Thermo-optics parameters measurements in photorefractive sillenite $Bi_{12}SiO_{20}$ crystals by thermal lens technique

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The present work reports on the thermo-optical properties of photorefractive sillenite $Bi_{12}SiO_{20}$ (BSO) crystals obtained by applying the Thermal Lens Spectrometry technique (TLS). This crystals presents one high photorefractive sensitivity in the region blue-green spectra, since the measurements were carried out at two pump beam wavelengths (514.5 nm and 750 nm) to study of the light-induced effects in this material (thermal and/or photorefractive). We determine thermo-optical parameters like thermal diffusivity (D), thermal conductivity (K) and temperature coefficient of the optical path length change (ds/dT) in sillenite crystals. These aspects, for what we know, not was studied in details up to now using the lens spectrometry technique and are very important against of the promising potentiality of applications these crystals in non linear optics, real time holography and optical processing data.

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1. Introduction

The photorefractive $Bi_{12}MO_{20}$ type crystals, where M: Ge, Si or Ti (here after BGO, BSO and BTO, respectively), known as sillenite, crystallizes in the cubic system with a I23 space group [1-9]. They display a fast photorefractive effect [1, 9], which renders them suitable as a potential reversible recording medium for real-time holography and holographic interferometry [10-11], optoelectronic sensors and image processing applications [1]. In last decades, many works were realized to determine photorefractive non linear optical parameters [12-18] in photorefractive crystals; however few are related to determination the thermo-optics parameters [19-22].

When a laser beam is passing through a photorefractive medium, we can to observe an inhomogeneous distribution of the refractive index, thus an optical damage will be created due to light-induced lens effect in the material. There are two mechanisms that can induce this optical damage depending whether the wavelength laser beam is absorbed or not by the medium. In order to laser beam non absorbed by material, an inhomogeneous distribution of refractive index can be created by photorefractive effect. In this case, the laser beam creates charge carriers that induce an inhomogeneous local electric field and the refractive index change by electro-optic effect, consequently, a light-induced lens and leading to a beam defocusing by photorefractive effect. Otherwise, when the laser beam is absorbed by material its beam profile produces an inhomogeneous temperature distribution, consequently, the thermal lens and leading to a beam focusing is created due the thermo-optical effects. In mediums with strongly thermo-optical and photorefractive effects both mechanisms could contribute for the optical damage and focusing or defocusing effects.

The thermo-optics properties, essentially, determine the thermal shock e stress conditions, thermal lens effects (focusing and defocusing), holographic thermal grating, among others. These properties are strongly dependents of physical properties as grating parameters, electronics polarizability, linear thermal expansion coefficient and phonon mean free path. The Thermal Lens Spectrometry (TLS) has established to be a valid method for study of the thermo-optics properties of diverse transparent optical materials [23-24]. Many works the TLS use to determine the thermo-optical properties in diverse materials, like photonics glass, polymers and crystals obtain important properties of these materials for applications in lasers physics [23-24].

As regards the photorefractive effect [2-8] incite the light-induced refractive index gradient (in other words, a light-induced optical path length change), and, consequently the focusing and defocusing effects of the Gaussian laser beam. The theoretical analysis of photorefractive effects was presented in Band Transport Model and the by Kukhtarev equations [1, 9].

In sillenite crystals, the intensity, wavelength and beam waist change shown that the different types of light-induced lens can be involved due thermo-optical and/or photorefractive effects.

In this work, we present quantitative values of the thermo-optical coefficients and qualitative PR properties of sillenite $Bi_{12}SiO_{20}$ (BSO) crystals. The analysis was carried out applying the light-induced lens technique, TL spectrometry with two beam (pump and probe) mode-mismatched configuration [20, 22-24]. Thermo-optical properties such as thermal diffusivity (*D*), thermal conductivity (*K*) *and* temperature coefficient of the optical path length change (*ds/dT*) of BSO crystals were determined. This technique has proved to be a valuable method to obtain thermo-optical properties of transparent materials. Although, the electro-optics and PR crystals were not studied in details up to now using TL spectrometry, their thermo-optical properties are important due to the promising potentiality of these crystals in nonlinear optics, solitons, optoelectronic, optical data storage and optical processing [1, 9].

