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| 14. ABSTRACT<br><br>CW fiber lasers pumped by laser diodes (LD) become the most reliable powerful systems for the wavelengths around 1.5 micron (Er-) and 1 micron (Yb-doped fiber lasers - YFL [1]) as well as for tunable 1.1-1.7 micron radiation (Raman fiber lasers - RFLs [2]). The main advantages of fiber lasers compared to conventional ones are obvious: efficient heat removal and high beam quality due to specific fiber geometry (thin and long fiber core in silica cladding), efficient and reliable LD pump injection, compact design of fiber bobbin, direct fiber delivery and hands-free operation due to special cavity mirrors - fiber Bragg gratings (FBGs), imprinted directly into the fiber. Recently 10 kW CW YFL system appears on the market (ILR-10000 by IPG Photonics Corp., USA). In spite of the outstanding advances in the IR region, the fiber lasers have not been yet applied for the visible and UV spectral range, where powerful ion lasers and DPSS (diode pumped solid state) lasers with frequency doubling remain to be the main laser sources at fixed lines and Ti:Sapphire and dye lasers as high-power tunable sources. |                       |                                |                                     |                                                                |                                                                 |
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# TECHNICAL REPORT

**PROJECT TITLE:** Frequency Doubling in Raman Fiber Lasers

**CRDF PROJECT #:** RUP1-1509-NO-05

**PROJECT DIRECTOR:** Sergey Babin

**PRINCIPAL  
ORGANIZATION:** Institute of Automation and Electrometry, SB RAS

**ADDRESS:** Ac. Koptug Ave. 1, Novosibirsk 630090, Russia

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## NOMENCLATURE

|      |                                         |
|------|-----------------------------------------|
| RFL  | Raman fiber laser                       |
| FWM  | four wave mixing                        |
| FBG  | fiber Bragg grating                     |
| YDFL | Ytterbium doped fiber laser             |
| CW   | continuous wave                         |
| SFM  | sum frequency mixing                    |
| SH   | second harmonic                         |
| PPLN | periodically poled lithium niobate      |
| PDT  | photo-dynamic therapy                   |
| IR   | infrared                                |
| SHB  | spectral hole burning                   |
| CARS | coherent anti-Stokes Raman spectroscopy |
| SBS  | stimulated Brillouin scattering         |
| LD   | laser diode                             |
| HR   | highly reflective                       |
| HT   | highly transmissive                     |
| Q    | quality factor                          |
| SMF  | single mode fiber                       |
| FWHM | full width at half maximum              |
| QPM  | quasi-phase matching                    |
| SHG  | second harmonic generation              |

## SUMMARY

The main goal of the project is to study basic physical mechanisms leading to spectral broadening in Raman fiber lasers, thus finding a way to control their output spectrum and make its frequency doubling efficient. The main goal for the 1<sup>st</sup> year of the project realization was to clarify the role of nonlinear optical effects in spectral broadening of phosphosilicate Raman fiber laser (RFL) output radiation. It has been shown that four-wave mixing (FWM) is the main reason of the broadening. Moreover, a FWM-induced stochasticity of the amplitude and the phase of each of  $\sim 10^6$  interacting longitudinal modes generated in a high-Q RFL cavity, results in specific hyperbolic secant shape of the spectrum and square-root power dependence of the spectral width. Such broadening is shown to be a unique example of a light wave turbulence in a fiber.

The main goal of the 2<sup>nd</sup> year of the project is to propose and realize techniques of driving the RFL spectral characteristics based on the developed model describing the RFL spectrum and obtained experimental results. Namely, to find opportunities to control (e.g. to narrow) the output spectrum by use of narrow-band cavity fiber Bragg gratings (FBGs) and to develop high-power CW RFL with tunable output at pumping it by a high-power CW Ytterbium-doped fiber laser (YDFL) for the sake of frequency doubling.

