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High average power generation in barium nitrate Raman laser

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Abstract The generation of low divergent ($M^2 \leq 1.5$) first and third Stokes radiation in a barium nitrate Raman laser with average powers of 11 W and 5 W, respectively, was demonstrated. The quantum conversion efficiency was up to 21%. The possibility of thermal lens compensation in such Raman lasers was shown.

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1 Introduction

Solid-state Raman converters are of interest lately [1–20]. They are compact and efficient and can convert femtosecond [8], picosecond [9, 10], and nanosecond [3–7, 12–17] laser pulses as well as a continuous-wave [11] laser radiation, allowing one to obtain radiation at wavelengths, where usual laser generation is not available or inefficient.

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Special attention is devoted to solid-state Raman lasers in which a solid-state Raman medium is placed in an optical cavity. The use of the cavity allows for a decreasing Raman generation threshold and divergence of converted radiation. Raman lasers can be pumped with radiation from the picosecond to the continuous-wave time scale. Most often these lasers are used for conversion of nanosecond laser pulses. Efficient generation of solid-state Raman lasers at nanosecond pumping was demonstrated for pump pulse energy from microjoule [16] level to hundreds of millijoules [13–15].

For some applications a high average power of converted radiation is required. Raman lasers allow for obtaining Stokes pulses with an energy higher than a hundred of millijoules [12, 14], while there is a problem to obtain radiation with a high average power. The problem is related to the heating of a Raman crystal during Raman conversion. Indeed, in the process of Raman conversion in each act of scattering one pump radiation photon is transformed to one first Stokes photon and one Raman vibration mode phonon:

$$h\nu_P = h\nu_S + h\nu_R, \tag{1}$$

where h is the Planck constant, ν_P, ν_S, ν_R are the frequencies of pump radiation, first Stokes radiation and of Raman vibration.

Expression (1) means that a part of pump energy is transferred to the energy of the excited vibration mode, which in its turn is converted to heat due to dissipation. The heating of the Raman medium will cause the thermal lens formation, because of the nonuniformity of the heating and cooling processes. The first Stokes generation strength (D_T) of the thermal lens can be estimated as [21]:

$$D_T = \frac{1}{f_T} = \left(\frac{dn}{dT}\right)\frac{1}{k_C}\frac{P_S}{\pi r_S^2}\frac{\nu_R}{\nu_S} \tag{2}$$

where f_T is the focal length of the thermal lens, k_C is the thermal conductivity of the Raman crystal, dn/dT is the thermo-optic coefficient, P_S is the average power of the first Stokes radiation, r_S is the first Stokes beam radius in the Raman crystal. In the expression (2) the crystal is assumed to be in a perfect thermal contact with a temperature-controlled heat sink.

Clearly, the heating of a Raman medium is small and its effect on Raman generation is negligible, if the average power of the converted radiation is small. However, when high average power Stokes radiation is generated the effect of heating can be substantial. Especially strong heating is observed for the IR spectral region. Indeed the ratio of energy converted to heat and first Stokes energy is equal to the frequency ratio ν_R/ν_S (see expression (2)); meanwhile the ratio is higher for longer pump wavelengths. In the same way for the cascade high order Stokes generation the heating is higher than for the first Stokes generation, because heat is emitted in each step of Raman conversion.

A thermal lens arising in a Raman crystal can make a cavity of a Raman laser unstable, which results in an increase of intracavity losses with a subsequent decrease of generation efficiency. This means that a thermal lens has a profound effect on Raman generation, if a cavity is on the stability limit and can easily get unstable. For instance [15], in a plane-plane cavity with a barium nitrate Raman crystal (vibration frequency equals to 1047 cm^{-1}) a strong effect of a thermal lens on Raman generation was observed at an average output power of only 0.5 W for the first (wavelength is 1.197 μm) and the second (wavelength is 1.369 μm) Stokes and 0.17 W for the third Stokes (wavelength is 1.599 μm). A pump (wavelength is 1.064 μm) average power was less than 3 W. It was found that the negative thermal lens in the crystal made the cavity of the Raman laser unstable, which resulted in drops for the first, second and third Stokes output power equal to 7%, 46% and 76%, respectively.

