Appl. Phys. B 73, 105–109 (2001) / Digital Object Identifier (DOI) 10.1007/s003400100619

Applied Physics B
Lasers
and Optics

A microchip-laser-pumped DFB-polymer-dye laser

T. Voss, D. Scheel, W. Schade*

Institut für Physik und Physikalische Technologien, TU Clausthal, Germany

Received: 1 March 2001/Revised version: 23 May 2001/Published online: 18 July 2001 –

Abstract. A miniaturized, high repetition rate, picosecond all solid state photo-induced distributed feedback (DFB) polymer-dye laser is described by applying a passively Q-switched and frequency-doubled Cr^{4+}:Nd^{3+}:YAG-microchip laser (pulse width $\Delta\tau = 540$ ps, repetition rate $\nu = 3$ kHz, pump energy $E_{\text{pump}} = 0.15$ μJ) as a pump source. A poly-methylmethacrylate film doped with rhodamine B dye serves as active medium. The DFB-laser pulses are temporally and spectrally characterized, and the stability of the thin polymer/dye film at high repetition rates is analyzed. The shortest DFB-laser pulses obtained have a duration of 11 ps. After the emission of 350 000 pulses the intensity of the DFB-laser output has decreased by a factor of two and the pulse duration has increased by a factor of 1.2. For single DFB-laser pulses of 20-ps duration the spectral bandwidth is measured to be $\Delta\lambda = 0.03$ nm, which is only 0.005 nm above the calculated Fourier limit assuming a Gaussian profile for the temporal shape of the pulses. Coarse wavelength tuning of the DFB laser between 590 and 619 nm is done by turning the prism. Additionally, a fine tuning of the DFB-polymer-laser wavelength is achieved by changing the temperature of the polymer/dye layer ($\frac{d\lambda}{dT} = -0.05$ nm/°C) in the range from 20 to 40 °C.

PACS: 42.70.Jk; 42.55.Mv; 42.60.By

Distributed feedback (DFB) dye lasers are well known as simple laser sources providing nearly bandwidth-limited picosecond pulses for spectroscopic applications [1–6]. Typically, dyes that are dissolved in organic solvents, e.g. methanol, are used as laser-active media. When pumped by excimer or Nd:YAG lasers, output powers of several nJ/pulse and repetition rates between 10–100 Hz are obtained.

Recently, the application of laser-active polymers and polymers doped with conventional laser dyes as active media for photo-induced distributed feedback lasers has been investigated. There are two different approaches towards a DFB-polymer laser: (1) the use of a laser-active polymer such as a ladder-type poly(*p*-phenylene) (LPPP) to prepare a thin film with a permanent DFB structure [7–9] and (2) the use of a thin film of a laser-active polymer or a polymer doped with a laser dye to generate a photo-induced distributed feedback structure for the duration of the pump pulse [10–14]. For the latter, the fabrication of the film is much easier, since no permanent structure has to be printed into the material. Furthermore, the DFB laser emits short pulses in the picosecond regime and its wavelength can easily be tuned within the gain profile of the polymer or the dye.

In this paper we report on the temporal and the spectral characterization of a DFB-polymer laser based on rhodamine-B-doped poly-methylmethacrylate (PMMA) as active laser medium. We show that a simple prism setup and a frequency-doubled Cr^{4+}:Nd^{3+}:YAG-microchip laser [15, 16] can be used to generate single DFB-laser pulses with a temporal width of typically 20 ps at repetition rates of up to 3 kHz. The pulse durations calculated by a numerical simulation based on a rate-equation model developed by Bor and Müller [17] are in good agreement with the experimental results. The dependence of the DFB wavelength on the temperature ($\lambda_{\text{DFB}} \sim n(T)$) can easily be used for a fine tuning or a stabilization of the output wavelength. Coarse wavelength tuning between 590 and 619 nm and the selection of a center wavelength is done by changing the angle of incidence of the pump beam. Furthermore, our results indicate that efficient pumping of the DFB structure by the frequency-doubled microchip laser at $\lambda = 532$ nm and $E_{\text{pump}} = 0.15$ μJ gives significantly larger lifetimes of the polymer/dye film compared to recent results applying ultraviolet pulses [14]. In this investigation a 50% decrease of the initial DFB-pulse intensity is obtained after 350 000 pulses.

