

Y. CHENG¹
H.J. ZHANG^{1,✉}
Y.G. YU¹
J.Y. WANG¹
X.T. TAO¹
J.H. LIU²
V. PETROV²
Z.C. LING³
H.R. XIA³
M.H. JIANG¹

Thermal properties and continuous-wave laser performance of Yb:LuVO₄ crystal

¹ State Key Laboratory of Crystal Materials, Institute of Crystal Materials, Shandong University, Jinan 250100, P.R. China

² Max-Born-Institute for Nonlinear Optics and Ultrafast Spectroscopy, Max-Born-Str. 2A, 12489 Berlin, Germany

³ Department of Physics, Shandong University, Jinan 250100, P.R. China

Received: 2 August 2006/Revised version: 9 October 2006
Published online: 5 December 2006 • © Springer-Verlag 2006

ABSTRACT A laser crystal of Yb:LuVO₄ with high optical quality was grown by the Czochralski technique. Its thermal properties including specific heat, thermal expansion coefficients, and thermal conductivities along the *a*- and *c*-axis have been measured for the first time. Continuous-wave laser output up to 3.5 W at 1031 nm was obtained at room temperature through end-pumping by a high-power diode laser. The corresponding optical conversion efficiency was 43% and the slope efficiency was 72%.

PACS 42.55.Xi; 66.60.+a

1 Introduction

Neodymium doped vanadate crystals have proven to be excellent laser materials [1–3]. Among these crystals, Nd:LuVO₄ is a relatively new one [3, 4], and its laser operation at 1.06 and 1.34 μm has been reported by our group [5, 6]. Recently, ytterbium (Yb) doped vanadate crystals, such as Yb:YVO₄ and Yb:GdVO₄, have also been developed as promising laser materials for building continuous-wave (cw) and ultrashort pulse sources in the 1 μm range [7–11].

Yb:LuVO₄, a new member of the Yb doped vanadates, appeared only very recently. Some preliminary laser results for room temperature cw operation have just been reported [12]. However, fundamental thermal properties like specific heat, thermal expansion coefficients, and thermal conductivity have not been studied yet. These thermal properties are very important for practical applications in laser oscillators and amplifiers, especially in the high-power regime, since the heat management sets the ultimate limit to the achievable power level. When a large temperature gradient is generated inside the laser crystal, this leads to thermal expansion, thermal lensing and thermally induced birefringence. These thermo-optic effects can seriously affect laser performance, modify resonator stability and even crack the crystals. Thermal properties are also important for the crystal growth and processing of active elements. If a crystal possesses large anisotropic thermal expansion coefficients, small specific heat and low

thermal conductivity, it may be easily cracked during its growth and processing when subjected to a large temperature gradient.

Thermal properties are therefore very important parameters in assessing the practical applications of any laser crystal. Furthermore, it is known that some thermal properties of laser host materials are affected by the doping process, e.g., the thermal conductivity of Yb:YAG is substantially reduced in comparison to that of undoped YAG. Therefore, it is important to measure all these thermal properties of Yb:LuVO₄ crystal. In this paper, we present our experimental results for the specific heat, thermal expansion coefficients, and thermal conductivities of Yb:LuVO₄, and also describe its cw laser performance at room temperature under high-power diode pumping.

2 Crystal growth and structure characterization of Yb:LuVO₄

Crystal growth of Yb:LuVO₄ has been achieved by the float zone method [13]. The most common technique in growing vanadate crystals is, however, the conventional Czochralski method, which can yield large size crystals with high optical quality. Following similar growth procedures reported recently for Nd:LuVO₄ crystals [14], we have grown large size Yb:LuVO₄ crystals.

Figure 1 shows one of Yb:LuVO₄ crystal boules grown from an *a*-oriented seed, the dimensions of which are about 18 × 35 × 25 mm³. The as-grown Yb:LuVO₄ crystal is jasmine, this color did not change after the crystal was annealed in a furnace at about 1100 °C for 10 h in air. Neither cracks nor small-angle boundaries were found inside this crystal boule. Its quality was checked by using a 5 mW He-Ne laser: no light scatter pellets were observed, indicating the high quality of the Yb:LuVO₄ crystal. Several samples of different sizes were prepared from this annealed crystal boule for use in the experiment.

