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HOLOGRAPHIC GRATING RECORDING IN "LYOTROPIC LIQUID CRYSTAL – VIOLOGEN" SYSTEM

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Abstract. *This work presents the results of nonlinear optical experiment run on the samples of lyotropic liquid crystal (LLC) with viologen admixtures. During the experiment we obtained dynamic grating recording on bilayered LLC-viologen samples and determined main characteristics of recoded gratings. It was found out that the recording takes place in a thin near-cathode coloured viologen layer. The analysis of kinetics of thermal gratings erasing showed that contribution of a thermal nonlinearity into general diffraction efficiency is negligible small. The last fact is connected with a separation of LLC-viologen samples under the action of an electric field and heat sink into the liquid crystal layer.*

Keywords: diffraction efficiency; diffraction grating; lyotropic liquid crystals; nonlinear optics; viologens.

1. Introduction

The article [Bordyuh, Polishchuk 2012] is devoted to a detailed investigation of optical and electrooptical properties of the new-created composite liquid crystalline materials based on lyotropic liquid crystal (LLC) of Potassium caprylate doped with viologen admixtures. It was shown that there is a possibility to manage electrooptical prorerties of the investigated composites. Furthermore, their structural peculiarities let us foretell nonlinear properties of LLC-viologen materials.

2. Analysis of investigations and publications

Nonlinear properties of liquid crystals still remain insufficiently known. However, studying of nonlinear properties of the lyotropic ionic Smectic formed by Potassium caproate and doped with polymethine dyes was presented in the works [Bugaychuk et al. 2005; Garbovskii et al. 2006; Klimusheva et al. 2006]. These works prove the possibility of using doped liquid crystals for diffraction grating recording.

The **aim** of the present work is to reveal nonlinear properties of LLC-viologen composites, realize grating recording, and study the main nonlinear optical characteristics of the investigated samples.

3. Materials and methods

Lyotropic Liquid Crystalline phase was formed at mixing powder of a Potassium caprylate ($C_7H_{15}COO^-K^+$) with water in 1:1 weight proportion at the room temperature (T = 293 K). Then samples of LLC were doped by the viologens of two types:

N,N'-diheptyl-4,4'-dipyridilium dibromide ($HD^{2+}2Br^-$);

N,N'-di(2-carboxyethil)-4,4'-dipyridilium dichloride ($CED^{2+}2Cl^-$).

The viologens differ in substitutes at Nitrogen atoms and counterions. X-ray investigations [Bordyuh, Polishchuk 2012; Bordyuh 2010] have shown that the created LLC obtains the structure of Smectic A. The Smectic ordering remains when doping liquid crystalline matrix by viologen admixtures (2% by weight).

Nonlinear optical investigations of LLC-viologen samples were conducted with using glass sandwich-cells whose inner surface was covered with ITO-electrodes.

In the case of external electric field application to the cells filled with LLC-viologen composite one can observe the colouration of the samples taking place due to a reduction of viologen molecules. Thus, one-electron reduction corresponds to the formation of radical cations and blue colouration, while two-electron reduction with subsequent dimerization goes with red colour of the samples [Bordyuh 2010].

Reduction of viologens takes place near cathode which serves as an electron donor for viologen molecules. The reduction results in a formation of a bilayer cell which consists of insoluble coloured layer of viologen reduction products and a liquid crystal layer.

The standart two-beam scheme was applied for diffraction grating recording in the coloured samples of LLC-viologen. The second harmonic radiation of pulse Q-switched Nd:YAP laser (TEM_{00}-mode, radiation wavelength λ = 539,8 nm, pulse duration τ = 20 ns, pulses frequency ν = 3 Hz) was used.

For studying the kinetics of gratings erasing in a microsecond time range recorded gratings were read by a testing nonpolarized radiation of continuous He-Ne laser (power P = 2 mW, λ = 632,8 nm).

4. Diffraction grating recording

Nonlinear optical experiments were realized for the coloured LLC samples doped with viologens $HD^{2+}2Br^{-}$ and $CED^{2+}2Cl^{-}$. The wavelength of a recoding laser radiation (λ = 539,8 nm) falls within an absorption band of LLC-viologen samples [Bordyuh et al. 2009]. Linearly polarized in a horizontal plane laser radiation was divided into two beams of about equal intensities, which were then converged on a sample at a given angle. Both the incident intensity and the intensity in the first diffraction order were registered by photodiodes. On the base of these measurements we studied the dependence of the self-diffraction efficiency on the converging angle and the laser radiation intensity.

The dynamic diffraction grating was recorded in consequence of a complex refraction index modulation in an interferential field of two laser beams converged on the sample. For all the samples the self-diffraction effect was observed; moreover, one laser pulse was enough for a grating recording. Usually we observed few diffraction orders in a self-diffraction regime. This fact along with the small value of Cook-Klein parameter (Q < 0,1) points at a thin grating recording.

