Continuous-wave ultraviolet generation at 320 nm by intracavity frequency doubling of red-emitting Praseodymium lasers

A. Richter, N. Pavel, E. Heumann, and G. Huber

Institute of Laser Physics, University of Hamburg, Luruper Chaussee 149, D-22761 Hamburg, Germany
andricht@physnet.uni-hamburg.de

D. Parisi, A. Toncelli, and M. Tonelli

NEST - Dipartamento di Fisica, Università di Pisa
Largo Pontecorvo 3, 56127 Pisa, Italy

A. Diening and W. Seelert

COHERENT Lübeck GmbH, Seelandstr. 9, 23569 Lübeck, Germany

Abstract: We describe a new approach for the generation of coherent ultraviolet radiation. Continuous-wave ultraviolet light at 320 nm has been obtained by intracavity frequency doubling of red-emitting Praseodymium lasers. Lasing at the 640-nm fundamental wavelength in $Pr:LiYF_4$ and $Pr:BaY_2F_8$ was realized by employing an optically pumped semiconductor laser at 480 nm as pump source. Using LiB_3O_5 as nonlinear medium, ~19 mW of ultraviolet radiation with ~9% optical efficiency with respect to absorbed power was reached for both laser crystals; the visible-to-ultraviolet conversion efficiency was 26% and 35% for $Pr:LiYF_4$ and $Pr:BaY_2F_8$, respectively.

OCIS Codes: (140.3580) Lasers, solid-state; (140.3610) Lasers, ultraviolet; (140.5560) Pumping; (140.7240) UV, XUV, and X-ray lasers.

References

1. B. Wu, N. Chen, C. Chen, D. Deng, and Z. Xu, "Highly efficient ultraviolet generation at 355 nm in LiB_3O_5," Opt. Lett. **14**, 1080-1081 (1989).
2. H. Kitano, T. Matsui, K. Sato, N. Ushiyama, M. Yoshimura, Y. Mori, and T. Sasaki, "Efficient 355-nm generation in CsB_3O_5 crystal," Opt. Lett. **28**, 263-265 (2003).
3. K. Mizuuchi, A. Morikawa, T. Sugita, K. Yamamoto, N. Pavel, and T. Taira, "Continuous-wave ultraviolet generation at 354 nm in a periodically poled $MgO:LiNbO_3$ by frequency tripling of a diode end-pumped $Nd:GdVO_4$ microlaser," Appl. Phys. Lett. **85**, 3959-3961 (2004).
4. L.B. Chang, S.C. Wang, A.H. Kung, "Efficient compact watt-level deep-ultraviolet laser generated from a multi-kHz Q-switched diode-pumped solid-state laser system," Opt. Commun. **209**, 397-401 (2002).
5. Y. Kaneda and S. Kubota, "Continuous-wave 355-nm laser source based on doubly resonant sum-frequency mixing in external resonator," Opt. Lett. **20**, 2204-2206 (1995).
6. R.G. Smart, D.C. Hanna, A.C. Tropper, S.T. Davey, S.F. Carter, and D. Szebesta, "CW room temperature upconversion lasing at blue, green and red wavelengths in infrared-pumped Pr^{3+}-doped fluoride fibre," Electron. Lett. **27**, 1307-1309 (1991).
7. T. Sandrock, H. Scheife, E. Heumann, and G. Huber, "High-power continuous-wave upconversion fiber laser at room temperature," Opt. Lett. **22**, 808-810 (1997).
8. R.G. Smart, J.N. Carter, A.C. Tropper, D.C. Hanna, S.T. Davey, S.F. Carter, and D. Szebesta, "CW room temperature operation of praseodymium-doped fluorozirconate glass fibre lasers in the blue-green, green and red spectral regions," Opt. Commun. **86**, 333-340 (1991).
9. A. Richter, H. Scheife, E. Heumann, G. Huber, A. Diening, and W. Seelert "Semiconductor laser pumping of a continuous-wave Pr^{3+}-doped ZBLAN fiber laser," Electron. Lett. **41**, 794-795 (2005).
10. T. Sandrock, T. Danger, E. Heumann, G. Huber, and B.H.T. Chai, "Efficient continuous-wave laser emission of Pr^{3+}-doped fluorides at room temperature," Appl. Phys. B **58**, 149-151 (1994).
11. T. Sandrock, E. Heumann, G. Huber, and B.H.T Chai, "Continuous-Wave Pr,Yb:$LiYF_4$ Upconversion Laser in the Red Spectral Range at Room Temperature," in *Advanced Solid State Lasers Conference*, Vol. 1 of OSA Trends in Optics and Photonics, S.A. Payne and C. Pollack, eds. (Optical Society of America,

