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# Dye adsorption induced nonvolatile reading of erasable polarization holograms in liquid crystal films

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**Abstract.** A nonvolatile reading of erasable polarization holograms in dye-doped liquid-crystal films using the same writing wavelength is presented. The recorded hologram can be easily erased with the illumination of one *p*-polarized wave; nevertheless, it becomes nonvolatile when reading the hologram with one *s*-polarized wave. We have proven the nonvolatile reading property is induced by dye adsorption on the substrate of the sample. © 2011 Society of Photo-Optical Instrumentation Engineers (SPIE). [DOI: 10.1117/1.3599517]

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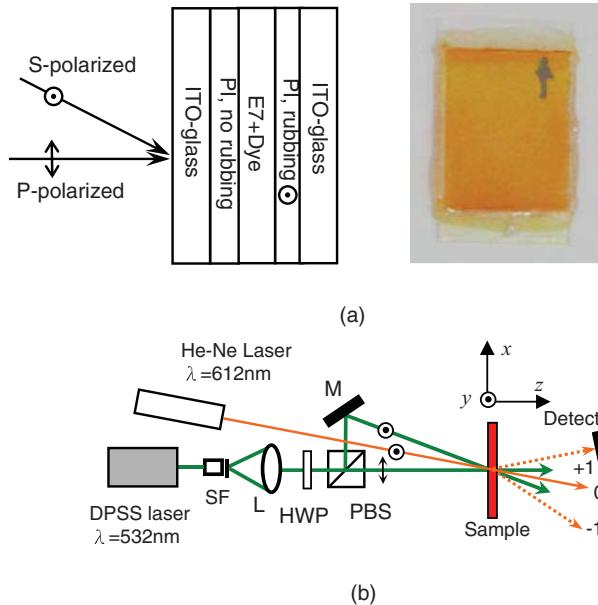
Photorefractive crystals<sup>1,2</sup> and azo-dye-doped liquid crystals<sup>3–5</sup> are currently attractive rewritable materials for holographic memory. However, the reversibility of these materials also leads to volatile readout, i.e., the recorded hologram would be gradually erased during the readout process. For photorefractive crystals, one method for overcoming the obstacle is using a fixing technique,<sup>1,2</sup> and the other solution is to read the hologram at a longer wavelength.<sup>6</sup> For azo-dye-doped liquid crystals, owing to their reversible process of trans–cis photoisomerization makes them become the promising rewritable materials for holographic optical recording. Even though it has been proven that the recorded holograms in azo-dye-doped liquid crystals can persist for a long time in the dark,<sup>5</sup> using the same recording wavelength for readout is much less discussed. Previous research groups still use a light with a longer wavelength far from the absorption band as a reading beam to avoid erasure effect. In this letter, we demonstrate our study on nonvolatile reading property of recorded holograms in azo-dye-doped liquid crystals using the same recording wavelength.

The dye-doped liquid crystal films used in this experiment were fabricated with nematic liquid crystal E7 and methyl red (MR). The mixing ratio, MR:E7, of these two components was 4:96 wt. %. Two glass slides coated with

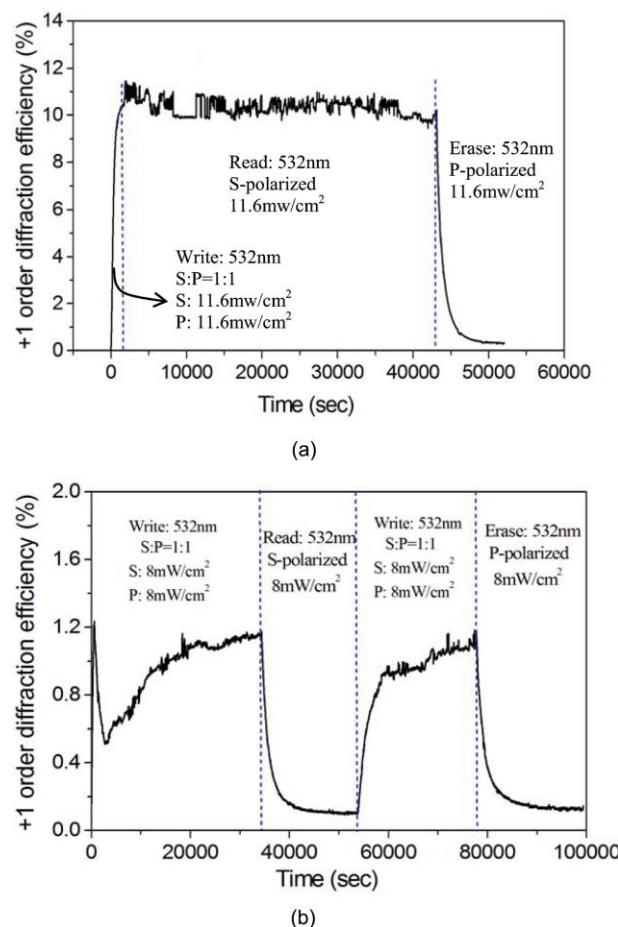
indium tin oxide were separated by 6 μm plastic spacers to fabricate an empty cell with dimension of 2 cm × 3 cm. Two glasses are coated with polyimide film, and only one of them is rubbed. The homogeneously mixed compound was then injected into the empty cell to form a dye-doped liquid crystal sample. The liquid crystal molecules were aligned with the rubbed surface. The sample structure is shown in Fig. 1(a). If irradiating an azo-dye-doped liquid crystal film with two orthogonally polarized waves, the polarization direction of the generated interference light field is periodically modulated in space. Then the azo-dye molecules are photoexcited from trans-state to cis-state and their molecular reorientation is orthogonal to the local polarization direction of the spatially modulated field. The reoriented dyes will diffuse and then adsorb onto cell substrates to generate a microgroove structure. The process is accompanied by liquid crystal (LC) reorientation, and accordingly a polarization hologram is generated.<sup>4</sup>