The lattice structure of the as-grown Yb:LuVO₄ single crystal was studied by using X-ray powder diffraction (XRPD) technique ($\lambda = 1.54184 \text{ \AA}$). Based on the XRPD data acquired, the unit cell parameters of Yb:LuVO₄ crystal were calculated through the TEROR program [15].

The XRPD peaks obtained for a sample of Yb_{0.015}Lu_{0.985}VO₄ are displayed in Fig. 2a. Figure 2b shows the JCPDS file for LuVO₄ crystal [16]. One can see that

✉ Fax: +86-531-88574135, E-mail: hjzhang@icm.sdu.edu.cn

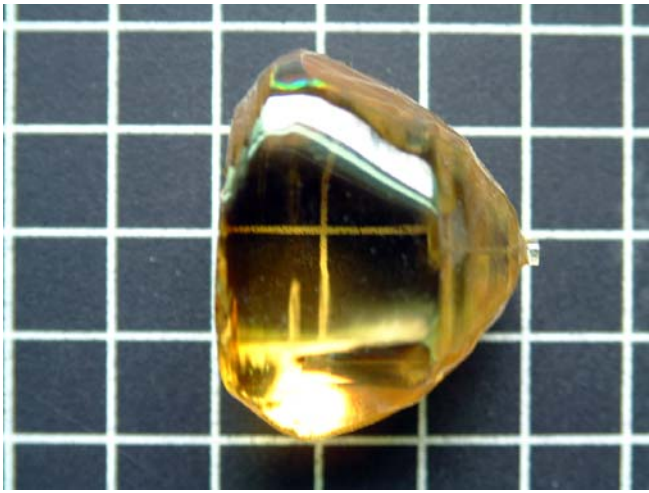


FIGURE 1 As-grown Yb:LuVO₄ crystal (the scale shown is 10 mm per division)

the diffraction peak positions and intensities of the grown Yb:LuVO₄ crystal are in accordance with the JCPDS file for LuVO₄, confirming that the grown Yb:LuVO₄ crystal possesses the structure of ZrSiO₄ with the space group of *I*4₁/amd. The unit-cell parameters calculated by the TREOR program are $a = b = 7.0389 \text{ \AA}$, $c = 6.2350 \text{ \AA}$, and $V = 308.920 \text{ \AA}^3$. These results are in close agreement with the data given in the JCPDS for LuVO₄ ($a = b = 7.0243 \text{ \AA}$, $c = 6.2316 \text{ \AA}$, and $V = 307.472 \text{ \AA}^3$).

3 Measurement of thermal properties

The thermal expansion coefficients and thermal conductivity, both being symmetrical second-rank tensors, possess only two independent nonzero components in their principal frames for Yb:LuVO₄, which belongs to the tetragonal crystallographic system. Accordingly, the corresponding tensors have a diagonal form

$$\begin{pmatrix} T_{11} & 0 & 0 \\ 0 & T_{22} & 0 \\ 0 & 0 & T_{33} \end{pmatrix}, \quad (1)$$

where T_{11} , T_{22} and T_{33} are the principal components with $T_{11} = T_{22}$.

The average linear thermal expansion coefficients along the a -, b - and c -axes can be calculated according to the following formula:

$$\alpha = \frac{\Delta L}{L_0} \frac{1}{\Delta T}, \quad (2)$$

where α is the average linear thermal expansion coefficient; L_0 is the sample length at T_0 (initial temperature); ΔL is the length change when the temperature changes from T_0 to T (final temperature); and $\Delta T = T - T_0$.