The use of a stable cavity [13, 21, 22], the stability of which is not so sensitive to negative thermal lenses, allows for generating radiation in barium nitrate with sufficiently higher average power. Third Stokes generation with an average power of 1.8 W [13] was generated in barium nitrate at a pump power of 6 W (wavelength is 1.064 μm), when a spherical-plane stable cavity (curvature radius of input mirror was 1500 mm, cavity length was 150 mm) was used. The focal length of the thermal lens formed in the crystal was estimated to be several meters and no sufficient effect on the third Stokes generation was found.

The low strength of the thermal lens in [13] was due to the fact that the Raman laser was pumped with pulses of high energy (300 mJ) and low repetition rate (20 Hz). This allowed for using a large pump beam (diameter was 4 mm), while the strength of a thermal lens decreases when the beam diameter increases (see expression (2)). When the pump radiation is focused strongly in a Raman crystal the focal length of a thermal lens is significantly shorter than that in [13] even at lower output powers. For instance, thermal lenses with focal lengths of about 10 cm were reported for strong focusing of pump radiation (beam diameter was 0.2 mm) in Raman crystals (barium nitrate, potassium gadolinium tungstate, and calcium tungstate were investigated) at a first Stokes output power of only 1 W [21, 23].

The necessity of such strong focusing arises from the fact that the main part of high average power lasers produces radiation pulses with a high repetition rate and a low energy. Small pump beam diameters are required to reach the Raman threshold and achieve efficient generation using the radiation of these lasers. On the one hand, small pump beam diameters ensure small Fresnel numbers resulting in low divergence of generated radiation, as compared to those obtained with a wide pump beam (for example in [13] the M^2 factor of the generated radiation was about 15). On the other hand, small beam diameters result in a high strength of the thermal lens (see (2)), which can lead to a decrease of generation efficiency and to an increase of the divergence of the output radiation. So a proper choice of pump radiation parameters as well as parameters of the Raman cavity is required to obtain a high average power Raman conversion with an appropriate generation efficiency and a low divergence of output Stokes radiation.

In this article Raman generation from a radiation of average power up to 60 W is investigated to our knowledge for the first time. The pump pulse repetition rate of 1.1 kHz and pulse energy of 50 mJ are chosen. A simple method of thermal lens compensation is demonstrated experimentally providing high average power Stokes generation with low divergence and an efficiency of more than 20%.

2 Description of the experiment

The experimental setup is shown in Fig. 1. As a pump source an actively Q-switched single-mode (the M^2 factor was close to 1) Nd:YAG laser system consisting of an oscillator and two amplifiers (wavelength is 1.064 μm) was used. The system was flash lamp pumped with a repetition rate of 100 Hz, producing a burst of 11 pulses per one shot (average repetition rate was 1.1 kHz). The duration of each pulse was about 50 ns, the distance between pulses was 40 μs. Figure 2a demonstrates an oscilloscope trace of one burst, an oscilloscope trace of a separate pulse is shown in Fig. 2b. The spectral width of the pump radiation was 0.02–0.03 cm^{-1}. It was measured by the use of a Fabry-Pérot interferometer and a CCD-camera.

The pump pulse energy incident in the Raman laser was controlled by an optical system consisting of a half-wave phase plate and a polarizer. This system allows changing the