#68599 - $15.00 USD
Received 1 March 2006; revised 3 April 2006; accepted 3 April 2006

Washington, D.C., 1996), pp. 550-553.
12. A. Richter, E. Heumann, E. Osiac, G. Huber, W. Seelert, and A. Diening, "Diode pumping of a continuous-wave Pr^{3+}-doped $LiYF_4$ laser," Opt. Lett. **29**, 2638-2670 (2004).
13. H. Kogelnik and T. Li, "Laser Beams and Resonators," Appl. Opt. **5**, 1550-1567 (1966).
14. J.A. Caird, S.A. Payne, P.R. Staber, A.J. Ramponi, L.L. Chase, and W.F. Krupke, "Quantum Electronics properties of the $Na_3Ga_2Li_3F_{12}$:Cr^{3+} laser," IEEE. J. Quantum Electron. **QE-24**, 1077-1099 (1988).
15. T. Baer, "Large-amplitude fluctuations due to longitudinal mode coupling in diode-pumped intracavity-doubled Nd:YAG lasers," J. Opt. Soc. Am. B **3**, 1175-1180 (1986).
16. R.G. Smith, "Theory of intracavity optical second-harmonic generation," IEEE J. Quantum Electron. **6**, 215-223 (1970).
17. A. Agnesi, A. Guandalini, and G. Reali, "Efficient 671-nm pump source by intracavity doubling of a diode-pumped Nd:YVO_4 laser," J. Opt. Soc. Am. B **19**, 1078-1082 (2002).
18. E. Osiac, E. Heumann, G. Huber, S. Kück, E. Sani, A. Toncelli, and M. Tonelli, "Orange and red upconversion laser pumped by an avalanche mechanism in Pr^{3+},Yb^{3+}:BaY_2F_8," Appl. Phys. Lett. **82**, 3832-3834 (2003).

1. Introduction

Currently, solid-state laser sources that emit in the ultraviolet (UV) spectral region use birefringent phase-matching or quasi phase-matching methods and a two-stage nonlinear frequency conversion process. The near infrared radiation of a solid-state laser is frequency-doubled by second harmonic generation (SHG) in the first step. Next, sum frequency generation (SFG) of the visible light with the residual infrared radiation is employed to obtain UV radiation at 355 nm [1-3] in case of Nd-lasers, or a second SHG stage is added to achieve UV at 266 nm [4]. Fairly good performances were obtained by operating the infrared lasers in pulsed mode [1, 2, 4], which granted high peak power. The low nonlinear coefficient for bulk nonlinear materials (a few pm/V) limits, however, the performances of continuous-wave (cw) UV lasers. Cw generation at 354 nm was reported recently by using quasi-phase-matching and cascading SHG and SFG in a periodically-poled MgO:$LiNbO_3$ crystal [3]. Still, the infrared-to-UV conversion efficiency of this device was as low as ~1%, mainly because the nonlinear crystals were placed outside of the laser cavity and a single-pass conversion scheme was used. Doubly resonant sum-frequency mixing of infrared and visible radiation in external resonators could be used to enhance the efficiency of cw UV generation [5]; the set-up is, however, complicated and single-longitudinal mode operation is necessary.

Due to an energy level scheme that enables several transitions in the red, orange, green, or blue spectral region, trivalent Praseodymium (Pr) ions are of great interest for obtaining laser emission in the visible spectrum. Cw operation at various wavelengths between 490 nm and 635 nm was demonstrated in Pr-doped fiber lasers since 1991 [6-8]. The excitation of the upper level was realized through an upconversion avalanche process, by pumping in the near infrared region at ~840 nm [6, 7], or directly, employing pumping sources with emission in the blue spectral range close to 480 nm [8, 9].

First cw laser emission of Pr:$LiYF_4$ (Pr:YLF) in the visible spectrum was demonstrated under pumping at ~460 nm, with an Ar-ion laser [10]. Lately, an upconversion avalanche pump process initiated by the 840-nm radiation of a Ti:sapphire laser was used to obtain efficient orange and red emission in Pr,Yb:$LiYF_4$ [11]. A diode laser pumped Pr:YLF laser was first demonstrated in 2004 [12]: the cw output power at 640 nm was, however, modest (~2 mW) due to the limited output power of the blue GaN diode laser used for pumping.

