



ELSEVIER

Available online at [www.sciencedirect.com](http://www.sciencedirect.com)

SCIENCE @ DIRECT®

Optics Communications 252 (2005) 39–45

OPTICS  
COMMUNICATIONS

[www.elsevier.com/locate/optcom](http://www.elsevier.com/locate/optcom)

# Wavelength dispersion of Verdet constants in chalcogenide glasses for magneto-optical waveguide devices

Yinlan Ruan <sup>a,b,\*</sup>, Ruth A. Jarvis <sup>b</sup>, Andrei V. Rode <sup>b</sup>, Steve Madden <sup>a,b</sup>,  
Barry Luther-Davies <sup>a,b</sup>

<sup>a</sup> Centre for Ultra-high-bandwidth Devices for Optical Systems, The Australian National University, Canberra ACT 0200, Australia

<sup>b</sup> Laser Physics Centre, The Australian National University, Canberra ACT 0200, Australia

Received 1 January 2005; received in revised form 16 March 2005; accepted 21 March 2005

## Abstract

The wavelength dispersion of the Verdet constants of bulk As–S, As–Se–S, Ge–As–Se and Ga–La–S-based chalcogenide glasses has been investigated at wavelengths between 675 and 1550 nm.  $\text{Ge}_{33}\text{As}_{12}\text{Se}_{55}$  was found to have the highest Verdet constant, 0.0485 min/G.cm at 1550 nm, 30 times that of silica. The high Verdet constant of  $\text{Ge}_{33}\text{As}_{12}\text{Se}_{55}$  is related to its smaller band gap energy compared with the other glasses. Further improvement of the Verdet constants of the Ge–Se-based chalcogenide glasses by engineering their compositions could make them promising materials for integrated magneto-optical devices.

© 2005 Elsevier B.V. All rights reserved.

PACS: 85.70.S

Keywords: Verdet constant; Chalcogenide glasses; Faraday rotation; Dispersion

## 1. Introduction

An increasing number of optical and laser devices require protection from back-reflected beams. This has an increased interest in new magneto-optic materials that can be used to make opti-

cal isolators based, for example, on the Faraday effect – the non-reciprocal rotation of the plane of polarisation of light when it passes through a transparent material in an external magnetic field. For bulk devices, magnetic garnet materials produce very large specific rotation and have been used widely in the fabrication of isolators in optical fibre networks. The move towards integration of active and passive optoelectronic devices to make photonic “chips” is now motivating research

\* Corresponding author. Tel.: +61-2-61254079; fax: +61-2-61250029.

E-mail address: [lan111@rsphysse.anu.edu.au](mailto:lan111@rsphysse.anu.edu.au) (Y. Ruan).

into magneto-optical waveguide devices that allow the fabrication of waveguide isolators or circulators.

Attempts to directly deposit and pattern crystalline magnetic garnet films onto semiconductor substrates suffer from lattice mismatch or growth-induced magnetic anisotropy [1]. This results in modal birefringence – a large difference between the refractive indices of the TE and TM guided modes. High modal birefringence is a problem because it produces oscillation of the principal axis of polarisation, rather than a linear increase of the rotation angle of linearly polarised light with distance, making it impossible to obtain the required Faraday rotation of 45° for an optical isolator. Overcoming these effects greatly increases the complexity of the fabrication.

An alternative to garnet films is to use amorphous glass films that, in principle, can have very low birefringence. However, the commonly used amorphous films used for integrated optical devices have too low magneto-optic activity. Presently, there is growing interest in amorphous chalcogenide glasses for use in integrated optics because their high non-linear refractive indices at infrared wavelengths suggest that they can be used to make chips capable of all-optical processing. Their high linear refractive indices and relatively strong dispersion in the infra-red also indicates that they should also have relatively high Verdet constants [2]. Previous investigations of the Faraday effect in chalcogenide glasses have been made by Qiu et al. [3] involving some As–S based glasses studied at several wavelengths in the visible region of the spectrum but well away from the wavelengths of interest for integrated optics applications. Measurements of magneto-optical rotation in gallium lanthanum sulphur ( $\text{Ga}_{28}\text{La}_{12}\text{S}_{42}\text{O}_{18}$ ) glasses were also performed by Pedroso et al. [4] at 543 nm, for which the Verdet constant was found to be as high as 0.2 min/G.cm.

To explore the interesting possibilities presented by chalcogenide films for integrated magneto-optical devices, we report here measurements of the dispersion of the Faraday rotation of several chalcogenide glasses at visible and near-infrared wavelengths.

