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The observation of the competition and suppression for stimulated Raman scattering near the potassium 4S–5P resonance

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Abstract. The competition of stimulated Raman scattering (SRS) between 4S–(5P)–5S and 4S–(3D)–5S channels has been observed as a laser is tuned near the 4S–5P resonance in potassium vapour. The experimental results show that SRS only occurs in the 4S–(5P)–5S channel. In this Raman scattering, suppression of the amplified spontaneous emission (ASE) (5S–4P) has been observed. The influence of the four-wave mixing (FWM) processes on the SRS is discussed. In the 4S–(5P)–3D channel, in the backward direction no detectable emission corresponding to SRS (4S–(5P)–3D) has been observed, so the forward (5P)–3D emission should be FWM, not SRS. The ASE (3D–4P) arises from the ASE (5P–3D), which can be observed in the backward direction.

1. Introduction

Stimulated Raman scattering (SRS) pumped with a XeCl laser near 6s7p in barium vapour was observed by Cotter and Zapka (1978), Manners (1983) and Glownia *et al* (1987). There are two possible channels in this process, one is $6s^2$–(6s7p)–6s7s and the other is $6s^2$–(6s7p)–6s5d. With the 350 ps pumping pulse, only the channel $6s^2$–(6s7p)–6s7s could be observed. However, with the 23 ns pumping pulse, only one other channel $6s^2$–(6s7p)–6s5d could be observed. Glownia *et al* explained this phenomenon as follows. In the transient region (350 ps pumping pulse), the gain coefficient g of SRS is proportional to the oscillator strength f. Due to $f_S(6s7s \rightarrow 6s7p) > f_D(6s5d \rightarrow 6s7p)$, only the 2.4 μm SRS ($6s^2$–(6s7p)–6s7s) occurs. In the steady-state region (23 ns pumping pulse), the gain is proportional to f/Γ, where Γ is the Raman linewidth. Since $\Gamma_S(6s7s)$ is significantly larger than $\Gamma_D(6s5d)$, we have $f_S/\Gamma_S < f_D/\Gamma_D$. Thus the SRS wavelength is switched to 475 nm corresponding to the $6s^2$–(6s7p)–6s5d channel.

Later this effect was explained by Malakyan (1990). The SRS gain coefficient is basically the same as that given by Cotter and Zapka (1978) and Glownia *et al* (1987). However, there still exists a difference between them. Malakyan includes a suppression term in the expression for the SRS intensity. The fields (ω_1, ω_2) induce new fields (ω_3, ω_4), which build up a second pathway of excitation of the $|3\rangle$ state, as shown in figure 1. The population on the $|3\rangle$ state is reduced by this four-wave mixing (FWM), the emissions related to the population of the $|3\rangle$ state, such as SRS and amplified spontaneous emission (ASE), are suppressed. If the phase matching of FWM is considered, the suppression can occur only in the forward

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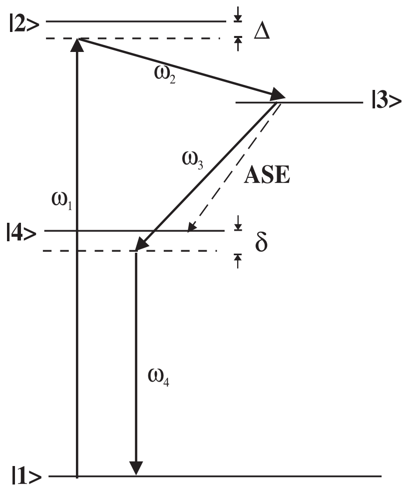

Figure 1. The energy level diagram for SRS and ASE suppressions related to FWM in Raman scattering. Δ and δ represent the detuning from the $|2\rangle$ and $|4\rangle$ states, respectively.

direction. The backward emissions for SRS and ASE are not influenced. Unfortunately, data for the linewidth Γ of barium are not currently available and no measurement in the backward direction for barium vapour has been made, so the competition and suppression for Raman scattering have not been exactly verified by experiment as yet. It is expected that similar experiments with alkali vapour will be performed, where the linewidth Γ is well known, to verify the above theory.