The experimental setup for generating a polarization hologram in dye-doped liquid crystal films is shown in Fig. 1(b). The polarization holographic gratings were generated by one *s*-polarized and one *p*-polarized plane wave. The rubbing direction of the sample was perpendicular to the incident plane of the experimental system. In this experimental setup, the incident angle of reference beam was kept at 45° in the air. An *s*-polarized He–Ne laser beam with wavelength of 612 nm was used to probe the grating. The intensity of the probe beam was about 50 μW/cm<sup>2</sup>. A detector was used to measure the diffracted power of the +1 order diffraction. In the writing process, the polarization holographic gratings were generated by two orthogonally polarized writing beams with equal intensity. In the reading process, we blocked the *p*-polarized recording beam, and an *s*-polarized recording beam with 11.6 mW/cm<sup>2</sup> intensity was incident on the sample. Then, in the erasable process, we blocked the *s*-polarized recording beam, and a *p*-polarized recording beam also with 11.6 mW/cm<sup>2</sup> intensity was incident on the sample. The dynamic change of diffraction efficiency measured by the probed beam during recording, reading, and erasable process is shown in Fig. 2(a). From Fig. 2(a), we can find when the hologram was illuminated with one *s*-polarized wave, the diffraction efficiency of the hologram was very stable during the continuous 6 h readout process. Once the hologram is illuminated with a *p*-polarized wave, the holographic grating is erased effectively.

For proving the effect of dye adsorption on a nonvolatile reading, we prepared another sample in which doped dye does not adsorb on a substrate. The new sample structure was the same with MR doped liquid crystals except MR dyes were replaced with D2 dyes. We repeated the writing, reading, and erasing process. The dynamic change of diffraction efficiency is shown in Fig. 2(b). We found that when the recorded hologram was either illuminated with one *s*-polarized wave or one *p*-polarized wave, the hologram would be erased. Apparently, nonvolatile reading property is not observed in D2-doped liquid crystals. Adsorbed dyes after the writing process can be observed by the following material analyses. First, we recorded one polarization hologram in each of the two different dye-doped samples. After that, the recorded samples were decomposed into two glass slides, and then the liquid crystals were removed from the material system. We found the residual MR dyes (red color) were adsorbed on the inner surface of the un-rubbed glass