In the 3<sup>rd</sup> year of the project, the work on a frequency doubling of the tunable RFL radiation generated around 1.3 micron has been performed. Physical mechanisms of the influence of the RFL spectral broadening on the frequency doubling efficiency in PPLN crystal have been studied. It has been discovered that even at significant spectral broadening (order of magnitude larger than an acceptance width of the crystal) the second harmonics (SH) output may increase linearly with increasing fundamental power. In the reporting period a model has been proposed that explains this phenomenon. The model takes into consideration sum-frequency mixing (SFM) processes inside the broad RFL spectrum. It has been shown that the SFM processes give the main contribution to the SH power, while the direct frequency doubling gives a vanishing value at large number of modes with random phases. Thus, efficient SH generation is possible for broadband Raman fiber lasers, which spectral width is sufficiently larger than the acceptance bandwidth of the crystal. In low-power domain the SH efficiency for multiple random modes appears even higher than that for a single mode of equal power. The developed SFM model describes quite well the SH spectrum and power measured at frequency doubling of 1.31- $\mu$ m Raman fiber laser. Laser radiation of >60 mW at 655 nm for about 7W RFL power has been generated.

## INTRODUCTION

It is well known, that in Raman fiber lasers having long (hundreds of meters) fiber cavity a significant spectral broadening occurs. As a result, the obtained efficiency of frequency doubling in RFLs is very low [1]. If high efficiency may be possible, tunable lasers operating in the yellow-red spectral range based on frequency-doubled RFLs will be very competitive due to their ultimate reliability and may replace the existing lasers, especially dye lasers in many applications such as laser spectroscopy, super-continuum generation, laser cooling and trapping, medicine (PDT: photo-dynamic therapy), astronomy (laser guide star source), printing, flow cytometry etc.

The main goal of the project is to study basic physical mechanisms leading to spectral broadening in Raman fiber lasers, thus finding a way to describe and to control their output

spectrum, more specifically, to develop configurations with unique spectral features suitable for efficient frequency doubling. For experimental realization of tunable source in near-IR we explore a RFL based on a  $\text{P}_2\text{O}_5$ -doped silica fiber [2]. The main advantage of the phosphosilicate fibers compared to conventional germanosilicate ( $\text{GeO}_2/\text{SiO}_2$ ) ones is 3 times larger Stokes shift, therefore is possible to convert powerful Yb-doped fiber laser (YDFL) pump radiation at wavelength 1.06-1.08 micron into 1.2-1.3 micron region in one step instead of 2-3 steps for germanosilicate fibers.

In the 1<sup>st</sup> and 2<sup>nd</sup> years of the project realization we studied physical mechanisms leading to spectral broadening in RFLs. First, we tested the Raman gain spectral profile saturation and spectral hole burning (SHB) effects at high pump and Stokes wave powers in RFL cavity by CARS technique and concluded that the Raman gain spectral profile is saturated homogeneously and no hole burning occurs even at high Stokes powers (several Watts) [3]. After that we studied the role of nonlinear effects, such as stimulated Brillouin scattering (SBS) and four-wave mixing (FWM). It has been shown experimentally, that SBS has no influence on the RFL spectral broadening in contrast to hypotheses existing in the literature. The role of four-wave mixing (FWM) has been studied both theoretically and experimentally [4, 5]. A theoretical model based on the wave kinetic equations applied earlier for description of a weak wave turbulence [6] has been developed and its comparison with the experiment for RFL with high-Q cavity has been made. The shape and power dependence of the intra-cavity Stokes wave spectrum appear in excellent quantitative agreement with predictions of the theory at various power levels. It is shown that FWM-induced stochasticity of the amplitude and the phase of each of  $\sim 10^6$  longitudinal modes generated in RFL cavity simultaneously is a unique example of a light wave turbulence in a fiber. At the same time, the results show that the spectral shape of the Stokes wave is defined by the nonlinearity, dispersion and effective FBGs losses profile. In case of the Gaussian FBG reflection spectrum, the intra-cavity TFL spectrum in high-Q cavity is shown to have hyperbolic secant shape having wide exponential tails that is undesirable at frequency doubling. Anyway, there are some possibilities of spectral shaping that is important for efficient frequency doubling. The model has been applied to the real RFL with low-Q cavity, its comparison with the experiment has been performed and output spectrum has been described and optimized [7]. It is also interesting to combine the doubling with frequency tuning. For that a tunable RFL with tuning range of about 50 nm around 1.3 micron has been developed [8].

