# Dark diffusional enhancement of holographic multiplexed gratings in phenanthrenequinone doped poly(methyl methacrylate) photopolymer*

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In this paper, we experimentally investigate the dark diffusional enhancement of the optimized multiplexed grating in the phenanthrenequinone doped poly (methyl methacrylate) (PQ-PMMA) photopolymer. The possibility of improving the holographic characteristics of the material through the dark enhancement is demonstrated. The optimal preillumination exposure and the optimal time interval between exposures are extracted to obtain the optimized diffraction efficiency, and their values are $3.4\times10^3$ mJ/cm$^2$ and 2 min, respectively. The dark enhancement of the multiplexed grating is presented as an effective method to improve the response region and the dynamic range and to prevent saturation of the material. The dependence of the phenanthrenequinone concentration on the increment of the refractive index modulation is quantitatively studied, which provides a significant basis for improving the homogeneity in the multiplexed gratings using a quantitative strategy. Finally, a simple experimental procedure using the dark enhancement is introduced to improve the homogeneity of the diffraction efficiency and to avoid the complex schedule exposure.

**Keywords:** photopolymer, holographic multiplexing, dark diffusional enhancement

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## 1. Introduction

Recently, the interest in photopolymers for holographic data storage application has increased.[1−5] Among those polymers, the phenanthrenequinone (PQ) doped poly(methyl methacrylate) (PMMA) photopolymer has attracted more attention due to its neglectable shrinkage, high thickness and good optical stability.[6−13] This material is widely used in optical data storage and relevant information processes.[14−17] The main disadvantage of this material is its low response. The dark diffusional enhancement, as one of the alternative methods, can relieve this primary limitation. The dark enhancement of grating after a short exposure is observed in the photopolymer.[12,18,19] It is attributed to the diffusion of the PQ molecules from the dark region to the bright region. Compared with the consecutive exposure, the short exposure can prevent the amplification of the holographic scattering and reduce the photosensitizer consumption in the exposure spot. The dark enhancement in a single grating as an optimized strategy for the holographic memory has been analysed in the photopolymer.[20] It provides a simple and efficient method to obtain a high and steady diffraction efficiency in the single grating.

In order to obtain a high storage density and a large storage capacity, several multiplexed gratings are generally recorded in the same spot of a sample. Furthermore, the multiplexing gratings in the photopolymer have significant applications in fabricating the photonic crystal and the photonic micro/nanostructure.[21−26] The homogeneity of the diffraction efficiency in the multiplexing is influenced by the recording procedure. The exposure schedule technique is a typical method to obtain a homogeneous grating.[16,27] However, the complicated calculation technique and the complicated experimental procedures of the schedule exposures restrict the appli-

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cability of the method to the photopolymers.[28] The dark diffusional enhancement of the multiplexed gratings is an efficient strategy to improve the holographic multiple characteristics of the PQ-PMMA photopolymer and can simplify the recording schedule. However, the diffusional enhancement of the multiplexed gratings attracts less attention. The analytical and the experimental investigations on the dark enhancement of the multiplexing are not sufficient. In this paper, a detailed experimental study on the dark enhancement of the holographic multiplexed grating in the PQ-PMMA photopolymer is presented. The holographic characteristics are improved and the corresponding physical mechanism is analysed.

## 2. Material and experimental setup

The PQ-PMMA photopolymer was composed of a poly (methyl methacrylate) (PMMA) host matrix and a phenanthrenequinone (PQ) photosensitizer. The PMMA host matrix was formed through the polymerization reaction of MMA monomers using the thermal initiator azo-di-iso-butyro-nitrile (AIBN). The thermal initiator AIBN and the PQ molecules were dissolved in a solvent of MMA with a weight ratio of 0.5:0.7:100. The solvent was then laid for about 3 h to eliminate the nitrogen produced by the thermal decomposition process of the AIBN. The mixture was poured into a glass mold and solidified at 60 °C for 120 h. After the thermal polymerization, the sample with a PQ concentration of 0.1 M and a thickness of 2 mm was prepared.