## 2. Theoretical background: the Faraday effect and the wavelength dependence of the Verdet constant

Linearly polarised light with a given plane of polarisation can be represented as a superposition of right and left circularly polarised light with a specific phase difference. The refractive indices,  $n^+$  and  $n^-$ , for the right-hand and the left-hand circularly polarised waves, respectively, are equivalent in the absence of a magnetic field. When the linearly polarised light passes through a diamagnetic material, parallel to the direction of the applied magnetic field  $H$ ,  $n^+$  and  $n^-$  diverge, causing the two polarisations to propagate with different velocities and phases. As a consequence, the plane of the polarisation of the linearly polarised light rotates through the angle macroscopically described by Zvezdin and Kotov [5]

$$\theta = \frac{\omega}{c}(n^+ - n^-)L = VHL, \quad (1)$$

where  $\omega$  is the angular frequency of light,  $c$  is light speed,  $L$  is the length of the light path in the medium, and  $V$  is known as the Verdet constant.

The mechanism for the Faraday effect can be explained by the magnetic-field-induced Larmor precession of electron orbits. Instead of one eigen-frequency of the electrons ( $\omega$ ), two arise, corresponding to the right-hand and the left-hand circular oscillations when there is a magnetic field in the medium. The difference between the resonance frequencies  $\omega^+$  and  $\omega^-$  results in a displacement of  $n^+(\omega)$  and  $n^-(\omega)$  relative to each other on the frequency scale:

$$n^\pm(\omega) \approx n(\omega) \pm \frac{dn}{d\omega} \frac{eH}{2mc}, \quad (2)$$

where  $e$  and  $m$  are the electron charge and mass, respectively, and  $n(\omega)$  is the refractive index of the material in the absence of the field  $H$ . Substitution of Eq. (2) into (1) yields the well-known Biquel formula:

$$V = \frac{e}{2mc^2} \lambda \frac{dn}{d\lambda}, \quad (3)$$

where  $\lambda = 2\pi c/\omega$  is the wavelength of the light. Eq. (3) indicates that the Verdet constant is linearly proportional to the dispersion  $dn/d\lambda$  of the material.

However, the dispersion of the refractive index is theoretically related to the electronic absorption spectrum through the Wemple equation based on the single electronic oscillator model in the region from UV to near-infrared as [2]

$$n^2 - 1 = \frac{E_d \cdot E_0}{E_0^2 - E^2}, \quad (4)$$

where  $n$  is the refractive index,  $E$  the photon energy,  $E_0$  the average electronic energy gap, and  $E_d$  the electronic oscillator strength. Larger refractive index arises from smaller  $E_0$  and/or large  $E_d$  and results in a larger dispersion throughout the glass system.

### 3. Experiments

Six different chalcogenide glasses,  $\text{As}_{40}\text{S}_{60}$  ( $\text{As}_2\text{S}_3$ ),  $\text{As}_{40}\text{S}_{45}\text{Se}_{15}$ ,  $\text{As}_{24}\text{S}_{38}\text{Se}_{38}$ ,  $\text{Ge}_{33}\text{As}_{12}\text{Se}_{55}$  (AMTIR-1),  $\text{Ga}_{27}\text{La}_{13}\text{S}_{41}\text{O}_{19}$  (GLSO), and  $\text{Ga}_{28}\text{La}_{12}\text{S}_{60}$  (GLS) were investigated. Silica was also characterised for comparison. The bulk samples were obtained from three different sources: bulk  $\text{As}_2\text{S}_3$  and AMTIR-1 were obtained from Amorphous Materials Inc. (Garland, Tx); the As–Se–S samples were prepared at CREOL at the University of Central Florida; and the GLS samples were prepared in-house using the process described in [6].

Thin films of  $\text{As}_2\text{S}_3$ ,  $\text{Ge}_{33}\text{As}_{12}\text{Se}_{55}$ ,  $\text{As}_{40}\text{S}_{45}\text{Se}_{15}$  and  $\text{As}_{24}\text{S}_{38}\text{Se}_{38}$ , were prepared from bulk samples using the Ultra-Fast Pulsed Laser Deposition (UFPLD) technique developed at the Australian National University [7,8]. UFPLD uses short, ps or sub-ps, low energy ( $\sim 0.1$  to  $10 \mu\text{J}$ ) pulses at MHz-range repetition rate. UFPLD produces high quality films free from contamination with particulates suitable for waveguide fabrication. However, the surface layer of the target is ablated with moderately high kinetic energy which densifies the resulting films and this results in some differences between the deposited films and bulk glasses as mentioned below.