In the 3<sup>rd</sup> year of the project, the work on a frequency doubling of the tunable RFL radiation generated around 1.3 micron has been started. Physical mechanisms of the influence of the RFL spectral broadening on the frequency doubling efficiency in PPLN crystal have been studied. It has been discovered that even at significant spectral broadening (order of magnitude larger than an acceptance width of the crystal) the second harmonics (SH) output may increase linearly with increasing fundamental power. In the reporting 12<sup>th</sup> period a model has been proposed that explains this phenomenon. The model takes into consideration sum-frequency mixing (SFM) processes inside the broad RFL spectrum. It has been shown that the SFM processes give the main contribution to the SH power, while the direct frequency doubling gives a vanishing value at large number of modes with random phases. A good qualitative and quantitative agreement of the model with the experimental data has been demonstrated. A comparison of the efficiency for the multi- and single frequency pumping has shown that in low-power domain the SH efficiency for multiple random modes may be even higher than that for a single mode of equal power. Laser radiation of >60 mW at 655 nm for about 7W RFL power has been generated.

## TECHNICAL DESCRIPTION

### 1. Experiment

The experimental setup is shown in Fig.1. We have utilized linear all-fiber scheme for the phosphosilicate RFL pumped by the Yb-doped fiber laser (YDFL), similar to that one used in tunable RFL configuration [8]. The YDFL is pumped by 3 laser diodes (LDs) and delivers up to 13.8 W at  $\sim 1.11 \mu\text{m}$  via output fiber Bragg grating (FBG) with high transmission ( $\text{HT}_{1.1}$ ). The RFL cavity is formed by two FBGs  $\text{HR}_{1.3}$  and  $\text{HT}_{1.3}$  with reflection coefficients 99% and 23% at  $1.31 \mu\text{m}$ , correspondingly, placed at the ends of 350-m long fiber. Large Stokes shift ( $\sim 40 \text{ THz}$ ) associated with phosphorus secures conversion from  $1.11$  to  $1.31 \mu\text{m}$  in one stage, see e.g. [2]. Additional FBG ( $\text{HR}_{1.1}$  - highly-reflective at  $1.11 \mu\text{m}$ ) is installed at the RFL output to implement a double pass pumping scheme. The RFL generates up to 7 W with  $\sim 50\%$  efficiency. Its output spectrum broadens significantly with increasing power, see Fig.2a, in spite of a narrowband ( $\sim 0.2 \text{ nm}$ ) output FBG ( $\text{HT}_{1.3}$ ). At high powers the spectrum acquires wide exponential tails with a central dip corresponding to the output FBG reflection. Mechanisms of the spectral broadening in RFLs have been clarified at previous stages: multiple four-wave mixing processes involving numerous longitudinal modes (up to  $10^6$  in  $\sim 1 \text{ km}$  long cavity) induce stochastic evolution of their amplitudes and phases [4, 5]. The weak wave turbulence model is shown applicable to RFLs with high-Q cavity and describes well their spectra, e.g. exponential shape and square-root growth of the width with power. However, this analytical model is not applicable directly to the RFL with narrowband low-reflection FBG which behavior deviates from the above: its linewidth grows almost linearly, see Fig.2c, while the power density tends to saturation at  $3.5 \text{ W/nm}$  level, see Fig. 2a, that seems to be crucial for frequency doubling efficiency.

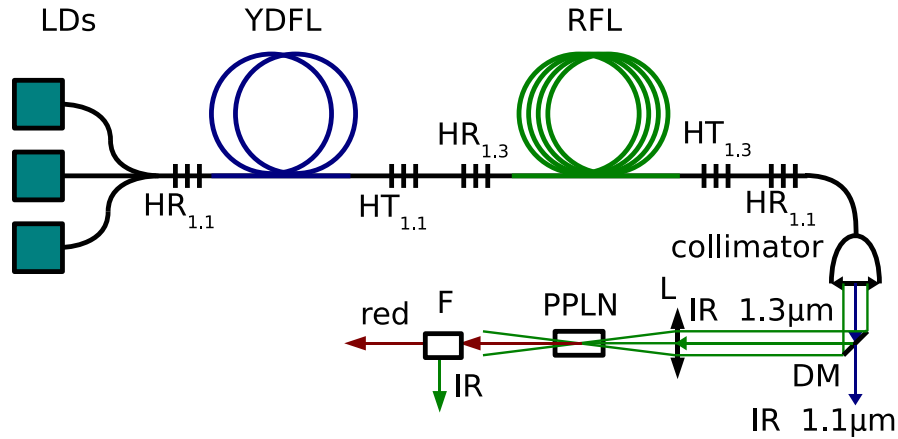


Fig. 1 Experimental setup.