The two-beam coupling setup with a 60° geometry was used to record unslanted transmission gratings, as shown in Fig. 1. The sample was placed on a computer-controlled motorized stage to allow the Bragg angle tuning of the probe beam. The rotating axis of the rotation stage was perpendicular to the recording beam plane. The recording beams intersected inside the sample with respect to the sample normal. The recording and the reconstructing beams operated at wavelengths of 532 nm and 633 nm, respectively. The angle-multiplexing method by rotating the sample was used to record a series of holographic transmittance gratings in the photopolymer. The angle difference between the multiplexed gratings was larger than 0.5° to avoid the cross talk.

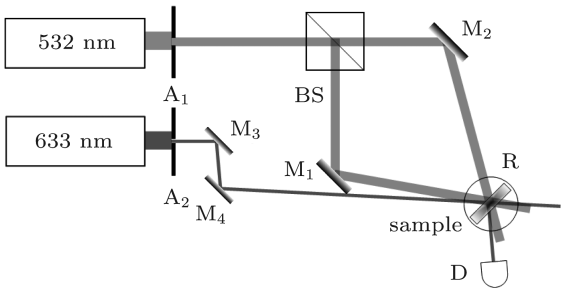



**Fig. 1.** Experimental setup for recording holographic multiplexed gratings. The $A_1$ and $A_2$ are apertures, S stands for the beam splitter, $M_1$, $M_2$, $M_3$ and $M_4$ are mirrors, R is a rotation stage, and D is a detector.

## 3. Photochemical mechanism

To clarify the dark diffusion process in the multiplexing after an exposure, the involved physical mechanism is analysed first. The complicated photochemical processes can be described as follows:[9,11,20]

$$\mathrm{PQ} \rightarrow {}^3\mathrm{PQ}, \tag{1}$$

$${}^3\mathrm{PQ} + \mathrm{HR} \rightarrow \mathrm{HPQ}^{\bullet} + \mathrm{R}^{\bullet}, \tag{2}$$

$$\mathrm{R}^{\bullet}_{n-1} + \mathrm{R} \rightarrow \mathrm{R}^{\bullet}_{n}, \tag{3}$$

$$\mathrm{HPQ}^{\bullet} + \mathrm{R}^{\bullet}_{n} \rightarrow \mathrm{HPQ}{-}n\mathrm{R}, \tag{4}$$

where R represents the MMA molecules ($n \geq 1$), $\mathrm{HPQ}^{\bullet}$ and $\mathrm{R}^{\bullet}$ denotes free radicals of PQ and MMA molecules, respectively, and ${}^3$PQ refers to the phenanthrenequinone in the triplet-excited state. The PQ consumption is related to the initiation reaction by

$$-\frac{\partial U(t)}{\partial t} = R_{\mathrm{i}}, \tag{5}$$

where $U$ denotes PQ's concentration, $R_{\mathrm{i}} = fk_{\mathrm{d}}U$ represents the rate of the initiation reaction, $f$ denotes the fraction of the radicals reacted due to the cage effect, $k_{\mathrm{d}}$ is the rate constant of the initiation reaction. Therefore, the temporal evolution of the primary PQ molecules and the corresponding photoproduct can be given respectively by[20]

$$U(t) = U_0 \exp(-fk_{\mathrm{d}}t) = U_0 \exp(-E/E_\tau), \tag{6}$$

$$P(t) = U_0 - U(t) = U_0 [1 - \exp(-E/E_\tau)], \tag{7}$$

where $U_0$ represents the initial concentration of the PQ molecule, $P(t)$ denotes the concentration of the photoproduct, $E$ is the exposure flux, $E_\tau = I_0/fk_{\mathrm{d}}$ is the exposure constant related to the polymerization rate and $I_0$ is the incident intensity. The diffusion of the PQ molecules under a short exposure is neglectable, because the diffusion process is separated

from the polymerization process due to the low diffusion coefficient.[29,30] Under illumination, the refractive index modulation is proportional to the density of the primary components at any point and it grows following the asymptotic curve