To deposit films for these experiments, a frequency doubled mode-locked Nd:YAG laser producing 6–7 W average power (70–80 nJ/pulse) at 532 nm, was directed at the bulk glass targets, at

a repetition rate of 76 MHz [7]. The beam was focused to a  $30 \mu\text{m}$  spot onto the bulk glass target, inside a vacuum chamber (base pressure  $< 10^{-6}$  Torr), to produce an ablated plume which was deposited on the 100-mm substrate positioned  $\sim 20$  cm from the target. The substrate wafer was rotated to ensure good ( $\pm 5\%$ ) thickness uniformity of the film. The substrates used were either pure silicon or silicon wafers oxidized with a  $2.4 \mu\text{m}$  silica layer to act as a buffer layer for waveguiding.

The refractive index  $n$  and extinction coefficient  $k$  of the chalcogenide films were measured in the wavelength range from 450 to 1650 nm using a commercial SCI FilmTek 4000 wafer mapper. The FilmTek system is based on spectrophotometry of the samples, using reflection fringes from the film at two different angles (normal to the surface,  $0^\circ$  and  $70^\circ$ ) to model the refractive index and thickness of the films according to a Lorentz–Tauc dispersion model. This technique also determines the value of the optical band gap energy, below which the absorption is assumed to be zero. The FilmTek 4000 provides  $n$  and  $k$  data across the full wavelength range of the measurement from which the dispersion can be calculated.

Absorption spectra of polished bulk samples were measured using a Cary 5000 UV–Vis spectrophotometer. From the absorption data, the optical band gap,  $E_g$ , could also be determined using the Tauc and Menth [9] method by plotting the function  $(\alpha h\nu)^{1/2}$  versus  $h\nu$ , where  $\alpha$  is the absorption coefficient ( $\text{cm}^{-1}$ ) and  $h\nu$  is the photon energy (eV). By fitting a straight line to the data, the intercept at  $\alpha = 0$  determines the energy of the band gap.

The system used to measure the Faraday rotation of chalcogenide glasses is shown in Fig. 1. A solenoid was driven at 30 Hz to give a maximum magnetic field of 460 G in the centre of the solenoid, where the polished glass samples were placed. The magnetic field was calibrated by using a sample of silica glass whose Verdet constant was taken as 0.01352 at 632.8 nm at room temperature [10]. The polarisation rotation of the input beam was determined with high sensitivity with the aid of a lock-in amplifier. Laser diode sources used had wavelengths of 675, 780, 1064, 1310 and 1550 nm. The measurement sequence was repeated to confirm reproducibility.

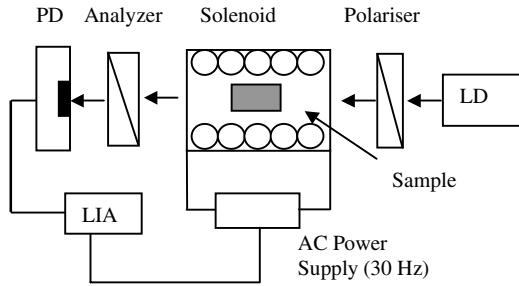


Fig. 1. Experimental setup for measuring magneto-optic rotation. PD, photodetector; LD, laser diode; LIA, lock-in amplifier.

#### 4. Results

The absorption spectra of bulk chalcogenides and silicon oxide glass are shown in Fig. 2(a). Fig. 2(b) shows the dispersion of the linear refractive indices of the chalcogenide films measured by FilmTek in the case of the films deposited by UFPLD and from [2] for the GaLaS and GaLaSO samples. Fig. 3 shows a sample Tauc plot used to determine the band gap energy of bulk  $\text{As}_2\text{S}_3$  from the absorption spectrum [9].