Recently, Domiaty *et al* (1994) and Deng *et al* (1997) observed the suppression in Raman scattering for sodium vapour excited near the 3S–4P resonance. Domiaty *et al* observed all the emissions except the 4P–3D emission in the SRS 3S–(4P)–3D channel and found suppression of the 3D–3P emission. Deng *et al* did a similar experiment and obtained the same result. However, neither of them discussed the competition between the two possible SRS channels 3S–(4P)–3D and 3S–(4P)–4S.

We observed suppression of stimulated hyper-Raman scattering (SHRS) and related ASE suppression for sodium and lithium vapours (Lu and Liu 1993, 1994). In this paper, we present observation of the competition of SRS between both the 4S–(5P)–5S and 4S–(5P)–3D channels for potassium vapour pumped near the 4S–5P resonance. We also observed the suppression of ASE (5S–4P) as Domiaty *et al* and Deng *et al* did in sodium vapour. In this experiment we also found some FWM processes, which could contribute to the forward emissions corresponding to the SRS. These processes are similar to the multiwave mixing processes excited by two-photon processes (Tsai and Lu 1990) in potassium vapour. Though SRS of potassium vapour had been reported by several authors (Cotter *et al* 1975, Bernage *et al* 1981), they did not observe SRS in the backward direction. In order to identify each emission, it is necessary to measure them in both the forward and backward directions.

2. Experiment

The set-up of the forward and backward measurements is shown schematically in figure 2. The measured radiation was generated by potassium vapour excited near the 4S–5P resonance (see figure 3) with the 404 nm pumping wave from the second harmonic generation of the 808 nm

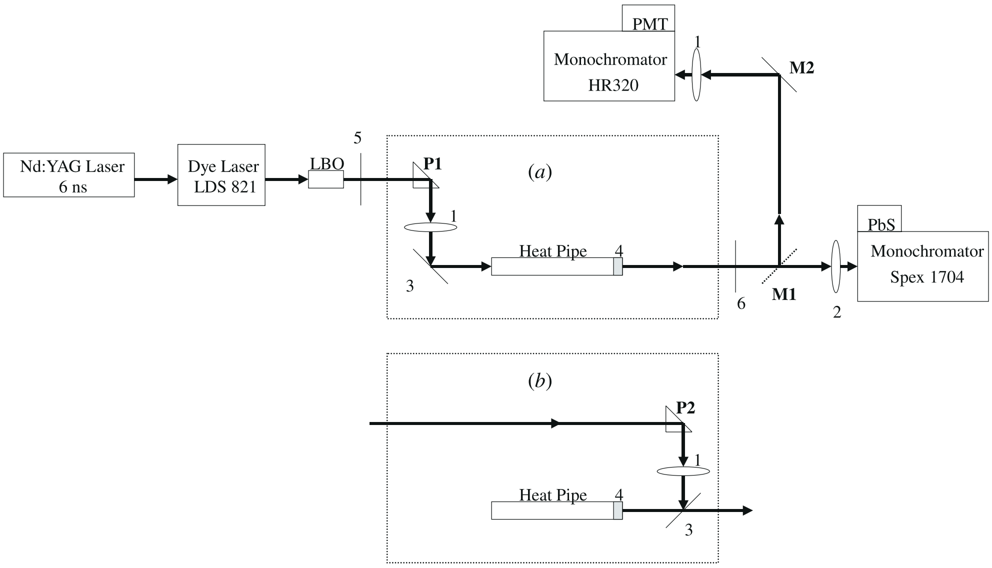

Figure 2. The experiment set-up of (*a*) forward and (*b*) backward measurements. The mirrors M1 and M2 are only used when we measure in the 0.7–1.3 μm range. The prism P1 is used for the forward measurements and the prism P2 for the backward measurements. The other components shown in this figure are: (1) BK7 lens; (2) IR grade quartz lens; (3) 45° dichroic mirror; (4) IR grade quartz window; (5) colour glass filter; and (6) ZnSe plate.