Experimental dependences (in logarithmic scale) of the diffraction efficiency on the intensity of an exciting laser radiation are shown in Fig. 1.

The grating period was Λ = 20 μm, the samples optical density was equal to D = 0,45-0,46 at the radiation intensity of I = 0,38 MW/cm^2. As it is seen from the given dependences $lg\eta = 2lgI$, measured in a self-diffraction regime diffraction efficiency depends quadratically on the intensity of recoding laser radiation. Such a dependence explicitly points at the third-order optical nonlinearity of the investigated LLC-viologen samples [Eichler et al. 1986]. Experimental data obtained as a result of a set of measurements with different samples showed satisfactory recurrence of their results [Bordyuh 2010].

Typical dependence of the optical density on the laser radiation intensity is shown in Fig. 2.

The similar dependence was observed for LLC doped with viologens $HD^{2+}2Br^{-}$ and $CED^{2+}2Cl^{-}$. As it is seen from the shown graph nonlinear absorption is insignificant in the intensities working range, thus the recorded gratings are mainly phase ones in both cases.

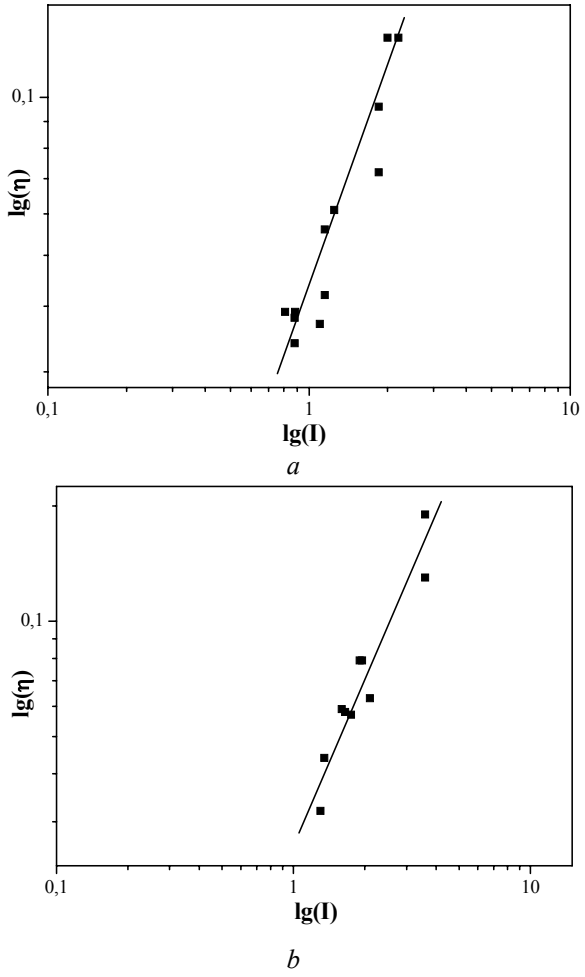

Fig. 1. Diffraction efficiency dependence (η) on the value of the laser radiation intensity (I) for the coloured samples of LLC-$HD^{2+}2Br^{-}$ (*a*) and LLC- $CED^{2+}2Cl^{-}$ (*b*)

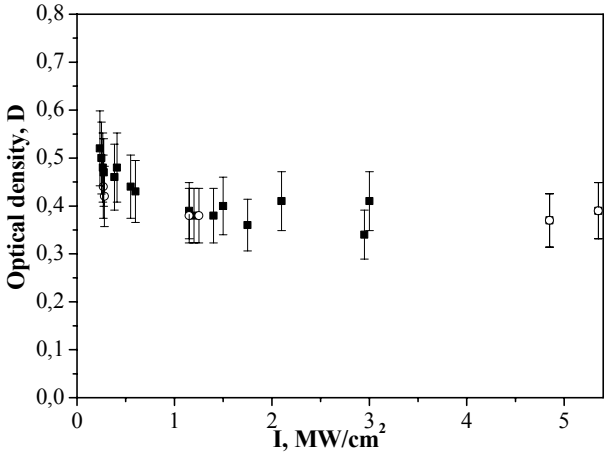

Fig. 2. Optical density dependence (D) on the value of the laser radiation intensity (I) for LLC-viologen samples:
■ corresponds to the measurements at an intensity increase;
○ corresponds to the measurements at an intensity decrease

5. Kinetics of gratings erasing

The investigations of gratings erasing kinetics were conducted in a microsecond time range. For this time range we observed an exponential time-dependence of the intensity in the first diffraction order (Fig. 3), which is typical for thermal gratings.