1 Experimental setup

The experimental setup is shown in Fig. 1. A frequency-doubled microchip laser is used as a pump source for a DFB-polymer laser. The microchip laser consists of a Cr^{4+}:Nd^{3+}:YAG crystal (ALPHALAS GmbH) which com-

*Corresponding author.
(Fax: +49-5323/723-600, E-mail: wolfgang.schade@tu-clausthal.de)

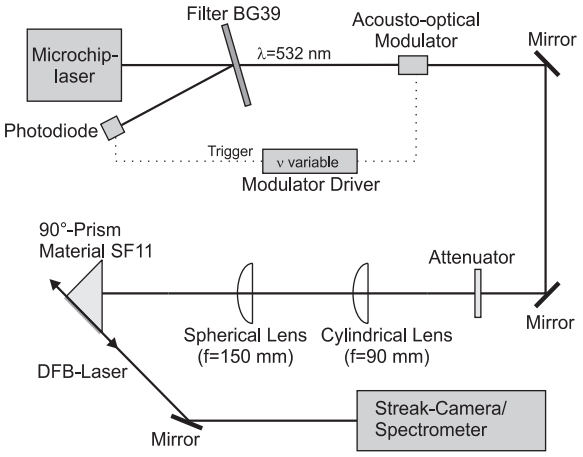

Fig. 1. Experimental setup for the microchip-laser-pumped DFB-polymer laser

bines the laser medium and the passive Q-switch in one crystal and is pumped by a continuous-wave (cw) diode laser (Osram SPL CG81, $P = 1$ W, $\lambda = 808$ nm) via a gradient-index lens. The pulsed output of the microchip laser ($\lambda = 1064$ nm) is frequency-doubled in a KTP crystal. A bandpass filter (Schott BG 39) is used for the separation of the 532-nm beam from the fundamental. The microchip laser typically generates pulses with a duration of 540 ps at 532 nm and a repetition rate of 3 kHz (see Fig. 2a). The pulse to pulse fluctuations in the output intensity are less than 2% (see Fig. 2b). The repetition rate of the passively Q-switched microchip laser is determined by the concentration of the Cr^{4+} ions, which act as a saturable absorber in the YAG crystal. An acousto-optical modulator is used as a pulse picker to tune the repetition rate between 10 Hz and 1.6 kHz.

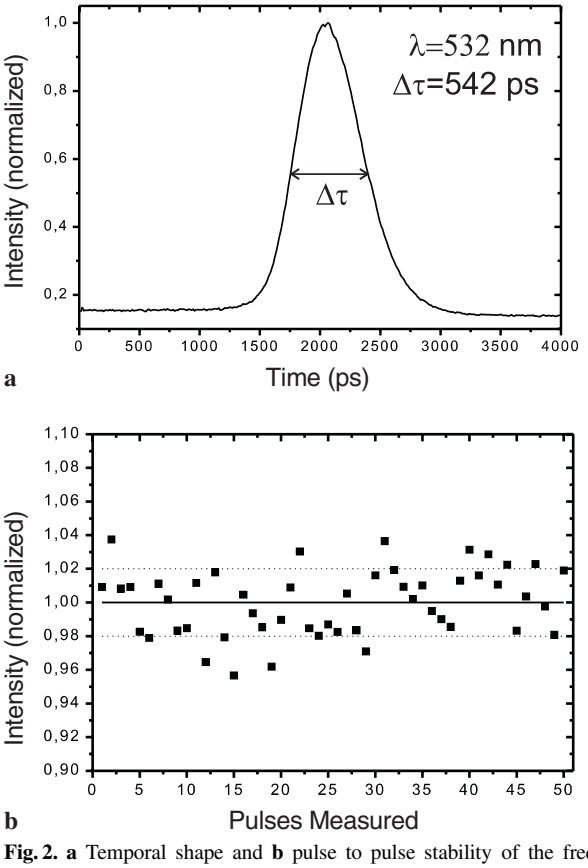

Fig. 2. a Temporal shape and **b** pulse to pulse stability of the frequency-doubled microchip laser