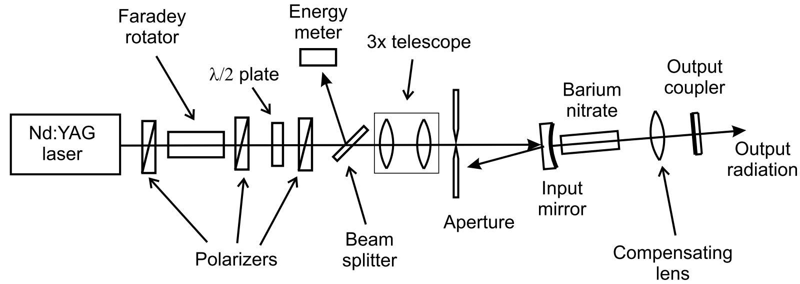

Fig. 1 Experimental setup

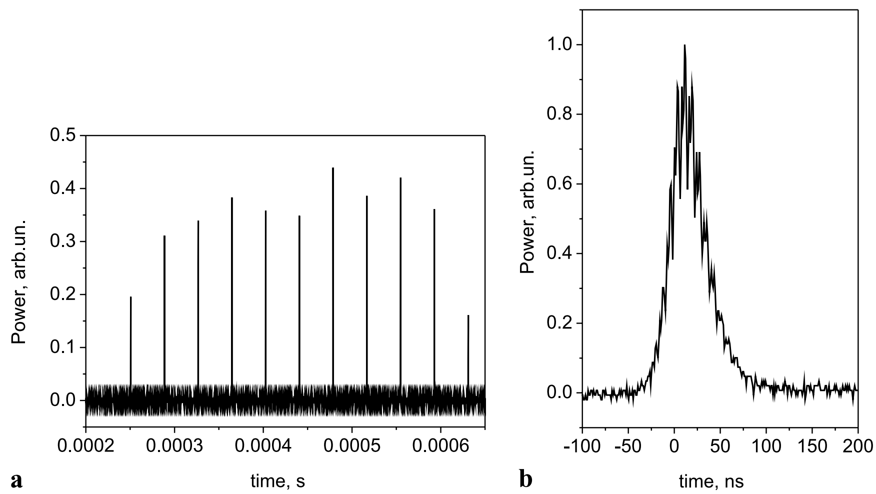

Fig. 2 Oscilloscope traces of the pump burst (**a**) and of a separate pulse in the burst (**b**)

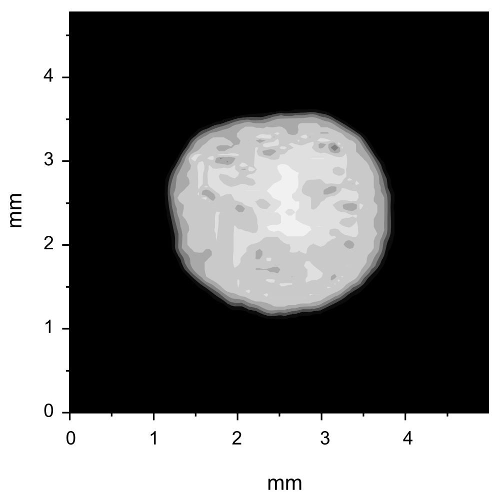

Fig. 3 Spatial distribution of pump radiation detected with black photopaper

pump energy continuously without any changes in the pump beam and pulse shape. The beam splitter directed a part of the pump radiation to a calibrated energy meter to measure the pump energy. The pump beam diameter incident to the Raman laser was decreased down to 2.5 mm with a telescope formed of 400 mm and 150 mm convex lenses. The pump beam profile at the entrance of Raman cavity is shown in Fig. 3.

The Raman laser investigated in our experiments consisted of a 7 cm long barium nitrate crystal placed in a cavity. Crystal facets were anti-reflection coated in the spectral region of 1–1.6 μm. No special cooling was used for the crystal. Our previous measurements showed that the crystal placement in an aluminum heat sink causes a stronger thermal lens and even the destruction of the crystal. The temperature of the crystal without special cooling was evidently higher, but the temperature gradient as well as the strength of the thermal lens was lower.

The cavity of the Raman laser was similar to the one described in [13] and was formed of a spherical input mirror (curvature radius of 1500 mm) and a flat output coupler. The input mirror transmitted highly the pump radiation and reflected the first (1.197 μm), the second (1.369 μm), and the third (1.599 μm) Stokes radiations. We used two different output couplers for the first and the third Stokes generation. The first one (OC1) reflected highly the pump radiation and 34% at the first Stokes wavelength, while the second one (OC2) reflected highly the pump, the first, and the second Stokes radiation and reflected 50% at the third Stokes wavelength. For all output couplers the cavity length of the Raman laser was 18 cm. A Faraday isolator was used to avoid feedback between the pump laser system and the Raman laser. In addition the cavity of the Raman laser was tilted at a small angle (about 5 degrees) too. An aperture was used to block the depleted pump radiation reflected from the Raman laser in a backward direction.

The power of the pump radiation and the converted radiation was measured with a Gentec PS-150 power meter. Oscilloscope traces were registered with a two channel digital real-time oscilloscope LECROY LT 364 connected to fast photodiodes for IR measurements. Beam profiles were registered with a photopaper and then scanned.