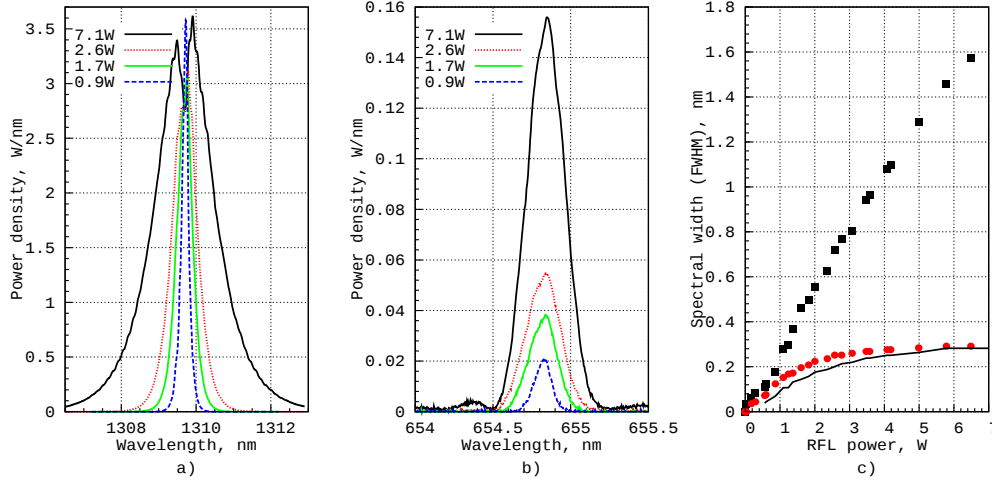


Fig. 2 Fundamental wave (a) and SH (b) output spectra measured at different RFL powers. (c) - Fundamental wave (boxes) and SH (circles) spectral widths together with SH width calculated from Eqs. 4, 5 (line).

Frequency doubling is performed in a simple and robust single-pass scheme with the 5% MgO-doped periodically poled LiNbO<sub>3</sub> (PPLN) crystal with poling length  $L \approx 8$  mm and specified poling period  $\Lambda \approx 12.73$   $\mu\text{m}$  that provides quasi-phase matching (QPM) at  $\sim 49^\circ\text{C}$  for 1.31- $\mu\text{m}$  operation. Corresponding QPM bandwidth amounts to  $\sim 0.6$  nm. The crystal has anti-reflection coatings for the fundamental and SH waves at both facets. RFL output radiation is collimated (see Fig.1), then cleaned by dichroic mirror DM and focused by lens L into the PPLN crystal placed in the oven. The generated red spectrum has typical sidelobes, Fig.2b. The main SH peak is narrow, its width (FWHM) approaches  $\sim 0.3$  nm at high powers, Fig.2c, in accordance with the QPM bandwidth. Although the RFL spectral width reaches  $\sim 1.6$  nm (3 times broader than QPM bandwidth), the SH power grows linearly up to  $> 60\text{mW}$  at  $\sim 7$  W RFL power, see Fig. 3. The obtained  $\sim 1\%$  efficiency is comparable with that for a single-frequency fundamental wave (results of calculations are shown by dashed line in Fig.3). In spite of quadratic growth, in the low-power domain the single-frequency model yields even lower values than the experiment. Thus we can conclude that not only direct frequency doubling contributes to the SH of the broadband RFL.