The pump beam is imaged on the polymer/dye film of the prism-DFB laser by a system of a cylindrical lens ($f = 90$ mm) and a spherical lens ($f = 150$ mm). The layer is prepared by first solving 20 mg rhodamine B dye and then 3.7 g PMMA pellets in a mixture of 10 ml chloroform and 0.8 ml DMSO (dimethylsulfoxide). This solution is afterwards spin-coated directly onto the side $\overline{AC}$ of a 90°-SF11 prism and baked for one hour at a temperature of $T = 80$ °C (see Fig. 3). The thickness of the layer is adjusted by changing the concentration of the PMMA in the solution, the temperature of the solution which determines the viscosity and by changing the rotational frequency of the spin-coater. The best performance of the DFB laser is obtained with films having a thickness between 1 and 3 μm. The prism material SF11 is chosen because of its high index of refraction $n = 1.795$ to ensure that the generated DFB-laser wavelengths are within the gain profile of the rhodamine B laser dye (refer to (1)).

The pump beam enters the prism through the side $\overline{AB}$. One part of the beam is refracted on the polymer/dye film directly, while the other part is totally reflected on the side $\overline{BC}$ of the prism. The two parts of the beam form an interference pattern in the polymer/dye film. The length of this pattern is approximately $L = 2.0$ mm and its height is $b = 0.1$ mm. This interference pattern acts as distributed feedback structure of the laser and only exists for the duration of the pump pulse (see Fig. 3). Since the wavelength selection is achieved by first-order Bragg scattering, the wavelength of the DFB-polymer laser in vacuum is given by the relation $\lambda = 2n_p\Lambda$, where Λ is the separation of the interference fringes [2]. In the case of the prism geometry which is used in this investigation, the wavelength of the DFB-polymer laser is given by [18]:

$$\lambda = \frac{n_p\lambda_p}{n\sin\gamma} \approx \frac{n_p\sqrt{2}\lambda_p}{n}\left[1 - \frac{\alpha}{n} + \frac{3}{2}\frac{\alpha^2}{n^2} + \dots\right]. \quad (1)$$

Here, $n_p = 1.49$ is the index of refraction of the polymer/dye film, $n = 1.79$ the index of refraction of the prism, $\lambda_p =$

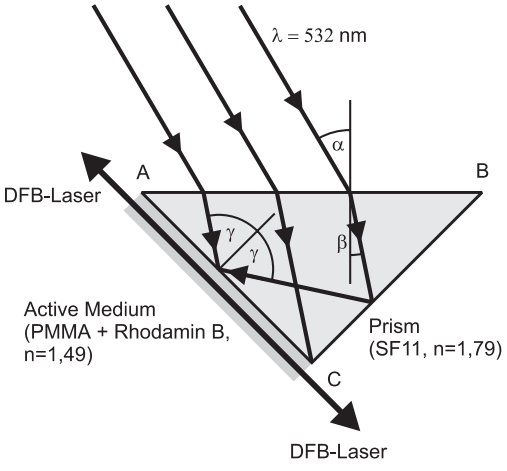

Fig. 3. Setup of the prism-DFB-polymer-dye laser

532 nm the wavelength of the pump beam, α the angle of incidence of the pump beam on the prism surface and γ the angle of incidence on the polymer/dye film. The expansion (1) holds for $|\alpha| \ll 1$. For this geometry only one standing wave can be generated in the DFB resonator. The corresponding wavelength can be calculated by (1). It further shows that for a fixed pump wavelength λ_p the wavelength of the DFB-polymer laser can be tuned by changing the angle of incidence α and/or the indices of refraction n and n_p.

