Efficient local fixing of photorefractive polymer hologram using a laser beam

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We propose and demonstrate a novel technique for efficient local fixing of photorefractive polymer hologram using a laser beam. In the technique, a CO₂ laser beam is used to heat the sample and a local hologram can be fixed easier than previous methods. By using thinner glass substrates for the photorefractive device, the hologram can be fixed efficiently and at much faster speed. The fixation efficiency can be greater than 80% and the hologram can be fixed in the order of a second. This technique is critical for dynamic holographic three-dimensional display and holographic data storage. © 2010 American Institute of Physics. [doi:10.1063/1.3361655]

Holographic stereography is one of the highest quality three-dimensional (3D) display methods and the most practical technique for displaying computer-generated synthetic images.^{1–3} A rewritable holographic stereogram system using photorefractive (PR) polymers allows dynamic recording and display and ease of scalability. In a rewritable holographic stereogram, the elementary holograms are written by raster scanning of the recording material or the optical head. It takes certain time to refresh the whole hologram array. During every period, each written hologram must be persistent, i.e., the hologram needs to hold for certain time with sufficient efficiency. Therefore, writing, reading, and erasing must be controllable. However, the conventional PR holograms decay rapidly and the decay time is in the same magnitude as the rising time, which is typically in the order of seconds or milliseconds, depending on the PR materials. To fix the holograms efficiently is very critical for this application and other applications of holographic data storage and represents a breakthrough progress.

Thermal fixing of refractive index gratings is one efficient way to improve the storage time of holograms in a PR material, in which a volume hologram is written at an elevated temperature and then stored at the ambient temperature for a longer time. Thermal fixing using an oven has been demonstrated in several inorganic crystals such as LiNbO₃, BaTiO₃, and KNbO₃, etc. The mechanism is based on ionic charge displacement in a crystal's lattice. A novel technique for local thermal fixing using a CO₂ laser has been demonstrated^{4,5} in LiNbO₃. With this technique, any hologram recorded in a local area can be selectively fixed and the fixation is due to generation of an ion grating. However, it took more than 15 min for the diffraction efficiency to reach the steady state and hence it is not fast enough for real time processing. In PR polymer, thermal fixing through conventional heating (e.g., with a thermoelectric cooler) has been demonstrated,⁶ but the diffraction efficiency was only about 2%. The mechanism of thermal fixing of gratings in PR polymers is different, and it is based on quasifreezing of the orientation of anisotropic chromophore molecules in a polymer matrix. The fixed gratings can be easily erased by el-

evating temperature inside the sample to the glass transition temperature (T_g) of the composite. However, for a holographic stereogram, it is impractical to use conventional heating method to selectively fix a local hologram. In addition, the fixation speed is slow. Here we propose to accomplish thermal fixing of gratings in a polymeric PR material using a laser. In the new technique, a CO₂ laser beam is used to heat the sample and a local hologram can be fixed easily. The fixation efficiency can be larger than 80%. Within the first 5 min, the diffraction efficiency can be higher than 72%. The hologram can be persistent for a relatively long time (in the order of several hours, here the data for 20 min are shown). By using thinner glass substrates, the hologram can be fixed efficiently and at much faster speed.

Our goal is to achieve real-time local thermal fixing of each individual elementary hologram recorded in a restricted region of the polymer. This requires heating the local area within which the temperature is above the T_g value, and while outside this area the temperature is below T_g . The speed for heating with a laser and that for cooling should be fast. We conducted both theoretical and experimental studies to determine the optimal condition for best diffraction efficiency. Figure 1 shows the basic geometry for recording and reading the hologram in PR polymer based on four-wave mixing and detailed information about the geometry can be found in Ref. 7. The hologram is written at 532 nm. The reference beam and the object beam interfere inside the PR

FIG. 1. (Color online) Basic geometry for recording and reading the hologram in PR polymer with a CO₂ laser beam for thermal fixing of the hologram.

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FIG. 2. (Color online) PR polymer composites with a low T_g . (a) Chemical structures of the composites. (b) Persistency of the material.

polymer, which is sandwiched between two indium-tin-oxide (ITO)-coated transparent substrates. An external electrical field is applied to the ITO electrodes. The grating is written based on the Pockels electro-optic effect and the oriented gas model. A reading beam counter propagating with the reference beam reads the hologram and reconstructs the recorded information. In our experiment, we used a laser beam at 633 nm to read the hologram and the direction of the reading beam is slightly different from the reference beam for Bragg matching.