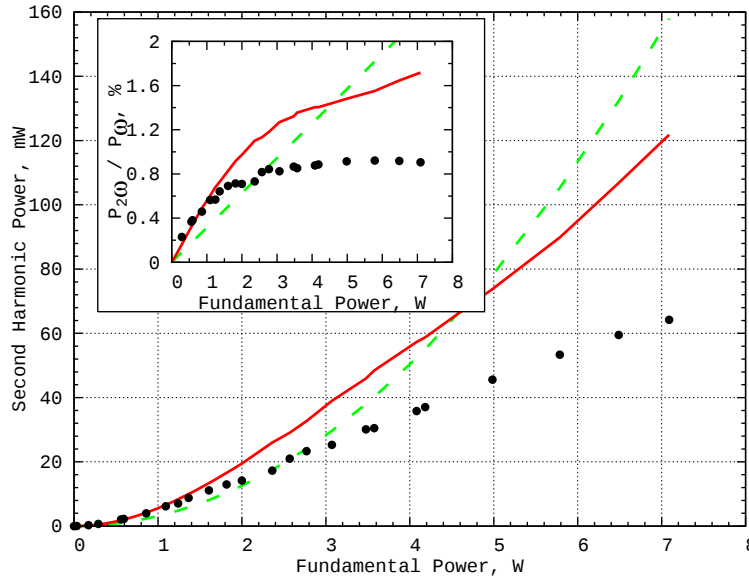


Fig. 3. Experimental data (points) and calculated SHG power  $P_{2\omega}$  versus  $P_\omega$  for single frequency (dashed line) and multiple frequency (solid line) fundamental wave. Inset: corresponding SHG efficiency  $P_{2\omega}/P_\omega$ .

## 2. Theoretical model

To explain the observed behavior, let us take into account sum frequency mixing (SFM) processes. In our description we follow methods of [9]. If the fundamental wave spectrum consists of  $N$  equidistant modes with frequencies  $\omega_{1j}$  ( $N \sim 10^6$  in the RFL), the second harmonic (SH) spectrum should consist of  $2N-1$  modes with frequencies  $\omega_{2j}$ . Even SH modes are generated only due to SFM, but odd modes have also direct SH contribution. Inset in Fig. 4 shows an example of the SHG for  $N=4$ .

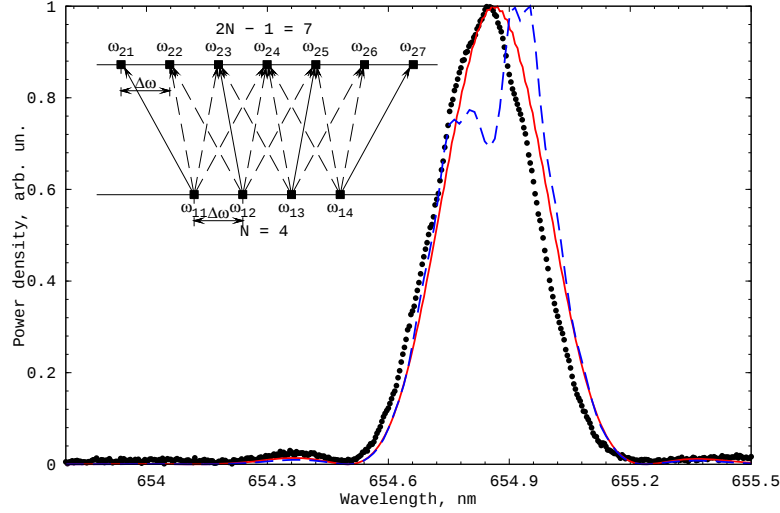


Fig. 4. Second harmonic spectrum: experiment at  $P_\omega \approx 7.1$  W (dots), calculation from Eqs. 4, 5 for direct frequency doubling (dashed line) and the same with sum-frequency mixing (solid line). Inset: an example of SHG for  $N=4$ .

The power conversion is described by  $2N-1$  equations:

$$\frac{\partial A_{2n}}{\partial z} + \frac{1}{u} \frac{\partial A_{2n}}{\partial t} = -i\sigma_2 \left( A_{1m}^2 \zeta_n + \sum_{j+k=n+1} \sum_{j \neq k} A_{1j} A_{1k} \right) \quad m = \frac{(n+1)}{2} \quad \zeta_n = \frac{[(-1)^{n+1} + 1]}{2}, \quad (1)$$

where  $A_{in}$  is the complex amplitude of  $n$ -th spectral mode of  $i$ -th harmonics,  $u$  is the group velocity,  $\sigma_2$  is the nonlinear coefficient. The first term in the brackets describes the direct frequency doubling, the second one corresponds to the SFM. From (1) one can find an intensity of  $n$ -th spectral component of the SH:

$$I_{2n} = (\sigma_2 z)^2 \left[ \left( a_{1m}^4 + 2a_{1m}^2 \sum_{j+k=n+1} \sum_{j \neq k} a_{1j} a_{1k} \times \cos(2\varphi_{1m} - \varphi_{1j} - \varphi_{1k}) \right) \zeta_n + \right. \\ \left. + 2 \sum_{j \neq k} \sum_{j \neq k} (a_{1j} a_{1k})^2 + 2 \sum_{j \neq k} \sum_{j \neq k} \sum_{p \neq q} \sum_{p \neq q} a_{1j} a_{1k} a_{1p} a_{1q} \times \cos(\varphi_{1j} + \varphi_{1k} - \varphi_{1p} - \varphi_{1q}) \right] \quad (2)$$