The pulses of the DFB-polymer laser are temporally analyzed using a streak camera (Optronis Optoscope, time resolution < 2 ps, repetition rate < 2.5 kHz). The spectral analysis is done with an optical multichannel analyzer (OMA4, EG&G Princeton Applied Research, spectral resolution of ≈ 0.1 nm) and a 0.5-m monochromator with a spectral resolution of ≈ 0.01 nm.

2 Temporal characterization of the DFB-polymer laser

The temporal behavior of the generated DFB-laser pulses is analyzed experimentally by recording streak images, and the measured results are compared to numerical simulations that apply a system of coupled differential equations given by Bor and Müller [17]:

$$\frac{\mathrm{d}N}{\mathrm{d}t} = I_p \sigma_p (N_0 - N) - \frac{\sigma_e c}{n_p} NQ - \frac{N}{\tau_c}, \tag{2}$$

$$\frac{\mathrm{d}Q}{\mathrm{d}t} = \frac{(\sigma_e - \sigma_a)\, c}{n_p} NQ - \frac{Q}{\tau_c} + \frac{\Omega N}{\tau_f}, \tag{3}$$

$$\tau_c = \frac{n_p L^3}{8\pi^2 c} \left[N (\sigma_e - \sigma_a) V \right]^2, \tag{4}$$

$$P_d = \frac{hcQ}{2\lambda\tau_c} Lab. \tag{5}$$

In these equations $N(t)$ describes the density of dye molecules in the first excited singlet state, $Q(t)$ the density of DFB photons, τ_c the average lifetime of the photons in the DFB resonator and P_d the output power of the DFB laser. N_0 is the density of dye molecules in the PMMA film, I_p the spatially averaged pump photon flux per unit area, σ_p the absorption cross section for the pumping wavelength for the ground state, σ_a the absorption cross section for the DFB laser wavelength for the first excited singlet state to the second, σ_e the stimulated emission cross section for the laser wavelength and τ_f the fluorescence lifetime of the first excited state. c is the speed of light in vacuum, L the length of the pumped volume, a the penetration depth of the pump light into the polymer/dye layer, b the height of the excited volume, V the visibility of the interference fringes and n_p the index of refraction of the polymer/dye layer. Ω determines the fraction of the spontaneous emission which propagates into the angular and the spectral ranges of the DFB-laser beam and h is Planck's constant. Whereas (2) and (3) hold for different types of lasers, the non-linearity of τ_c in (4) is responsible for the typical self-cavity-dumping of the DFB laser, which makes the laser emit short pulses in the picosecond regime.

The equations (2) to (5) are solved numerically for parameters describing the PMMA/rhodamine B film [19, 20] and are referred to in Table 1. The results of the simulation for two different values of the pump energy are shown in Figs. 4a and 5a. With $E_{pump} = 0.09\,\mu$J the DFB laser is operated just slightly above the laser threshold. Then only a single pulse with a duration of $\Delta\tau = 21$ ps is emitted. When increasing the pump energy to $E_{pump} = 0.15\,\mu$J the simulation