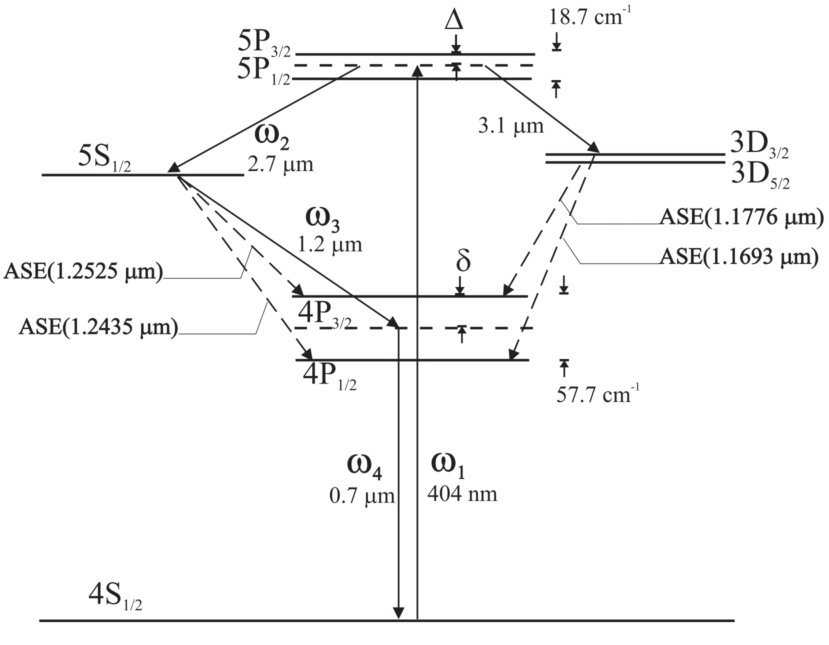

Figure 3. Partial energy level diagram of potassium. Δ and δ represent the detuning from the $5P_{3/2}$ and $4P_{3/2}$ states, respectively.

dye laser with an LBO crystal. The pulsed dye laser (Quanta Ray, PDL-1) was pumped by a frequency-doubled Nd:YAG pulsed laser (Continuum, Surelite II). The parameters for the 404 nm were a pulse duration of 6 ns, 220 μJ pulse energy, 0.2 cm^{-1} linewidth and 10 Hz repetition rate. The potassium vapour was enclosed in a heat pipe (Dunning and Hulet 1996) and heated to a temperature of about 643 K with a 30 cm heated zone. Argon gas at 2.5 Torr pressure was used as a buffer gas in the heat pipe. IR grade quartz was used as the exit window of the heat pipe to avoid absorption at 2.7 μm. The pumping wave was focused into a spot with a diameter of 0.5 mm at the centre of the heat pipe.

In the backward measurement, a 45° dichroic mirror was used to reflect the pumping beam into the heatpipe and to transmit the emissions generated from the potassium vapour. This 45° dichroic mirror is specially made to transmit the radiation in the 730 nm–4 μm wavelength range and reflect radiation in the 404 nm wavelength range. In order to eliminate the 808 nm emission from the pulsed dye laser, a colour glass filter (Mells Griot Co., BG12) was put in front of the heatpipe. In both the forward and backward measurements, a ZnSe plate was put behind the heatpipe to shield against the strong 404 nm pumping wave and transmit the emissions generated from the heatpipe. The output emissions in the region from the visible to 1.3 μm were detected using a photomultiplier tube (Hamamatsu R5108) after a 0.32 m monochromator (Jobin-Yvon HR 320) and in the region 1.3–3.2 μm were detected using a PbS photoconductor (Hamamatsu P3258) after a 1 m monochromator (Spex 1704). These signals from the detectors were fed into a boxcar averager (NF BX531) and processed.

The ASE, SRS, FWM involved in this experiment can be identified by the measurements in the forward and backward directions, as shown in figure 2.