In this work we report cw UV generation at 320 nm by intracavity frequency-doubling of Pr:YLF and Pr:BaY_2F_8 (Pr:BYF) lasers operating at 640 nm. The power scaling at the red fundamental wavelength was realized by using an optically pumped semiconductor laser (OPS) emitting at 480 nm as pump source. Employing LiB_3O_5 (LBO) nonlinear material in a single folded cavity, about 19 mW of UV radiation was obtained for both laser crystals, at an efficiency of ~9% with respect to absorbed pump power. This the first intracavity SHG demonstration resulting in cw UV laser radiation, to the best of our knowledge.

Received 1 March 2006; revised 3 April 2006; accepted 3 April 2006

2. Experimental: Results and discussion

A sketch of the Pr laser set-ups is shown in Fig. 1. The Pr:YLF crystal (0.7-at.% doping level and 5.7-mm length) has a high-reflectivity (HR, transmission $T < 0.1\%$) coating for the 640-nm lasing wavelength (λ_ω) and a high-transmission (HT, $T > 98\%$) coating for the 480-nm pumping wavelength (λ_p) on the input side (M1); the opposite crystal's surface was antireflection (AR) coated for λ_ω. The V-type resonator, presented in Fig. 1(a), consists of M1 and two concave mirrors M2 and M3 of 100 mm and 50 mm radius of curvature, respectively. The distances M1-to-M2 and M2-to-M3 were 92 mm and 146 mm, respectively. The mirror M2 was HR coated at λ_ω and HT ($T > 97\%$) at 320 nm ($\lambda_{2\omega}$).

An uncoated Pr:BYF crystal (0.8-at.% Pr, 3-mm thick) was also used in the experiments and placed in a modified V-resonator as shown in Fig. 1(b). The mirror M1 has a 50-mm radius of curvature and is HR coated for λ_ω as well as HT at λ_p; the M1-to-M2 distance was 140 mm and the Pr:BYF crystal was placed 45 mm apart of M1. For these experiments the Pr:YLF crystal was provided by Coherent Lübeck GmbH, Lübeck, Germany, whereas the Pr:BYF laser medium was grown at Pisa University, Italy. However, Pr:YLF laser crystals can be grown in our laboratory by Czochralski method, too.

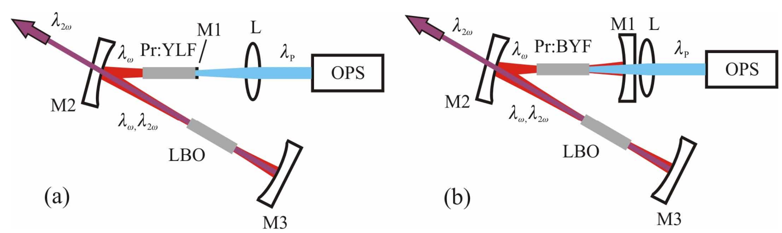

Fig. 1. Schematics of the (a) Pr:YLF and (b) Pr:BYF lasers pumped by a cw OPS source and intracavity frequency-doubled by LBO nonlinear crystals. L: lens; M1, M2, M3: mirrors.

The optical pumping was accomplished with a cw frequency-doubled OPS laser (Coherent Lübeck GmbH, Lübeck, Germany). The pump source provided a nearly diffraction-limited (M^2~1) and linear polarized beam at $\lambda_p = 480$ nm with a maximum power of ~310 mW. The lens L with a focal length of 50 mm was used to focus the pump beam into the laser crystals to a spot-size of ~39-µm radius; ~18% of the pumping light was lost on the coupling optics placed between the OPS and the laser media. The pump polarization was in the *c*-direction (π polarization) for Pr:YLF and in the *b*-direction for Pr:BYF, such that ~84% of the incident blue light was absorbed in each Pr-doped medium. Operation at 640 nm was obtained without the nonlinear crystal in the laser cavity and by using M3 mirrors with various output couplers *T*. The ABCD formalism for Gaussian beams [13] was used to determine the laser beam waist in the laser crystals. These waists were 58 µm and 62 µm in case of Pr:YLF and Pr:BYF, respectively.