3 First Stokes generation

Firstly, the Raman laser with the OC1 output coupler was investigated. In the laser the generation of first Stokes radiation was obtained. The dependence of the average output power of the first Stokes on the pump average power is presented in Fig. 4a. No significant second Stokes generation was observed. The first Stokes average output power was about 8 W at a pump power of 60 W. It corresponds to a quantum conversion efficiency of more than 14% (Fig. 4b demonstrates the dependence of the quantum efficiency on the pump average power). The high average output power of the first Stokes was accompanied with a high divergence and M^2 factor of the first Stokes beam. The divergence was about 11 mrad, which corresponds to a M^2 factor of about 25. The far-field angle distribution of the first Stokes beam is seen in Fig. 5a.

The high divergence of the first Stokes beam was caused by a negative thermal lens formed in the Raman crystal. The action of the thermal lens in a Raman active crystal can be described as an additional thick lens arisen in a cavity. The cavity with the thick lens is equivalent to an empty cavity with equivalent g-parameters (g_i^*) [24]:

$$g_i^* = g_i - D_T \cdot d_i \left(1 - \frac{d_i}{r_i}\right), \tag{3}$$

where g_1 and g_2 are the g-parameters of the cavity without the thermal lens:

$$g_i = 1 - \frac{d_1 + d_2}{r_i}, \tag{4}$$

$$d_i = x_i - \frac{L_{CR}}{2}\frac{n-1}{n}, \tag{5}$$

where r_1 and r_2 are the curvature radiuses of input and output mirrors, d_1 and d_2 are the distances of the principal planes of the thermal lens to the mirrors, x_1 and x_2 are the distances from the crystal center to the input and output mirrors, L_{CR} is the Raman crystal length, n is the refraction index of the crystal.

Expression (3) shows that the thermal lens changes the g-parameters of the Raman laser cavity. The condition of cavity stability is

$$0 < g_1 g_2 < 1. \tag{6}$$

A thermal lens changing the g-parameters of a cavity may make the cavity unstable. For example, a plane-plane cavity ($g_1 = g_2 = 1$) gets unstable even at slight negative thermal lens ($f_T < 0$), which is in a good agreement with results presented in [15]. The stable cavity used in our experiment ($r_1 = 1500$ mm, $r_2 = \infty$, $L_{CR} = 7$ cm, $n = 1.55$ [25], $x_1 = 6$ cm, $x_2 = 12$ cm, $g_1 = 0.897$, $g_2 = 1$) was less sensitive to a thermal lens. However, at a strong negative thermal lens the cavity can also get unstable. Expressions (3) and (6) show that the cavity gets unstable when the focal length of

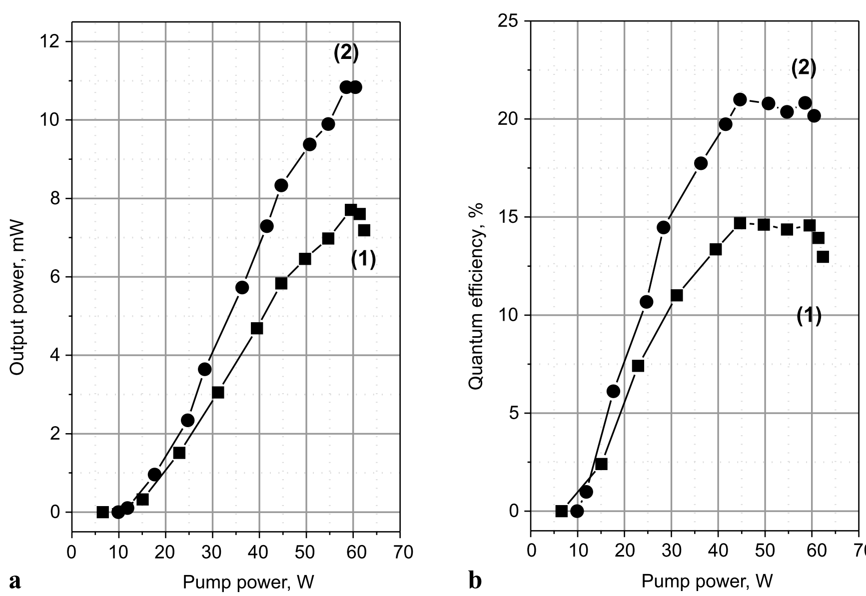

Fig. 4 Dependencies of the first Stokes average power (**a**) and quantum efficiency (**b**) of generation on the pump average power. 1—The cavity without addition lens, 2—the cavity contains additional lens with focal lengths of 500 mm