$$\Delta n\,(t) = C_1 - C_2 \exp\left(-E/E_\tau\right) \tag{8}$$

with $C_1$ and $C_2$ being two constants. After a series of gratings are recorded in the sample, the refractive index modulation resulted from the recording and the dark diffusion processes evolves in the following way for $t > t_{\rm e}$ ($t_{\rm e}$ is the exposure time),

$$\begin{aligned}\Delta n\,(t - t_{\rm e}) &= \Delta n\,(t_{\rm e}) + \Delta n_{\rm D}\,(t - t_{\rm e}) \\ &= \Delta n\,(t_{\rm e}) + C_{\rm PQ} U_{\rm D}\,(t - t_{\rm e})\,, \end{aligned} \tag{9}$$

where $C_{\rm PQ}$ is the coefficient related to the polymeric component and subscript D indicates the diffusion process.

# 4. Results and discussion

Several important aspects, such as the preillumintaion and the time interval between consecutive exposures, are crucial to characterizing the dark enhancement of the multiplexed gratings. To optimize the performance of the PQ-PMMA photopolymer, the influence of the parameters on the diffraction efficiency of the multiplexed gratings is analysed in the experiments. We focus our attention on the characteristics of the multiplexed grating for the dark enhancement.

## 4.1. Preillumination

At the beginning of the recording process, the diffraction efficiency of the grating is very low, because oxygen or other inhibitors are present in the material.[30,31] The low diffraction efficiency is a disadvantage in the multiplexing, so the inhibition process should be shortened. The preillumination is one of the most effective methods to prevent the inhibition process and enhance the sensitivity.[16,32,33] Before the grating formation, the preillumination can provide sufficient energy to reduce the concentration of the inhibitors, therefore the corresponding photosensitivity can be enhanced.

In order to investigate the influence of the preillumination on the dark enhancement of the grating, the sample was illuminated by a collimated laser at wavelength of 532 nm before recording the gratings. The various exposure energies used for the preillumination were $2.3\times10^3$ mJ/cm$^2$, $3.4\times10^3$ mJ/cm$^2$, $5.7\times10^3$ mJ/cm$^2$ and $7.9\times10^3$ mJ/cm$^2$ respectively. When the preillumination was turned off, a single unslanted transmittance grating was immediately recorded by using an identical exposure of $1.1\times10^4$ mJ/cm$^2$. Then the initial and the saturated diffraction efficiencies after the dark enhancement were measured for optimizing. The experimental results are shown in Table 1, from which it can be seen that the maximal initial and saturation diffraction efficiencies were obtained at the exposure energy of $3.4\times10^3$ mJ/cm$^2$. The obvious increase in the diffraction efficiency is ascribed to the preillumination. The corresponding preillumination exposure was selected for the following multiplexing experiments.

**Table 1.** Diffraction efficiencies with various preillluminations.

| Preillumination /$10^3$ mJ·cm$^{-2}$ | Initial diffraction efficiency/% | After 6000 s diffraction efficiency /% |
|---|---|---|
| 0 | 5.2 | 30.8 |
| 2.3 | 5.9 | 33.2 |
| 3.4 | 7.6 | 36.3 |
| 5.7 | 7.2 | 34.0 |
| 5.7 | 6.8 | 30.0 |

## 4.2. Time interval between consecutive exposures

Another important parameter in the multiplexing process is the dark time interval between consecutive exposures. An appropriated time interval can accelerate the dark reaction and bring an optimal concentration of active radicals for the formation of the consecutive gratings. In order to optimize the dark time interval, we multiplexed several gratings with an identical exposure and observed the dependence of the diffraction efficiency on the time interval. The same experimental procedure was repeated for several different time intervals, namely, 10 s, 1 min, 2 min, and 5 min, respectively.