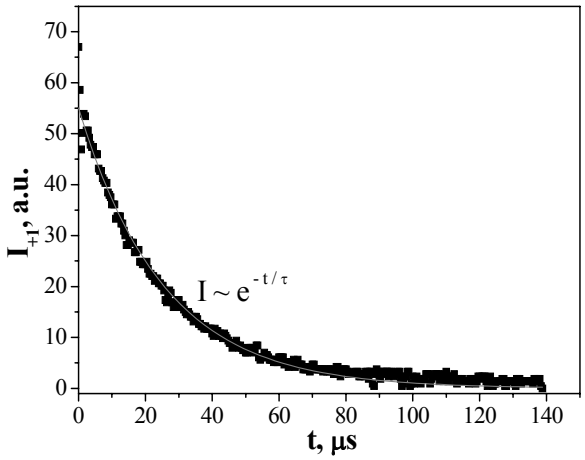

Fig.3. Kinetics of gratings erasing in a microsecond time range for LLC-viologen samples

Calculated for a grating period of Λ = 15 μm decay constant is equal to τ = 25 μs. The comparison of the diffraction efficiency of thermal gratings η_T with the diffraction efficiency in a self-diffraction regime η (Fig. 1) testifies that $\eta_T << \eta$ ($\eta/\eta_T > 100$). This means that diffraction gratings recorded in a self-diffraction regime are not determined by a thermal mechanism.

Resembling results were obtained in the works [Klimusheva et al. 2006; Bugaychuk et al. 2006] for a dynamic grating recording in bilayer cells "dye film – lyotropic ionic liquid crystal (LILC)". In these works small diffraction efficiency of thermal gratings was explained as following: during nanosecond recording times thermal gratings appear in a thin LILC layer near the border between dye film and liquid crystal.

Similarly, the most probable cause of thermal gratings small diffraction efficiency lays in a separation of the sample under the action of an external electric field. In this case thermal gratings appear near the place of contact between LLC layer and photosensitive viologen layer. This contact provides effective rejection of the energy absorbed by viologens into the LLC layer. In that way the absence of an excessive heating of the coloured viologen film prevents the formation of a stationary relief. That is, the separation of a sample explains the dynamic nature of recoded gratings.

In bilayer cell LLC performs one more important role: in consequence of its high viscosity and elastic characteristics liquid crystal makes impossible the diffusion process of coloured viologen reduction products to the LLC volume. This helps to preserve the morphology of viologen film. Formation of the photosensitive film on the cathode also leads to an increase in diffraction efficiency in comparison with the case when admixtures are uniformly distributed in the whole volume of a liquid crystal [Bugaychuk et al. 2006].

Mechanism of the optical nonlinearity, just as diffraction grating recording in LLC-viologen composites could relate with both nonlinear polarization of delocalized π-electrons of viologens and transformations of photosensitive viologen molecules in the field of an intense laser radiation. This problem demands careful analysis and additional calculations, which are to consider in the following works.

6. Conclusions

1. The diffraction grating recording by nanosecond laser pulses was obtained and studied for the LLC-viologen composites. It was determined that recording takes place in a self-diffraction regime and has dynamical nature.

2. It was ascertained that LLC-viologen composites are characterized by the third-order optical nonlinearity and the recorded gratings are mainly phase ones. It was proved that the recording takes place in a thin coloured layer of viologen reduction products, which is formed on the cathode under the action of an electric field.

3. It was specified that the residual thermal gratings have diffraction efficiency which is much less than the efficiency in a self-diffraction regime. Small diffraction efficiency of thermal gratings relates with their appearance in the thin LLC layer near the place of contact between liquid crystal and coloured viologen film.

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Г.Б. Бордюг[1], А.П. Поліщук[2]. Голографічний запис в системі "ліотропний рідкий кристал – віологен"
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Наведено результати нелінійно-оптичного експерименту, проведеного для зразків ліотропного рідкого кристалу (ЛРК) з домішками віологенів. Експериментально здійснено запис динамічних дифракційних ґраток на двошарових зразках ЛРК-віологен. Установлено, що запис відбувається в тонкому прикатодному забарвленому шарі віологенів. Визначено основні характеристики записаних ґраток. Проведено аналіз кінетики затухання залишкових теплових ґраток. Розглянуто вклад теплової нелінійності в дифракційну ефективність зразків.
Ключові слова: віологени; дифракційна ґратка; дифракційна ефективність; ліотропні рідкі кристали; нелінійна оптика.

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Приведены результаты нелинейно-оптического эксперимента, проведенного для образцов лиотропного жидкого кристалла (ЛЖК) с примесями виологенов. Экспериментально получена запись динамических дифракционных решеток на двухслойных образцах ЛЖК-виологен. Установлено, что запись происходит в тонком прикатодном окрашенном слое виологенов. Определены основные характеристики записываемых решеток. Проведен анализ кинетики затухания остаточных тепловых решеток. Рассмотрен вклад тепловой нелинейности в дифракционную эффективность образцов.
Ключевые слова: виологены; дифракционная решетка; дифракционная эффективность; лиотропные жидкие кристаллы; нелинейная оптика.

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