Lloyd's mirror interferometer for holographic recording in photorefractive crystals

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ABSTRACT: We report the use of Lloyd's mirror interferometer for holographic recording in photorefractive material. The experimental results show that perturbations in the diffraction efficiency considerably decrease due the feature of that system; and, as a consequence, fast (electron-based) and slow (hole-based) gratings were recorded in a nominally undoped photorefractive Bi₁₂TiO₂₀ crystal. Measurement of the response times were determined as a function of light intensity for the 0-1.2 mW/cm² range using the 532 nm laser wavelength. Parameters that characterize the crystal were also determined and compared with available data in the literature in order to assess the reliability of this technique.

Palabras clave: Interferometry; Optical recording; Photorefractive materials

REFERENCES AND LINKS

- [1] P. Günter. Photorefractive Effects and Materials — Topics in Applied Physics: Photorefractive Materials and Their Applications I and II, ed. by P. Günter, J.-P. Huignard. Volume 61 and 62, Springer-Verlag (Berlin, Heidelberg), 1988.
- [2] J. Frejlich. Photorefractive Materials: Fundamental Concepts, Holographic Recording, and Materials Characterization. Wiley-Interscience, New York, 2006.
- [3] G.C.Valley. Erase rates in photorefractive materials with two photoactive species. Appl. Opt., 22:3160–3164, 1983.
- [4] J.P. Huignard and A. Marrakchi. Two-wave mixing and energy transfer in Bi₁₂SiO₂₀ crystals: application to image amplification and vibrational analysis. Opt. Lett., 6:622–624, 1981.
- [5] L.Solymar, D.J.Webb, and A.Grunnet-Jepsen. The Physics and Applications of Photorefractive Materials. Clarendon Press, Oxford, 1996.
- [6] Igor A. Sokolov. Adaptive photodetectors: novel approach for vibration measurement. Measurement, 27:13–19, 2000.
- [7] M.P. Georges, V.S. Scaufaire, and Ph.C. Lemaire. Compacta and portable holographic camera using photorefractive crystals. Applications in various metrological problems. Appl. Phys. B, 72:761–765, 2001.
- [8] Ph. Lemaire and M. Georges. Photorefractive materials and their applications 3, volume 115 of Springer Series in Optical Sciences, chapter Dynamic holographic interferometry: Devices and applications, pages 223–251. P. Günter and J-P. Huignard, 2007.
- [9] Freschi, A.A., De Vicente, F.S. Short-time kinetics of photorefractive gratings recorded under active phase control: an experimental method to materials characterization. Appl. Phys. B, 126, 16 (2020).
- [10] A.A. Freschi, F.A. Callegari, F.S. De Vicente, M.R.R. Gesualdi, Sign of the dominant charge carriers in photorefractive crystals determined by a phase-locked holographic technique, Optical Materials, Volume 48, 2015.
- [11] R. Montenegro, A. Shumelyuk, R. Kumamoto, J. F. Carvalho, R. C. Santana, and J. Frejlich. Vanadium-doped photorefractive titanosillenite crystal. Appl. Phys. B, 95:475–482, 2009.
- [12] V. Marinova, M. L. Hsieh, S. Lin, and K. Y. Hsu. Effect of ruthenium doping on the optical and photorefractive properties of Bi₁₂TiO₂₀ single crystals. Opt. Commun., 203:377–380, 2002.
- [13] J.Li, X.H.Li, F.Y.Lu, H.F.Wang, S.Z.Yuan, Y.Zhu, and X.Wu. Erasure of photorefractive gratings in barium titanate. J.Appl.Phys., 76:7541–7544, 1994.