Figure 2 shows the diffraction efficiencies of the multiplexed gratings with various time intervals. Several multiplexed gratings were recorded with an identical exposure of $1.1\times10^3$ mJ/cm$^2$ by rotating the sample. After recording the gratings in the sample, the diffraction efficiency of the last grating was close to zero. It means that the sample reached saturation after the multiplexing recording, and a further recording could not be achieved in the same spot. Different time

intervals brought out obvious differences in the multiplexing. At time intervals around 2 min and 5 min, a high diffraction efficiency was obtained in the experiment. These results could be explained by the dark reaction and the diffusion mechanism that occurred at the period of the time intervals. The appropriated time interval could accelerate the dark reaction of the phenanthrenequinone radicals and the diffusion of the phenanthrenequinone molecules from the dark region to the bright region.

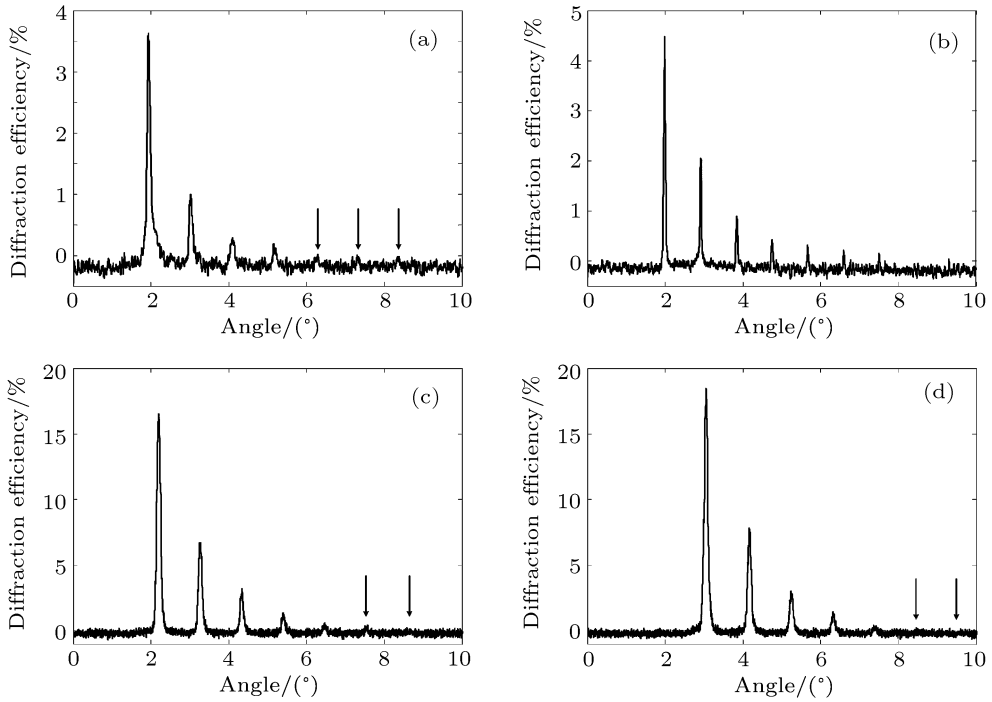



**Fig. 2.** Diffraction efficiencies of multiplexed gratings with time intervals of (a) 10 s, (b) 1 min, (c) 2 min, and (d) 5 min.

In our sample, the primary photochemical reaction is the photoattachment between the PQ and the polymer matrix. Therefore, the dark reaction is determined by the lifetime of the PQ radical. The radical lifetime is defined to be the radical concentration divide by the photoinitiated reaction rate, i.e.,

$$\tau_{\text{lifetime}} = \frac{U^{\bullet}}{R_{\text{i}}} = \frac{E_{\tau}}{I_0} \cdot \frac{U^{\bullet}}{U} = \tau \frac{U^{\bullet}}{U}, \tag{10}$$

where $\tau$ is a time constant, and $U^{\bullet}$ denotes the concentration of the PQ radicals. According to Eq. (6), the rate of the initiation reaction is $R_{\text{i}} = fk_{\text{d}}U = UI_0/E_{\tau} = U/\tau$. The $\tau$ is approximately 340 s under the consecutive exposure of intensity 50 mW/cm$^2$.[20] The value of $U^{\bullet}/U$ is approximately 50%, which comes from a theoretical estimation based on Eq. (6).[20] Therefore, the lifetime of the radical can be calculated, and it is approximately equal to 170 s. This time is close to the optimal experimental result, which demonstrates that the radical lifetime and the dark reaction are the primary factors influencing the optimization of the time interval.

