Photorefractive polymer device with video-rate response time operating at low voltages

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The high-voltage bias required for video-rate compatible, efficient operation of a photorefractive polymer composite is reduced from 6–8 to 1.3 kV. At this low voltage, the device can hold erasable Bragg holograms with 80% efficiency in addition to having a video-rate response time. The transition of the hologram's state from thick to thin is analyzed in detail. © 2006 Optical Society of America

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Photorefractive (PR) polymers are alternatives to their inorganic or semiconductor counterparts owing to their low cost, ease and flexibility of fabrication, large size, and superior performance.[1] The photorefractive figure of merit in organic composites can be an order of magnitude more than those of inorganic materials.[1,2] In the past decade and half, there has been considerable effort to improve both steady-state and dynamic performance of the materials[1–8] to bring them close to their potential applications, such as real-time optical processing.

Limitations of these organic materials for direct practical applications are the necessity for use of high bias voltages (3–10 kV for a typical 100 μm thick sample) and their slow response time. The high voltage is necessary for orienting nonlinear molecules, separating the charges, and facilitating their transport in the matrix because of the low conductivity of organic materials. Several materials such as amorphous organic glasses and polymer dispersed liquid crystals have recently been employed to reduce the required voltages.[5] For amorphous glass the operating voltage has been reduced but the response time is much longer, of the order of seconds.[2] The drawbacks of polymer-dispersed liquid crystals are a high degree of scattering and slow response times.[5] Here we present both a material and a device approach to reducing the operation voltage of a photorefractive polymer to the 1 kV level without sacrificing high diffraction efficiency and fast response time.

In commonly used guest–host photorefractive polymer systems, one can enhance the effective index modulation by doping the matrix with a larger amount of polar, nonlinear optical molecules called chromophores and reducing the glass-transition temperature by adding more of a suitable plasticizer agent. However, loading of large amounts of polar molecules can cause potential disadvantages such as phase separation, crystallization, and reduced mobility. The choromophore mixture approach can provide large birefringence without resulting in phase separation. Increasing the plasticizer content, however, will increase the inert volume, therefore worsening the dynamic properties.

To complement the work on the materials side, one can investigate a thin PR device at or near minimal Bragg requirement conditions. As stated in Kogelnik's groundbreaking paper,[9] the Q factor can be used to draw the boundary line between Bragg (one diffracted order at Bragg incidence) and Raman–Nath (several diffracted orders) regimes:

$$Q = 2\pi\lambda d/n\Lambda^2, \tag{1}$$

where λ is the vacuum wavelength, d is the thickness of the film, n is the mean refractive index, and Λ is the grating spacing. Bragg diffraction requires that $Q \gg 1$. One can adjust the thickness and grating spacing values but still keep the Q value above 1 to take advantage of high Bragg diffraction efficiency.

The diffraction efficiency is a function of thickness and of bias voltage as well. The simplified form of the diffraction efficiency in a transmission geometry can be written as[9]

$$\eta \propto \sin^2\left[\frac{\pi\Delta n(E)d}{\lambda(\cos\alpha_1 \cos\alpha_2)^{1/2}}\right], \tag{2}$$

$$\Delta n \propto EE_{sc} \propto (E)^P = (V/d)^P, \tag{3}$$

where d is the thickness of the film, λ is the wavelength and α_1 and α_2 are the writing beam angles in the polymer, E_{sc} is the space-charge field, and V is the bias voltage. The index modulation (Δn) has a power-law dependence on the field, where $P \sim 1.9–2.0$. Additionally, Δn depends on the grating spacing through a sublinear relationship.

To get the same diffraction efficiency from both 105 and 20 μm devices, one should apply voltages with a ratio of $V_{105\,\mu m}/V_{20\,\mu m} = (d_{105\,\mu m}/d_{20\,\mu m})^{(p-1)/p} \approx 2.3$. This reduction in thickness will bring the operating voltage from a typical 3 kV level to $\sim$1.3 kV (or 65 V/μm). At the same time, as the operating field is larger than that of a 105 μm thick sample (30 V/μm), the device will have faster dynamics.