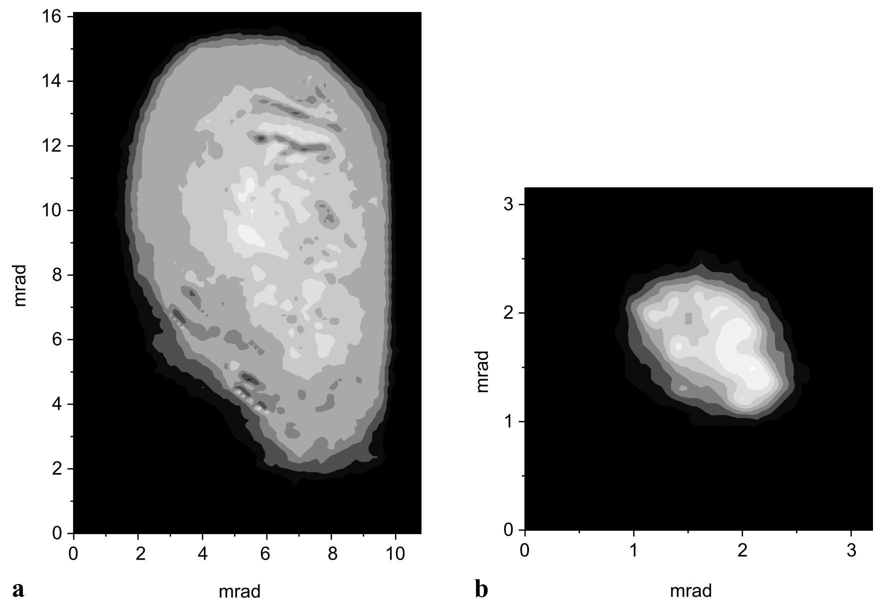

Fig. 5 Far-field angle distribution of the first Stokes output radiation for cavity without compensation of a thermal lens (**a**) and with compensation (**b**)

the negative thermal lens is shorter than 1.4 m (the strength is $-0.7\ \text{m}^{-1}$). In experiments the cavity got unstable at a first Stokes output power of about 6 W (it was observed by a sharp increase of divergence). This means that the thermal lens strength of $0.7\ \text{m}^{-1}$ occurred at the first Stokes power of 6 W, while expression (2) shows that a power of 4 W is already sufficient for this value of the thermal lens strength. The mismatch of the estimated and experimental values of the first Stokes power is probably because expression (2) was derived under the assumption of perfect cooling, while in the experiments no special cooling was used. Secondly, the expression (2) does not take into account the effect of gain focusing [26], which can partially compensate for the thermal lens effect.

When the cavity gets unstable additional geometrical losses (L_{geom}) arise [24]:

$$L_{\text{geom}} = 1 - \frac{1}{M_A^2}, \tag{7}$$

where M_A is the cavity magnification:

$$M_A = \left|2g_1^* g_2^* - 1\right| + \sqrt{\left(2g_1^* g_2^* - 1\right)^2 - 1}. \tag{8}$$

Assuming that the thermal lens strength is proportional to the first Stokes power and taking into account that it was $-0.7\ \text{m}^{-1}$ at the power of 6 W we estimated the thermal lens strength at the first Stokes power of 8 W to be $-0.9\ \text{m}^{-1}$. Using the expressions (3), (7) and (8) we found that the magnification was equal to 1.4, while the geometrical losses were equal to 50%. The high geometrical losses significantly decreased the feedback induced with the output coupler, which possesses a low reflectivity as it is (34%). As a result, the effect of the cavity on the Raman generation was weak and single-pass generation occurred. The Fresnel number [27] for the generation was about 60, which led to the high value of M^2 factor of the output radiation.

