INFLUENCE OF TEMPERATURE ON REFRACTIVE INDICES OF $BaLaGa_3O_7$ AND $SrLaGa_3O_7$*

W. Ryba-Romanowski, S. Gołąb

Institute of Low Temperature and Structure Research, Polish Academy of Sciences
Okólna 2, 50-950 Wrocław, Poland

and M. Berkowski

Institute of Physics, Polish Academy of Sciences, Warszawa, Poland

(Received October 1, 1992)

A simple theoretical approach based on Kramers–Krönig relations predicts well the dispersion and birefringence of $BaLaGa_3O_7$ and $SrLaGa_3O_7$. In the whole transparency region the birefringence of both compounds is too low to offset dispersion in the process of a second harmonic generation, thus the crystals cannot be made phase matchable. Birefringence of $BaLaGa_3O_7$ and $SrLaGa_3O_7$ is stable with respect to the temperature region of 300–550 K. The refractive indices increase linearly at a rate of 2×10^{-5} K^{-1} with increasing temperature.

PACS numbers: 42.70.–a

1. Introduction

Temperature-dependent variation of refractive indices is one of material's properties which affects the performance of nonlinear crystals and optically pumped laser crystals and glasses.

Detailed knowledge of dispersion and birefringence of nonlinear crystal in a wide temperature region is needed to determine the conditions of operation such as limits of temperature tuning or temperature control requirements. On the other hand, the optical pumping generates an appreciable quantity of heat which gives rise to non-uniform temperature distribution within a laser active medium. Resulting gradients of refractive indices influence strongly the output beam quality,

*The work was supported by the Committee for Scientific Research under grant Nr 2 0358 91 01.

therefore the appropriate methods of compensation for optical distortion should be chosen.

Single crystals of $BaLaGa_3O_7$ (BLGO) and $SrLaGa_3O_7$ (SLGO) investigated in this work belong to a large family of compounds of general chemical formula ABC_3O_7, where A = Ca, Sr, Ba; B = La, ... Gd and C = Al, Ga. All these compounds form tetragonal crystals belonging to the space group $P\overline{4}2_1m$, hence one may expect that they will exhibit piezoelectric and optically nonlinear properties.

Low threshold lasing has been obtained in the single crystals of BLGO and SLGO doped with neodymium [1, 2]. Investigation of spectroscopic [1, 3] and physical [4] properties of BLGO:Nd indicate that these crystals are promising for application as laser active materials.

A threshold for laser action in the recently obtained single crystals of SLGO:Nd is found to be considerably lower than in BLGO:Nd mainly because SLGO:Nd crystals are less susceptible to formation of built-in strains.

The goals of this work are as follows: (1) assessment of suitability of BLGO and SLGO crystals for frequency doubling and (2) evaluation of material constants which determine the thermally induced distortion of BLGO:Nd and SLGO:Nd laser output.

Previously reported data on dispersion and birefringence of BLGO at room temperature [5] will be used for the discussion.

2. Experiments and results

The single crystals of BLGO and SLGO have been grown by the Czochralski method using the system of automatic control of crystal diameters. Details of preparation and growing technique are described elsewhere [6]. Samples for measurements have been cut out from carefully chosen parts of the crystals which were free from built-in strains. Plane parallel plates have been prepared with the optical axis in the plane of the plates and their absorption at room temperature has been measured with a Varian 2300 spectrophotometer. Refractive indices were determined using the method of refraction in prisms cut out from the crystals. In these measurements the samples were placed in variable temperature heater whose temperature was controlled within ±2K. Argon ion laser and helium neon laser have been used as sources of monochromatic light of wavelengths: 476.5, 488.0, 496.5, 514.0 and 632.8 nm. The desired plane of light polarization has been selected by means of a quarter wave plate followed by a Glan–Taylor polarizer. Accuracy of refractive index measurements is estimated to be not worse than ±0.0002. The results of the measurements are presented in Figs. 1–4.