The band gap energies, determined by the Tauc method for bulk samples and calculated by FilmTek for thin film samples, are summarized in Table 1. With the same composition, the band gap

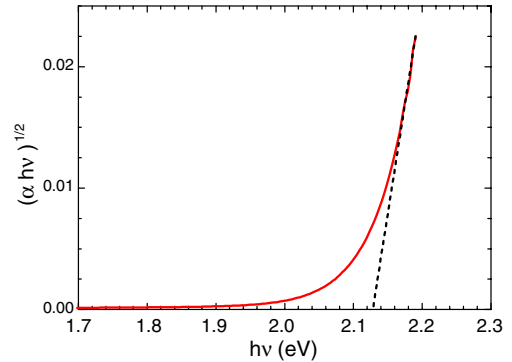


Fig. 3. Tauc plot for  $\text{As}_2\text{S}_3$ .

energy,  $E_g$ , was found to be higher in the thin film samples than in the bulk, with the exception of  $\text{Ge}_{33}\text{As}_{12}\text{Se}_{55}$ . The difference between the band gap in the films and bulk forms can be related to the fact that they have been created in very different conditions which result in different bond structure [11]. In fact it is a general characteristic of chalcogenides that a wide range of bond structures can form for the same stoichiometry and this has a strong effect on the band edge and hence refractive index and dispersion of the material. For example, Raman spectroscopy has revealed significant differences in the bond structure present in  $\text{As}_2\text{S}_3$  films compared with bulk samples [12].

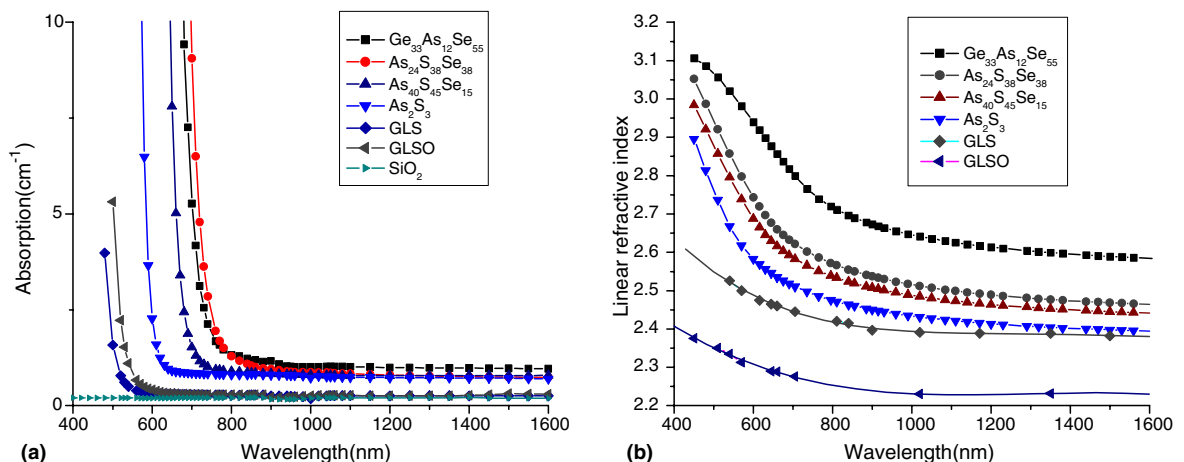


Fig. 2. Dispersion of absorption and refractive index of the chalcogenide glasses: (a) absorption; (b) refractive index dispersion.

Table 1  
Band gap for bulk samples and films

	Bulk (Cary)		Film (FilmTek)	
	Band gap (eV)	Band-edge (nm)	Band gap (eV)	Band-edge (nm)
GLSO	2.66	467		
GLS	2.40	516		
As <sub>2</sub> S <sub>3</sub>	2.12	585	2.24	553
As <sub>40</sub> S <sub>45</sub> Se <sub>15</sub>	1.84	673	1.98	626
As <sub>24</sub> S <sub>38</sub> Se <sub>38</sub>	1.72	720	1.92	646
Ge <sub>33</sub> As <sub>12</sub> Se <sub>55</sub>	1.66	745	1.64	756

Nevertheless, the linear refractive indices and absorption of chalcogenide materials also strongly depend on their chemical compositions, and it can be seen from Fig. 2(a) and Table 1 that the band-edge wavelength  $\lambda_g$  increases with increasing Se and Ge content. As the heavier Se or Ge atoms substitute the lighter S atoms, the linear refractive indices and band-edge wavelength of the materials increase.

Fig. 4(a) shows the Verdet constants for the bulk samples, measured at different wavelengths. In the infrared, the Verdet constants of the different glasses increase in the sequence:  $V_{\text{SiO}_2} < V_{\text{GLSO}} < V_{\text{GLS}} < V_{\text{As}_2\text{S}_3} < V_{\text{As}_{40}\text{S}_{45}\text{Se}_{15}} < V_{\text{As}_{24}\text{S}_{38}\text{Se}_{38}} < V_{\text{Ge}_{33}\text{As}_{12}\text{Se}_{55}}$ , which is the same sequence for the decrease in band gap energy. This confirms that the reduction of the

band gap  $E_g$  and increase in refractive index enhances the Verdet constants of the glasses for wavelengths where the materials are transparent as predicted by Eqs. (3) and (4).