3. Results and discussion

In this experiment, we observed the 2.7 μm emission, as shown in figure 4, in both the forward and backward directions. Because the 2.7 μm emission changed wavelength when the laser was tuned, it could be identified mainly as SRS. However, in the 3.1 μm region, we observed forward emission which changed wavelength as the laser was tuned, but did not find any corresponding backward emission even if we opened the slit to 1 mm, as shown in figure 4. When the slit was opened to 1 mm in the measurement of 3.1 μm backward emission, we observed either the 3.14 μm ASE ($5P_{3/2}$–$3D_{5/2}$) or the 3.16 μm ASE ($5P_{1/2}$–$3D_{3/2}$), depending on the laser tuning, as shown in figure 5. We also found that if the laser was tuned near the $4S_{1/2}$–$5P_{3/2}$ resonance only the ASE ($3D_{5/2}$–$4P_{3/2}$) was observed, and if the laser was tuned near the $4S_{1/2}$–$5P_{1/2}$ resonance only the ASE ($3D_{3/2}$–$4P_{1/2}$) was observed. We can see that the ASE ($5P_{3/2}$–$3D_{5/2}$) coexists with the ASE ($3D_{5/2}$–$4P_{3/2}$), and the ASE ($5P_{1/2}$–$3D_{3/2}$) coexists with the ASE ($3D_{3/2}$–$4P_{1/2}$). Furthermore, when the laser was detuned from $5P_{3/2}$ with $\Delta > 28.7$ cm^{-1}, both the ASEs (5P–3D) and (3D–4P) disappeared but the 3.1 μm forward emission was still there, as shown in figures 4(*c*) and 5(*c*). All the above results indicate that the 1.1 μm ASEs (3D–4P) arise from the ASEs (5P–3D), which populate the 3D states. If we consider that no detectable 3.1 μm SRS emission is observed in the backward direction, as shown in figures 4 and 5, the tunable forward 3.1 μm emission should be not the SRS (5P)–3D but the FWM. Actually, we observed the emissions 544.1, 559.7 and 576.2 nm, generated from the coupling of the pumping laser photon with two IR photons corresponding to the 2.7 and (or) 3.1 μm wavelengths, as shown in table 1 and figure 6. From the coupling schemes, we can see that some tunable 3.1 and 2.7 μm forward emissions come from the above FWM processes.

From the above analyses, we conclude that SRS only occurs in the left channel 4S–(5P)–5S due to the competition between both the SRS channels. According to the SRS gain $g \sim f/\Gamma$

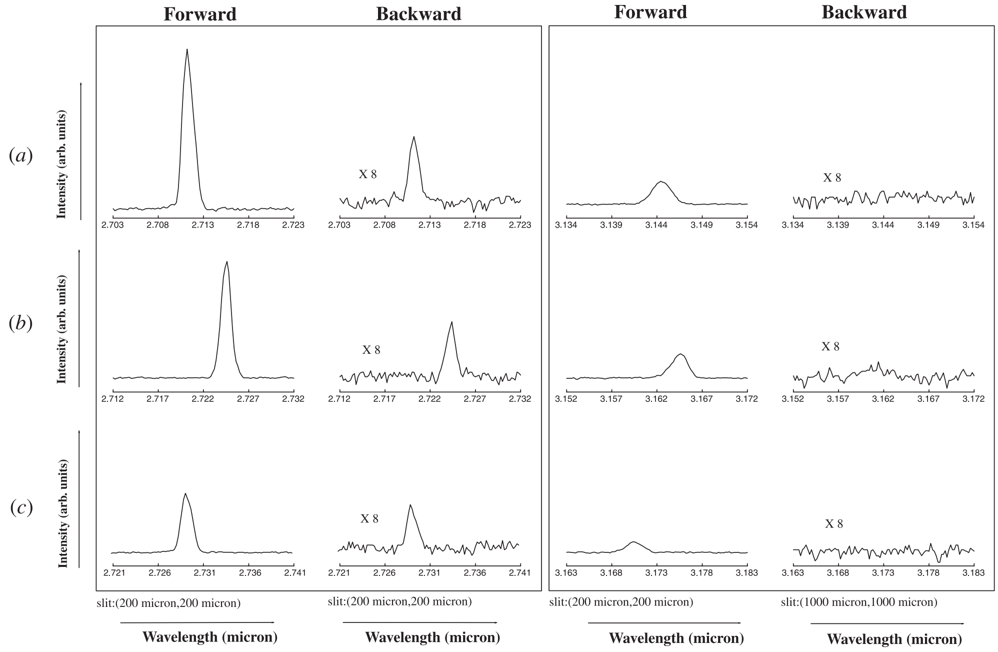

Figure 4. The spectra for 2.7 and 3.1 μm emissions in the forward and backward directions as the laser is tuned at (*a*) $\Delta = 5$ cm^{-1} (5 cm^{-1} below $5P_{3/2}$); (*b*) $\Delta = 23.7$ cm^{-1} (5 cm^{-1} below $5P_{1/2}$); (*c*) $\Delta = 28.7$ cm^{-1} (10 cm^{-1} below $5P_{1/2}$).