- [14] Ivan de Oliveira, Renata Montenegro, and Jaime Frejlich. Hole-electron electrical coupling in photorefractive materials. *Appl. Phys. Lett.*, 95:241908, 2009.
- [15] L. Boutsikaris, S. Mailis, and N.A. Vainos. Determination of the photorefractive parameters of $\text{Bi}_{12}\text{SiO}_{20}$ by study of the dynamic behavior of complementary gratings. *J. Opt. Soc. Am. B*, 15:1042–1051, 1998.
- [16] I. Aubrecht, H.C. Ellin, A. Grunnet-Jepsen, and L. Solymar. Space-charge field in photorefractive materials enhanced by moving fringes: comparison of hole-electron transport models. *J. Opt. Soc. Am. B*, 12:1918–1923, 1995.
- [17] F. P. Strohkendl, J. M. C. Jonathan, and R. W. Hellwarth. Hole-electron competition in photorefractive gratings. *Opt. Lett.*, 11:312–314, 1986.
- [18] K. Buse. Light-induced charge transport processes in photorefractive crystals I: Models and experimental methods. *Appl. Phys. B*, 64:273–291, 1997.
- [19] Ivan de Oliveira and Jaime Frejlich. Dielectric relaxation time measurement in absorbing photorefractive materials. *Opt. Commun.*, 178:251–255, 2000.
- [20] A.A. Freschi and J. Frejlich. Adjustable phase control in stabilized interferometry. *Opt. Lett.*, 20:635–637, 1995.
- [21] Cédric Thizy, Marc Georges, Philippe Lemaire, Yvan Stockman, and Dominic Doyle. Phase control strategies for stabilization of photorefractive holographic interferometer. In Pierre Slangen and Christine Cerruti, editors, *Speckle06: Speckles, From Grains to Flowers*, volume 6341, pages 364 – 369. International Society for Optics and Photonics, SPIE, 2006.
- [22] Emilio J. Ambite and Luis Arizmendi. Feedback-controlled recording and fixing of photorefractive holograms in reflection geometry on lithium niobate crystals. *J. Opt. Soc. Am. B*, 28:1161–1167, 2011.
- [23] Haitao Jiang, Zhe Zhuang, Xiong Zhang, Cheng Li, Hu Chen, Aijie Fan, Shuai Chen, Xuguang Luo, Yiping Cui, Omnidirectional investigation of two-dimensional periodic nanostructure arrays by a two-axis Lloyd's mirror interferometer, *Results in Physics*, Volume 18, 2020.
- [24] James Leibold, Ribal Georges Sabat, Fabrication of micrometer-scale surface relief gratings in azobenzene molecular glass films using a modified Lloyd's mirror interferometer, *Optical Materials*, Volume 96. 2019.
- [25] Yuki Shimizu, Kazuki Mano, Hiroki Murakami, Shunsuke Hirota, Hiraku Matsukuma, Wei Gao, Design optimization of a non-orthogonal two-axis Lloyd's mirror interferometer for fabrication of large-area two-dimensional scale gratings, *Precision Engineering*, Volume 60, 2019, Pages 280-290.
- [26] Lin yong Qian, Bin Sheng, Yuan shen Huang, Jin zhong Ling, Rui jin Hong, Da wei Zhang, Bang lian Xu, Bai cheng Li, Zheng ji Ni, and Song lin Zhuang. Tilted and axis-shift lloyd's mirror system for recording low-density and large-area holographic grating. *Optik*, 125(3):1287–1291, 2014.
- [27] C. Lu and R.H. Lipson. Interference lithography: a powerful tool for fabricating periodic structures. *Laser & Photonics Reviews*, 4(4):568–580, 2010.
- [28] Ishan Wathuthanthri, Weidong Mao, and Chang-Hwan Choi. Two degrees-of-freedom lloyd-mirror interferometer for superior pattern coverage area. *Opt. Lett.*, 36(9):1593–1595, May 2011.
- [29] G.C.Valley. Simultaneous electron/hole transport in photorefractive materials. *J. Appl. Phys.*, 59:3363–3366, 1986.
- [30] C.H. Kwak and S.Y. Park and E-H Lee. Intensity dependent two-wave mixing at large modulation depth in photorefractive BaTiO_3 crystal. *Opt. Commun.*, 15:315–322, 1995.
- [31] Ivan de Oliveira, J.F. Carvalho, and Jaime Frejlich. Resonance running hologram velocity nonlinearity dependence upon light intensity in photorefractive crystals. *Appl. Phys. Lett.*, 102:251913–1–251913–3, 2013.
- [32] I. de Oliveira and J. Frejlich. Light intensity dependendent Debye screening length in undoped photorefractive titanosillenite crystals. *J.Appl.Phys.*, 112:113523, 2012.
- [33] I. de Oliveira and Agnaldo A. Freschi and Igor Fier and J. Frejlich. Stabilized photorefractive running holograms, with arbitrarily selected phase shift, for material characterization. *Opt.Mater.Express*, 2:228–234, 2012.

