Comparative investigation of long-wave infrared generation based on $ZnGeP_2$ and CdSe optical parametric oscillators*

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Long-wave infrared (IR) generation based on type-II (o→e+o) phase matching $ZnGeP_2$ (ZGP) and CdSe optical parametric oscillators (OPOs) pumped by a 2.05 μm Tm,Ho:$GdVO_4$ laser is reported. The comparisons of the birefringent walk-off effect and the oscillation threshold between ZGP and CdSe OPOs are performed theoretically and experimentally. For the ZGP OPO, up to 419 mW output at 8.04 μm is obtained at the 8 kHz pump pulse repetition frequency (PRF) with a slope efficiency of 7.6%. This ZGP OPO can be continuously tuned from 7.8 to 8.5 μm. For the CdSe OPO, we demonstrate a 64 mW output at 8.9 μm with a single crystal 28 mm in length.

Keywords: long-wave infrared generation, ZGP/CdSe optical parametric oscillator, birefringent walk-off effect, oscillation threshold

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1. Introduction

Lasers operating in the atmospheric transmission window of 8–12 μm are of great important for many applications, including spectra, chemical monitoring, gas analysis, biomedical applications, remote sensing and infrared (IR) countermeasures. However, due to the lack of a direct laser gain medium to obtain this long-IR laser radiation, an alternative way of producing the laser in this wavelength range is to use the nonlinear optical frequency down-conversion of shorter wavelength sources based on high-quality infrared transmitting crystals with good nonlinear properties, such as CdSe, GaSe, $AgGaSe_2$ and $ZnGeP_2$.[1−5] In particular, the very high nonlinear optical coefficient of $ZnGeP_2$ (ZGP, $d_{\rm eff}$ = 75 pm/V), combined with its excellent optical, mechanical and thermal properties, makes it the most promising nonlinear medium for optical parametric oscillators (OPOs) to generate high-power mid-IR and long-IR laser outputs.[4−7] Another potentially excellent nonlinear medium for long-wave IR generation is CdSe. Although it has a much smaller nonlinear optical coefficient ($d_{\rm eff}$ = 18 pm/V), long (> 5 cm) crystals can be grown routinely, and it possesses small birefringent walk-off.[2,3] In addition, its transparency range is 0.75–25 μm, wider than $ZnGeP_2$ (0.75–12 μm[8]), and its optical loss is extremely low (< 0.01 cm^{-1}) in the 1–10 μm range.[3] As well as an excellent nonlinear medium, some of the other parameters of the laser, such as pump diameter, pump spectral linewidth, pump angular divergence, cavity length, crystal length and birefringent walk-off effect, should also be considered to obtain the high efficiency and low threshold oscillation for the long-infrared OPO output.[9−11]

In this paper, we report on long-wave IR generation based on type-II (o→e+o) phase matching ZGP and CdSe OPOs pumped by a pulsed 2.05 μm Tm,Ho:$GdVO_4$ laser. In particular, the birefringent walk-off effect and the oscillation threshold are compared theoretically and experimentally. For the ZGP OPO, an up to 419 mW output at 8.04 μm is obtained under 8 kHz pump pulse repetition frequency (PRF) with a slope efficiency of 7.6%. This ZGP OPO can be continuously tuned from 7.8 to 8.5 μm. For the CdSe OPO, only 64 mW output is obtained at 8.9 μm due to the short length of the crystal used in the experiment.

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2. OPO threshold analysis

To obtain the high-power long-wave IR generation, on the one hand the pump laser should have a narrow linewidth, a small angular divergence and a high brightness, and on the other the OPO cavity should be designed to minimize the oscillation threshold. In this section, we focus on the difference in oscillation threshold between ZGP and CdSe OPOs, and quantitatively point out that the CdSe OPO has a much higher oscillation threshold than that of the ZGP OPO due to its small nonlinear optical coefficient. Therefore, the CdSe OPO always requires a much longer crystal or a dual-crystal for the walk-off compensation to reduce the oscillation threshold.[2,12]