The thermal conductivity components of Yb:LuVO₄ can be calculated from the measured thermal diffusion coefficients along a - and c -axes with the following expression

$$\kappa = \lambda \rho C_p, \quad (3)$$

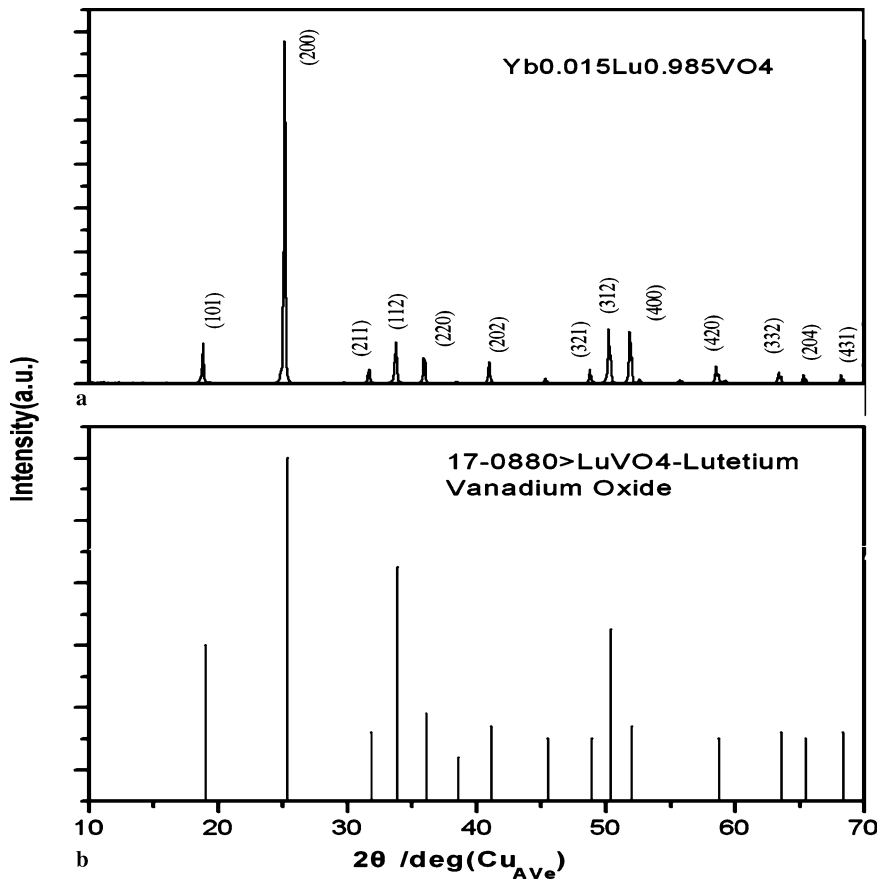


FIGURE 2 X-ray powder diffraction patterns: (a) as-grown Yb:LuVO₄ crystal; (b) JCPDS file:17-0880

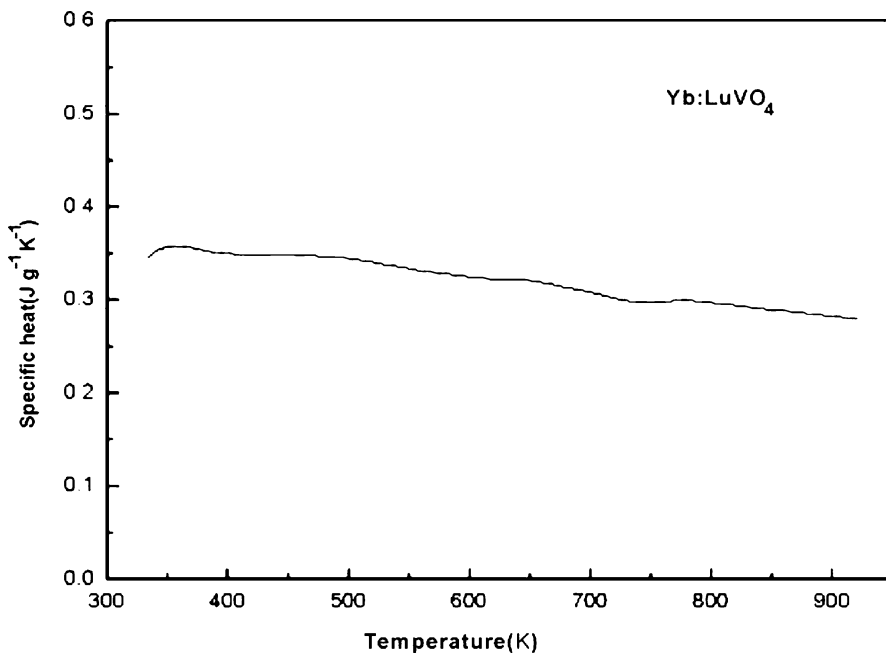


FIGURE 3 Specific heat of Yb:LuVO₄ crystal versus temperature

where λ is the thermal diffusivity coefficient, ρ the density, and C_p the specific heat.