1. Introduction

Photorefractive crystals are photoconductive, electro-optic materials that allow records of diffraction gratings in the volume of the sample [1, 2]. The incidence of a sinusoidal light pattern on the sample excites charges carriers (electron and/or hole) from photoactive centers (localized states inside the band gap) to the conduction band (electron) or the valence band (hole). This process produces a spatial modulation of charge carriers that in turn generate an index of refraction modulation [3]. The photorefractive crystals have potential applications in different areas, such as image processing, optical devices, vibration and deformation sensors and photoelectric conversion devices [4, 5, 6, 7, 8]. The presence of hole-electron competition has been reported for different photorefractive materials [9, 10, 11, 12, 13]. As such, hole-electron competition in those materials gives rise to fast and slow gratings [12, 13, 14, 15].

The response time of photorefractive materials is an important parameter as that depends on conductivity [16, 17]. By measuring the response time, it is possible to determine the conductivity without any kind of electrodes. Although the conductivity of photorefractive crystals could be determined using direct electric techniques, the use of holographic strategies is far easier and more reliable. However, it is important to mention that while such techniques are useful, the experimental setup is very sensitive to environmental perturbations. This makes it difficult to obtain reproducible holographic recording.

As such, in order to obtain reproducible holographic recording, several researchers use holographic stabilized techniques [18, 19, 20]. However, the use of such techniques is made difficult by the need of specific equipment.

The use of compact interferometers for holographic recording is an alternative to stabilized techniques. One of such instruments is the Lloyd's mirror interferometer that have smaller arms compared to other similar devices, such as the transmission geometry interferometer. Lloyd's mirror interferometer has been widely used for the fabrication of micrometer-scale periodic structures [21, 22, 23, 24, 25, 26] due to its simplicity in configuration and to the control of pattern of interference fringes.

In this paper, we use the Lloyd's mirror interferometer to record holographic gratings in undoped $\text{Bi}_{12}\text{TiO}_{20}$ (BTO) photorefractive crystals. We show that the interferometer allows us to record stable and reproducible holograms in the volume of the sample. In addition, we also compare the recording holographic process using Lloyd's mirror interferometer with a typical interferometer by transmission geometry.

2. Theory

The recording of fast and slow gratings in photorefractive crystals is based on the two-photoactive centers model one center (donors) for electrons and the other (acceptors) for holes [27]. The action of light intensity on the ionized density is also considered [8]. The space-charge field evolution based on electron and hole charge carriers are respectively described by the equations below [2],

$$\tau_{sc1} \frac{\partial E_{sc1}}{\partial t} + E_{sc1} + \kappa_{12} E_{sc2} = -m E_{\text{eff}1} \quad (2.1)$$

$$\tau_{sc2} \frac{\partial E_{sc2}}{\partial t} + E_{sc2} + \kappa_{21} E_{sc1} = -m E_{\text{eff}2} \quad (2.2)$$

with the parameters,

$$E_{\text{eff}j} = \frac{i E_{Dj}}{1 + K^2 l_{sj}^2} \quad (2.3)$$

$$\tau_{scj} = \frac{\epsilon \epsilon_0 k_B T h \nu}{q^2 L_{Dj}^2 \alpha \Phi_j I} \frac{1 + K^2 L_{Dj}^2}{1 + K^2 l_{sj}^2} \quad (2.4)$$

$$(2.5)$$

$$\kappa_{12} = \frac{1}{1 + K^2 l_{s1}^2} \quad \text{and} \quad \kappa_{21} = \frac{1}{1 + K^2 l_{s2}^2}$$

where $i = \sqrt{-1}$ and the subindex $j=1$ is always for electrons and $j=2$ for holes, with E_D being the diffusion field, k_B the Boltzmann constant, T the absolute temperature, ϵ_0 the vacuum permittivity, Φ_j is the quantum efficiency for photoelectron generation, q the absolute value of the electron electric charge, $K = 2\pi/\Lambda$ is the value of hologram vector with Λ the period of spatial fringe pattern, ϵ the dielectric constant, $h\nu$ the photonic energy of the light, I the light intensity in the input plane inside the crystal and L_D, l_s are the diffusion length and the Debye screening length that are changed by light intensity; as such, it is possible to write these as [28, 29, 30],

$$L_{Dj} = \frac{L_{D0j}}{\sqrt{1 + \frac{I}{I_{sat}} \frac{N_{Dj}}{N_A}}} \quad (2.6)$$

$$l_{sj} = l_{s0j} \sqrt{\frac{I_{sat}}{I} \frac{N_A}{N_{Dj}}} \quad (2.7)$$

with $I_{sat} = \frac{\gamma_{Rj} N_A}{S_j}$ and N_A the density of nonphotosensitive negative ions that compensate for the electric charge of the (positively ionized) electron acceptors of density N_D^+ in the as-grown crystal, N_{Dj} the total photoactive centers density, S_j is the filled trap cross-section for photoelectron excitation, and γ_{Rj} is the recombination constant.