2. Theoretical considerations

The TL effect is created when the excitation laser beam passes through the sample and the absorbed energy is converted into heat, consequently local temperature change $\Delta T(r,t)$, changing the optical path length (Δs) and producing a lens like optical element. The propagation of a probe beam laser through the TL will result in either a spreading ($ds/dT < 0$) or a focusing ($ds/dT > 0$) of the beam, depending mainly on the sample temperature coefficients of electronic polarizability, stress, and thermal expansion. The TL effect can be treated through the calculation of the temporal evolution of the sample temperature profile $\Delta T(r,t)$, caused by a Gaussian intensity distribution of the excitation beam. The aberrant model was generalized to the dual beam mode-mismatched configuration by Shen *et. al.* [23], and the following expression for the intensity in the far field was obtained:

$$I(t) = I(0)\left\{1 - \frac{\theta}{2}\tan^{-1}\left[\frac{2mV}{\left[(1+2m)^2+V^2\right]\frac{t_c}{2t} + 1 + 2m + V^2}\right]\right\}^2 \quad (1)$$

Where

$$m = \left(\frac{w_p}{w_e}\right)^2 ; \quad V = \frac{Z_1}{Z_{cp}}, \quad Z_{cp} \ll Z_2$$

Here, w_p and w_e are respectively the probe and pump beam radii at the sample position, Z_1 is the distance between the probe beam waist and the sample, Z_2 is the distance between the sample and the detector, Z_{cp} is the confocal distance of the probe beam, and $I(0) = I(t)$ when the transient time t or θ is zero. The TL transient signal amplitude θ is approximately the phase difference of the probe beam at $r = 0$ and $r = \sqrt{2}\, w_e$ induced by TL, given by [22, 24]:

$$\theta = \frac{P_{abs}}{K\lambda_p}\frac{ds}{dT} \quad (2)$$

where λ_p is the probe beam wavelength and $P_{abs} = P_{in}\alpha_e L_{eff}$ is the absorbed excitation power, in which P_{in} is the incident pump power, α_e is the optical absorption coefficient at the excitation wavelength, $L_{eff} = [1-\exp(-\alpha_e.L)]/\alpha_e$ is the sample effective thickness, and *L* the sample thickness. For liquids, in Eq. (2), the temperature coefficient of refractive index (*dn/dT*) must be used instead of *ds/dT*. On the other hand, in solids two other terms jointly with *dn/dT* contribute to *ds/dT* [22, 24]:

$$\frac{ds}{dT} = \frac{dn}{dT} + \alpha_T(n-1)(1+\nu) + \frac{n^3}{4}Y\alpha_T(q_\perp + q_{//}) \quad (3)$$

where α_T is the linear thermal expansion coefficient, *n* is the refractive index, ν is the Poisson number, Y is the Young modulus, and $q_\perp$ and $q_{//}$ are the stress-optical coefficients for perpendicular and parallel orientations relative to the excitation beam polarization. The term $\Delta\varphi_T = \alpha_T(n-1)(1+\nu)$ represents the bulging of the sample end-faces and $\Delta\psi_T = \frac{n^3}{4}\alpha_T Y(q_\perp + q_{//})$ the thermal stress. Equation (3) is valid for "thin disk geometry", where the sample diameter is much larger than L, which corresponds approximately to our experimental conditions.

The temporal evolution of the TL signal depends on the characteristic TL signal response time, t_c, given by [22-24]:

$$t_c = w_e^2/4D \quad (4)$$

where D is the thermal diffusivity, which is related to the thermal conductivity by K= ρcD, where c is the specific heat and ρ is the density.