where  $a_{ij}$  and  $\phi_{ij}$  are the real amplitudes and phases of corresponding modes, respectively. The equation can be simplified for free-running modes, when all the phases  $\phi_{ij}$  are random:

$$I_{2n} = (\sigma_2 z)^2 \left[ a_{1m}^4 \zeta_n + 2 \sum_{j \neq k} (a_{1j} a_{1k})^2 \right] \quad (3)$$

Eqs.1-3 are valid in the case of exact phase matching for all interacting waves. One can modify Eqs.1-3 taking into account finite width of the phase matching (PM):

$$I_{2n} = (\sigma_2 z)^2 \left[ a_{1m}^4 \zeta_n \text{sinc}^2(\Delta k_{mm} z / 2) + 2 \sum_{j \neq k} \sum_{l \neq m} (a_{1j} a_{1l} \text{sinc}(\Delta k_{jk} z / 2))^2 \right], \quad (4)$$

where  $\Delta k_{jk} = k(\omega_j + \omega_k) - k(\omega_j) - k(\omega_k)$  is the wave vector mismatch.

It is also necessary to take into account specifics of the periodically-poled crystals (with poling period  $\Lambda$ ). It is known, that the transition from PM to QPM can be done by:

$$z \rightarrow \frac{2}{\pi} z, \quad \Delta k \rightarrow \left( \Delta k - \frac{2\pi}{\Lambda} \right) \frac{\pi}{2}, \quad (5)$$

One also needs to take into account Gaussian beam profile [10], which leads to modification of conversion coefficient  $(2\sigma_2 L / \pi)^2$  at single frequency and polarization to

$$\eta = \frac{P_{2\omega}}{(P_\omega)^2} = \frac{16\pi^2 d_{\text{eff}}^2 L}{\lambda_\omega^3 n_\omega n_{2\omega} \varepsilon_0 c} h, \quad (6)$$

where  $d_{\text{eff}} = 2d_{33}/\pi$  is the effective nonlinear coefficient,  $n_j$  is the corresponding refractive index,  $\varepsilon_0$  is the permittivity of vacuum,  $c$  is the speed of light, and  $h \sim 1$  is the Boyd-Kleinman focusing factor. For  $d_{33} = 27$  pm/V and  $L = 8$  mm  $\eta = 1.24\%$  / W at optimal focusing.

### 3. Comparison of the model with the experiment

Using Eqs.4-6 we have calculated an expected SH spectrum using the measured RFL spectra (Fig.2a). To obtain results at a reasonable computational time, we have reduced number of modes taking the effective mode spacing of 4.4 GHz ( $\sim 0.025$  nm) and have checked that a further increase in number of modes does not lead to significant changes in SH power values ( $\leq 1\%$ ). Two cases have been compared: direct frequency doubling (first term in Eq.4) and frequency doubling assisted by SFM (both terms in Eq.4). The SH spectrum calculated for direct frequency doubling has a very low amplitude. It is normalized to 1 at maximum for a comparison (Fig. 4, dashed line). This spectrum “copies” the dip in the fundamental wave spectrum (Fig.2a), that is absent in the measured SH spectrum. At the same time, calculations considering SFM (Fig. 4, solid line) provide a good coincidence with the experimental shape. Calculated second harmonic spectral width (FWHM) is also in a good agreement with the measured one, see Fig. 2c. Thus, in presence of multiple frequencies in the fundamental wave the SFM makes the main contribution to the SH power.

For a quantitative comparison of the calculated and measured power values, we take into account that for the randomly polarized RFL laser a half of the power corresponds to one linear polarization component. In addition, we have found a more exact value of the poling period ( $\Lambda = 12.54$   $\mu\text{m}$ ) by fitting positions of the two first minima in the SH spectra. Slight difference with the specified period may be associated with temperature dependence and MgO doping while we use the refraction index data for pure lithium niobate. Under these conditions, the SH power values calculated by summing up the power of individual modes (4) in frames of the described SFM model (solid line in Fig.3) are in very good agreement with the experimental points in low-power domain ( $\leq 2$  W), confirming the fact that the multimode SH power is higher than single-frequency one. At higher powers one can see a significant deviation of the experimental points from the theory, which can be attributed to the poorer beam quality ( $M^2 \approx 1.3$ ) and non-uniform crystal heating induced by the SH light absorption (see [18] and references therein). Nevertheless, the SFM model describes the experimental data quite well, especially a character of the dependence both for the SH power and efficiency (see inset in Fig.4), that is quite different from that for the single-frequency model.