A thermo-mechanical analyzer (TMA, Perkin-Elmer) and a differential scanning calorimeter (DSC) were employed to measure the thermal expansion coefficients and the specific heat. The crystal used for thermal expansion coefficients was $5.97 \times 6.50 \times 7.12 \text{ mm}^3$ ($a \times b \times c$). The measurement was performed in a temperature range from 25 to 300 °C, and the thermal expansion coefficients were calculated to be $\alpha_a = \alpha_b = 6.35 \times 10^{-6}$, and $\alpha_c = 13.3 \times 10^{-6} \text{ K}^{-1}$, along the a -, b - and c -axis, respectively. One can see the considerable anisotropy in the thermal expansion of Yb:LuVO₄ crystal. This implies that crystals of Yb:LuVO₄ must be cooled down to room temperature at a low rate after the growth; otherwise cracks will easily occur inside the crystals. In our crystal-growing process, the cooling rate was chosen to be 30–50 °C/h to obtain high-quality crystals of large size.

Figure 3 shows the variation of the measured specific heat of Yb:LuVO₄ with temperature. It is seen that the specific heat changes approximately linearly with temperature, decreasing smoothly from 0.34 to 0.28 J g⁻¹K⁻¹, when the temperature is increased from 50 to 500 °C. This behavior is different from that observed in other Nd-doped vanadate crystals [17, 20, 21], further study should be done to explain this in the future.

The thermal diffusivity coefficient was measured with the laser flash method (NETZSCH LFA 447 Nanoflash), from which the thermal conductivity can be calculated. Two thin Yb:LuVO₄ plates of size $6 \times 6 \times 2 \text{ mm}^3$ cut along the a - and c -axes were used in the thermal diffusivity measurement.

Figure 4 shows the temperature dependence of the thermal conductivities of Yb:LuVO₄ crystal along the a - and c -axis (κ_a, κ_c), determined by use of (3) (the crystal density is about 6.33 g/cm³). As expected, the magnitude of the thermal conductivity is greatly reduced when the crystal temperature rises from 29 to 290 °C: κ_a decreases from 5.14 to 1.96 Wm⁻¹K⁻¹, and κ_c from 5.78 to 2.19 Wm⁻¹K⁻¹. From this figure one

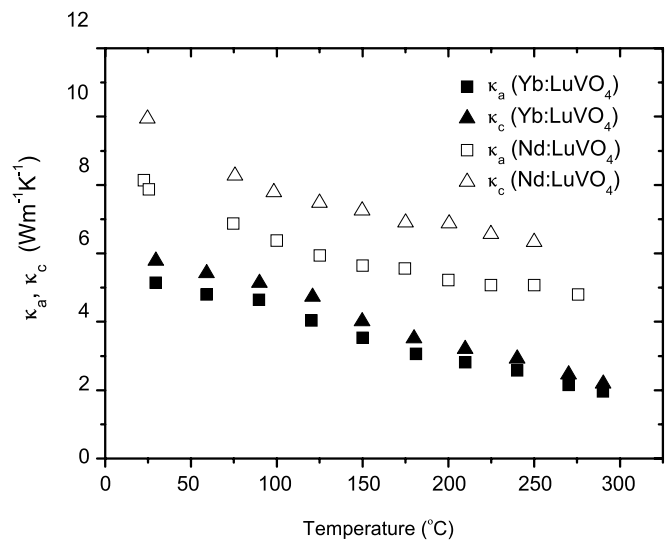


FIGURE 4 Thermal conductivity of Yb(1.56%):LuVO₄ and Nd(0.9%):LuVO₄ crystals along the a - and c -axes

also notices that for the whole temperature range in which the measurement was conducted, the thermal conductivity along the c -axis is larger than along the a -axis by a factor of about 1.12. This means that the anisotropy of thermal conductivity is not significant in Yb:LuVO₄ crystals.