Table 1. The observed and calculated wavelengths, and the corresponding generation processes for the FWM processes, which contribute to the forward tunable 2.7 and 3.1 μm emissions.

	This work (nm)	Theory (nm)	Coupling scheme
(*a*)	544.1 ± 0.2 nm	544.1	$\omega_p - 2\omega((5P)\text{–}3D)$
(*b*)	559.7 ± 0.2 nm	559.6	$\omega_p - \omega((5P)\text{–}5S) - \omega((5P)\text{–}3D)$
(*c*)	576.2 ± 0.2 nm	576.1	$\omega_p - 2\omega((5P)\text{–}5S)$

(Cotter and Zapka 1978, Glownia *et al* 1987), we estimate the ratio of $g_{2.7\,\mu\text{m}}/g_{3.1\,\mu\text{m}}$ to be 8.9. Here we take the oscillator strength to be 1.48 for 5P–5S and 0.14 for 5P–3D (Lindgard and Nielsen 1977). At a vapour pressure of several Torr, the pressure-broadened linewidth is much smaller than the Doppler linewidth (Demtröder 1981). So we take the Γ as the Doppler linewidth $\Gamma_{2.7\,\mu\text{m}} = 1.75 \times 10^{-3}$ cm^{-1} and $\Gamma_{3.1\,\mu\text{m}} = 1.48 \times 10^{-3}$ cm^{-1} at an operating temperature of 643 K. According to the theory mentioned before, we expect that only the 2.7 μm SRS can be observed. It agrees with this experimental result.

From figure 7, we find that the forward 1.2 μm ASE (5S–4P) in the left channel 4S–(5P)–5S is almost completely suppressed. This suppression could always be observed in the range of laser detuning $\Delta = -5$–23.7 cm^{-1}. If the forward ASE (5S–4P) is suppressed, the forward 2.7 μm SRS 4S–(5P)–5S, as shown in figure 4, should also be suppressed. However, in this experiment it is difficult to determine how much forward SRS is suppressed by the intensity comparison between the forward and backward SRS. The reason is that the 2.7 μm (5P)–5S forward emission contains both SRS and FWM emissions. Because of the FWM processes listed in table 1, the forward 2.7 μm (5P)–5S emission is stronger than the backward one even if the 2.7 μm forward SRS is suppressed. In other words, both SRS and FWM emissions are

