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Continuous-wave solid-state two-Stokes Raman laser

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***Abstract*. We report an experimental study of the first cw solid-state Raman laser operating simultaneously at the frequencies of the first and second Stokes components. Simultaneous generation is ensured by a cavity with an enhanced finesse at both Stokes frequencies. The threshold pump powers for the first (3.4 W) and second (3.67 W) Stokes components suggest that the second Stokes generation follows a cascade mechanism. We demonstrate for the first time Raman conversion with intensity stability exceeding the pump radiation stability and show that this approach may find application in Raman spectroscopy.**

Keywords: *cw lasing, Raman laser, Stokes components, solid-state laser.*

Laser radiation conversion is among the most important applications of stimulated Raman scattering (SRS). As distinct from other nonlinear methods, Raman conversion allows simultaneous generation on several spectral lines at frequencies above or below the laser frequency and is capable of covering broad spectral regions with tunable or fixed frequencies [1 – 3]. Up to several tens of components can be generated [4]. All the results in question were obtained with pulsed laser pumping. Fundamental aspects of pulsed SRS were analysed in classic works by S.A. Akhmanov et al. [5, 6], who took into account the transient nature of the process and excitation nonmonochromaticity.

Raman conversion of cw laser radiation is usually considered a nontrivial problem because, to reach the SRS threshold, high power, up to hundreds of kilowatts, is needed, which is difficult to achieve with cw lasers. In recent years, significant advances have been made in cw Raman generation owing to the use of ultrahigh-finesse cavities [7] or high-Raman-gain solid media in combination with relatively high-finesse cavities [8, 9]. In those studies, major attention was paid to generation of the first Stokes component under various conditions and optimisation of the process. At the same time, generation at the frequencies of several Raman components is also of interest because multifrequency sources have many potential applications.

CW anti-Stokes emission from a Raman laser in hydrogen was demonstrated by Brasseur et al. [10]. Anti-Stokes Raman conversion, including that in Raman lasers [11], is usually thought of as a four-wave-mixing process. In contrast to anti-Stokes generation, the generation of higher order Stokes componenets may follow both parametric and cascade mechanisms. In the latter case, the fulfilment of phase matching conditions does not complicate the generation process and enables higher order Stokes generation in the Raman laser cavity even when the Raman medium has considerable dispersion. Significant progress in this direction has been made with Raman fibre lasers [12]. To our knowledge, cw lasing at two Stokes components in solid-state Raman lasers has not been reported in the literature.

In this paper, we present the first experimental results on cw generation at two Stokes wavelengths in a solid-state Raman laser pumped with the 514.5-nm argon-ion laser line. The approach we use to achieve generation takes advantage of scattered radiation accumulation in the Raman laser cavity at both the first and second Stokes wavelengths.

The experimental setup was described in detail elsewhere [8]. The pump source used was an argon-ion laser (Spectra Physics Model 2085), which ensured up to 10 W of output power at 514.5 nm. The width of its multimode lasing spectrum was about 0.1 cm^{-1}. The laser beam was similar in cross-sectional profile to the TEM_{00} mode.

The concentric Raman laser cavity was formed by two mirrors with a radius of curvature of 105 mm, spaced about 23 cm apart. The reflectivity of the input cavity mirror was 99.44 % at 543.8 nm (first Stokes component) and 98.74 % at 576.7 nm (second Stokes component), and its 514.5-nm transmission was 87.2 %. The reflectivity of the output mirror was 99.6 %, 99.94 % and 99.85 % at 576.7, 543.8 and 514.5 nm, respectively. The pump beam was mode-matched to the cavity by the focusing system.

The Raman medium used was a 68-mm-long barium nitrate crystal. Its faces had antireflection coatings for the pump and Stokes wavelengths in the range 490 – 580 nm. The choice of the medium was prompted by the high Raman gain of barium nitrate, 47 cm GW^{-1} at 532 nm [13]. Barium nitrate is an optically isotropic material with a cubic structure. Its broad transparency region (0.35 to 1.8 μm) enables stimulated Raman conversion in the visible and IR spectral regions. It has high optical quality. The frequency

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shift of the strongest Raman line of barium nitrate is 1047 cm^{-1}, and its spectral width is about 0.4 cm^{-1}. The output spectral characteristics of the Raman laser were analysed with an SPEX 1404 spectrometer equipped with a liquid-nitrogen-cooled CCD detector (Photometrics SDS 9-9000/CH270). The temporal behaviour of the output radiation was analysed using photodiodes and a Tektronix TDS 220 oscilloscope (100 MHz, 1 GS s^{-1}), and its power was measured with a Molectron laser power meter.

The onset of Raman generation was inferred from the emergence of lines at both Stokes frequencies in the emission spectrum. A typical generated spectrum is presented in Fig. 1. The frequency separation between the observed lines corresponds to the Raman shift in barium nitrate (1047 cm^{-1}).