For the recording material, a copolymer material TPD/Cbz/DCST (5/5/1) is used as the transport matrix for better stability. F-DCST serves as both the sensitizer and the non-linear optical chromophore. For recording at low T_g value (room temperature), a plasticizer ECz is used. The chemical structures of the composites for the low- T_g sample are shown in Fig. 2 and the transient response and decay of the hologram is presented. It indicates that the composites has good sensitivity at 532 nm and is promising for combining thermal fixing for longer persistence. In order to incorporate laser thermal fixing, the material needs to have a higher T_g value. In this case, there is no plasticizer, and the composition of the high- T_g material is TPD/Cbz/DCST (5/5/1) (62.5 wt %) and F-DCST (37.5 wt %) with a T_g value about 316 K (43 °C).

For implementation of the laser thermal fixing, the laser beam is normally incident on the sample (Fig. 1). No device is available to directly measure the temperature in the thin film layer. In order to achieve the desired temperature in the local area, controlling the power and exposure time of the laser beam is extremely important. Therefore, simulation must be performed before the experiment to determine the heating time based on the power of the CO₂ laser and the thermal properties of the substrates.

The time- and space-dependent temperature fields in laser local heating can be theoretically predicted by solving the following heat transfer equations:⁸

$$\frac{\partial}{\partial t} \theta(r, z, t) - \frac{K}{C} \nabla^2 \theta(x, y, z, t) = \frac{1}{C} g(r, z, t),$$

$$\theta(r, z, t = 0) = 0, \quad (1)$$

where the temperature rise above the ambient temperature is denoted by $\theta(x, y, z, t)$, C , and K are the specific heat and the heat conductivity of the substrate, respectively, and $g(r, z, t)$ is the power delivered to the unit volume by the laser. The equation can be solved with particular boundary conditions.

FIG. 3. (Color) Example of the spatial temperature distribution inside the PR device with 1 mm-thick substrates.

Since the layer of the PR polymer is very thin (100 μ m or less), the temperature of this layer is approximated by that of the inner surface of the front substrate. Assume the laser is a Gaussian beam with a diameter of 5 mm (for practical application of holographic display, the beam diameter should be smaller). Assume the thickness of both substrates (BK7) is 1 mm. Figure 3 shows the spatial temperature distribution at 5 s with an incident CO₂ laser power of 1.3 W. The temperature in the center (the location of the polymer) is about 323 K (50 °C), which is in the temperature range required for fixing the hologram in our PR polymer. In experiment, in order to optimize the diffraction efficiency, the standard geometry is chosen⁷ and an electronic shutter is used to control the exposure time of the CO₂ laser. Figure 4 shows the experimental demonstration of the efficient local fixing of the hologram in the high- T_g PR polymer described above. In the beginning, the hologram was recorded for about 1.5 min at room temperature with the applied voltage and the diffracted signal is low. Then the sample was heated by the CO₂ laser

FIG. 4. Experimental demonstration of the efficient local fixing of the hologram in a PR polymer.

FIG. 5. Experimental demonstration of the efficient local fixing of the hologram in a PR polymer.

beam for about 7 s and naturally cooled down for 45 s. Then the two writing beams are blocked with the voltage on. As seen from the figure, the hologram can be persistent for a relatively long time. There is no significant drop of the diffracted signal. Please note that if the voltage is turned off, the diffraction efficiency is much lower. In this case, when the voltage is turned on again, the diffracted signal will be back.

The thickness of the glass substrates can greatly affect the heating condition due to the time needed for heat transfer from the external surface to the inner surface of the substrate. For example, if the thickness of the substrates is reduced from 1 to 0.5 mm, the required exposure time of the laser is significantly reduced. Simulation shows that by heating the sample with a 2 W CO₂ laser beam for about 1.5 s, the desired temperature at the PR polymer can be achieved. Figure 5 shows the experimental result of the most efficient local fixing of the hologram with the 0.5-mm-thick glass substrates. Again in the beginning, the hologram was recorded for about 1.5 min at room temperature and the efficiency is low. Then the sample was heated by the CO₂ laser beam for about 2.5 s and naturally cooled down for 30 s.

After fixing, the peak diffraction is about 93% (The internal diffraction efficiency is calculated as the ratio of the power of the diffracted beam and the total power of the diffracted beam and the transmitted beam). When the two writing beams are blocked, the grating can be persistent for a relatively long time. Within the first 5 min, the diffraction efficiency is higher than 72%. This indicates that the fixation efficiency (the ratio of the diffraction efficiency after fixation to that before fixation) is more than 80%. To our knowledge, this is the best result up to now. The grating lifetime is more than 2 h, which is suitable for practical applications and is a significant step in this field.

We have proposed and demonstrated a novel technique for efficient local fixing of PR polymer hologram using a laser beam. In the new technique, a CO₂ laser beam is used to heat the sample and a local hologram can be fixed easily. Both simulation and experiment have been performed. Fixing efficiency of 80% has been demonstrated. By using glass and sapphire with particular thickness as the substrates for the PR device, the hologram can be fixed even more efficiently and at much faster speed. This technique is critical for dynamic holographic 3D display and holographic data storage. Other techniques may also be studied for the possibility of further improving the fixing speed.

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