Here, we use the analytical expression derived by Brosnan and Byer[13] to estimate the oscillation thresholds of ZGP and CdSe OPOs. Although the result from the analytical expression is slightly higher than that from a numerical calculation, it is of adequate accuracy for design purposes. The analytical expression for the threshold pump fluence $F_{\rm th}$ (J/cm^2) of a pulse pumping a singly resonant oscillator (SRO) is given by

$$F_{\rm th} = \frac{2.25\tau}{k g_{\rm s} \zeta^2}\Big(\frac{L}{2\tau c}\ln\frac{P_n}{P_0} + 2\alpha l + \ln\frac{1}{\sqrt{R}} + \ln 2\Big)^2, \quad (1)$$

where τ is the $1/e^2$ pump pulse width, k is the nonlinear coupling constant, $g_{\rm s}$ is the signal beam spatial mode coupling coefficient, ζ is the effective parametric gain length, L is the optical cavity length, c is the velocity of light, α is the absorption coefficient at the signal wavelength, l is the crystal length and R is the reflectivity of the output coupler. The nonlinear coupling constant and the signal beam spatial mode coupling coefficient, $g_{\rm s}$, are defined as

$$k = \frac{8\pi^2 d_{\rm eff}^2}{\lambda_{\rm s}\lambda_{\rm i} n_{\rm s} n_{\rm i} n_{\rm p} \varepsilon_0 c}, \quad (2)$$

$$g_s = \frac{1}{1 + (w_{\rm s} + w_{\rm p})^2}, \quad (3)$$

where $d_{\rm eff}$ is the effective nonlinear coefficient, ε_0 is the permittivity of free space, $\lambda_{\rm s}$ and $\lambda_{\rm i}$ are the wavelengths of the signal and idler waves, respectively, $n_{\rm s}$, $n_{\rm i}$ and $n_{\rm p}$ are the refractive indices of the three waves, and $w_{\rm p}$ and $w_{\rm s}$ are the beam radii of the signal and pump waves, respectively. It should be pointed out that the effective parametric gain length of ζ is not equal to the actual crystal length due to the birefringent walk-off effect, and can be expressed as

$$\zeta = l_{\rm w}\,{\rm erf}\Big(\frac{\sqrt{\pi}}{2}\frac{l}{l_{\rm w}}\Big), \quad (4)$$

where $l_{\rm w}$ is the walk-off length given by

$$l_{\rm w} = \frac{\sqrt{\pi}}{2}\frac{w_{\rm p}}{\rho}\sqrt{\frac{w_{\rm p}^2 + w_{\rm s}^2}{w_{\rm p}^2 + w_{\rm s}^2/2}}, \quad (5)$$

with ρ being the walk-off angle. The signal radius $w_{\rm s}$ can be numerically solved from Eq. (6), in which the $w_{\rm p}$, $w_{\rm s}$ and $\lambda_{\rm s}$ are related by

$$\Big(\frac{\pi}{2L\lambda_{\rm s}}\Big)w_{\rm s}^6 + w_{\rm s}^2 - \frac{w_{\rm p}^2}{2} = 0. \quad (6)$$

The walk-off angle ρ is given by[14]

$$\tan\rho = n^2\Big[\Big(\frac{k_x}{n^{-2} - n_x^{-2}}\Big)^2 + \Big(\frac{k_y}{n^{-2} - n_y^{-2}}\Big)^2 + \Big(\frac{k_z}{n^{-2} - n_z^{-2}}\Big)^2\Big]^{-1/2}, \quad (7)$$

where k_x, k_y and k_z are the wave propagation directions of k, n is the refractive index in direction k, and n_x, n_y and n_z are the principal axis refractive indices. Combining Eqs. (1)–(7), we can easily obtain the SRO oscillation thresholds for the ZGP and CdSe crystals.

2.1. ZGP OPO

For the type-II phase matching ZGP OPO (with o, e and o polarizations for idler, signal and pump, respectively), the walk-off arises by the e polarization of the signal. Here we assume that the signal is at 2.75 μm and the idler is at 8.04 μm with a 2.05 μm pump. The corresponding phase matching angle ($\theta_{\rm pm}$) is 65.4° with $\varphi = 45°$, where φ is the azimuth angle measured from x. With the help of the Sellmeier equation derived by Zelmon and Hanning,[15] the calculated walk-off angle is 9.4 mrad. The OPO is assumed to resonate with $R = 100\%$ at the signal and totally couple out the idler. The effective parametric gain length of ζ is calculated as a function of the crystal length, and threshold pump fluence $F_{\rm th}$ is calculated as a function of the pump beam radius. The $1/e^2$ pump pulse width τ, the crystal absorption coefficient α at the signal wavelength, and the nonlinear optical coefficient $d_{\rm eff}$ in the calculation are assumed to be 85 ns (corresponding to a full width at half maximum (FWHM) of about 50 ns), 0.03 cm^{-1} and 75 pm/V, respectively. For threshold pump fluence $F_{\rm th}$, the results at three different lengths of crystals, 12, 17 and 20 mm, are compared. The OPO physical cavity length in the calculation is assumed to be 5 mm longer than the crystal length, i.e. 17, 22, and 25 mm, respectively.