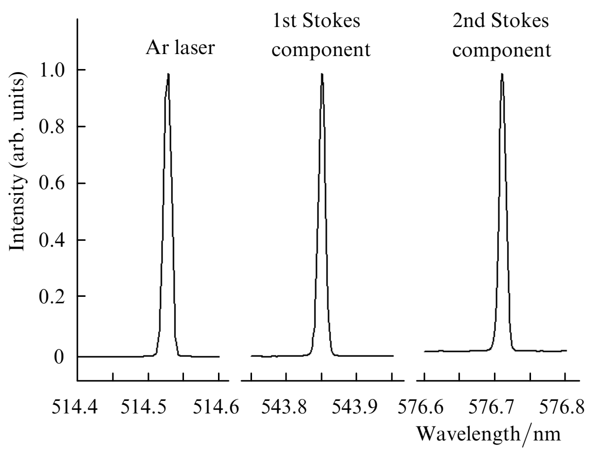

Figure 1. Emission spectrum of the Raman laser. The intensities of the spectral components are scaled to give a peak height of unity.

Figure 2 shows the output power as a function of pump power for the first (P_{st1}) and second (P_{st2}) Stokes components of the Raman laser. The threshold pump power for first Stokes generation is $P_p = 3.4$ W. At higher pump powers, P_{st1} gradually increases, with saturation at a level of 5–6 mW at $P_p = 6.5$ W. P_{st2} varies with P_p in a different way: generation begins at a pump power of 3.67 W, and P_{st2} reaches 21.7 mW with no saturation. According to our estimates, the powers of the first and second Stokes modes in the cavity are 20 and 11 W, which corresponds to intensities of 260 and 140 kW cm^{-2}.

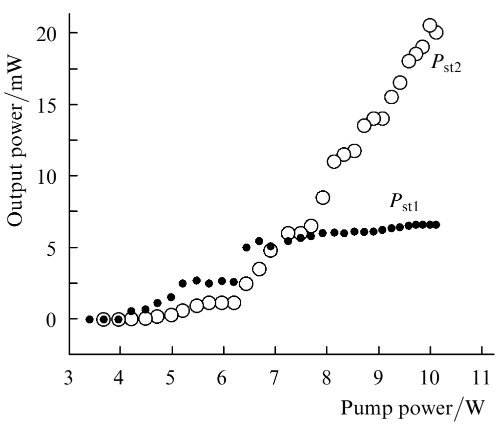

Figure 2. Output power as a function of pump power for the first and second Stokes components.

The well-resolved thresholds for first and second Stokes generation (3.4 and 3.67 W) and the lack of conical wave generation lead us to assume cascade Raman generation in our experiments. The first Stokes radiation then pumps the second. Accordingly, the threshold for second Stokes generation exceeds that for the first Stokes generation.

Figures 3 and 4 show oscilloscope traces illustrating the temporal behaviour of the two Stokes generations at the highest excitation power. The traces were obtained using a mechanical beam chopper. It is seen in Fig. 3 that, under the conditions of this study, the first Stokes signal differs in temporal behaviour from the pump signal. At a pump rise time of about 1 ms, the leading edge of the first Stokes signal is three orders of magnitude narrower (about 2 μs). The emergence of the scattered radiation signal is accompanied by strong relaxation oscillations, similar to those reported earlier for cw hydrogen [14] and barium nitrate [14] Raman lasers operating only on the first Stokes wavelength. The Stokes signal does not replicate characteristic pump fluctuations. The difference in temporal behaviour between the first and second Stokes signals can be understood in terms of the energy balance of the interacting waves, which may, in particular, lead to P_{st1} stabilisation at a level corresponding to the threshold for second Stokes generation. The ratio of the standard deviation in intensity to its average over ~ 100 μs, examined as a stability criterion, was 4 % for the argon-ion laser output beam and 2 % for the first Stokes output beam. These estimates were obtained for the last millisecond in the above oscilloscope traces.

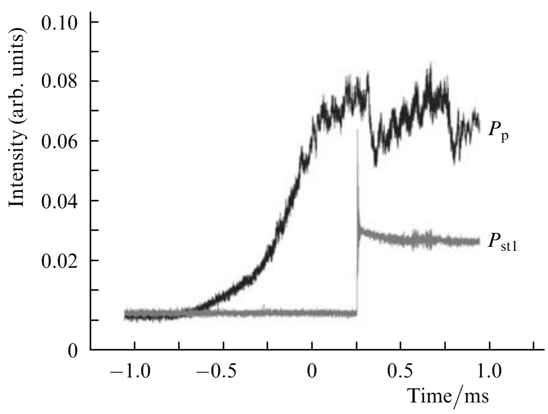

Figure 3. Oscilloscope traces illustrating typical temporal behaviour of the pump intensity and first Stokes intensity at the highest excitation power (10 W).

As distinct from those in Fig. 3, the oscilloscope traces of the first and second Stokes signals in Fig. 4 show simultaneous development of relaxation oscillations. The first Stokes signal rapidly reaches a steady level, while stabilisation of the second Stokes component takes more than 150 μs, which may be due to the higher cavity losses at the second Stokes frequency.