2.1. Laser emission at the 640-nm fundamental wavelength

Figure 2 presents the output power at 640 nm versus the absorbed power at 480 nm for the Pr:YLF crystal. The best performances were achieved with M3 of $T = 2.0\%$: the threshold was 37 mW, the slope efficiency with respect to the absorbed power (η_{sa}) was 40% and 72.4 mW of red light was measured for 217 mW of absorbed pump power. A Findlay-Clay analysis gave round-trip residual losses L_i below 0.02 (~0.017), while the coefficient K_c that relates the small-signal gain to the absorbed power was ~0.50 W^{-1}.

The output characteristics obtained with the Pr:BYF laser medium are shown in Fig. 3(a). A cw power of 51 mW at 640 nm for 205 mW absorbed pump power resulted for M3 with $T = 0.6\%$; the laser threshold power and η_{sa} were 33 mW and 30%, respectively. The losses L_i

#68599 - $15.00 USD
Received 1 March 2006; revised 3 April 2006; accepted 3 April 2006

were 0.01 and the coefficient K_c was evaluated to be ~0.19 W^{-1}. One can see that the residual losses L_i were slightly higher for Pr:YLF than those for Pr:BYF in spite of using uncoated Pr:BYF: it seems that the uncoated Pr:BYF end faces act as a low loss Fabry-Perot and that the greater length of the Pr:YLF crystal accounts for the higher losses in this material. From a Caird plot of the inverse slope efficiency versus inverse T of the output coupler [14] the limiting internal slope efficiency (η_0) was evaluated as 0.53 and 0.56 for Pr:YLF and Pr:BYF, respectively. Taking into account the pump, laser beam spot-sizes and confocal parameters, the laser-to-pump beam overlap efficiency (η_m) for the Pr:YLF and Pr:BYF was deduced to be 0.69 and 0.72, respectively. Thus, the maximum slope efficiency that could be obtained in our set-up, $\lambda_p/\lambda_\omega \times \eta_m$, is ~0.52 for Pr:YLF and ~0.54 for Pr:BYF. The agreement with η_0 is quite good, if one considers that for the procedure only four output mirrors were used in the experiments. A general view of the Pr:BYF laser is shown in Fig. 3(b).

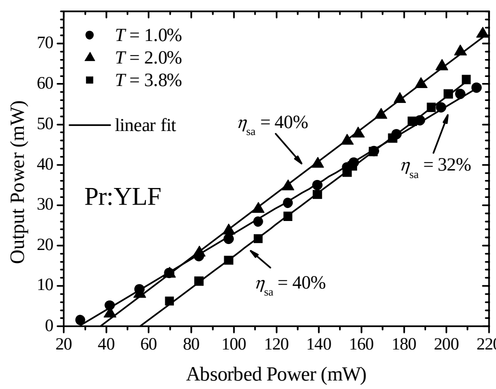

Fig. 2. Output power at 640 nm vs. the absorbed pump power at 480 nm of Pr:YLF at various output couplings T of mirror M3.

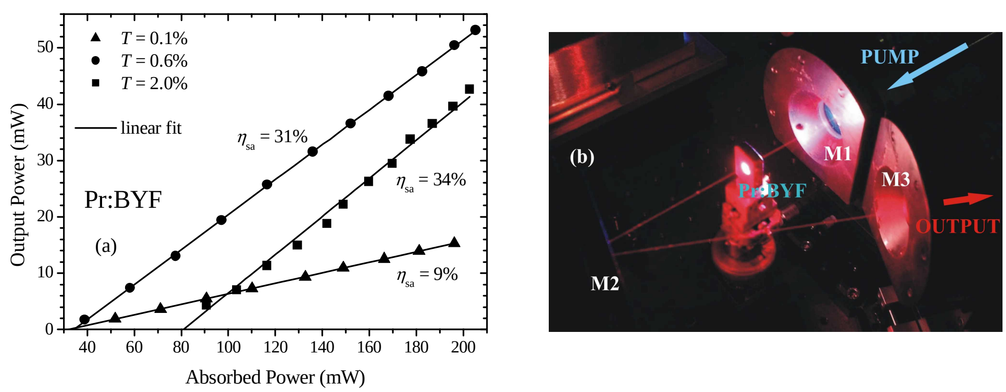

Fig. 3. (a) Output power at 640 nm vs. the absorbed power at 480 nm for the Pr:BYF laser and (b) a general view of the red emitting Pr:BYF laser.