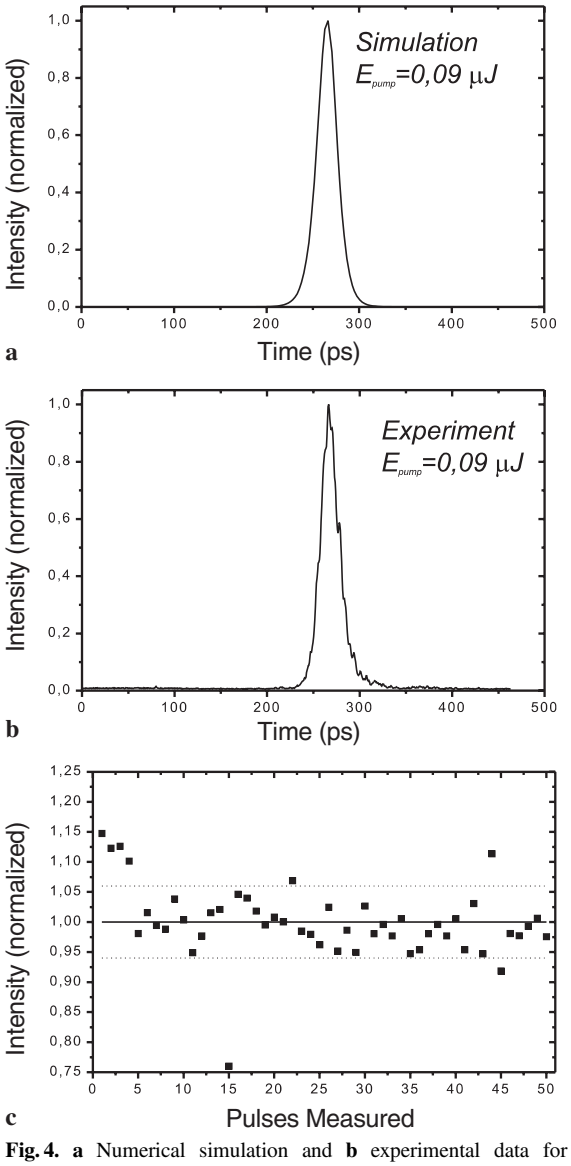

Fig. 4. **a** Numerical simulation and **b** experimental data for $E_{pump} = 0.09\,\mu$J. Duration of the single pulse obtained with a Gaussian fit: **a** $\Delta\tau = 22$ ps and **b** $\Delta\tau = 21$ ps. **c** Pulse to pulse stability of the DFB-laser output for single-pulse operation

Table 1. Values used for the numerical simulation [19, 20]

N_0	σ_p	σ_e	σ_a	τ_f	ϱ	η	c	V
7.6×10^{24} m^{-3}	3.2×10^{-20} m^2	1.5×10^{-20} m^2	$< 0.7 \times 10^{-20}$ m^2	3.9×10^{-9} s	1.18 kgm^{-3}	1.49	3×10^8 ms^{-1}	1

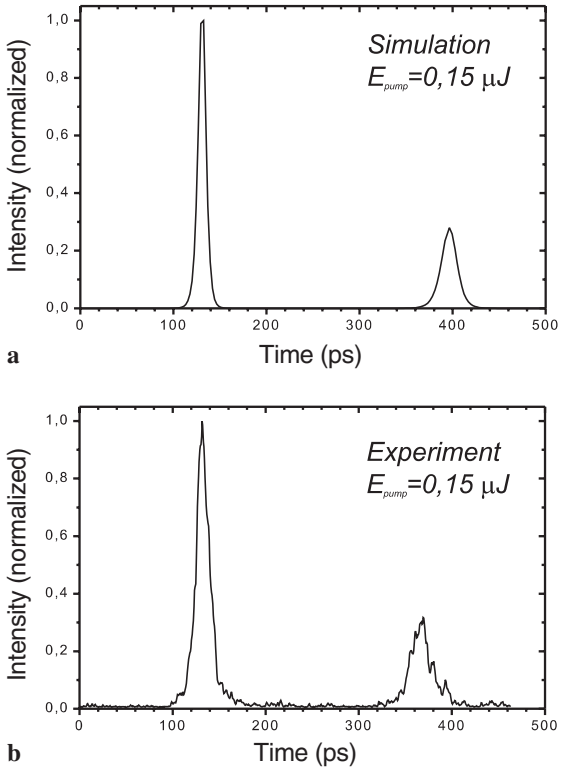

Fig. 5. **a** Numerical simulation and **b** experimental data for $E_{\text{pump}} = 0.15\,\mu\text{J}$. Duration of the first pulse obtained with a Gaussian fit: **a** $\Delta\tau = 9$ ps and **b** $\Delta\tau = 13$ ps

predicts two DFB-laser pulses, with the first pulse having a duration of $\Delta\tau = 9$ ps. The second pulse is emitted 260 ps after the first and has a duration of 17 ps.