3. Discussion

Experimental data in Fig. 1 indicate that refractive indices for SLGO are slightly lower than those for BLGO, but for both compounds the refractive indices depend on wavelength in a very similar way. Therefore, the simplified theoretical approach based on Kramers–Krönig relation should account as well for experimen-

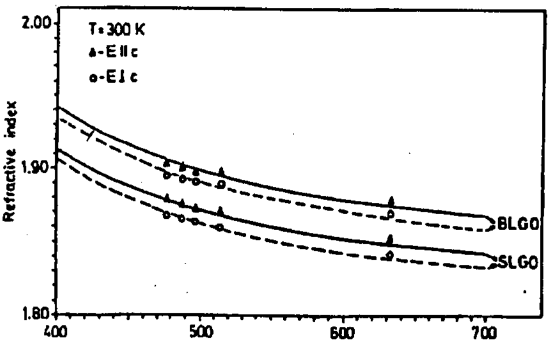

Fig.1

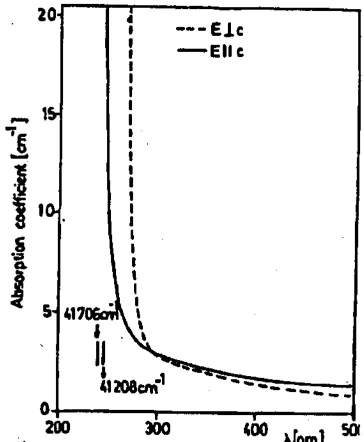

Fig.2

Fig. 1. Dependence of refractive indices of $BaLaGa_3O_7$ and $SrLaGa_3O_7$ on wavelength. See text for the explanation of theoretical lines.

Fig. 2. Absorption spectrum of $SrLaGa_3O_7$ in visible and UV region. Arrows indicate the absorption edges for two polarizations.

tal data of SLGO as it does in the case of BLGO. Kramers–Krönig relation

$$\varepsilon_{ij}^{(1)}(\omega) = \delta_{ij} + \frac{2}{\pi}\int_0^\infty \frac{\omega'\varepsilon_{ij}^{(2)}(\omega)}{\omega'^2 - \omega^2}\mathrm{d}\omega', \tag{1}$$

where $\varepsilon_{ij}^{(1)}$ and $\varepsilon_{ij}^{(2)}$ denote real and imaginary parts of the dielectric constant tensor and ω is the light frequency, may be written for two polarizations

$$\varepsilon_{\parallel}^{(1)}(\omega) = 1 + \frac{2}{\pi}\int_0^\infty \frac{\omega'\varepsilon_{\parallel}^{(2)}(\omega)}{\omega'^2 - \omega^2}\mathrm{d}\omega',$$
$$\varepsilon_{\perp}^{(1)}(\omega) = 1 + \frac{2}{\pi}\int_0^\infty \frac{\omega'\varepsilon_{\perp}^{(2)}(\omega)}{\omega'^2 - \omega^2}\mathrm{d}\omega'. \tag{2}$$

we take into account that

$$\varepsilon_{\parallel}^{(1)} = n_{\parallel}^2 - \frac{\alpha_{\parallel}^2 c^2}{4\omega^2}, \qquad \varepsilon_{\perp}^{(1)} = n_{\perp}^2 - \frac{\alpha_{\perp}^2 c^2}{4\omega^2},$$
$$\varepsilon_{\parallel}^{(2)} = \frac{c}{\omega}\alpha_{\parallel} n_{\parallel}, \qquad \varepsilon_{\perp}^{(2)} = \frac{c}{\omega}\alpha_{\perp} n_{\perp}, \tag{3}$$

where n denotes the refractive index and α denotes the absorption coefficient. In Eqs. (2) and (3) the subscripts ($\parallel$) and ($\perp$) indicate the light polarization parallel and perpendicular to the optical axis of the crystal. Refractive indices may be calculated using these relations, provided the absorption coefficient in the whole frequency region is known. Taking into account the absorption spectra of SLGO (see Fig. 2) we will assume that the absorption coefficient for both polarizations is zero below the absorption edges denoted by $\omega_{\parallel}$ and $\omega_{\perp}$ and constant from the absorption edges to infinity. Under this assumption we obtain from Eqs. (2) and (3)

$$n_{\parallel}^2 = 1 + \frac{2c}{\pi}\alpha_{\parallel} n_{\parallel}\int_{\omega_{\parallel}}^\infty \frac{\mathrm{d}\omega'}{\omega'^2 - \omega^2}, \quad n_{\perp}^2 = 1 + \frac{2c}{\pi}\alpha_{\perp} n_{\perp}\int_{\omega_{\perp}}^\infty \frac{\mathrm{d}\omega'}{\omega'^2 - \omega^2}. \tag{4}$$