From the refractive index dispersion curves shown in Fig. 2(b), the Verdet constants of the chalcogenide glasses were calculated using Eq. (3) and are plotted in Fig. 4(b). The curves predict the highest Faraday rotation closest to the band edge and a monotonically decreasing Verdet constant above the band gap wavelength, which is the same behaviour as exhibited by the experimentally measured values. In addition, both the measured and predicted values follow the same sequence of Verdet constant versus glass composition. The dependence of the Verdet constant on glass composition demonstrates that the values in the infrared could be increased by engineering the glass composition, for example by increasing the content of Se or Ge, and subsequently decreasing the band gap energy.

In the telecommunication wavelengths of 1310 and 1550 nm, Ge<sub>33</sub>As<sub>12</sub>Se<sub>55</sub> showed the maximum magneto-optical rotation with a Verdet constant of 0.0485 min/G.cm at 1550 nm, 30 times of that of silica. We compared our results with experimental values from the literature [2]. The Verdet constant of Ga<sub>27</sub>La<sub>13</sub>S<sub>41</sub>O<sub>19</sub> at 544 nm is predicted by extrapolating its fitting line in Fig. 4(a) and

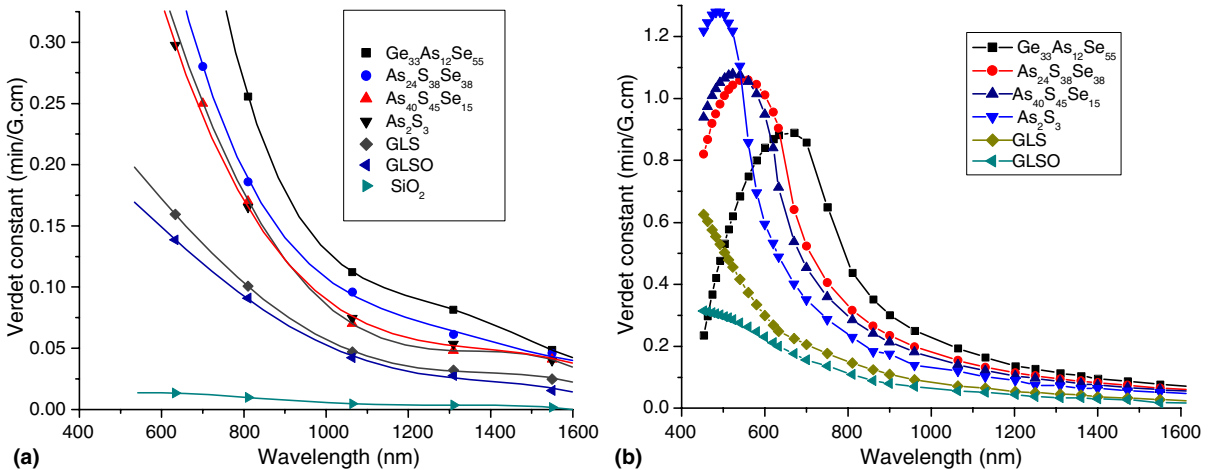


Fig. 4. Wavelength dependence of the Verdet constants of chalcogenide glasses: (a) experimental measurement; (b) theoretical calculation.

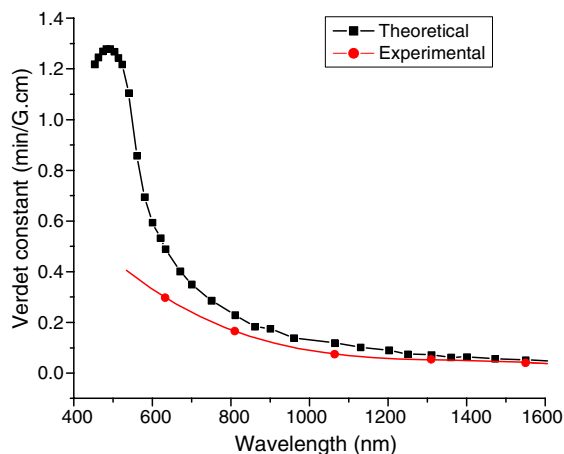


Fig. 5. Comparison of the measured and calculated Verdet constant of  $\text{As}_2\text{S}_3$  material.

was about 0.17 min/G.cm. The practical measured value for  $\text{Ga}_{28}\text{La}_{12}\text{S}_{42}\text{O}_{18}$  at 544 nm was 0.2 min/G.cm from [2]. The 15% difference may come from different composition.