The corresponding experimental data are shown in Figs. 4b and 5b. A good agreement between experiment and simulation in the predicted pulse durations, their spacing to each other and their relative intensities is obtained. Typical single-pulse durations are $\Delta\tau = 20$ ps. When increasing the pump energy from $E_{\text{pump}} = 0.09\,\mu\text{J}$ to $E_{\text{pump}} = 0.15\,\mu\text{J}$ (the highest pump energy that is obtained with the present experimental setup), a less intense second pulse with a duration of 21 ps appears 240 ps after the first pulse, while the duration of the first pulse decreases from $\Delta\tau = 21$ ps to $\Delta\tau = 13$ ps. When changing the repetition rate between 20 and 800 Hz no significant change of the temporal output characteristics of the DFB-polymer-dye laser is measured.

In Fig. 4c the pulse to pulse fluctuation in the output intensity of the DFB-polymer-dye laser is shown for 50 single-shot measurements. While the intensity of the microchip laser shows a fluctuation of $\pm 2\%$ (which is the standard deviation; refer to Fig. 2b) the pulse to pulse stability of the DFB-laser output is $\pm 6\%$ (see Fig. 4c).

3 Spectral characterization of the DFB-polymer laser

The wavelength of a photo-induced DFB-polymer-dye laser can easily be tuned by changing the angle of incidence γ of the pump beam on the polymer/dye film (refer to (1)) [12–14]. In the present experiment a tuning range of the wavelength from 618.8 to 590.0 nm is obtained. The spectral bandwidth of a 20-ps single DFB-laser pulse is measured to be $\Delta\lambda = 0.03$ nm (resolution of the spectrometer ≈ 0.01 nm). This is only 0.005 nm above the theoretically calculated Fourier limit $\Delta\lambda = \frac{0.41\lambda^2}{c\Delta\tau} = 0.025$ nm assuming a Gaussian-shaped single pulse of 20-ps duration.

A fine tuning of the DFB-polymer-laser wavelength by temperature control of the polymer/dye film was also investigated [27]. Our results show that by changing the temperature of the prism and of the polymer/dye film from 20 to 41 °C the DFB wavelength can be tuned from 616.2 nm to 615.2 nm (see Fig. 6). A linear fit to the measured data gives a dependence of the wavelength on the temperature of

$$\frac{\mathrm{d}\lambda}{\mathrm{d}T} = -0.05\frac{\text{nm}}{\text{°C}}. \tag{6}$$

Therefore, a coarse tuning of the wavelength can be done by changing the angle γ, and a fine tuning of about 1 nm is achieved by controlling the temperature of the prism and the polymer/dye film.

The dependence of the DFB-laser wavelength on the temperature is calculated by expanding the indices of refraction in (1) in powers of $(T - T_0)$. For fixed values of γ and λ_{p} the wavelength is approximately given by

$$\lambda\,(T) = C\frac{n_0 - n_1\,(T - T_0)}{n_2 + n_3\,(T - T_0) + n_4\,(T - T_0)^2}. \tag{7}$$

As a starting point for the simulation the measured wavelength $\lambda = 616$ nm for a temperature $T_0 = 24$ °C is chosen. Using (7) the constant C is determined with these initial values to be $C = 741.1$ nm. The constants n_0 and n_1 are used for the approximation of the index of refraction of PMMA. The constants $n_2 - n_4$ characterize the prism material SF11. These constants are taken from [21, 22] and are listed in Table 2. The results of this calculation (line) together with the experimental data (dots) are shown in Fig. 6.