For other hand, when a Gaussian beam propagates in a PR medium, the inhomogeneous beam profile generates an inhomogeneous refractive index inducing a focusing and/or defocusing effect. A complementary analysis of the light-induced effects involved in refractive index change on PR medium were accomplished by Buse *et al* [12] and Kukhtarev *et al* [13], where they considered other effects as the thermo-optical and the pyroelectric ones. Then, the total refraction index change due to linear electro-optic effect is given by [1,9,12]:

$$\Delta n(r,t) = -\frac{1}{2}n^3 r^{eff}\Delta E(r,t) \quad (5)$$

where *n* is the refractive index, r^{eff} is the effective electro-optic coefficient, and ΔE(r,t), considering both the

Kukhtarev equations [1,9,12,14,21,22] and linear electro-optic effect, is given by:

[illegible] (6)

in which, $\Delta E_{Dif}(r,t)$ is the diffusion field, $\Delta E_0(r,t)=V_{ext}/L$ is the extern electric field (drift field), $\Delta E_{FV}(r,t)$ is the photovoltaic field, $\Delta E_{Pyro}(r,t)$ is the pyroelectric field, I_e and I_d are respectively the excitation and dark intensities (I_d is the intensity at which photoconductivity is equal to the conductivity in the dark).

The space-charge field effects in light-induced lens on PR sillenite medium was studied by Ryf *et al* [14], who presented a theoretical treatment of this index change due to the Gaussian beam incidence in PR materials. The photorefractive sillenite crystals are not photovoltaic and pyroelectric materials, then the refraction index change is due a diffusion field or a extern electric filed.

3. Experimental details

The sillenite $Bi_{12}SiO_{20}$ (BSO) crystal used in this work were grown the melt by the Czochralski method. The crystal presents transverse electro-optic configuration and high optical quality. We used in our measures one $Bi_{12}SiO_{20}$ (BSO) crystal ($10.00\times10.00\times3.28\ mm^3$).

The TL experiment was performed using the dual beam mode-mismatched configuration, where the apparatus is described in detail elsewhere [20, 22-245]. The absorption of a relatively intense excitation beam generates the TL heat profile. The induced phase shift, which is proportional to θ, is measured by the weak probe beam that counter-propagates nearly collinear with the excitation one. The TL signal is obtained by measuring the on-axis probe beam intensity in the far field (an iris was put in front of the photodiode detector to select the central part of the probe beam). A He-Ne laser was used as probe beam (λ_p= 632.8 nm, w_p = 180 μm) and either a Ti:sapphire (λ_e =750 nm, w_e = 46 μm) or an Ar^+ (λ_e= 514.5 nm, w_e = 33 μm) laser was applied as excitation beam. The TL sensitivity is maximized using a probe beam waist much larger than the excitation one ($w_p >> w_e$), which characterizes the mode-mismatched configuration.

4. Results and Discussions

We analyze each separately case for the measures with different λ_e. Leaving of the fact of that the sillenite crystal has one high photorefractive sensitivity in the region blue-green spectra, we measure first at *750nm* for to obtain thermo-optic parameters and neglected the PR effect. Figure 1 shows the time-resolved and normalized TL signal for BSO at $\lambda_e=750\ nm$. The optical absorption coefficient of BSO is $A_e= 0.27\ cm^{-1}$[19,24]. The sample was performed under condition of open circuit, without extern electric field and the parallel [001] axis the direction of propagation of the pump beam. The transient curve was adjusted by Eq.(1) and the values obtained were of the parameter $t_T=(0.89\pm0.09)ms$ *and* $\theta_T=-(0.037\pm0.008)mrad$. Using t_T and ω_e values, we calculated the thermal diffusivity for the BSO crystal as being $D_T=(5.8\pm0.1)\times10^{-3}\ cm^2/s$. Knowing the value of $\rho c= 0.251JKcm^{-3}$*[26]*, we calculated the thermal conductivity as $K_T=(1.46\pm0.06)\times10^{-3}W/Kcm$. The procedure was repeated for diverse measures varying the power of the pump beam and the interval of acquisition time. These values are in accordance with the literature data: $D_T=5.8x10^{-3}cm^2/s$*[19]* and $K_T=1.5x10^{-3}W/Kcm$*[25]*. Figure 2 presents the curve of θ_T x P_e for the BSO crystal, with dependence of θ_T decreasing linearly with the increase of the P_e. The theoretical fitting was performed with θ_T (Eq. 6) and using the average values of θ_T, we calculated $(ds/dT)_{EXP}=(0.40\ \pm0.05)\times10^{-6}K^{-1}$. These results of the thermo-optic parameters are showed in Tables 1 and 2. We performed measurements with the crystal turned in 90 degrees to observe polarization effects, where we verify that the results undefended of the polarization of beams in this configuration.