Figure 1 shows the calculated effective parametric gain lengths each as a function of the crystal length under three different pump beam radii, 0.42, 0.22, and 0.18 mm. It is obvious that the effective parametric gain length is shorter than the crystal length due to the birefringent walk-off effect, and it becomes even shorter under a smaller pump beam radius. Figure 2(a) shows the calculated threshold energy fluencies each as a function of the pump beam radius. As shown in Fig. 2(a), the threshold energy fluence is approximately 0.23 J/cm^2 for the 17 mm long ZGP crystal when the pump beam radius is 0.4 mm. The corresponding threshold pump power is about 5.8 W if the pump pulse repetition frequency is 10 kHz, as shown in Fig. 2(b). All these values are related to the actual ZGP OPO experiment and will be discussed in the next section.

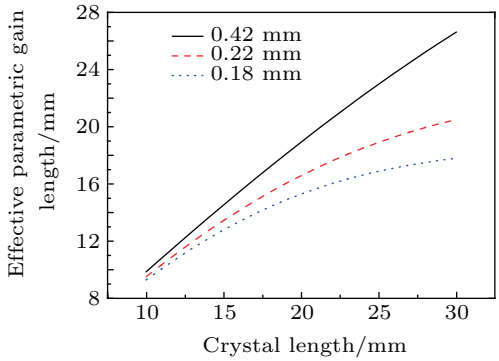


Fig. 1. (colour online) Effective parametric gain length as a function of crystal length for type-II phase-matching ZGP crystals with a phase matching angle of 65.4°.

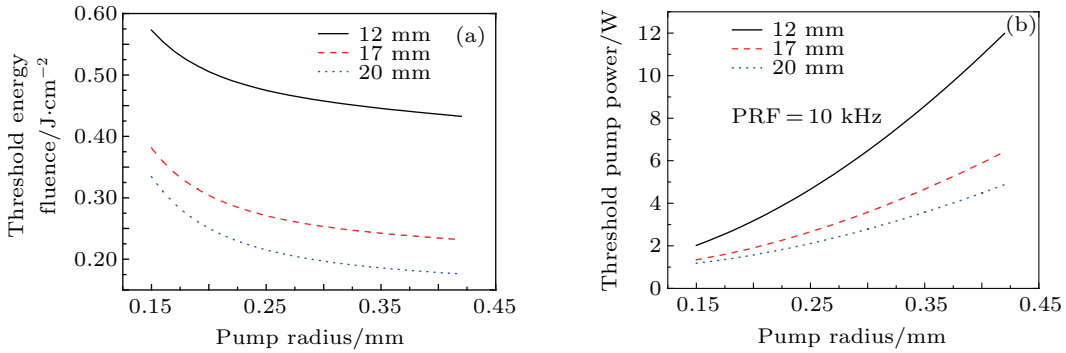


Fig. 2. (colour online) (a) Calculated threshold energy fluence and (b) threshold pump power as functions of pump radius for ZGP OPO at a 10 kHz pulse repetition frequency.

2.2. CdSe OPO

The signal of the CdSe OPO is assumed to be at 2.66 μm, and the idler is at 8.9 μm with the pump at 2.05 μm. The corresponding phase matching angle ($\theta_{\rm pm}$) is around 84° with φ=45°. With the help of the Sellmeier equation derived by Bhar,[16] the calculated walk-off angle is 1.7 mrad for phase matching angle 84°. It is obvious that the walk-off angle of CdSe is much smaller than that of ZGP. One reason is that the phase matching angle of CdSe is close to the non-critical phase matching case. Another reason is that CdSe has relatively low intrinsic birefringence compared with that of ZGP. Using the same assumption and the calculation procedure for ZGP, we show the calculated effective parametric gain length and the threshold in Figs. 3 and 4, respectively. As shown in Fig. 3, the effective parametric gain lengths are only slightly shorter than the crystal length, even with the smallest pump radius of 0.18 mm. This indicates that we can use a long CdSe crystal to obtain an efficient parametric gain despite the small nonlinear optical coefficient. From Fig. 4, we can see that the 60 mm long CdSe has a comparative threshold level with that of the 20 mm long ZGP. With 0.4 mm pump radius, the threshold pump power of the 30 mm long CdSe is about 16 W, as shown in Fig. 4(b), which is nearly 4 times that of the 20 mm long ZGP shown in Fig. 2(b). It is unrealistic to reduce the pump radius to lower

