

# Photo-dynamics of polarization holographic recording in spirooxazine-doped polymer films

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## Abstract

Polarization holographic gratings were recorded in 6-cyano-1,3,3-trimethylspiro[indolino-2,3-[3H]naphtha-[2,1-b][1,4]oxazine] (SO-1)-doped PMMA films. A photochemical equilibrium was observed in holographic gratings recorded by (s,s) polarized He–Ne beams with accompanying UV irradiation. For comparison, (s,p) polarization holographic recordings were also performed. The observed maximum value of the diffraction efficiency of (s,s) gratings was higher than that of (s,p) gratings. The mechanism of the photo-dynamics of the two polarization holographic recordings for SO-1 was investigated in detail.

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## 1. Introduction

Spirooxazines (SOs) and spiropyranes (SPs) represent an important class of photochromic compounds [1–7]. In contrast with spiropyranes, SOs exhibit high light stability and fatigue resistance [8,9], which provide the possibility of practical applications in lenses of variable optical density, displays, filters, and optical-storage devices. Photochromism in SOs and SPs arises from carbon-oxygen (C<sub>spiro</sub>–O) bond cleavage in the colorless spiro form upon UV-light excitation and subsequent isomerization to colored open forms, called photomerocyanines (PMCs). PMCs can revert to the spiro forms thermally or photochemically. Hence, it is possible to perform holographic recording with visible light for SOs pre-irradiated by UV-light. Previously, most studies of SOs focused on photochromism and optical storage employing UV, Ar<sup>+</sup>, or CO<sub>2</sub> lasers as the writing beams [10–

12]. Recently, photo-induced birefringence and polarization holographic recording for spirooxazines was accomplished with red light [13]. However, little is known about the photodynamic mechanism of polarization holographic recording for SOs.

In this work, polarization holographic recording for 6-cyano-1,3,3-trimethylspiro[indolino-2,3'-[3H]naphtha-[2,1-b][1,4]oxazine] (SO-1) doped PMMA films irradiated by UV-light was accomplished by 632.8 nm He–Ne laser light. The mechanism of the photo-dynamics of polarization holographic recording for SO-1 was investigated in detail.

## 2. Experiment

### 2.1. Materials and preparation

The chemical structures of 6-cyano-1,3,3-trimethylspiro[indolino-2,3-[3H]naphtha-[2,1-b][1,4]oxazine] (SO-1) and the corresponding UV-induced photomerocyanine (PMC)

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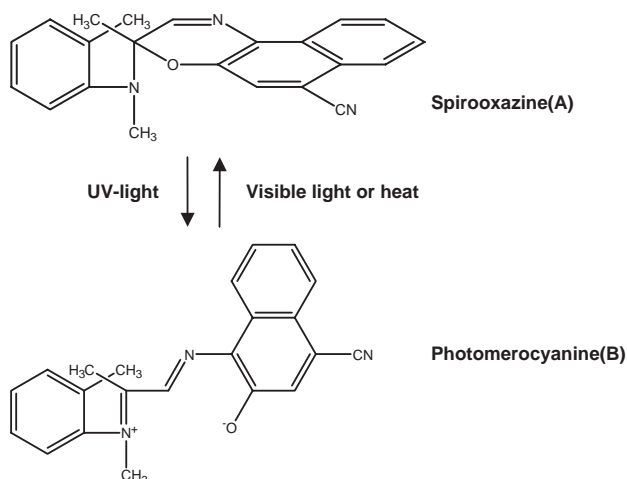


Fig. 1. Chemical structure of SO-1 and corresponding UV-induced PMC form.

are shown in Fig. 1. SO-1 was synthesized with microwave irradiation [14]. Commercially available poly(methyl methacrylate) (PMMA) was used without further purification. Both SO-1 and PMMA were dissolved in  $\text{CHCl}_3$  and then cast on a clean glass substrate. After solvent evaporation, the resulting composite film was used for the measurement of polarization holographic recording. The pre-set dye concentration was 10 wt.%, and typical film thickness was  $10 \pm 1 \mu\text{m}$ , as measured with a Precision Ellipsometer.