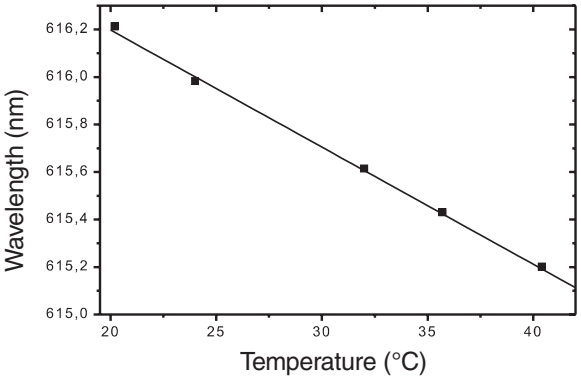

Fig. 6. Fine tuning of the DFB wavelength by changing the temperature of the prism and polymer/dye film from 20 to 41 °C The *dots* correspond to the experimental data. The *line* is the result of the calculation using (7)

Table 2. Values for the refractive indices of PMMA and SF11 [21, 22]

n_0	$n_1\left[\frac{1}{\text{K}}\right]$	n_2	$n_3\left[\frac{1}{\text{K}}\right]$	$n_4\left[\frac{1}{\text{K}^2}\right]$
1.492	1.1×10^{-4}	1.795	1.12×10^{-5}	1.18×10^{-8}

4 Stability of the polymer/dye film

Practical applications of DFB-polymer-dye lasers are limited by the lifetime of the polymer/dye film. Recently reported results indicate [14, 20, 23–26] that photodegradation and diffusion of the dye molecules probably caused by the local heating of the layer by the pump pulse contribute to a decrease of the output power. Most of these experiments are performed at repetition rates between 5 and 20 Hz. They show a 'lifetime' of several tens of thousand pulses depending on the wavelength and the power of the pump laser, where the 'lifetime' is defined as the number of pulses after which the output power has decreased to 50% of its initial value.

In the present investigation a typical 'lifetime' of 350 000 pulses for the PMMA/rhodamine B film is obtained with a microchip laser as pump source for the DFB-polymer-dye laser (see Fig. 7). Furthermore, our results show that a change of the repetition rate between 20 and 800 Hz has no significant influence on the stability of the polymer/dye film. Additionally, we analyzed the stability of the polymer/dye film at different temperatures. Between 6 and 45 °C no significant influence on the 'lifetime' is measured. However, at higher temperatures the performance of the laser dye significantly decreases and the 'lifetime' is lowered, which is in agreement with previous results [27]. This indicates that at moderate temperatures only the local heating caused by the pump beam contributes to the photodegradation of the laser dye, whereas the global temperature of the polymer/dye film has no significant influence on the stability.

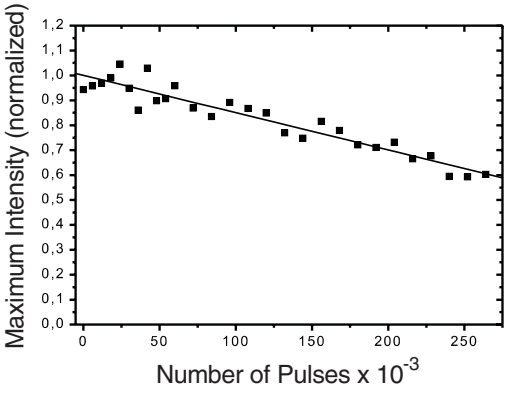

Fig. 7. Stability of the polymer/dye film, $\nu = 100$ Hz, $T = 22$ °C, $E_{\text{pump}} = 0.15$ μJ

5 Summary

We have shown that a miniaturized microchip-laser-pumped DFB-polymer laser with an active medium that consists of a PMMA layer doped with rhodamine B dye molecules generates single laser pulses with a duration of $\Delta\tau = 20$ ps at repetition rates of up to 3 kHz. The layer can easily be fabricated by the spin-coating method, and only a prism and a system of two lenses are necessary for the DFB-laser setup. The wavelength of the almost bandwidth-limited prism-DFB-laser pulses can be tuned over a 30-nm range by rotating the prism, and a fine tuning over a range of about 1 nm and a stabilization of the DFB wavelength is achieved by controlling the temperature of the prism and the polymer/dye film. The temporal characteristics of the DFB-polymer-laser output are quantitatively verified by computer simulations using a simple rate-equation system that is successfully applied to a DFB-polymer-dye laser. Compared to previous experiments the 'lifetime' of the PMMA/rhodamine B film film is increased to 350 000 pulses when using the frequency-doubled output of a passively Q-switched microchip laser as pump source. The photophysical properties of the layer concerning the characterization of the photo-induced DFB structure and the photodegradation of the laser dye are currently under investigation with a near-field scanning optical microscope.