In according to section 2, *ds/dT* is given by four contributions: *dn/dT*, end-face curvature, thermal stress and pyroelectric term. However, the sillenite crystal is not pyroelectric crystals. Then, an analysis of the thermal effect in the BSO crystals, *dn/dT* is positive; thus we would have a focusing of beam (*ds/dT>0)*, that until are it's agree with our experimental results (Fig. 2). Using the BSO literature room-temperature data, $dn/dT= -34.5\times10^{-6}K^{-1}$*(at 632.8nm)*; $\alpha(n_o-1)(1+\nu)= 33.1\times10^{-6}K^{-1}$, where $\alpha= 16.0\times10^{-6}K^{-1}$, $n_o=2.615$ and $\upsilon=0.28$; and, $n_0^3Y\alpha\, q^{eff}/4= 1.86\times10^{-6}K^{-1}$, where $q^{eff}\sim 0.031$ and $Y = 0.84$. Using these data in Eq. 8 we calculated $(ds/dT)_{cal}= 0.46\times10^{-6}K^{-1}$. This result is agree to a $(ds/dT)_{EXP}=(0.40\ \pm0.05)\times10^{-6}K^{-1}$. The measures performed at $\lambda_e=514.5nm$ in sillenite crystals.

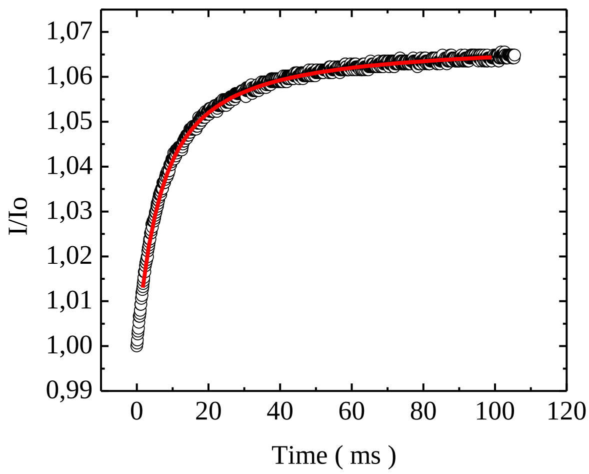

Fig. 1. Transient TL signal for BSO crystals for pump beam at λ_e= 750 nm. The values obtained from the curve fitting were t_T = (0.89±0.09) ms and θ_T=-(0.037±0.008)mrad.

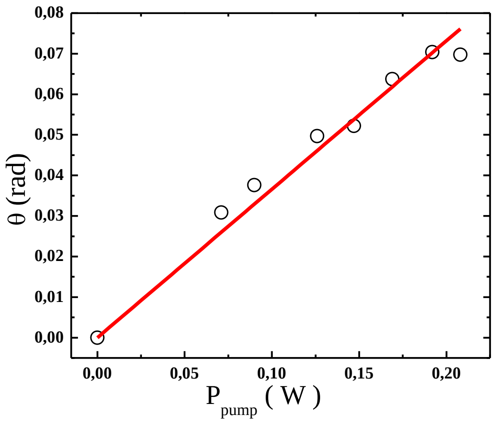

Fig. 2. Curve of θ_T x P_e for the BSO crystal and λ_e = 750 nm.

Figs. 3 and 4, present the highest photorefractive sensitivity of the light-induced fields, and it compels them to make a new analysis. More specifically the diffusion field, therefore the other light-induced fields in the sample can be neglected. Therefore the disturbance of the index will have now the strong dependent of the intensity of the pump beam $I_e(r)$ *at* λ_e due to thermo-optic and the diffusion regimen photorefractive effects dependence. How it is waited the disturbance in the index due $\Delta T(r,t)$ provokes in the light-induced lens a focusing effect of the beam, as can be verified for stops λ_e*=750.0nm* and for λ_e*=514.5nm* in an interval of time of the order of ms. However, it for λ_e*=514.5nm*, and intervals of time larger than ms, we observed a defocusing effect in the probe beam it can be explained by the strong influence of the diffusion regimen of the photorefractive effect in the gradient of refractive index in the illuminated region (to see section 2.3).