The obtained results can be easily understood taking into account that the RFL modes are stochastic, i.e. they have random phases and amplitudes. Summing up the power of  $N$  random modes within QPM bandwidth results in factors  $N$  and  $N^2$  for the first and the second terms in Eq.4, correspondingly, both proportional to squared RFL mode power  $(P_\omega/N)^2$ . So the relative contribution of the direct doubling  $1/N$  is vanishingly small at  $N \gg 1$ . Moreover, the second term in Eqs.3,4 describing SFM takes factor 2 after transition from Eq.2 to the stochastic case (with gaussian statistics). The enhancement coefficient is  $(2-1/N)$  and tends to 2 at  $N \gg 1$ . Let's focus an attention on this fact, which means that the conversion efficiency for the multi-frequency radiation is higher than that for single-frequency one. This interesting effect is discussed in theory starting from classical paper [11] (see also [9] and citation therein), here it is convincingly confirmed in the experiment with the RFL at low powers. At high powers the input spectrum becomes broader than QPM bandwidth, so an additional factor proportional to the ratio of the mode number within QPM to the total mode number leads to relative reduction of the resulting SH power below the single-frequency curve. At linear growth of the RFL modes number with power, the SH power should grow nearly linearly.

## RESULTS/CONCLUSIONS

We have implemented narrow-band FBGs in RFL in order to narrow the RFL output spectrum and studied frequency doubling in PPLN crystal with this laser. In spite of narrow-band FBGs, the RFL output linewidth grows almost linearly and reaches  $\sim 1.6$  nm at  $\sim 7$ W that is  $\sim 3$  times of the PPLN crystal acceptance width. Nevertheless, the second harmonic (SH) power appears to grow linearly without saturation though the spectral density of the fundamental wave is saturated. Analyzing the results we proposed a mechanism that may explain such a behavior. We proved that SH generation for radiation comprising multiple modes, the sum-frequency mixing (SFM) gives the main contribution to the SH power, while the direct frequency doubling gives a vanishing value at large number of modes. Thus, efficient SH generation is possible for broadband Raman fiber lasers, which spectral width is sufficiently larger than QPM bandwidth of the crystal. In low-power domain the SH efficiency for multiple random modes appears even higher than that for a single mode of equal power. The developed SFM model describes quite well the SH spectrum and power measured at frequency doubling of 1.31- $\mu$ m Raman fiber laser. Note that the frequency doubling of fiber lasers generating at multiple frequencies was studied in experimental works [12, 13] and was compared with calculations, but not accurately. In [12] a single-frequency model has been used with a correction of the input power according to the QPM bandwidth. In [13] the multi-frequency nature of Yb fiber lasers was taken into account, but without "randomization", as a result factor 2 was missed.

In our experiments, the red laser radiation of  $\sim 100$  mW at 655 nm (that may be also tunable in 620-660 nm range) has been generated from the Raman fiber laser. This is already enough for the most of biomedical applications (PDT, flow cytometry, confocal microscopy etc.) and this robust fiber source may be a good alternative to conventional dye and diode lasers. To increase the efficiency further, one can use a linearly polarized RFL like in [9]. Another parameter is the crystal length, but calculations in frames of the SFM model have shown that there is no significant enhancement owing to a QPM bandwidth reduction at the lengthening. Techniques of fiber laser tuning and doubling have been applied also to the Yb-doped fiber laser and tunable green laser of  $>100$  mW @ 540-560 nm has been demonstrated [14].

So the goals of the project have been reached and it may be treated as completed.

During the reporting period paper [14] has been published in Laser Physics Journal and paper [15] has been submitted to Optics Express. S.A.Babin is invited to present the invited talk based on the project results [16] on the conference ICMAT 2009 to be held in Singapore (June 28-July 3, 2009).

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## **APPENDICES**

Manuscripts of papers [14, 15] and conference abstract [16].