2.a. Diffraction Efficiency

The formulation of the diffraction efficiency η accounting for the hole-electron competition can be written as [2]

$$\eta \propto |E_{sc}^T|^2 \quad \text{with} \quad E_{sc}^T = E_{sc1} + E_{sc2} \quad (2.8)$$

where E_{sc1} and E_{sc2} are the solutions of set Eqs.(2.1-2.2) and with that we were able to write the diffraction efficiency as,

$$\eta = \left| A_f \left(1 - e^{-\frac{t}{\tau_f}} \right) + A_s \left(1 - e^{-\frac{t}{\tau_s}} \right) \right|^2 \quad (2.9)$$

with A_f and A_s are the amplitudes of the fast and slow gratings, respectively, τ_f and τ_s their corresponding space-charge response times that are given by,

$$\tau_f \approx \tau_{sc1} \quad (2.10)$$

$$\tau_s \approx \frac{\tau_{sc2}}{1 + \kappa_{12} \kappa_{21}} \quad (2.11)$$

with

$$\tau_{sc1} = \frac{\epsilon \epsilon_0 k_B T h \nu}{q^2 L_{D1}^2 \alpha \Phi_1 I} \frac{1 - e^{-\alpha d}}{\alpha d} (1 + K^2 L_{D1}^2) \quad (2.12)$$

$$\tau_{sc2} = \frac{\epsilon \epsilon_0 k_B T h \nu}{q^2 L_{D2}^2 \alpha \Phi_2 I} \frac{1 - e^{-\alpha d}}{\alpha d} (1 + K^2 L_{D2}^2) \quad (2.13)$$

where the approximations in Eqs.(2.10) and (2.11) derive from the assumption $\tau_s \gg \tau_f$ and $K^2 l_s^2 \ll 1$ and considering that the Maxwell relaxation times varies along of thickness of the sample due the light absorption.

3. Lloyd's Interferometer

Lloyd's interferometer consists of a mirror fixed perpendicularly to the sample as shown in Fig.1.

Fig.1. Lloyd's interferometer. Input 532 nm wavelength laser beam; **OBJ**, objective lenses ($f=4.5\text{mm}$); **L**, lenses ($f=15\text{ cm}$); **S**, sample (BTO) and **M**, Mirror.

In Lloyd's interferometer, one of the beams I_R travels directly to the sample, while the other I_S is reflected onto the sample by a mirror. These two beams form an interference pattern on the sample as depicted in Fig.2.

Fig.2. Schematic illustration of the recording setup: **S** is the $\text{Bi}_{12}\text{TiO}_{20}$ (**BTO**) crystal, I_R and I_S are the intensity of the two incident recording beams, whereas I_R^t and I_S^t are the transmitted beams, I_R^d and I_S^d the diffracted beams behind the crystal. **P** is the polarizer and **D** the photodetector. The inset shows the corresponding $[110]$ and $[001]$ axis directions.

4. Experiments and Results

Holograms were recorded using Lloyd's interferometer as shown in Fig.1, on a nominally undoped titanosillenite photorefractive $\text{Bi}_{12}\text{TiO}_{20}$ (BTO) crystal ($\alpha=0.85\text{ mm}^{-1}$ and $d=2.5\text{ mm}$ thickness) using the 532 nm wavelength line with the wave vector $\vec{K}$ ($K = 16.7\mu\text{m}^{-1}$) parallel to the $[110]$ axis and with $[001]$ crystal axis perpendicular to the incident plane. In order to obtain uniform irradiances over the entire surface of the sample and the mirror, the incident beam was expanded and collimated. Because of the very low diffraction efficiency involved (less than 0.1% for the BTO) self-diffraction can be neglected and the diffracted light ($I_S^d \propto \eta$) can be used to describe the holographic dynamics [2]. The hologram was recorded under different total light intensities ($I=I(0)=I_R+I_S$) while taking into account a ratio of $I_R/I_S \approx 12$ and the results are shown in Fig.3 with the continuous curve chosen to best fit Eq.(2.9).