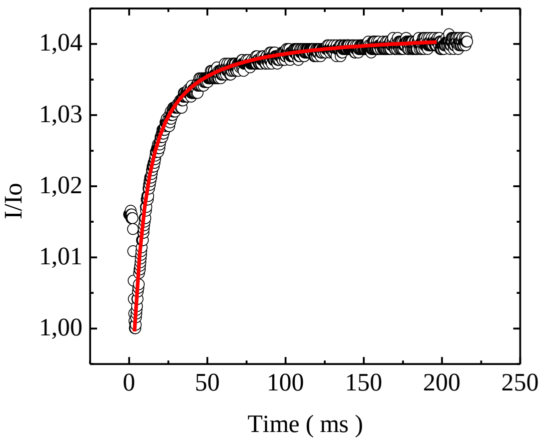

Fig. 3. Transient TL signal for BSO crystals for pump beam at λ_e= 514.5 nm.

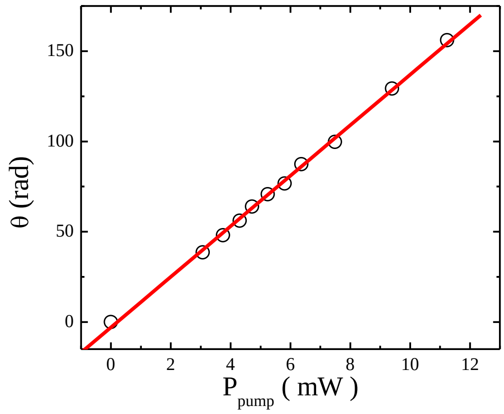

Fig. 4. Curve of θ_T x P_e for the BSO crystal and λ_e = 514.5 nm.

If neglected the photorefractive signal and adjust by Eq.(1), we are obtain the results of the thermo-optic parameters obtained for measures performed at λ_e*=514.5nm* in BSO crystal, showed in Tables 1 and 2. These results are agree to measures performed at λ_e*=750nm*. Then, the results performed at TLS technique are basically the same ones and in agreement with the values found in literature.

Table 1. Results of thermal diffusivity (D) and conductivity (K) for BSO and BTO crystals.

Sample	λ_e (nm)	tc (cm^{-1})	D (10^{-4} K^{-1})	ρc	K (10^{-4} K^{-1})
BSO	514.5	(0.47 ± 0.03)	(5.8 ± 0.1)	(0.25)	(14.6 ± 0.2)
	750.0	(0.89 ± 0.05)	(5.8 ± 0.6)	(0.25)	(14.5 ± 0.6)

Table 2. Results of temperature coefficient of the optical path length change (ds/dT) and temperature coefficient of the refractive index change (dn/dT) for BSO crystal

Sample	λ_e (nm)	α (cm^{-1})	Leff (cm)	ds/dT (10^{-6} K^{-1})	dn/dT (10^{-4} K^{-1})
BSO	514.5	(0.97 ± 0.03)	(0.28 ± 0.2)	(0.47 ± 0.04)	(34.5 ± 0.2)
	750.0	(0.27 ± 0.05)	(0.31 ± 0.5)	(0.40 ± 0.09)	(34.4 ± 0.2)

5. Conclusions

The light-induced lens technique, based in the thermal lens spectrometry model, was applied for the first time to investigate the focusing effect (or defocusing effect), the thermo-optics and photorefractive properties for sillenite $Bi_{12}SiO_{20}$ crystals. We determine thermo-optic parameters as *D, K, ds/dT* and we discussed the thermo-optical and photorefractive analysis in sillenite crystals. The values of thermal diffusivity and conductivity for these crystals obtained with the TLS technique are basically the same ones and in agreement with the values found in literature. The values of the temperature coefficient of the optical path length change (*ds/dT)* and temperature coefficient of the refractive index change (*dn/dT)* were obtained, few works present the experimental values of ds/dT and dn/dT in these crystals. For other side, in wavelengths where the sillenite crystals has strong photorefractive sensitivity (blue-green spectra) we observed a combination of the focusing and defocusing effects, and the light-induced field effect need to be considered in the variation of the optical path. Finally, this method is an easy-to-implement approach to determine quantitative values of the thermo-optical parameters (theses parameters are of relevance importance in PR materials) and is promising for thermal and photorefractive analysis in photorefractive and photosensitive materials.

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