For comparison, Fig. 4 also illustrates the variation with temperature of the thermal conductivities of Nd:LuVO₄, determined with the same technique using samples of the same sizes. In comparison with Yb:LuVO₄, Nd:LuVO₄ has approximately 1.5–2.0 times larger thermal conductivities over the temperature range of measurement. It can also be seen that the trend of reduction in the magnitude of the thermal conductivity with temperature rising is more pronounced in the case of Yb:LuVO₄ in comparison to Nd:LuVO₄. In general it can be expected that Yb-doping affects the crystal lattice vibrations and hence the thermal conductivity to

crystals	Nd:YVO ₄ ^{a,b,c}	Nd:GdVO ₄ ^{d,e}	Nd:LuVO ₄ ^{f,g} (0.9 at. %)	Yb:GdVO ₄ ^h (2 at. %)	Yb:LuVO ₄ ^f (1.56 at. %)	YbVO ₄ ^k
Thermal conductivity (W/mK)	<i>c</i> : 5.23, or 11 <i>a</i> : 5.10	<i>c</i> : 11.4 <i>a</i> : 10.1	<i>c</i> : 9.94 <i>a</i> : 8.14	<i>c</i> : 8.1 <i>a</i> : 7.1	<i>c</i> : 5.78 <i>a</i> : 5.14	<i>c</i> : 5.1 <i>a</i> : 3.9
Thermal expansion coefficient (10 ⁻⁶ /K)	<i>a</i> : 2.2	<i>a</i> : 1.5	<i>a</i> : 1.7	–	<i>a</i> : 6.3	<i>a</i> : 2.1
Specific heat at 330 K (cal/mol K)	<i>c</i> : 8.4	<i>c</i> : 7.3	<i>c</i> : 9.1	–	<i>c</i> : 13.3	<i>c</i> : 8.3
(J/g K)	24.6	32.6	31.6		23.58	25.47
Density (g/cm ³)	0.50	0.50			0.34	0.37
	4.22	5.45	6.33	5.45	6.33	6.17

^a Ref. [17]

^b Ref. [8] Nd doping concentration is 0.5 at. %

^c Ref. [18]

^d Ref. [19] Nd doping concentration is 0.7 at. %

^e Ref. [20]

^f this work

^g Ref. [21]

^h Ref. [7]

^k Ref. [22]

TABLE 1 Thermal mechanical properties of Yb- and Nd-doped vanadates at room temperature (the indicated doping level refers to the crystal)

a much less extent than Nd-doping, because compared to Nd-ion, Lu-ion is much closer in size and mass to Yb-ion. The opposite trend observed here is attributed to the different doping levels in the Yb:LuVO₄ (1.56 at. % in the crystal) and Nd:LuVO₄ (0.9 at. % in the crystal) samples used in the present study and the small thermal conductivity of YbVO₄ crystal [22]. Some of the important thermal properties of Yb- and Nd-doped vanadates at room temperature are summarized in Table 1. From this table, we can see that the thermal conductivities of Yb doping vanadate crystals are smaller than the Nd doping ones. (One can compare the thermal conductivity of Nd:GdVO₄ and Yb:GdVO₄ crystal, and the difference is about 3–4, and our measured results of Yb:LuVO₄ and Nd:LuVO₄ are almost the same.).

4 cw laser performance

The cw laser performance of Yb:LuVO₄ using a sample from the same boule was studied and a high-power high-brightness fiber-coupled diode laser (200 μm fiber core diameter, 0.2 NA, Apollo) with an output of 50 W of output power at emission wavelengths of 974–983 nm, depending on the output level, was used as pumping source. A 1.5 mm thick, uncoated *a*-cut Yb:LuVO₄ crystal, fixed to a water-cooled copper holder, was placed into a plano-concave resonator which was arranged in a hemispherical configuration. A 25 mm radius-of-curvature concave mirror with a transmittance of $T = 1%$ at 1030 nm served as the output coupler. The total physical cavity length was about 26 mm. The pumping light was focused by a re-imaging unit and delivered through the plane mirror upon the laser crystal with a spot radius of about 100 μm.