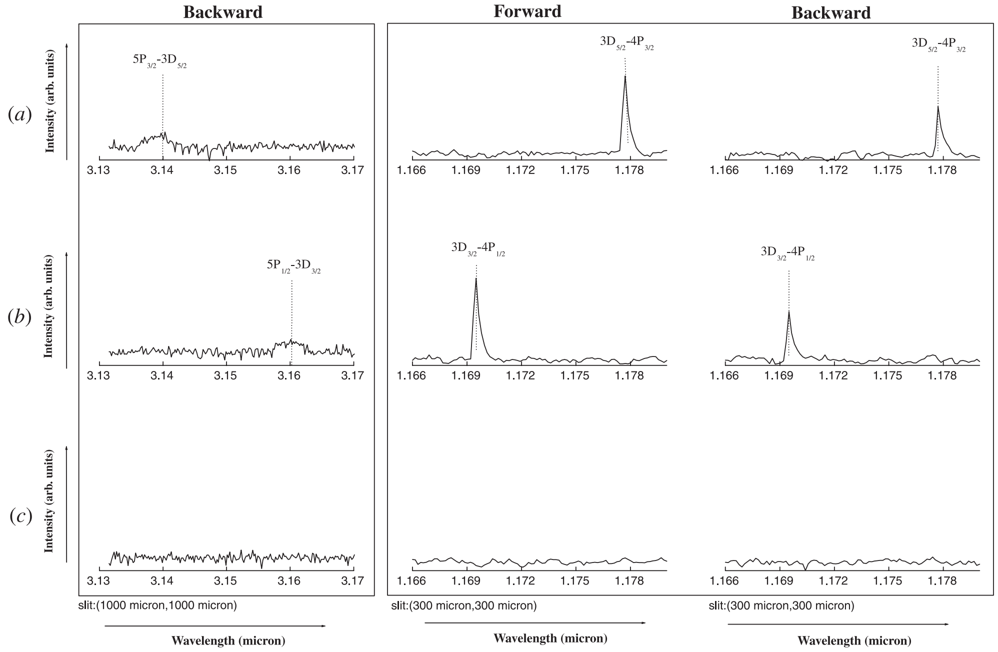

Figure 5. The 3.1 μm ASEs (5P–3D) in the backward direction and 1.1 μm ASEs (3D–4P) in both directions as the laser is tuned at (*a*) $\Delta = 5$ cm^{-1} (5 cm^{-1} below 5P$_{3/2}$); (*b*) $\Delta = 23.7$ cm^{-1} (5 cm^{-1} below 5P$_{1/2}$); (*c*) $\Delta = 28.7$ cm^{-1} (10 cm^{-1} below 5P$_{1/2}$). The broken lines indicate the resonant wavelengths. For the measurement of the 3.1 μm backward emissions, the sensitivity is five times higher than that in figure 4.

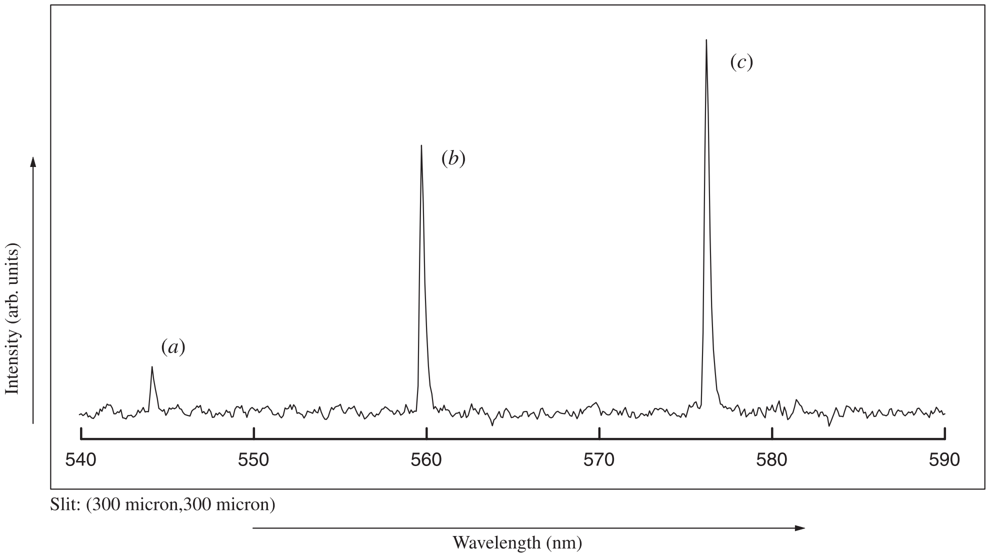

Figure 6. The spectra related to the FWM processes as the laser is tuned at $\Delta = 23.7$ cm^{-1}.

tunable and have the same wavelength, we cannot distinguish one from the other in the forward direction. So we cannot verify the SRS suppression quantitatively. Here the FWM processes