## 4.3. Dark enhancement of holographic multiplexed grating

The experiment to study the dark enhancement of the multiplexed grating in the polymer was conducted by taking into account the previous analyses. After a series of gratings were recorded in the sample, the recording beams were turned off. In order to study the dark enhancement process of the multiplexed grating, the profiles of the Bragg selectivity of all gratings were scanned simultaneously by rotating the sample.

The saturation diffraction efficiencies obtained 6000 s after the exposure in the multiplexing with various time intervals are shown in Fig. 3. The diffraction efficiencies are amplified, which is attributed to the dark diffusion of the PQ molecules from the dark region into the bright region. When the recording beams are turned off, the dark reaction of the PQ radicals is restricted by their limited lifetime, which is approximately 170 s. Compared with the time of 6000 s in the dark enhancement experiment, the lifetime of the

active radical is short, which leads to the fact that the dark reaction can be neglected after the exposure. Therefore, the PQ diffusion is dominated during the all-dark amplification of the multiplexed grating.

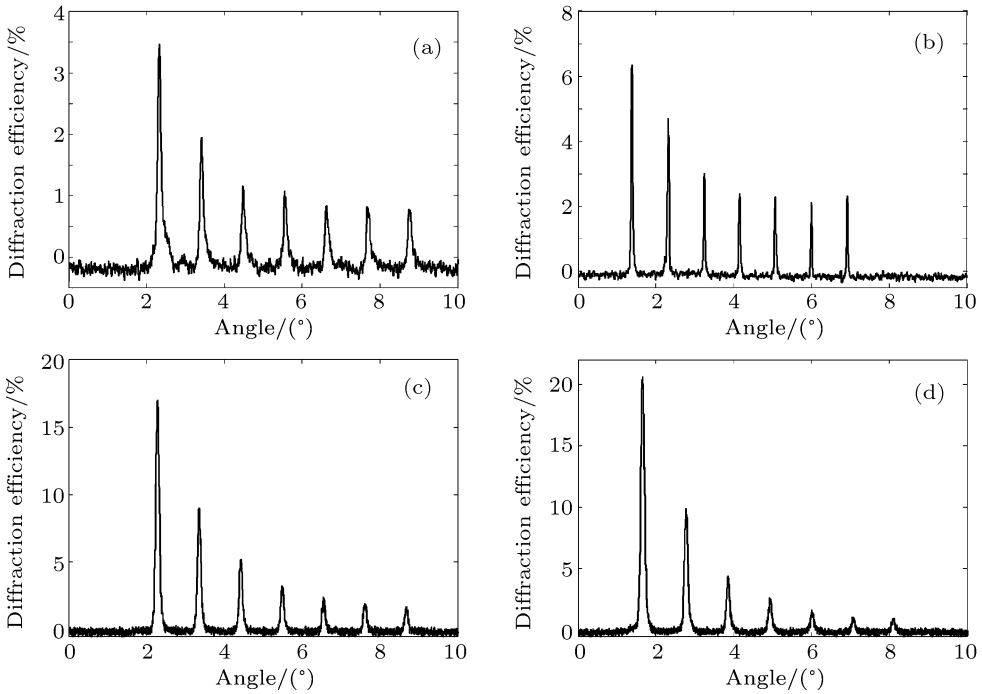



**Fig. 3.** Diffraction efficiencies of multiplexed gratings with time intervals of (a) 10 s, (b) 1 min, (c) 2 min and (d) 5 min obtained 6000 s after the exposure.