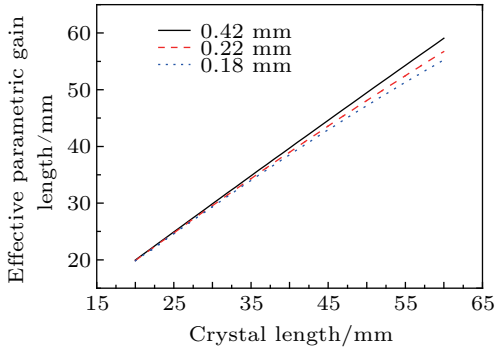


Fig. 3. (colour online) Effective parametric gain length as a function of crystal length for type-II phase matching CdSe crystals with a phase matching angle of 84°.

the oscillation threshold since this increases the risk of crystal damage. As a result, the CdSe OPO always requires a much longer crystal or a dual-crystal for walk-off compensation to reduce the oscillation threshold as pointed out previously.

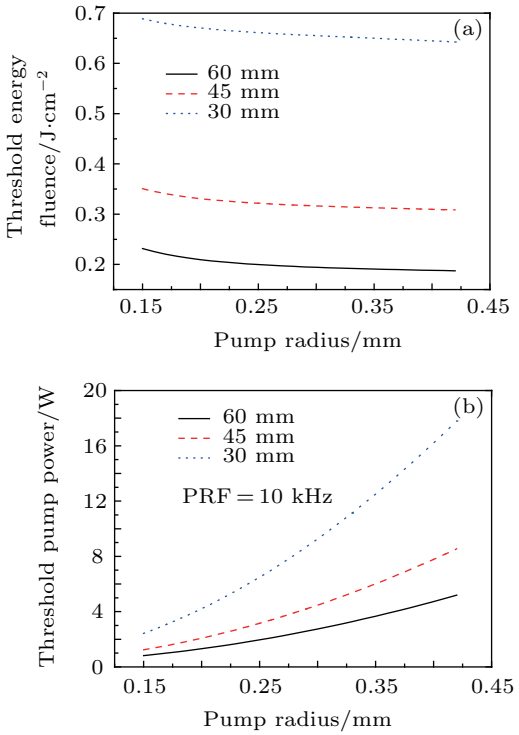


Fig. 4. (colour online) (a) Calculated threshold energy fluence and (b) threshold pump power as functions of pump radius for CdSe OPO at a 10 kHz pulse repetition frequency.

3. Experimental setup and results

A schematic diagram of the experimental setup is shown in Fig. 5. The pump source is a Q-switched Tm,Ho:$GdVO_4$ laser with a 10 kHz pulse repetition rate. This Tm,Ho:$GdVO_4$ laser has been used previously to generate more than 14 W output from a ZGP OPO tuned to the 3–5 μm region.[17] In our present experiment, the Tm,Ho:$GdVO_4$ laser can only deliver 17 W output at 2.05 μm due to some unexpected contaminations in the laser cavity. The Tm,Ho:$GdVO_4$ laser has a good beam quality with an M^2 value less than 1.1 and a minimum pulse width (FWHM) of 30 ns. The OPO cavity consists of two mirrors, M1 and M2, where M1 has high reflectivity in the signal wavelength range of 2.6–2.9 μm, a moderate reflectivity of 23% for the 8 μm idler, and high transmission (> 99%) for the 2.05 μm pump. Mirror M2 has the same coatings as mirror M1, and is used as an output coupler. Mirror M3 is a dichroic mirror, and is used to transmit the idler and reflect the pump. To determine the wavelength of the OPO output, the idler is passed through a 300 mm grating monochromator (WDZ30) and then detected using an HgCdTe detector, which is combined with a 350 MHz digital oscilloscope (wavejet 332, Lecroy). The blazed wavelength of the monochromator is 10 μm. The ZGP crystal used in the experiment is 5.2 mm × 5.5 mm in cross section and 16.4 mm in length. Both surfaces of the crystal are flat and antireflection-coated for the pump, signal and idler wavelength bands. The crystal is cut at $\theta = 65.4°$ for type-II phase matching (with e, o and o polarizations for the signal, idler and pump, respectively). The measured absorption coefficient for the 2.05 μm o-light pump of the ZGP crystal is about 0.09 cm^{-1}. The pump diameter in the input surface of the ZGP crystal is measured to be approximately 0.8 mm by using the 90/10 knife-edge technique, and the ZGP OPO cavity length is kept at 22 mm. The CdSe crystal is 6 ×8 mm in cross section and 28 mm in length, and the nominal cut angle of θ, given by the supplier, is 78°. Both surfaces of the crystal are flat and antireflection-coated for the pump, signal and idler wavelength bands. The measured absorption coefficient for the 2.05 μm o-light pump of the CdSe crystal is about 0.12 cm^{-1}. This relatively large absorption coefficient indicates that the CdSe crystal quality is not as good as expected. The CdSe OPO cavity length is increased to 32 mm and the pump radius is reduced to 0.21 mm to lower the oscillation threshold, as shown in Fig. 4(b).