The additional geometrical losses besides the increase of the M^2 factor decreased also the Raman generation efficiency. In order to decrease the output beam M^2 factor and to increase the generation efficiency it is necessary to keep the cavity stable regardless of the thermal lens. To compensate for the action of the negative thermal lens an additional focusing lens was placed inside the Raman laser cavity between the output coupler and the output facet of crystal. It is easy to show using the ABCD technique [24] that the combination of the lens and the flat output coupler is equivalent to a spherical mirror with the curvature radius (r_2^*) placed at the distance of (x_2^*) from the crystal center:

$$r_2^* = \frac{f^2}{f - x_3}, \tag{9}$$

$$x_2^* = x_2 + \frac{x_3^2}{f - x_3}, \tag{10}$$

where f is the focusing length of the additional lens, x_3 is the distance between the focusing lens and the output coupler.

The focal length of the additional lens was 500 mm; the facets of the lens were antireflection coated at 1.064 µm wavelength. The lens was placed at a distance of 60 mm from the output coupler. It is seen from (9) and (10) that the lens is equivalent to an output coupler with the curvature radius of 570 mm located at the distance of 12.8 cm from the crystal centre. The lens keeps the cavity stable until the strength of the negative thermal lens is lower than $2.9\ \text{m}^{-1}$. The M^2 factor of the first Stokes radiation generated in the

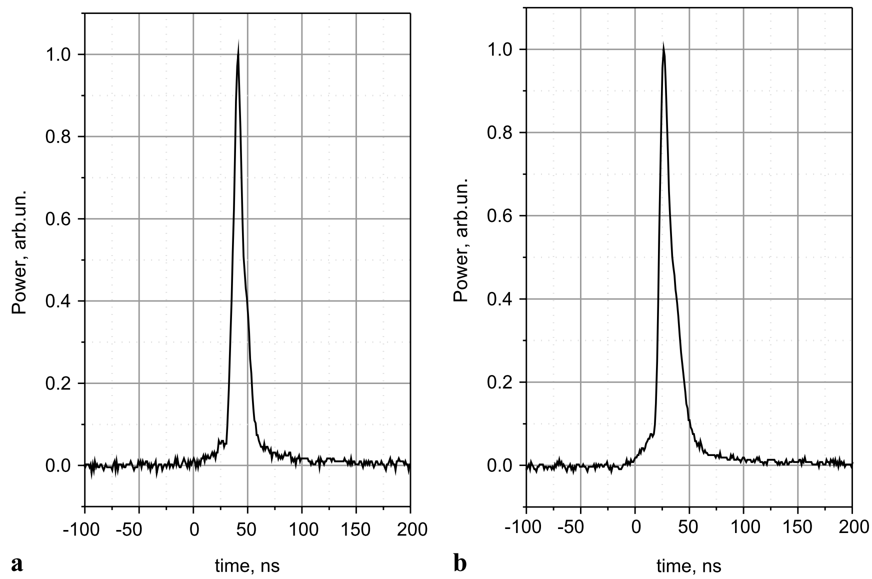

Fig. 6 Oscilloscope traces of the first (**a**) and the third Stokes (**b**) pulses

cavity was 1–1.5 even at the highest pump power (60 W) used in the experiment. The divergence was about 1.7 mrad.

Besides the decrease of the M^2 factor the compensation of the thermal lens led to an increase of the output average power and the generation efficiency. The dependencies of the first Stokes output power and the quantum efficiency on the pump average power are seen in Fig. 4. The first Stokes average power was 11 W at the pump power of 60 W, which corresponds to the quantum efficiency of 21%. All pulses in the pump train (see Fig. 2a) were converted to first Stokes radiation. The duration of a separate first Stokes pulse depended on the pump power and was about 11 ns at the maximal pump power. It means that the first Stokes peak power (0.9 MW) was close to the pump peak power (1.1 W). A typical oscilloscope trace of a first Stokes pulse is presented in Fig. 6a.

Thus the use of the compensation lens kept the cavity of the Raman laser stable at first Stokes generation. It allowed for generating the radiation with low M^2 factor and the highest average power, to our knowledge.