There are two important factors that contribute to the dark enhancement of the multiplexing grating in the material. One is the dark attachment of residual PQ radicals with the polymer matrix after the illumination and the other is the PQ diffusion from the dark region into the bright region. The dark chemical reaction, which contributes to the enhancement of the gratings by the formation of more photoproducts, is limited by the PQ radical lifetime. As a result, the dark reaction is responsible for the enhancement of the grating in a short period after the exposure (at the scale of 100 s). Compared with the dark reaction, the PQ diffusion is a slow process and is responsible for the long-term enhancement of the multiplexing gratings.[4]

The dynamic range of a material is a significant parameter for describing the capacity of the material in the holographic multiplexing. When each grating in the multiplexed holographic storage has a diffraction efficiency different from the others, the dynamic range $M\#$ of the material can be defined as the summation of cumulative grating strengths of the recorded gratings,[34]

$$M\# = \sum_{i=1}^{N} \sqrt{\eta_i}, \tag{11}$$

where $N$ represents the number of gratings that can be recorded in the material, $\eta_i$ is the diffrafction efficiency of $i$-th grating. The corresponding dynamic ranges at the initial moment when the beams were turned off and after 6000 s dark development when the sample had reached the steady state are shown in Table 2. It is found that the dynamic range remarkably increases after 6000 s in the dark. There is an optimal dynamic range at the 2-min time interval.

**Table 2.** Dynamic ranges with various time intervals.

| Time interval | Initial $M\#$ | 6000 s $M\#$ |
|---|---|---|
| 10 s | 0.65 | 0.99 |
| 1 min | 0.92 | 1.48 |
| 2 min | 1.21 | 1.53 |
| 5 min | 1.18 | 1.45 |

After a series of gratings were recorded in the sample, the recording beams were turned off. The Bragg selectivity profiles of all gratings were simultaneously

scanned by rotating the sample. The cumulative grating strength (i.e., dynamic range $M\#$) was calculated by selecting the peak value as the diffraction efficiency of a grating. After a given period, the same procedure was repeated for obtaining the evolution of the dynamic range. The temporal evolution of the dynamic range in the multiplexed gratings with 2 min recording time interval is shown in Fig. 4(a). The symbols refer to the experimental data and the solid line represents the fitting curve using an exponential function. Figure 4(b) shows a typical dark development curve of the refractive index modulation in the single grating after a short exposure. The corresponding exposure energy is $1.5\times10^4$ mJ/cm$^2$. The good agreement between the experimental data and the fitting curve indicates that the dark enhancements of the dynamic range and the modulation depth in the single grating are induced by the same mechanism, i.e., the PQ diffusion.

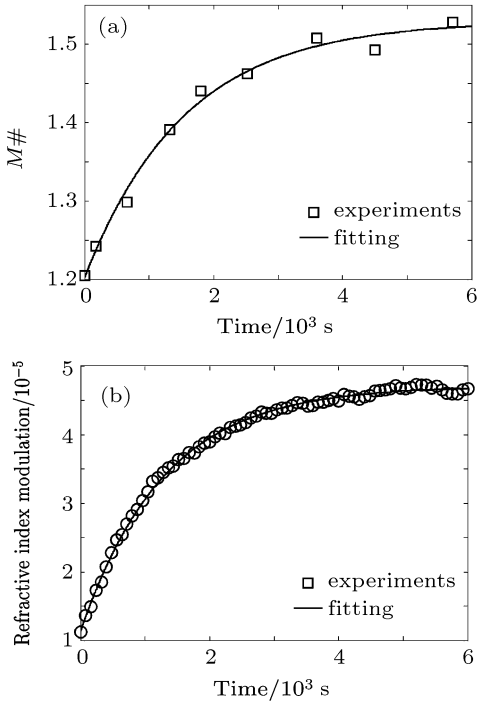



**Fig. 4.** (a) Temporal evolution of dynamic range in multiplexed grating with 2 min recording time interval after the exposure, and (b) temporal evolution of refractive index modulation in a single grating after the exposure. The beams are turned off at $t = 0$.