Figure 3: Time evolution of the diffraction efficiency (η) during the holographic recording for different light intensities. The continuous curve is the best fitting with Eq.(2.9) leading $A_I=0.296$, $A_S=0.147$, $\tau_I=5.53$ s and $\tau_S=36.7$ s.

In order to compare the stability of Lloyd's interferometer, a different experiment involving holographic recording in the same BTO sample was carried out, but using a typical interferometer by transmission geometry. Results are shown in Fig.4. The analysis of data shows that the diffraction efficiency η is much more unstable for the holographic recording by transmission geometry than for Lloyd's interferometer.

Figure 4: Time evolution of the diffraction efficiency (η) during the holographic recording for different laser power beams using a typical interferometer by transmission geometry.

In order to verify the stability of Lloyd's interferometer in Fig.5 it is shown, $\bar{\eta}_s/\Delta\eta_s$, as a function of laser beam power, where $\bar{\eta}_s$ is the average of the signal during the steady-state and $\Delta\eta_s$ the respective square deviation. The symbols (●) and (▲) corresponding to Lloyd's interferometer and the classical geometry (transmission) respectively. Furthermore, the parameter τ_s it was determined from data fitting with Eq.(2.9) of data shown in Fig.4, and the results (■) are shown in Fig.5. It is important to stress that the effect of the instability during the recording process is more important on the recorded of the slow grating and thus producing unreliable data for the parameter τ_s as shown in Fig.5.

Figure 5: Parameter $\bar{\eta}_s/\Delta\eta_s$ as a function of laser beam power for (●) Lloyd's interferometer and (▲) for classical geometry (transmission) respectively. The symbol (■) corresponding to the parameter τ_s for the slow grating recorded using the classical geometry.

From data fitting with Eq.(2.9), as shown by the continuous curve in Fig.3, the parameters τ_f and τ_s were obtained to different light total intensity $I(0)$. Results are shown in Fig.6 where the continuous curves are the best fitting with Eq.(2.12) and (2.13) respectively, leading to the best fitting parameters shown in Table 1.

Figure 6: Plotting the inverse of $\tau_{f,s}$ vs light intensity $I(0)$. The continuous curve is the best fitting with Eq.(2.12) (left) and Eq.(2.13) (right). The resulting best fitting parameters are: $L_{D0}^f = 0.18 \mu\text{m}$, $\Phi_f = 0.19$ and $I_{\text{sat}} \frac{N_A}{N_D} = 7.94 \text{ W/m}^2$ for the fast grating and $L_{D0}^s = 0.31 \mu\text{m}$, $\Phi_s = 0.036$ and $I_{\text{sat}} \frac{N_A}{N_D} = 0.44 \text{ W/m}^2$ for slow grating.

TABLE 1. Parameters from fitting of data in Fig.5 and 6.

Parameter	Fast grating	Slow grating
L_{D0}	$0.184 \mu\text{m}$	$0.31 \mu\text{m}$
Φ	0.19	0.036
$I_{\text{sat}} \frac{N_A}{N_D}$	7.94 W/m^2	0.44 W/m^2

The results in Table 1 show the most reliable parameters found for electrons and holes. These parameters, at least for electrons, are not far from those ($L_{D0}=0.18\text{ }\mu\text{m}$, $\Phi_1=0.2$ and $I_{sat}\frac{N_A}{N_D}=7\text{ W/m}^2$) already published [26] for the same sample. Besides that, the parameters for holes are roughly coherent with those reported ($L_{D2}=0.485\text{ }\mu\text{m}$ and $\Phi_2=0.011$) in Ref.[33] but for a different wavelength (514 nm).

5. Conclusions

This paper shows that using Lloyd's interferometer for recording holographic in photorefractive crystals produces more stable stationary holograms. The stability produced by Lloyd's interferometer allowed for the detection of hole (slow grating) even with their expected lower concentration. Detection of fast (based on the electron) and slow (based on the hole) gratings was experimentally verified and allowed for the independent determination of parameters for both types of charge carriers, as reported in Table 1. The parameters obtained in this work are in good agreement with data obtained from other techniques.

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