2.2. UV generation at 320 nm

Cw UV generation at 320 nm was obtained through intracavity SHG by conventional birefringent phase matching. Because of a large acceptance in temperature (~9.3 K·cm) and of a moderate nonlinear coefficient (d_{eff}~0.54 pm/V), Lithium Triborate LBO was chosen as nonlinear medium. The LBO (provided by Ekspla Company, Lithuania) was designed for type I non-critical phase-matching ($\theta = 90^o$, $\phi = 53.5^o$) and operated at room temperature. Nonlinear LBO crystals of various lengths (3, 5, and 8 mm) were mounted in the M2-M3 arm of the resonator into the second beam waist. Both LBO's surfaces were AR coated for 320 nm

#68599 - $15.00 USD Received 1 March 2006; revised 3 April 2006; accepted 3 April 2006

and 640 nm. In these experiments M3 was HR ($T < 0.02\%$) coated for λ_{ω} and $\lambda_{2\omega}$. The UV radiation was coupled out through mirror M2 (as presented in Fig. 1).

Figure 4 shows the UV output power versus the absorbed pump power for the Pr:YLF laser. With an LBO crystal of 3 mm thickness the maximum power at 320 nm was 7.8 mW, whereas a 5-mm long LBO increased the UV power to 13.4 mW. The highest cw UV output power of 19 mW was obtained with an 8-mm long LBO nonlinear crystal: as 216 mW of pump power were absorbed in Pr:YLF, the optical-to-optical efficiency with respect to the absorbed power was ~9%. Moreover, based on results of Fig. 2(a), the conversion of the available fundamental power from the visible to the UV, $\eta_{\text{V-UV}}$ (the ratio between the maximum UV power and the red power obtained for $T = 2\%$ output mirror at the same pumping level) was determined to be 26%.

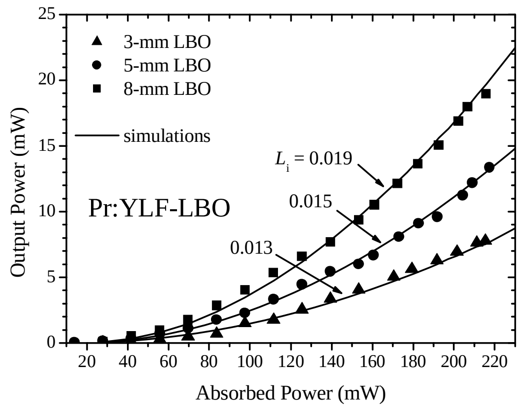

Fig. 4. Cw UV power at 320 nm vs. the absorbed pump power for the Pr:YLF laser with LBO crystals of various length. Continuous lines are simulations (according to the text).

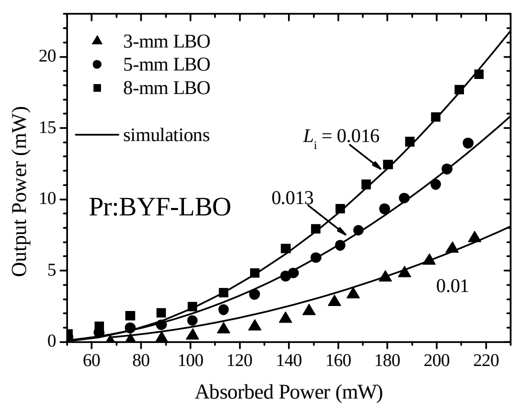

Fig. 5. Cw UV power at 320 nm vs. the absorbed pump power for the Pr:BYF laser.

The UV output power obtained with the Pr:BYF laser crystal is shown in Fig. 5. With the 8-mm long LBO we measured 18.9 mW at 320 nm: the overall efficiency with respect to absorbed power was ~9%, almost identical with that obtained with the Pr:YLF laser, but at an increased $\eta_{\text{V-UV}}$ of 36%. Cw UV radiation of 7.3 mW and 13.9 mW was obtained with the 3- and 5-mm thick LBO, respectively. The temperature of operation was ~23°C. A measurement of the power stability during 5 hours of operation resulted in peak-to-peak fluctuations of ±3%. These variations were attributed to mechanical instabilities of the laser set-up and to some weak output fluctuations of the OPS pump source. The noise of the UV radiation was recorded with a fast photodiode and an oscilloscope: it showed chaotic fluctuations of several MHz with a modulation depth up to 34%, similar to the well-known "green problem" [15].