## 2.2. UV-Vis measurements

Fig. 2 shows the UV-Vis spectra of PMMA films containing SO-1(a) and PMC(c), obtained with a Perkin-Elmer Lambda 900 spectrophotometer over the range of 190–3200 nm. The absorption bands at 365 nm correspond to  $\pi-\pi^*$  electronic transition for both SO-1 and PMC, while the absorption near 633 nm corresponds to  $n-\pi^*$  electronic transition and intermolecular charge-transfer of PMC [15]. For comparison, the UV-Vis spectrum of SO-1 in dilute solution is also shown (b). The absorption maximum ( $\lambda_{\text{max}}=650 \text{ nm}$ ) in the visible wavelength of the PMC-doped PMMA film is red-shifted by 86 nm relative to that in

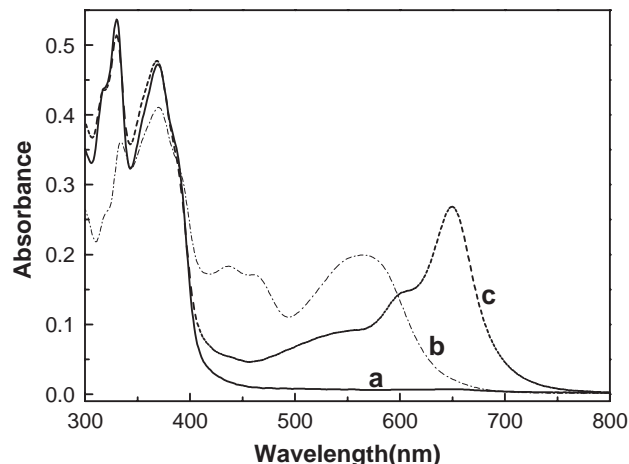


Fig. 2. UV-Vis spectrum of PMMA film doped with 10 wt.% SO-1 (a), the corresponding spectrum after UV-induced isomerization of SO-1 to PMC (b), and the spectrum of SO-1 doped in  $\text{CHCl}_3$  after UV irradiation with the concentration of 0.01 mg/ml (c).

dilute solution (564 nm), indicating a very strong J-aggregation of PMC molecules in the polymer films [16].

## 2.3. Optical setup

Holographic recording measurements were made possible by two interferential laser beams irradiating the sample, as shown in Fig. 3. The major elements of the experimental setup were as follows. A linearly polarized He-Ne laser (632.8 nm) generated the writing and reading beams. A half-wave plate was used to rotate the polarization of the He-Ne beam. The diameter of the laser beams was  $\sim 0.3 \text{ cm}$ , and the intersecting angle between the beams was  $\sim 10^\circ$ . A mercury-arc lamp (125 mW) was used to irradiate the sample with homogenous, incoherent light at 365 nm ( $\sim 1 \text{ mW/cm}^2$  at the sample location). The diffractive signals were registered on a CCD camera interfaced with a computer.

## 3. Results and discussion

Fig. 4 shows time evolution of the first-order diffractive signal of the holographic gratings generated by two He-Ne

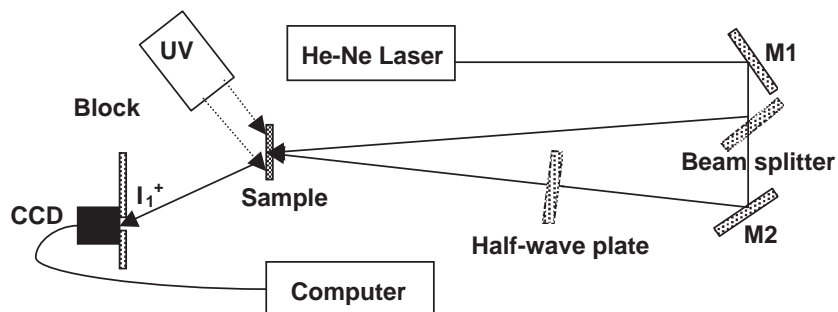


Fig. 3. Experimental configuration for the recording of holographic gratings by two He-Ne laser beams, where M denotes a mirror, P a polarizer, CCD a charge coupled device (sensitive region from 350 to 900 nm).

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