Figure 5 shows the relationship between the output power and the absorbed pump power obtained with the Yb:LuVO₄ laser. The laser reached its threshold at an absorbed pump power of 3.3 W. The output of the laser was linearly polarized parallel to the *c*-axis of the Yb:LuVO₄ crystal (π polarization), in consistency with the previous work [12]. At an absorbed pump power of 8.1 W, the laser produced an output power of 3.5 W at an emission wavelength of 1031 nm, resulting in an optical conversion efficiency of 43%. The slope

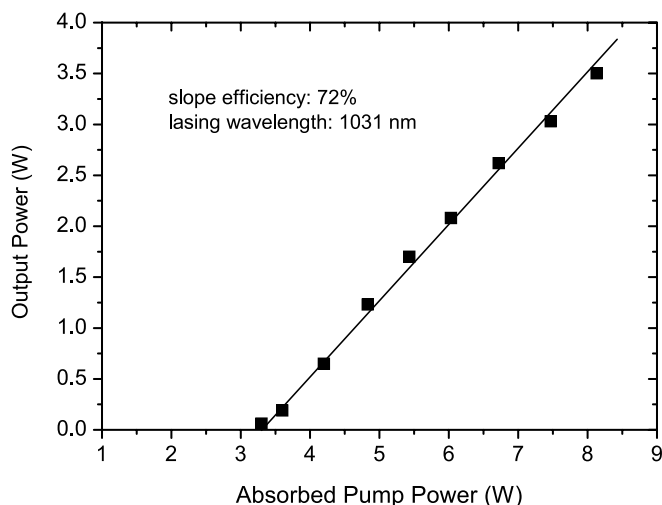


FIGURE 5 Output power versus absorbed pump power of the diode-pumped Yb:LuVO₄ laser

efficiency within the whole operational range was determined to be 72%. From the presented output–input dependence it can be seen that no output saturation occurs at the highest pump level applied in the experiment, suggesting that further power scaling should be possible. (Under the present cooling conditions used in the experiment, the maximum output power generated by the Yb:LuVO₄ laser was limited. To scale the laser to higher output power, it is essential to use a more efficient cooling system.)

5 Conclusions

Yb:LuVO₄ crystals of high optical quality were grown by the Czochralski method. It was confirmed by X-ray powder diffraction that they possess the structure of ZrSiO₄. The thermal expansion coefficients, specific heat and thermal conductivities of Yb:LuVO₄ have been determined experimentally. Room temperature high power cw laser operation of Yb:LuVO₄ crystal was achieved under diode pumping, generating an output power of 3.5 W with an optical efficiency of 43% and a slope efficiency as high as 72%.

ACKNOWLEDGEMENTS This work is supported by the National Natural Science Foundation of China (No. 50572054, No. 50590401 and No. 50325311), the Excellent Young Teachers Program of MOE, P.R. China, and the Grand for the State Key Program of China (2004CB619002).