Expressing ω by the wavenumber $\Lambda = 1/\lambda$, $\alpha_{\|}n_{\|}$ by $a_{\|}$ and $\alpha_{\perp}n_{\perp}$ by $a_{\perp}$, we get after integration

$$n_{\|}^2 = 1 - \frac{a_{\|}}{2\pi^2\Lambda}\ln\frac{\Lambda_{\|} - \Lambda}{\Lambda_{\|} + \Lambda}, \qquad n_{\perp}^2 = 1 - \frac{a_{\perp}}{2\pi^2\Lambda}\ln\frac{\Lambda_{\perp} - \Lambda}{\Lambda_{\perp} + \Lambda}. \tag{5}$$

The following fitting parameters have been determined for BLGO: $\Lambda_{\perp} = 40900\ \mathrm{cm}^{-1}$, $\Lambda_{\|} = 41425\ \mathrm{cm}^{-1}$, $a_{\perp} = \alpha_{\perp}n_{\perp} = 9.4866 \times 10^5\ \mathrm{cm}^{-1}$, $a_{\|} = \alpha_{\|}n_{\|} = 9.7430 \times 10^5\ \mathrm{cm}^{-1}$ [5].

It can be concluded on the basis of absorption measurements that the absorption edges for two polarizations in SLGO are shifted towards shorter wavelengths by about 300 cm^{-1} with respect to BLGO. The fitting parameters evaluated for SLGO: $\Lambda_{\perp} = 41208\ \mathrm{cm}^{-1}$, $\Lambda_{\|} = 41706\ \mathrm{cm}^{-1}$, $a_{\perp} = 9.1695 \times 10^5\ \mathrm{cm}^{-1}$, $a_{\|} = 9.4337 \times 10^5\ \mathrm{cm}^{-1}$, confirm this observation. The refractive indices calculated according to the relations (5) for BLGO and SLGO are indicated by solid and dotted lines in Fig. 1. The birefringence δ defined as $\delta = n_{\|} - n_{\perp}$ has been calculated also, and traced in Fig. 3.

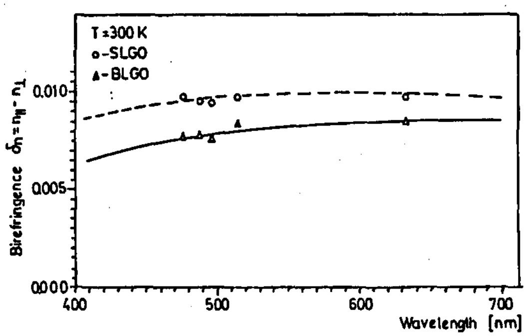

Fig. 3. Dependence of birefringence of $BaLaGa_3O_7$ and $SrLaGa_3O_7$ on wavelength. See text for the explanation of theoretical lines.

With these results the suitability of SLGO and BLGO crystals for a second harmonic generation of laser radiation may be estimated. It is well known that the efficiency of the second harmonic generation may be significant only when the phase velocity of fundamental wave can be made equal to the phase velocity of the second harmonic wave. An effective method of providing of equality of phase velocities utilizes the fact that the dispersion can be offset by the natural birefringence of crystals. The phase matching conditions, which determine the appropriate polarization and direction of propagation in positive uniaxial crystals are given by

$$\begin{aligned} n_{2\omega}^{\mathrm{o}} &= n_{\omega}^{\mathrm{e}}(\theta), && \text{(type I)} \\ n_{2\omega}^{\mathrm{o}} &= (1/2)[n_{\omega}^{\mathrm{e}}(\theta) + n_{\omega}^{\mathrm{o}}] && \text{(type II)} \end{aligned} \tag{6}$$

and

$$n^{\mathrm{e}}(\theta) = n^{\mathrm{o}}n^{\mathrm{e}}/[(n^{\mathrm{o}})^2\sin^2\theta + (n^{\mathrm{e}})^2\cos^2\theta]^{1/2},$$

where n^o and n^e are refractive indices for ordinary and extraordinary waves, θ is a phase matching angle. It is easy to verify that the values of refractive indices calculated using Eq. (5) cannot fulfill the conditions (6) for any fundamental wavelength within the transparency region of SLGO and BLGO. Moreover, the phase matching cannot be achieved by temperature tuning since the birefringence of both compounds is stable with respect to temperature. It can be seen in Fig. 4 that indices of refraction of SLGO and BLGO for both polarizations increase with increasing temperature. Solid lines in this figure indicate a linear dependence with

