CUBIC SUSCEPTIBILITY AND HYPERPOLARIZABILITY OF THE LYOTROPIC LIQUID CRYSTAL–VIOLOGEN SYSTEM

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Nonlinear optical properties of the "lyotropic liquid crystalviologen" system are investigated. The system shows electrochromism under the action of an external electric field. For the colored samples, the recording of dynamic holographic gratings is obtained, by using the pulse laser method. The main characteristics of a recording on such samples are studied. On their base in view of the electrooptical properties of the system, the mechanism of the recording of gratings is proposed. The recording is realized due to a change in the polarizability of the π -electron system of viologen derivatives under the action of an intense laser radiation. The main nonlinear optical parameters (n₂, $\chi^{(3)}$, γ) of the system under study are determined.

1. Introduction

It is known that dipyridyl quaternary salts (viologens) in solutions [1,2], polymeric matrices [3], liquid crystals [4], and in the crystalline state [5] exhibit various physicochemical properties such as electro-, thermo-, and photochromism. Complexes based on viologens could be used as redox indicators, light-sensitive systems, electron transfer catalysts, color filters of variable density, *etc.*

"Lyotropic liquid crystal-viologen" systems show electrochromism under the action of an external electric field. Earlier [4], main electrooptical characteristics of these systems were studied, and peculiarities of the redox reactions of viologens in a liquid crystal matrix were determined.

The present work considers both the nonlinear optical properties of the system and the holographic recording under the action of a pulse laser radiation on samples of a lyotropic liquid crystal with admixtures of viologens.

2. Materials and Experimental Methods

The lyotropic liquid crystal (LLC) matrix is formed by potassium caprylate with water (the chemical formula is $C_7H_{15}COOK:H_2O$) in 1:1 proportion. As electrochromic admixtures, we chose two viologens, namely: (1) N,N'diheptyl-4,4'-dipyridilium dibromide (HD²⁺2Br⁻) and (2) N,N'-di(2-carboxyethil)-4,4'-dipyridilium dichloride (CED²⁺2Cl⁻) which differ by substitutes near nitrogen atoms and counterions. The common structural formula for both viologens is as follows:

$$R - N^{+}$$
 $N^{+} - R 2A^{-}$

where R is a substitute, A is a counterion. In the case of $HD^{2+}2Br^{-}R = C_{7}H_{15}$, $A^{-} = Br^{-}$, and $R = (CH_{2})_{2}COOH$, $A^{-} = Cl^{-}$ for $CED^{2+}2Cl^{-}$. The content of viologens in the samples is 2% by weight.

The structure of a viologen-doped LLC had been investigated in [6] using a method of small-angle X-ray scattering. Studied LLC-viologen samples have structure of smectic A. Admixture concentrations of 2–4% by weight do no break the LC ordering, and viologen molecules are built into the LLC matrix.

The LLC matrix performs significant functions in the investigated samples: 1) as a consequence of a planar orientation of the LLC, admixture molecules are also plane-oriented; this facilitates the electric field-stipulated adsorption of viologen molecules at the cathode with the further formation of a photosensitive layer of viologen's reduction products; 2) high viscosity and elastic characteristics of LLC promote an increase of the lifetime of colored reduction products and the maintenance of a layer morphology during holographic recording; 3) a heat contact between the viologen layer

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