4 Third Stokes generation

Third Stokes generation was also obtained and investigated experimentally. The output coupler OC2 was used in the Raman laser for this purpose. Firstly the third Stokes generation was investigated in the cavity without a compensating lens. Similar to the first Stokes generation a strong effect of the thermal lens on the Raman generation was observed. The beam divergence of the third Stokes radiation was high (10 mrad) as well as the M^2 factor (10). The angle distribution of the output radiation is presented in Fig. 7a. In addition, in contrast to the first Stokes generation, the thermal lens adverse effect on the output average power and the generation efficiency was dramatic. Figure 8 (curve 1) shows the dependence of the average output power and the quantum generation efficiency on the pump average power. Unfortunately the first and second Stokes powers were not measured because of the high reflectivity of the output coupler at these wavelengths. Figure 8 demonstrates that the third Stokes efficiency and the average output power were very low. The output power was only 0.36 W at the pump power of 33 W, which corresponds to a quantum efficiency of 1%. A further increase of the pump power up to 38 W led to a decrease of the third Stokes average power down to 0.21 W.

Such a strong effect of the thermal lens on the output power and the efficiency is due to the fact that at the given output power the thermal lens arising at third Stokes generation is stronger (the quantum defect and thereafter the heating is three times higher) than that for first Stokes generation. Moreover, the negative effect of the instability of the cavity on third Stokes generation is very strong. It is well known [7, 12] that the cascade third Stokes generation requires low losses at first and second Stokes wavelength. Additional losses arising in an unstable cavity (see expression (7)) decrease dramatically the third Stokes output power and efficiency, in contrast to first Stokes generation, that can be generated quite efficiently in an unstable cavity [4].

We inserted an additional compensating lens in order to maintain the cavity stable and to obtain efficient generation. Focusing lenses with the focal lengths of 1000 mm, 250 mm, 200 mm, and 125 mm were used. These lenses keep the cavity stable if the focal length of the thermal lens is shorter than 570 mm, 190 mm, 150 mm, and 88 mm respectively. Dependencies of the output average powers and the quantum efficiencies on the pump average power when the com-

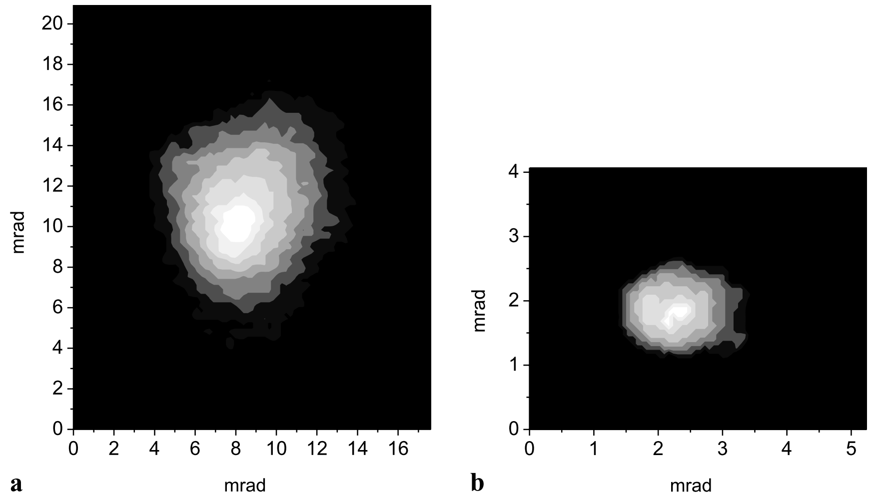

Fig. 7 Far-field angle distribution of the third Stokes output radiation for the cavity without compensation of a thermal lens (**a**) and with compensation (**b**)