To assess the long-term stability of Raman laser parameters, the two Stokes beams and argon-ion laser beam were used to measure spontaneous Raman scattering spectra of hematin. This substance is known to be a weakly scattering medium, and a long-term signal accumulation is needed to obtain its Raman spectrum. Each of the spectra

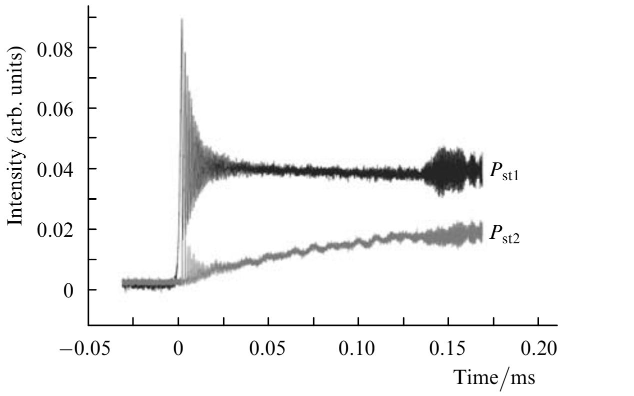

Figure 4. Oscilloscope traces illustrating typical temporal behaviour of the first and second Stokes intensities at the highest excitation power (10 W).

displayed in Fig. 5 was obtained via data acquisition for 4 h. The good quality of the spectra points to high stability of Raman generation parameters. Moreover, these results suggest that a Raman laser operating at two Raman wavelengths, in combination with an argon-ion laser, may provide additional spectroscopic information due to the possibility of tuning the excitation wavelength closer to resonance.

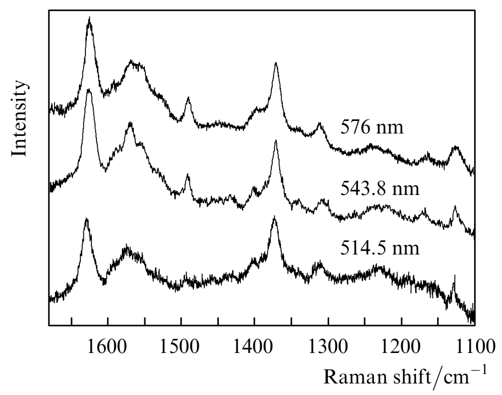

Figure 5. Raman spectra of hematin under Raman and argon-ion laser excitation.

Thus, we demonstrated the first cw solid-state Raman laser operating simultaneously at frequencies of two Stokes components. Simultaneous development of first and second Stokes generation enhances the stability of the first Stokes component. These results are, to out knowledge, the first to demonstrate the stability of Raman-converted radiation exceeding the pump radiation stability.

References

1. Frey R., Pradere F. *Opt. Commun.*, **12**, 98 (1974).
2. Wilke V., Schmidt W. *Appl. Phys.*, **17**, 477 (1979).
3. Apanasevich P.A., Batishche S.A., Ganzha V.A., Grabtchikov A.S., Kamach Yu.E., Kozlovskii Yu.N., Malevich N.A., Mostovnikov V.A., Ovchinnikov V.M., Orlovich V.A. *Izv. Akad. Nauk SSSR, Ser. Fiz.*, **47**, 1551 (1983).
4. Kawano H., Hirakawa Y., Imasaka T. *Appl. Phys. B*, **65**, 1 (1997).
5. Akhmanov S.A., Drabovich K.N., Sukhorukov A.P., Chirkin A.S. *Zh. Eksp. Teor. Fiz.*, **59**, 485 (1970).
6. Akhmanov S.A., D'yakov Yu.E., Pavlov L.I. *Zh. Eksp. Teor. Fiz.*, **66**, 520 (1974).
7. Brasseur J.K., Repasky K.S., Carlsten J.L. *Opt. Lett.*, **23**, 367 (1998).
8. Grabtchikov A.S., Lisinetskii V.A., Orlovich V.A., Schmitt M., Maksimenka R., Kiefer W. *Opt. Lett.*, **29**, 2524 (2004).
9. Demidovich A.A., Grabtchikov A.S., Lisinetskii V.A., Burakevich V.N., Orlovich V.A., Kiefer W. *Opt. Lett.*, **30**, 1701 (2005).
10. Brasseur J.K., Roos P.A., Carlsten J.L. *J. Opt. Soc. Am. B*, **17**, 1223 (2000).
11. Roos P.A., Meng L.S., Murphy S.K., Carlsten J.L. *J. Opt. Soc. Am. B*, **21**, 357 (2004).
12. Papernyi S., Karpov V., Clements W. *Optical Fiber Communication Conf., OSA Techn. Dig. Series* (Opt. Soc. Am., 2001) paper WDD15.
13. Karpukhin S.N., Stepanov A.I. *Kvantovaya Elektron.*, **13**, 1572 (1986) [*Sov. J. Quantum Electron.*, **16**, 1927 (1986)].
14. Brasseur J.K., Roos P.A., Repaky K.S., Carlsten J.L. *J. Opt. Soc. Am. B*, **16**, 1306 (1999).