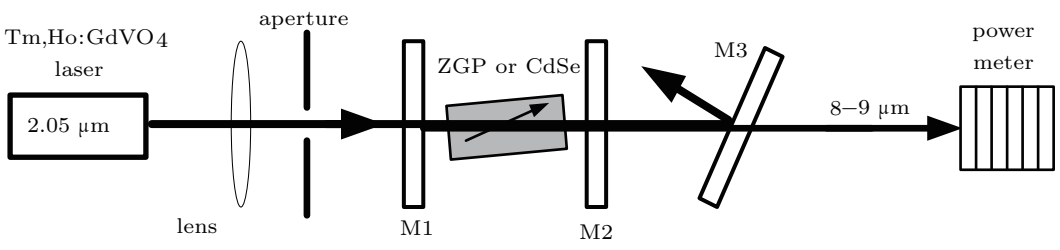


Fig. 5. The experimental setup for the ZGP and CdSe OPOs.

Figure 6(a) shows the ZGP OPO output power as a function of the incident pump power at 8, 10, and 12 kHz pulse repetition frequencies, respectively, when the wavelength is tuned at 8.04 μm. At 8 kHz, the maximum output power is 419 mW with a slope efficiency of 7.6% and an optical conversion efficiency of 4.9%. The incident pump power is limited, since the pump input surface coatings will be damaged if the pump power is further increased. The experimental threshold pump energy fluences are

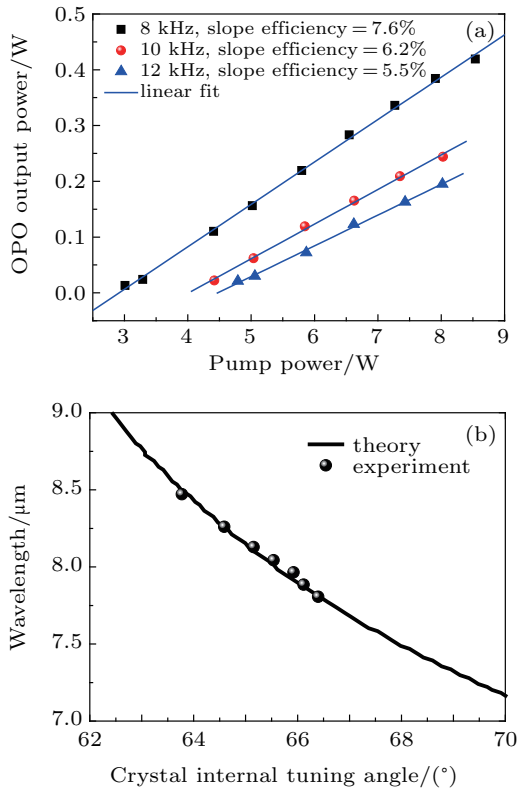


Fig. 6. (colour online) (a) The idler output power at 8.04 μm of the ZGP OPO as a function of incident pump power under different pulse repetition frequencies, and (b) the idler wavelength tuning curve between 7.8 and 8.5 μm; the theoretical curve is based on the Sellmeier equation.[15]

Table 1. ZGP OPO lasing performance at 8.04 μm.

PRF/kHz	Threshold pump pump power /W	Threshold energy fluence /J·cm^{-2}	Maximum output /mW (slope efficiency /%)
8	3.01	0.15	419 (7.6)
10	4.42	0.17	244 (6.2)
12	4.79	0.16	195 (5.5)

0.15, 0.17, and 0.16 J/cm^2 for the pulse repetition frequencies of 8, 10, and 12 kHz, respectively, as shown in Table 1, which are in reasonable agreement with the calculated data shown in Fig. 2. The idler can be continuously tuned from 7.8 to 8.5 μm, as shown in Fig. 6(b).