We have applied the principle described above to reduce the effective voltage requirement for high-performance operation of a polymer PR device. The

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composite is based on a hole transport polymer (PATPD), which has a polyacrylate backbone and a well-known hole-transporting tetraphenyldiaminobiphenyl-type pendant group attached through an alkoxy linker.[10] The hole-transporter PATPD was doped with a mixture of two nonlinear optical chromophores, 4-homopiperidinobenzylidenemalononitrile (7DCST) and (*N*,*N*-di-*n*-butylaniline-4-yl) *l*-dicyanomethylidene-2 cyclohexene (DBDC), with C_{60} as a sensitizer for charge generation at the operating wavelength of 633 nm, and the plasticizer, *N*-ethyl carbazole (ECZ). The mixture approach is used because of its better phase stability. Composites prepared included PATPD:DBDC/7:DCST:ECZ/C_{60} (39.3:40:10:10:0.7 wt. %). We prepared samples by laminating 20 and 105 μm thick layers between glass slides with indium tin oxide electrodes. The absorption coefficient of the film was 70 cm^{-1} at 633 nm.

To characterize the PR properties of the samples we performed degenerate four-wave mixing experiments in the standard tilted-sample geometry. Two interfering *s*-polarized beams (633 nm) of equal fluence (0.5 W/cm^2 each) were used to write the grating, and a weak counterpropagating *p*-polarized beam probed the efficiency of the grating. The writing beams were incident onto the sample at an interbeam angle of 20° in air, and the sample's surface was tilted 60° relative to the writing beam bisector, resulting in a grating period of 3.1 μm. Under these conditions, Kogelnik's Q value, based on Eq. (1), is 4.7, which is still in the Bragg (thick) hologram regime. Figure 1 shows the percentage of the diffracted probe beam that is a function of the applied external field under steady-state conditions for 20 and 105 μm thick devices. The external efficiency (when reflection and absorption losses are included) of the device was 58%. Using the present approach, we were able to reduce the voltage required for overmodulation (peak of diffraction efficiency) of diffraction efficiency by a factor of 4 in a single chromophore containing a composite reported earlier.[10]

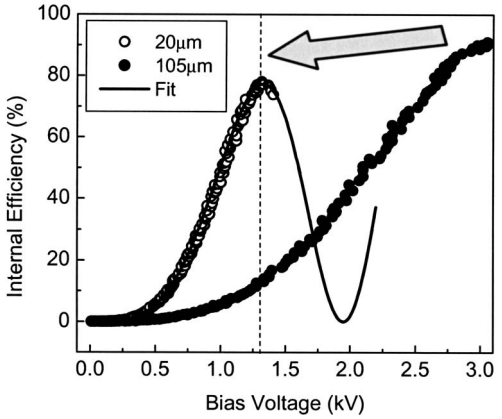

Fig. 1. Steady-state diffraction efficiency for both 20 and 105 μm thick devices as the bias voltage is increased. When the device's thickness is reduced by five times, the operating voltage has dropped by a factor of 2.3. The peak internal diffraction efficiency of the 20 μm device was 80%. External efficiency was ~=58%, which is higher than that of a similar thick device because of the low absorptance of the thin device. The index modulation was 0.012 at 1.3 kV.

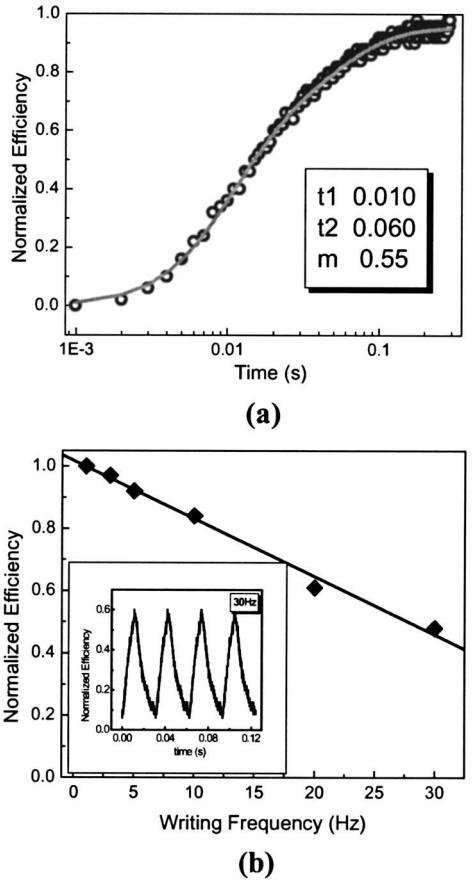

Fig. 2. (a) Typical growth of diffraction efficiency when one of the writing beams is switched on. The line is a biexponential fit. The total writing beam irradiance was 1 W/cm^2, and a bias voltage of 1.4 kV was applied through the device. (b) Net diffraction efficiency recorded as one writing beam is modulated at 1–30 Hz (full cycle write and erase) frequency. Inset, response of the photorefractive device at a 30 Hz frame rate.