Fig. 5 shows the measured Verdet constants for bulk  $\text{As}_2\text{S}_3$  glass compared with values calculated from the Becquerel equation with the dispersion measured for an  $\text{As}_2\text{S}_3$  film. The experimental values are 20–40% lower than those calculated from the dispersion data most probably due to differences in the band edge and the dispersion between the bulk samples and films mentioned above.

Recently, we have successfully fabricated and characterised  $\text{As}_2\text{S}_3$  and  $\text{Ge}_{33}\text{As}_{12}\text{Se}_{55}$  rib waveguides with low loss of 0.25 dB/cm at 1550 nm [13]. Using the best values for the Verdet constant at telecommunications wavelengths which lie between 0.05 and 0.1 min/G.cm, and a field of 6.5 kG, which is readily achievable over cm distances using rare earth magnets, we can calculate that the path length for 45° rotation is between 4.2 and 8.4 cm. The waveguide loss would then be in the range of 1–2 dB. Because of the high refractive index difference available between core and cladding with chalcogenides, waveguide bend radii can be as small as 500  $\mu\text{m}$  with negligible loss already confirmed by our recent experiments, and it should therefore be able to coil the waveguides and create a compact magneto-optic device.

## 5. Conclusion

The dispersion of the Verdet constants of bulk chalcogenide glasses has been experimentally measured and was found to be in agreement with values calculated by the Becquerel equation. The results show that the Verdet constants increase with the decrease of band gap energy  $E_g$ . The  $E_g$  can be tuned via the material composition. The glasses containing Se and Ge showed the lower band gap energies and are the most promising materials for waveguide magneto-optical devices and they present the potential of further improving their Verdet constants by engineering their compositions.

## Acknowledgements

The authors thank Dr. Kathleen Richardson and Mr. Cedric Lopez from the University of Central Florida for supplying various glass samples and Ms. Anita Smith for both preparation and fabrication of samples at Florida and ANU. The authors also acknowledge the assistance of Dr. Graham Atkins for making FilmTek measurements. Funding from the Australian Research Council through its Discovery, Federation Fellow and Centres of Excellence programs is gratefully acknowledged.

## References

- [1] R. Wolfe, V.J. Fratello, M. McGlashan-Powell, *J. Appl. Phys.* 63 (80) (1988) 3099.
- [2] H. Yayama, S. Fujina, K. Morinaga, H. Takebe, S.W. Hewak, D.N. Payne, *J. Non-Cryst. Solids* 239 (1998) 187.
- [3] J. Qiu, H. Kanbara, H. Nasu, K. Hirao, *J. Ceram. Soc. Jpn.* 106 (1998) 236.
- [4] C.B. Pedroso, E. Munin, A.B. Villaverde, J.A. Medeiros Neto, N. Aranha, L.C. Barbosa, *Opt. Eng.* 38 (2) (1999) 214.
- [5] A.K. Zvezdin, V.A. Kotov, *Modern magneto-optics and magneto-optical materials*, in: J.M.D. Coey, D.R. Tilley (Eds.), Institute of Physics Publishing, London, 1997.
- [6] J. Wang, J.R. Hector, D. Brady, D. Hewak, W. Brocklesby, M. Kluth, R. Moore, D. Payne, *Appl. Phys. Lett.* 71 (1997) 1753.
- [7] A.V. Rode, A. Zakery, M. Samoc, R.B. Charters, E.G. Gamaly, B. Luther-Davies, *Appl. Surf. Sci.* 197–198 (2002) 481.

- [8] A. Zakery, Y. Ruan, A.V. Rode, M. Samoc, B. Luther-Davies, *J. Opt. Soc. Am. B* 20 (2003) 1844.
- [9] J. Tauc, A. Menth, *J. Non-Cryst. Solids* 8–10 (1972) 569.
- [10] C.Z. Tan, J. Arndt, *Physica B* 233 (1997) 1.
- [11] D. Arsova, *J. Phys. Chem. Solids* 57 (1996) 1279.
- [12] A. Schulte, C. Rivero, K. Richardson, K. Turcotte, V. Hamel, A. Villeneuve, T. Galstian, R. Vallee, *Opt. Commun.* 198 (2001) 125.
- [13] Y. Ruan, W. Li, R. Jarvis, N. Madsen, A. Rode, B. Luther-Davies, *Opt. Exp.* 12 (2004) 5140.