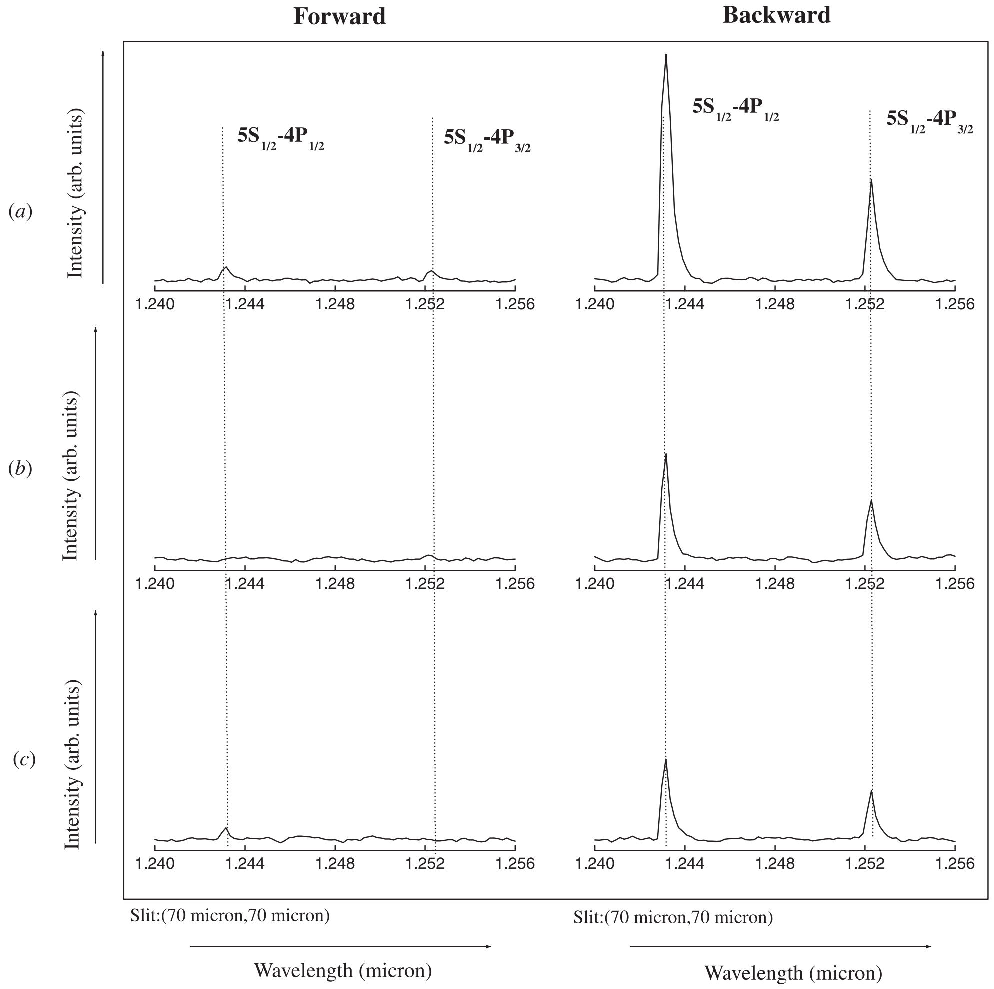

Figure 7. The ASEs (5S–4P) in the forward and backward directions as the laser is tuned at (*a*) $\Delta = -5\,\mathrm{cm}^{-1}$ ($5\,\mathrm{cm}^{-1}$ above $5P_{3/2}$); (*b*) $\Delta = 10\,\mathrm{cm}^{-1}$ ($10\,\mathrm{cm}^{-1}$ below $5P_{3/2}$); (*c*) $\Delta = 23.7\,\mathrm{cm}^{-1}$ ($5\ \mathrm{cm}^{-1}$ below $5P_{1/2}$). The broken lines indicate the resonant wavelengths.

do not satisfy the phase-matching condition, but they can still happen due to the large gain from the resonant enhancement (Zhang *et al* 1984). These FWM processes cannot cause SRS suppression because the forward SRS can only be suppressed by the FWM induced by SRS itself (Malakyan 1990).

4. Conclusion

In this experiment, we observed the SRS emissions for the two possible channels, 4S–(5P)–5S and 4S–(5P)–3D, for potassium vapour excited near 4S–5P resonance. The results show that only the 2.7 μm SRS (4S–(5P)–5S) emission could be observed in both directions. SRS only occurs in the 4S–(5P)–5S channel that agrees with the theory mentioned above. Since the 2.7 μm forward emission contains both the SRS and FWM emissions, we cannot estimate how much forward SRS emission is suppressed. However, we found the forward 1.2 μm ASEs

(5S–4P) to be almost completely suppressed in the 4S–(5P)–5S channel. This result proves that the suppression effect does exist in the forward 2.7 μm SRS.

For the 4S–(5P)–3D channel, in the backward direction we did not observe any detectable 3.1 μm SRS (4S–(5P)–3D), but did observe the 3.14 and 3.16 μm ASEs (5P–3D). We believe that these ASEs (5P–3D) populate the 3D state, which generates the 1.1 μm ASEs (3D–4P). The observed tunable forward 3.1 μm emission should come from the FWM processes and not from SRS.

Acknowledgments

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