In order to demonstrate the amplification of the dynamic range, which is ascribed to the diffusion of the PQ molecules, the time constant related to the diffusion coefficient is calculated. At a given spatial frequency, the relationship between the time constant and the diffusion coefficient can be express as

$$\tau = \frac{1}{DK^2}, \tag{12}$$

where $D$ is the diffusion coefficient, $K = 2\pi/\Lambda$, $\Lambda = 0.5$ μm is the fringe spacing calculated by $\Lambda = \lambda/2n' \sin\theta$ based on the recording angle 60° outside the sample, $\lambda$ is the wavelength of the reroding beams and $n'$ is the refractive index of the sample. The diffusion coefficients in the multiplexed grating and the single grating are calculated by using Eq. (12) to be $2.3\times10^{-18}$ m$^2$/s and $4.3\times10^{-18}$ m$^2$/s, respectively. The value of the diffusion coefficient is similar to the reported ones in the PQ-PMMA polymer.[20] The results demonstrate that the dark enhancement of the multiplexed gratings can be attributed to the PQ molecule diffusion from the dark region into the bright region.

The FWHMs (full width at half maximums) of the angle response in the multiplexed gratings were measured for evaluating the effect of the diffusion process. The FWHM evolution of the multiplexed gratings is shown in Fig. 5, only angle responses of No. 1 and No. 6 grating orders are analyzed for simplicity. It is found that the angle response widths of different gratings are similar and the dark diffusion of the PQ molecules does not enhance the angle width. The slight difference is attributed to the experimental uncertainty. The similar angle widths are responsible for the unchanged effective thickness of the polymer. It implies that the storage density and the storage capacity of a sample do not change when the dark amplification is used to improve the diffraction efficiency of the multiplexed grating.

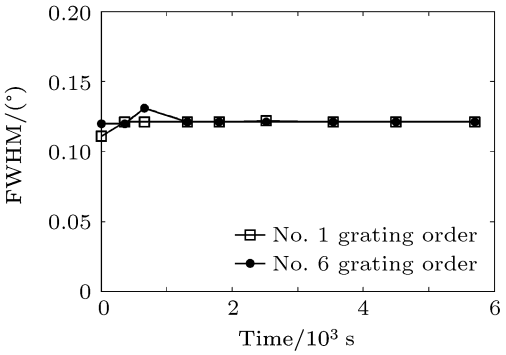



**Fig. 5.** Temporal evolutions of FWHMs in multiplexed gratings with 2 min recording time interval.

After recording several gratings, we calculate the cumulative grating strength to analyse the response region of the material. The cumulative grating strength of the holographic material is an important parameter for describing the multiplexing characteristics of the material. The cumulative grating strength

as a function of the cumulative exposure energy is calculated by summing the square roots of the diffraction efficiencies and is shown in Fig. 6. Figure 6(a) shows the cumulative strength at the initial time after the exposure and Fig. 6(b) exhibits the cumulative strength 6000 s after the exposre when the steady state has been reached. It can be seen that the cumulative grating strength reaches saturation when a series of gratings are recorded in the material, as shown in Fig. 6(a). However, the cumulative grating strength is close to the linear region after the 6000 s dark development, as shown in Fig. 6(b). It means that the response region of the material is far from saturation after the dark enhancement and more gratings could be recorded in the same spot of the sample.

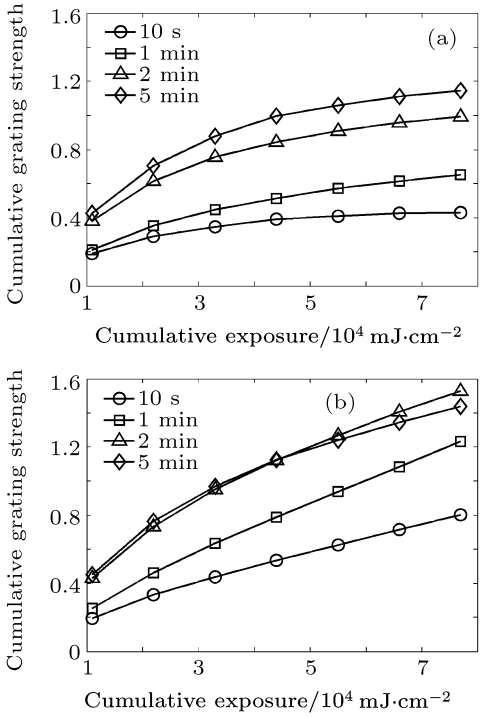



**Fig. 6.** Cumulative grating strengths versus cumulative exposure with different time intervals (a) at the initial time after the exposure and (b) 6000 s after the exposure when the steady state has been reached.

