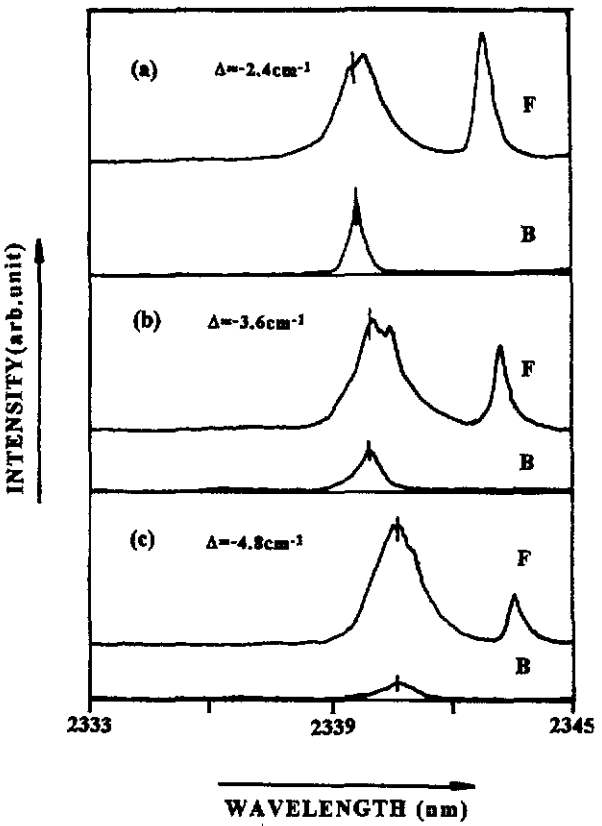

Figure 4. Emission spectra measured in both the forward (upper trace) and backward (lower trace) directions at detunings (*a*) $\Delta = -2.4\ cm^{-1}$, (*b*) $\Delta = -3.6\ cm^{-1}$ and (*c*) $\Delta = -4.8\ cm^{-1}$ from the 4D two-photon resonance. Here the vapour pressure is ~2.4 mbar.

stop are shown in figures 4(*a*)–(*c*), respectively, where the upper trace is for the forward emission and the lower trace for the backward. At the same detunings as above, the results measured with a 4 mm aperture or a 5 mm beam stop in the forward direction are shown in figures 5(*a*)–(*c*). Note that in figures 4(*a*)–(*c*) the peak at ~2.342 μm on the right-hand side of each upper trace should be generated by another parametric process because it only appears in the forward direction. This line was also observed by Hartig (1978). The mechanism generating this line is not yet clear. We also note that for detuning to the low energy side of the resonance the ASE can not be observed and only the SHRS exists in the backward measurement, as shown in figures 4(*a*)–(*c*). In figures 4(*a*)–(*c*) the broad-band signal on the short wave side of each upper trace should be considered to be from the SHRS and FWM processes. From figures 5(*a*)–(*c*) it can be seen that the broad-band signal is slightly reduced by the 4 mm aperture but

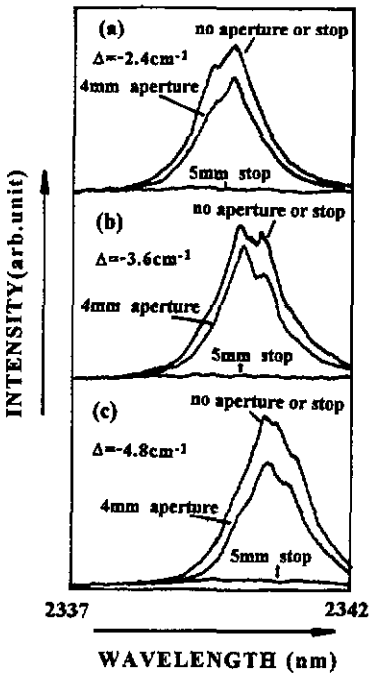

Figure 5. Forward emission spectra measured without any aperture or beam stop, with a 4 mm aperture, and with a 5 mm beam stop at detuning (*a*) $\Delta=-2.4$ cm, (*b*) $\Delta=-3.6\ \text{cm}^{-1}$ and (*c*) $\Delta=-4.8\ \text{cm}^{-1}$. The vapour pressure is ~2.4 mbar.

almost completely eliminated by the 5 mm beam stop. It means that the signal copropagates with the laser beam and no angle phase-matched FWM emission exists. From these figures we also find that the broad-band signal actually contains two peaks. According to the theoretical analysis (see figure 1), the left-hand one is produced by the SHRS, with a wavelength exactly the same as the backward SHRS wavelength, the right-hand one is from the axially phase-matched FWM process. From these measurements it seems that by increasing the detuning to the low energy side of the two-photon resonance the SHRS process becomes dominant, namely, the gain of the axially phase-matched FWM process decreases more rapidly than that of the SHRS process. The reason for the absence of the angle phase-matched FWM signal is that it could be considered to be too weak to be detected, or due to the large phase-matching angle, which can not be accepted by the monochromator in this experiment.

In this experiment the laser intensity I_p is $5.6\times10^8\ \text{W cm}^{-2}$. The sodium oscillator strengths $f_{3S_{1/2}-4P_{3/2}}$, $f_{4P_{3/2}-4D_{5/2}}$ and $f_{4P_{3/2}-4D_{3/2}}$ are taken from Wiese *et al* to be 0.0094, 0.82 and 0.091, respectively. The two-photon Rabi frequency for the 3S–4D transition is calculated with equation (3) and we have $\Omega_2=412I_p$. If the detuning, Δ, from the 3S–4D two-photon resonance is taken to be $2.4\ \text{cm}^{-1}$, from equation (2) we have $R=25$, that is much larger than one. This calculation confirms that the forward SHRS should be observed if the condition given by equation (2) is satisfied. Equation (1) is evaluated in the circumstance of this experiment, where the laser linewidth of $0.25\ \text{cm}^{-1}$ is dominant. The number density of sodium and the pathlength are taken to be 10^{14}–$10^{16}\ \text{cm}^{-3}$ and 30 cm, respectively. We find that the condition given by equation (1) is also well satisfied.

4. Conclusion

In this experiment the SHRS emission associated with the excitation of the $4P_{3/2}$ state of sodium has been observed in both the forward and backward directions as the pumping laser was detuned from the 3S–4D two-photon resonance. The results lead to the conclusion that though the forward SHRS emission is greatly reduced by the destructive interference effect, it can still occur with a positive gain and be observed if the condition given by equation (2) is satisfied. The angle phase-matched FWM emission, whose wavelength is expected to be very close to that of the SHRS emission and difficult to resolve spectrally, can be well distinguished spatially from the SHRS emission with an aperture or a beam stop. Actually, we find that at large detuning to the low energy side of the two-photon resonance the angle phase-matched FWM is not observed and the SHRS process becomes dominant. Our results verify that the condition given by equation (2) works well though it is derived with some approximations.

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