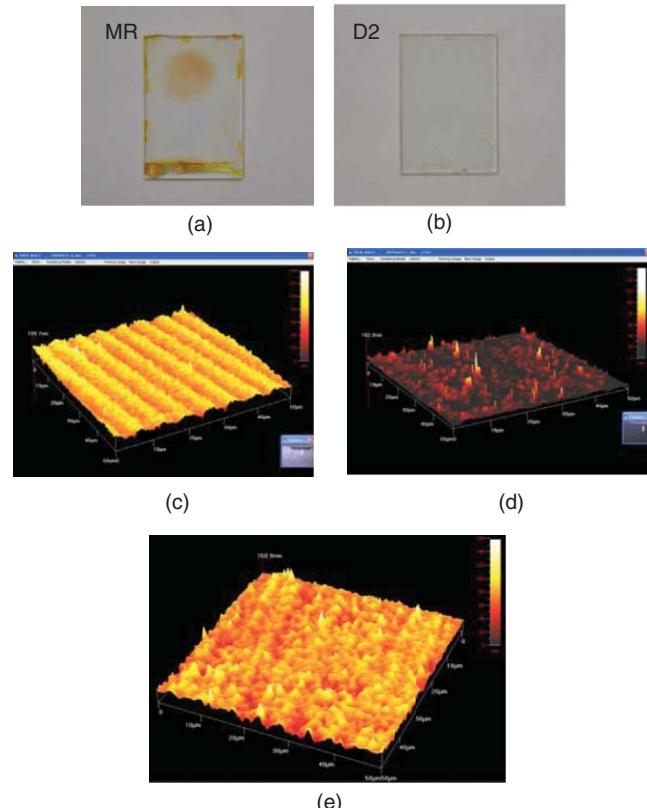


**Fig. 1** (a) Sample structure of a dye-doped liquid crystal film. (b) Schematic diagram of the experimental setup. SF; spatial filter, L; lens, HWP; half-wave plate, PBS; polarized beam splitters, M; mirrors.



**Fig. 2** (a) Evolution of +1 order diffraction efficiency during a write-read-erase process for a MR-doped liquid crystal film. (b) Evolution of +1 order diffraction efficiency during a write-read-write-erase process for a D2-doped liquid crystal film.

substrate within the recording area. However, the D2 dyes were not adsorbed on the glass substrate, and therefore the substrate looks clear within the recording area. Figures 3(a) and 3(b) show the pictures of different conditions of dye adsorption on these two samples. We further observed the surface relief structures of the adsorbed dyes from an atomic force microscope (AFM). Figure 3(c) shows the surface relief structures of the adsorbed MR dyes. We found the adsorbed MR dyes were bonded on a substrate with a periodic microgroove structure. This surface relief structure led to the alignment of liquid crystal molecules and maintains the stability of grating. Contrarily, as shown in Fig. 3(d) we found there were no D2 dyes adsorbing on the substrate. Lacking the adsorption structure, the newly generated orientations or the liquid crystals became unstable, and the recorded hologram was easily destroyed by the reading beam. In addition, Fig. 3(e) shows the AFM observation result after the erasing process in an MR-doped sample. After the erasing process, although diffraction disappeared, we still found the similar period microgroove structure on the substrate. However, the dye molecules on the microgroove structure have undergone the reverse process of cis-trans photoisomerization. They were re-excited by the *p*-polarized erase wave and their reorientation became parallel to the *s*-polarization direction. The whole liquid crystal molecules would align with the new orientation owing to guest-host effect and, accordingly, the polarization grating was erased.



**Fig. 3** (a) Adsorption conditions of MR dyes on the substrate. (b) Adsorption conditions of D2 dyes on the substrate. (c) Surface relief structures of the adsorbed MR dyes observed from an atomic force microscope. (d) AFM observation on substrate in D2-doped sample. (e) AFM observation on substrate after the erasing process in the MR-doped sample.

The mechanism of nonvolatile reading with *s*-polarization light and erasure with *p*-polarization light can be associated with adsorption of dyes and reorientation of the adsorbed dyes. Before recording the polarization hologram, the dye and liquid crystal molecules are aligned with the rubbed surface such that they are all parallel to the *s*-polarized direction. During recording process, the azo-dye molecules are photoexcited from trans-state to cis-state and they orientate from the *s*-polarized direction. Then the photoexcited dye molecules are diffused and adsorbed onto cell substrates with their new orientation to generate a microgroove structure. The process will continue until reaching the saturated state, i.e., all the dye molecules which can participate in this mechanism have been reoriented and adsorbed on the substrate. Liquid crystal molecules nearby the unrubbed substrate will align with the adsorbed dyes, and those nearby the rubbed substrate will align along the *s*-polarized direction, and therefore a twisted nematic grating structure is generated. In the reading process, illumination with *s*-polarized light does not harm the recorded hologram because there no more dye molecules will be reoriented, and those adsorbed dyes have already been reoriented in a stable direction for *s*-polarized light. During the erasure process, the dye molecules on the microgroove structure were excited by the *p*-polarized erase wave and their reorientation became orthogonal to the *p*-polarization direction. Reorientation of the adsorbed dyes leads to the orientation of the whole liquid crystal molecules going back to the initial state, i.e., all molecules are parallel to the *s*-polarized direction and, therefore, the twisted nematic grating structure disappeared.

In conclusion, we have presented the nonvolatile reading of erasable polarization holograms in MR-dye-doped liquid crystal films using the same writing wavelength. We find an *s*-polarized wave during the recording process can be used for nonvolatile readout and a *p*-polarized wave can be used to erase the holographic gratings effectively. Nonvolatile reading in dye-doped liquid crystal films is attributed to the dye adsorption effect, and destructive readout appears when the doped dyes do not adsorb on the substrate.

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