The dynamics of the index modulation buildup can be extracted from a biexponential function fit that is correlated to the growth of the space-charge field:

$$\Delta n \propto A[1 - m\exp(-t/t_1) - (1-m)\exp(-t/t_2)]. \quad (4)$$

Here A is a constant, m is a weighing factor and t_1 and t_2 are time constants. Figure 2(a) shows the evolution of the diffraction efficiency after the second writing beam is switched on at time $t=0$. The curve represents the fit to the data. Remarkably fast dynamics with a characteristic first time constant of 10 ms (at 1.4 kV bias voltage) are observed, indicating possible application of these composites in systems that operate at video rates. To test the performance of current devices for video-rate compatibility, we modulate one of the writing beams at 1–30 Hz full-cycle (on and off) frequency [Fig. 2(b)]. The efficiency has dropped to only half of its cw value at 30 Hz. Therefore this device can operate simply with high efficiency and video-rate refresh rates without requiring impractical bias voltages.

The transition from thick (Bragg) to thin (Raman–Nath) holograms is not sharp. Higher diffraction orders are strong when $Q \ll 1$ (Raman–Nath state). In this state, the maximal diffraction efficiency possible in the first order is 34%.[11] When one varies the grating spacing to scan the Q value through the transition region, the peak efficiency starts to drop gradually (Fig. 3). There were no visible higher orders below 3.5 μm grating spacing. Above that, higher orders started to become visible. Above 5.2 μm, the higher orders became stronger and contributed to a more than 50% reduction in the intensity of the first-order diffracted beam. The overmodulation field is simply shifted owing to the increased grating spacing, which can be explained through the Kuktharev model.[12] The index modulation (amplitude of the sinusoidal modulation of the index) was 0.012–0.018, increasing with the grating spacing. When the index modulation is large, the regime of the grating can be reliably specified if the parameter ρ [(Ref. 13), first defined by Nath] is utilized. It is defined as $\lambda^2/\Lambda^2 n \Delta n_1$, where Δn_1 is the index modulation. Based on this parameter, the Raman–Nath regime necessitates that $\rho \leqslant 1$. At a grating spacing of 3.1 μm, $\rho = 2$, and at 5.2 μm, $\rho = 0.5$, in agreement with our experimental results.

Note that the photorefractive nature of the hologram was confirmed through two-beam coupling measurements.[14] For equal-intensity writing beams the net gain, Γ, was more than 400 cm^{-1} at 1.3 kV.

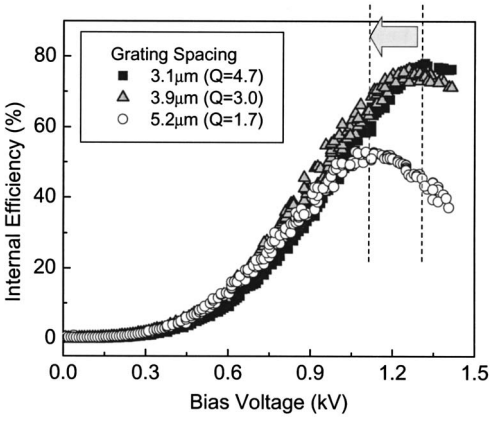

Fig. 3. Drop in peak efficiency as the state of the hologram is changed from thick (Bragg) to thin (Raman–Nath). The grating spacing is increased to vary Kogelnik's Q value. The shift in peak efficiency reveals growth in index modulation with increased grating spacing.

In summary, we have reduced the high bias voltage required for the operation of guest–host PR polymers to lower levels. We accomplished this reduction by doping the PR composite with two nonlinear optical chromophores and successfully reducing the device's thickness to the minimal Bragg condition. The current device can be operated at low voltages, maintaining the video-rate response time. Even some useful diffraction efficiency, of 20%, can be achieved at a practical voltage of 750 V. The photorefractive behavior of the device is characterized in the transition region between thin and thick holograms.

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