Acknowledgements. The authors wish to thank Heinz Christian Rost for the preparation of the polymer/dye films.

References

1. C.V. Shank, J.E. Bjorkholm, H. Kogelnik: Appl. Phys. Lett. **18**, 395 (1971)
2. Z. Bor: Opt. Commun. **29**, 103 (1979)
3. F.P. Schäfer: Laser Optoelektronik **2**, 95 (1984)
4. H. Kogelnik, C.V. Shank: J. Appl. Phys. **43**, 2327 (1972)
5. Z. Bor, A. Müller, B. Racz: Opt. Commun. **40**, 294 (1982)
6. S. Szatmari, B. Racz: Appl. Phys. B **43**, 173 (1987)
7. C. Kallinger, M. Hilmer, A. Haugeneder, M. Perner, W. Spirkl, U. Lemmer, J. Feldmann, U. Scherf, K. Müllen, A. Gombert, V. Wittwer: Adv. Mater. **10**, 920 (1998)
8. A. Haugeneder, M. Neges, C. Kallinger, W. Spirkl, U. Lemmer, J. Feldmann, M.-C. Amann, U. Scherf: Appl. Phys. B **66**, 389 (1998)
9. A. Haugeneder, M. Hilmer, C. Kallinger, M. Perner, W. Spirkl, U. Lemmer, J. Feldmann, U. Scherf: J. Appl. Phys. **85**, 1124 (1999)
10. M. Maeda, Y. Oki, K. Imamura: IEEE J. Quantum Electron. **QE-33**, 2146 (1997)
11. Y. Oki, K. Ohno, M. Maeda: Jpn. J. Appl. Phys. **37**, 6403 (1998)
12. X. Zhu, S. Lam, D. Lo: Appl. Opt. **39**, 3104 (2000)
13. V. Dumacher, L. Rocha, C. Denis, C. Fiorini, J.-M. Nunzi, F. Sobel, B. Sahraoui, D. Gindre: J. Opt. A: Pure Appl. Opt. **2**, 279 (2000)
14. M. Fukuda, K. Mito: Jpn. J. Appl. Phys. **39**, 5859 (2000)
15. J.J. Zayhowski: Opt. Lett. **21**, 588 (1996)
16. B. Braun, F.X. Kärtner, U. Keller, J.-P. Meyn, G. Huber: Opt. Lett. **21**, 405 (1996)
17. Z. Bor, A. Müller: IEEE J. Quantum Electron. **QE-22**, 1524 (1986)
18. S. Chandra, N. Tekeuchi, S.R. Hartmann: Appl. Phys. Lett. **21**, 144 (1972)
19. A. Tagaya, S. Teramoto, E. Nihei, K. Sasaki, Y. Koike: Appl. Opt. **36**, 572 (1997)
20. A. Deshpande, E. Namdas: J. Lumin. **91**, 25 (2000)
21. K. Horie, H. Ushiki, F.M. Winnik: *Molecular Photonics* (Wiley-VCH, Weinheim 2000) p. 162
22. Datasheet from *Schott Glaskatalog* (2000)
23. W. Hu, H. Ye, C. Li, Z. Jiang, F. Zhou: Appl. Opt. **36**, 579 (1997)
24. A. Costela, I. Garcia-Moreno, J. Barroso, R. Sastre: Appl. Phys. B **70**, 367 (2000)
25. S. Popov: Appl. Opt. **37**, 6449 (1998)
26. A. Costela, I. Garcia-Moreno, J.M. Figuera, F. Amat-Guerri, R. Mallavia, M.D. Santa-Maria, R. Sastre: J. Appl. Phys. **80**, 3167 (1996)
27. W. Schade, B. Garbe, V. Helbig: Appl. Opt. **29**, 3950 (1990)