#68599 - $15.00 USD Received 1 March 2006; revised 3 April 2006; accepted 3 April 2006

In order to evaluate the UV power ($P_{2\omega}$) as a function of the absorbed power at 480 nm (P_a) the plane-wave model introduced by Smith [16] was used. Considering that the optical pumping induces no thermal effect into the laser medium, the second-harmonic power can be written as [16, 17]:

$$P_{2\omega} = \frac{\pi\omega_1^2}{8k} I_S^2 \left\{ -\left(k + \frac{L_i}{I_S}\right) + \left[\left(k + \frac{L_i}{I_S}\right)^2 + 4\frac{k}{I_S}\left(2K_C P_a - L_i\right)\right]^{1/2} \right\}^2, \tag{1}$$

with the saturation intensity $I_s = h\nu_\omega/(\sigma_e\tau_f)$ of the 640-nm transition. The parameter k is the effective nonlinearity of the LBO nonlinear crystal:

$$k = \frac{4\pi^2}{\lambda_\omega^2} Z_0 \frac{d_{eff}^2 \ell^2}{n_2^3} \frac{\omega_1^2}{\omega_2^2} \beta, \tag{2}$$

where $Z_0 = 377\ \Omega$ represents the vacuum impedance, ℓ is the LBO length, $n_2 = 1.53$ is the refractive index at $\lambda_{2\omega}$, ω_1 and ω_2 are the beam waists in the laser medium and LBO, respectively, and $\beta \sim 2$ is a factor that takes into account the phase mismatch between the fundamental and the second harmonic wave in the second pass in the nonlinear crystal.

For the Pr:YLF configuration, the radius of the laser beam at the LBO position was evaluated as 63 µm and 72 µm in the tangential and sagittal plane, respectively, resulting in an average beam radius of ~67 µm. For the Pr:BYF laser, this parameter was 64 µm (61 µm in the tangential plane and 67 µm in the sagittal plane). Previous spectroscopic measurements [10,12,18] concluded that the emission cross section σ_e is 2.62×10^{-19} cm^2 in Pr:YLF ($E\perp c$, σ-polarization) and 2.0×10^{-19} cm^2 for Pr:BYF ($E\|b$ emission), with the fluorescence lifetime τ_f of 33 µs and 21 µs, respectively. An evaluation of K_c and losses L_i was made in Section 2.1. It is known, however, that the Findlay-Clay analysis is not very accurate in evaluation of the losses. On the other hand, intracavity frequency generation is very sensitive to resonator losses that directly limit the achievable internal fundamental power. Even small errors in determination of L_i, which may be increased by the insertion of the LBO nonlinear crystal for the intracavity frequency-doubled laser, can give large disagreement between experimental and theoretical results. For these reasons, we kept K_c constant and changed L_i around the values determined previously. Figures 4 and 5 show the theoretical modeling of the UV power by using the results of Eq. (1) as continuous lines: For example, a good fitting of the experimental results was obtained for $L_i \sim 0.019$ in the case of Pr:YLF and $L_i \sim 0.016$ for the Pr:BYF laser medium using the 8-mm long LBO crystal.

3. Conclusion

In conclusion, 19 mW of UV continuous-wave radiation at 320 nm with ~9% optical-to-optical efficiency with respect to absorbed power has been demonstrated by intracavity frequency doubling of Pr-lasers operating at the fundamental wavelength of 640 nm. To the best of our knowledge this is the first demonstration of such a 320 nm UV SHG laser and the first cw UV laser radiation source obtained by a single intracavity frequency doubling process. We expect efficient UV generation at various wavelengths, such as 360 nm, 303 nm, or 261 nm, by operating the Pr-lasers on other transitions in the visible spectral range. The UV-lasers are simple, robust, efficient, and offer many wavelengths in the visible and UV spectral region. The application potential of these lasers lies for instance in measurement techniques (cw, short wavelengths) and fluorescence microscopy.

Acknowledgments

N. Pavel acknowledges a post-doc scholarship of Alexander von Humboldt Foundation, Germany. He is on leave from the National Institute for Lasers, Plasma and Radiation Physics, Quantum Electronics Laboratory, Bucharest R-76900, Romania.