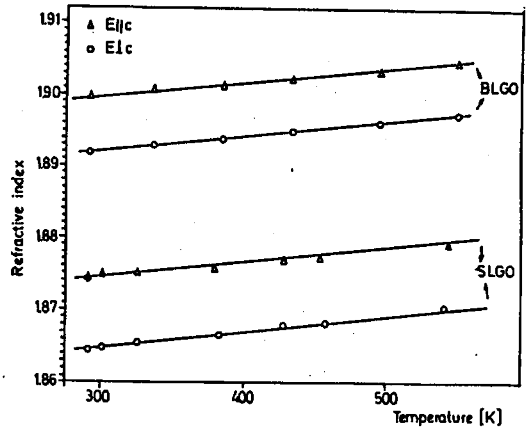

Fig. 4. Influence of temperature on refractive indices of $BaLaGa_3O_7$ and $SrLaGa_3O_7$. Wavelength 488 nm. Solid lines indicate a linear dependence with $\mathrm{d}n/\mathrm{d}T = +2 \times 10^{-5}\ \mathrm{K}^{-1}$.

$\mathrm{d}n/\mathrm{d}T = +2 \times 10^{-5}\ \mathrm{K}^{-1}$ and are in good agreement with experimental data. The value of $\mathrm{d}n/\mathrm{d}T$ obtained is higher than those reported for commonly used laser crystals and glasses, therefore in the design of BLGO:Nd and SLGO:Nd lasers the compensation for optical distortion may be necessary.

In the case of uniformly pumped cylindrical laser rod the most important is the temperature-dependent radial variation of refractive index causing the thermal lensing. This perturbation is equivalent to the effect of a spherical lens of focal length f [7]:

$$f = \frac{KA}{P}\left(\frac{1}{2}\frac{\mathrm{d}n}{\mathrm{d}T}\right)^{-1}, \tag{7}$$

where K is thermal conductivity of laser active material, A is rod cross-sectional area and P is the total heat dissipated in the rod.

Thermal lensing in BLGO:Nd (or SLGO:Nd) and in YAG:Nd may be compared using Eq. (7) and the relevant material constants. With reported data for YAG:Nd — $K = 0.13$ W/cm K [7], $\mathrm{d}n/\mathrm{d}T = 7.3 \times 10^{-6}\ \mathrm{K}^{-1}$ [8] and for BLGO:Nd — $K = 0.11$ W/cm K [4], $\mathrm{d}n/\mathrm{d}T = 2 \times 10^{-5}\ \mathrm{K}^{-1}$ the expected focal length of thermally induced lens is shorter in BLGO:Nd by a factor of three than in YAG:Nd.

References

[1] W. Ryba-Romanowski, B. Jeżowska-Trzebiatowska, W. Piekarczyk, M. Berkowski, *J. Phys. Chem. Solids* **49**, 199 (1988).

[2] W. Ryba-Romanowski, S. Gołąb, G. Dominiak-Dzik, M. Berkowski, *Mater. Sci. Eng. B, Solid-State Mater. Adv. Technol.*, in press.

[3] W. Ryba-Romanowski, S. Gołąb, J. Hanuza, M. Berkowski, *J. Phys. Chem. Solids* **50**, 685 (1989).

[4] W. Ryba-Romanowski, S. Gołąb, M. Berkowski, *Acta Phys. Pol. A* **79**, 501 (1991).

[5] M. Berkowski, M.T. Borowiec, K. Pataj, W. Piekarczyk, W. Wardzyński, *Physica B* **123**, 215 (1984).

[6] W. Piekarczyk, M. Berkowski, G. Jasiołek, W. Ryba-Romanowski, M. Chabiera, Patent PRL No. 260309 (1986); US Patent No. 4, 820, 445 (1989).

[7] W. Koechner, *Solid State Laser Engineering*, Springer-Verlag, New York, Heidelberg, Berlin 1976.

[8] P.H. Klein, W.J. Croft, *J. Appl. Phys.* **38**, 1603 (1967).