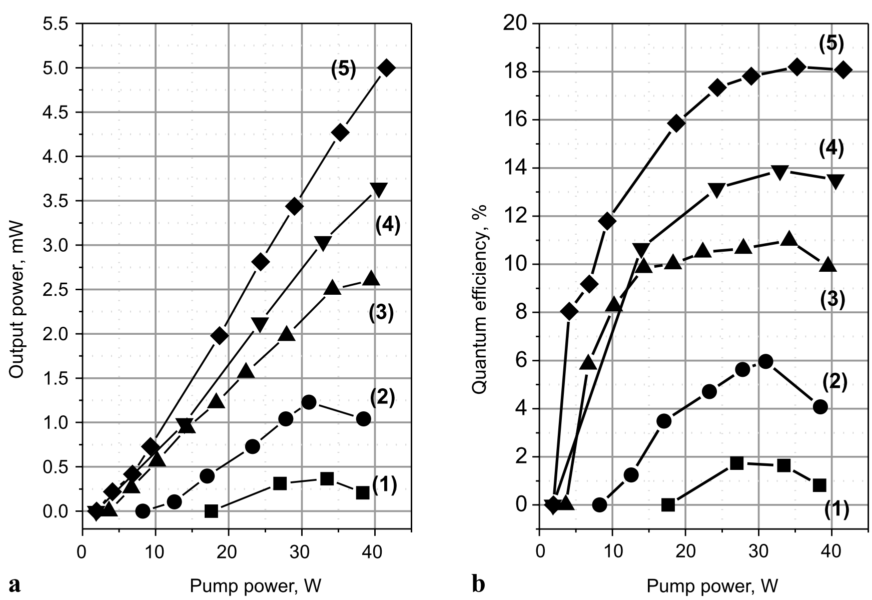

Fig. 8 Dependencies of the third Stokes average power (**a**) and quantum efficiency (**b**) of generation on the pump average power. 1—The cavity without addition lens, 2–5—the cavity contains additional lenses with focal lengths of 1000 mm (2), 250 mm (3), 200 mm (4), and 125 mm (5)

pensating lenses were used are presented in Fig. 8. It is seen that the lenses allow for obtaining sufficiently more effective third Stokes generation than before.

For the lenses with focal lengths of 1000 mm, 250 mm, and 200 mm at a certain third Stokes output power the cavity got unstable, which resulted in a decrease of the quantum efficiency and in an increase of the divergence. This means that the strength of the thermal lens, which depends on the third Stokes power, is high enough to get the cavity unstable in spite of the compensating lens. The use of a stronger additional lens increases the strength of the thermal lens required to get the cavity unstable, increasing also the third Stokes power that can be obtained.

The highest third Stokes output power (5 W) was obtained when the focusing lens with the focal length of 125 mm was used. The pump power was 40 W, the quantum efficiency was 18%. The M^2 factor was 1–1.5 at all pump powers (the divergence was about 1.8 mrad, the far-field angle distribution is seen in Fig. 7b). It means that the cavity was stable even at a pump power of 40 W. However, further increase of the pump power led to the destruction of the barium nitrate crystal due to overheating.

As well as for the case when the first Stokes was generated, all pulses in the pump burst (see Fig. 2a) were converted to third Stokes radiation. The duration of a separate third Stokes pulse in the burst depended on the pump power and was 12 ns at the maximal power. A typical oscilloscope trace of the pulse is shown in Fig. 6b. The pulse duration of 12 ns means that the third Stokes peak power was 0.4 MW, which corresponds to a conversion of the pump peak power (0.7 MW) to the third Stokes peak power of about 50%.

Thus the thermal lens at third Stokes generation is sufficiently stronger than that for the first Stokes generation, and its adverse effect on Raman generation can be dramatic. The use of an additional lens allows for compensating the thermal lens effect and increasing the output average power and the efficiency as well as decreasing the M^2 factor. Spherical mirrors with appropriate curvature radiuses can be used instead of the combinations of the additional focusing lenses and the output coupler used in our experiments. However, as is seen from the previous discussion, the strength of a thermal lens depends on many generation parameters (in particular, on the average power of the radiation generated, which in its turn depends on the thermal lens strength). Thus it is very difficult to predict the value of the thermal lens strength and to choose mirrors with appropriate curvature radius. The insertion of additional lenses allows for using only one pair of cavity mirrors and set of typical lenses, and thus is more flexible.

5 Conclusion

High average power generation of the first and third Stokes radiation in the barium nitrate Raman laser was obtained and investigated. The possibility of compensation of the negative thermal lens arisen in barium nitrate was demonstrated. First and third Stokes radiation with the average output powers of 11 W and 5 W were generated as a result of the thermal lens compensation with the M^2 factor of about 1–1.5. The quantum efficiencies were 21% and 18%, respectively. The efficiencies can be increased by further optimization of mirror reflectivities and by the use of compensating lenses with better AR coatings in order to decrease losses at Stokes wavelengths.

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