For the CdSe OPO, due to the fact that only single 28 mm long crystals are available in our experiment, the pump radius is reduced to 0.21 mm to achieve the oscillation threshold. The threshold pump power is found to be 5.9 W at 10 kHz and 7.02 W at 12 kHz. The corresponding threshold energy fluences are 0.85 and 0.84 J/cm^2, respectively, which are also in reasonable agreement with the calculated data shown in Fig. 4. For the 12 kHz pulse repetition frequency, the maximum output power is 64 mW with 7.6 W incident pump power. When the pump power is further increased, the crystal coatings are found to be damaged. The output wavelength is around 8.9 μm, with a spectrum bandwidth (FWHM) of nearly 30 nm, as shown in Fig. 7.

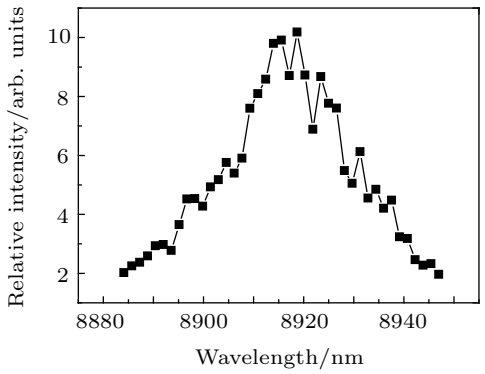


Fig. 7. The idler output spectrum of CdSe OPO.

4. Conclusion

We performed comparisons of the birefringent walk-off effect and the threshold performance between ZGP and CdSe OPOs theoretically and experimentally in this paper. With a comparative crystal length, the CdSe OPO threshold is much higher than that of the ZGP OPO. However, this can be circumvented by using a long CdSe crystal, as CdSe has a much smaller birefringent walk-off effect, or by adopting a dual-crystal for the walk-off compensation to reduce the threshold and achieve a high efficiency operation. For the ZGP OPO, up to 419 mW output power at 8.04 μm is obtained at the 8 kHz pump pulse repetition frequency (PRF) with a slope efficiency of 7.6%. This ZGP OPO can be continuously tuned from 7.8 to 8.5 μm. For the CdSe OPO, only 64 mW output is obtained at 8.9 μm due to the short-length crystal used in the experiment.

References

[1] Bianchi A and Garbi M 1979 *Opt. Commun.* **30** 122

[2] Allik T H, Chandra S, Rines D M, Schunemann P G, Hutchinson J A and Utano R 1997 *Opt. Lett.* **22** 597

[3] Vodopyanov K L 1998 *Opt. Commun.* **150** 210

[4] Lippert E, Rustad G, Arisholm G and Stenersen K 2008 *Opt. Express* **16** 13878

[5] Li G, Yao B Q, Duan X M, Zhu G L, Wang Y Z and Ju Y L 2010 *Chin. Phys. Lett.* **27** 014207

[6] Budni P A, Pomeranz L A, Lemons M L, Schunemann P G, Pollak T M and Chicklis E P 1998 *Advanced Solid-State Lasers* (Washington D.C.: Optical Society of America) p. 90

[7] Elder I 2009 *Proc. SPIE* **7325** 73250I

[8] Haidar S, Miyamoto K and Ito H 2004 *Opt. Commun.* **241** 173

[9] Liu J H, Liu Q and Gong M L 2011 *Acta Phys. Sin.* **60** 024215 (in Chinese)

[10] Ma J, Liu Y and Zhang L J 2011 *Acta Phys. Sin.* **60** 024211 (in Chinese)

[11] Ding X, Sheng Q, Chen N, Yu X, Wang R, Zhang H, Wen W, Wang P and Yao J 2009 *Chin. Phys.* B **18** 4314

[12] Zakel A, Wagner G, Alford W and Carrig T 2005 *Advanced Solid-State Photonics, Technical Digest* (Washington D.C.: Optical Society of America)

[13] Brosnan S and Byer R 1979 *IEEE J. Quantum Electron.* **15** 415

[14] Brehat F 1989 *J. Phys.* B: *At. Mol. Opt. Phys.* **22** 1891

[15] Zelmon D E and Hanning E A 2001 *J. Opt. Soc. Am.* B **18** 1307

[16] Bhar G C 1976 *Appl. Opt.* **15** 305

[17] Zhu G L, Ju Y L, Wang T H and Wang Y Z 2009 *Chin. Phys. Lett.* **26** 034208