REFERENCES

- 1 R.A. Fiefls, M. Birnbaum, C.L. Fincher, *Appl. Phys. Lett.* **51**, 1885 (1987)
- 2 T. Jensen, V.G. Ostroumov, J.-P. Meyn, G. Huber, A.I. Zagumennyi, I.A. Scherbakov, *Appl. Phys. B* **58**, 373 (1994)
- 3 C. Maunier, J.L. Doualan, R. Moncorge, A. Speghini, M. Bettinelli, E. Cavalli, *J. Opt. Soc. Am. B* **19**, 1794 (2002)
- 4 H. Zhang, H. Kong, S. Zhao, J. Liu, J. Wang, Z. Wang, L. Gao, C. Du, X. Hu, X. Xu, Z. Shao, M. Jiang, *J. Cryst. Growth* **256**, 292 (2003)
- 5 J. Liu, H. Zhang, Z. Wang, J. Wang, Z. Shao, M. Jiang, H. Weber, *Opt. Lett.* **29**, 168 (2004)
- 6 H. Zhang, J. Liu, J. Wang, X. Xu, M. Jiang, *Appl. Opt.* **44**, 7439 (2005)
- 7 J. Petit, B. Viana, P. Goldner, D. Vivien, P. Louiseau, B. Ferrand, *Opt. Lett.* **29**, 833 (2004)
- 8 C. Kränkel, D. Fagundes-Peters, S.T. Fredrich, J. Johannsen, M. Mond, G. Huber, M. Bernhagen, R. Uecker, *Appl. Phys. B* **79**, 543 (2004)
- 9 V.E. Kisel, A.E. Troshin, N.A. Tolstik, V.G. Shcherbitsky, N.V. Kuleshov, V.N. Matrosov, T.A. Matrosova, M.I. Kupchenko, *Opt. Lett.* **29**, 2491 (2004)
- 10 V.E. Kisel, A.E. Troshin, V.G. Shcherbitsky, N.V. Kuleshov, V.N. Matrosov, T.A. Matrosova, M.I. Kupchenko, F. Brunner, R. Paschotta, F. Morier-Genoud, U. Keller, *Opt. Lett.* **30**, 1150 (2005)
- 11 A.A. Lagatsky, A.R. Sarmani, C.T.A. Brown, W. Sibbett, V.E. Kisel, A.G. Selivanov, I.A. Denisov, A.E. Troshin, K.V. Yumashev, N.V. Kuleshov, V.N. Matrosov, T.A. Matrosova, M.I. Kupchenko, *Opt. Lett.* **30**, 3234 (2005)
- 12 J. Liu, X. Mateos, H. Zhang, J. Wang, M. Jiang, U. Griebner, V. Petrov, *Opt. Lett.* **30**, 3162 (2005)
- 13 M. Higuchi, T. Shimizu, J. Takahashi, T. Ogawa, Y. Urata, T. Miura, S. Wada, H. Machida, *J. Cryst. Growth* **283**, 100 (2005)
- 14 S. Zhao, H. Zhang, J. Liu, J. Wang, X. Xu, Z. Zhao, J. Xu, M. Jiang, *J. Cryst. Growth* **279**, 146 (2005)
- 15 L. Eriksson, M. Westdahl, *J. Appl. Cryst.* **18**, 367 (1985)
- 16 Joint Committee for Power Diffraction Standards diffraction file: 17-0880
- 17 H.J. Zhang, L. Zhu, X.L. Meng, Z.H. Yang, C.Q. Wang, W.T. Yu, Y.T. Chow, M.K. Lu, *Cryst. Res. Technol.* **34**, 1011 (1999)
- 18 B.H.T. Chai, G. Loutts, J. Lefaucherur, X.X. Zhang, P. Hong, M. Bass, I.A. Shcherbakov, A.I. Zagumennyi, In: *Adv. Solid-State Lasers*, ed. by T.Y. Fan, B.H.T. Chai, Vol. 20 of OSA Trends in Optical and Photonics Series (Opt. Soc. of Amer., Washington DC, 1994) pp. 41
- 19 L.J. Qin, X.L. Meng, H.Y. Shen, L. Zhu, B.C. Xu, L.X. Huang, H.R. Xia, P. Zhao, G. Zheng, *Cryst. Res. Technol.* **38**, 793 (2003)
- 20 H. Zhang, J. Liu, J. Wang, C. Wang, L. Zhu, Z. Shao, X. Meng, X. Hu, M. Jiang, Y.T. Chow, *J. Opt. Soc. Am. B* **19**, 18 (2002)
- 21 S. Zhao, H. Zhang, J. Wang, H. Kong, X. Cheng, J. Liu, J. Li, Y. Lin, X. Hu, X. Xu, X. Wang, Z. Shao, M. Jiang, *Opt. Mater.* **26**, 319 (2004)
- 22 Y. Yu, Y. Cheng, H. Zhang, J. Wang, X. Cheng, H. Xia, *Mater. Lett.* **60**, 1014 (2006)