When the dark enhancement in the multiplexed grating has reached the steady state, the increment of the refractive index modulation of each grating can be approximately described by

$$\Delta n\left(t_{\mathrm{s}}-t_{\mathrm{e}}\right)-\Delta n\left(t_{\mathrm{e}}\right)\propto C_{\mathrm{PQ}}U_{\mathrm{D}}\left(t_{\mathrm{s}}-t_{\mathrm{e}}\right)$$
$$=C_{\mathrm{PQ}}U_{0}-C_{\mathrm{PQ}}U\left(t_{\mathrm{e}}\right), \tag{13}$$

where $t_{\mathrm{s}}$ is the time needed to reach the steady state. In the multiplexing, the increment represents the difference between the refractive index modulations of each recording grating at the initial moment and after the 6000 s dark development. The PQ consumption is a cumulative process by multiplexing the exposure energy. Based on Eq. (13), there is an approximately linear relationship between the increment of the refractive index modulation and the residual PQ concentration. After recording each grating, the consumption of the PQ molecules can be calculated by using Eq. (6), in which the exposure constant is $E_{\tau}=1.7\times10^{4}$ mJ/cm$^{2}$. Therefore, the increment of the refractive index modulation versus the residual PQ concentration can be obtained, which is shown in Fig. 7. The result indicates a good agreement between the experiment and the theory. The curve provides a quantitative description of the relationship between the increment of each grating and the corresponding PQ concentration (i.e., cumulative exposure energy). Consequently, by selecting an appropriate exposure energy and an appropriate procedure, the diffraction efficiency of the multiplexed gratings can be improved efficiently through the dark diffusional enhancement.

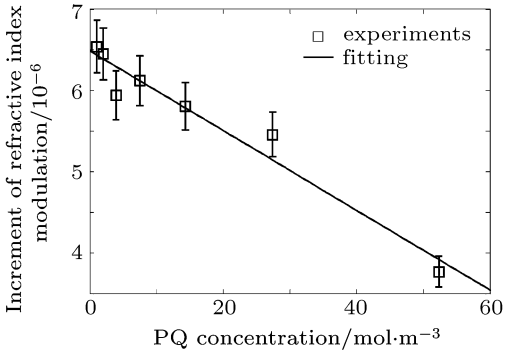



**Fig. 7.** Increment of refractive index modulation as a function of residual PQ concentration through the dark enhancement. The experiment error is 5%.

In order to obtain the homogeneous diffraction efficiency in the multiplexing and to simplify the recording procedures, the differences of the increment of the diffraction efficiency in the multiplexing are investigated. Based on Figs. 1 and 2, it is obvious that the increment of the weak grating is larger than that of the strong grating. The increment of the weak grating becomes stronger as the exposure energy increases. Consequently, the difference in the diffraction efficiency between the consecutive gratings can be efficiently reduced by the dark amplification. According to the above analyses, we recorded several gratings with an identical exposure of $1.1\times10^{3}$ mJ/cm$^{2}$, the corresponding experimental results are shown in Fig. 8. Figures 8(a) , 8(c) and 8(b), 8(d) show the two

separate experimental results, respectively. In both experiments, only the angle intervals are different, the other experimental conditions are identical, which includes the preillumination of $3.4\times10^3$ mJ/cm$^2$ and the time interval of 2 min. Figures 8 (a) and 8(b) show the diffraction efficiencies in the multiplexing at the initial time after the exposure. Figures 8 (c) and 8(d) indicate the saturation values of the diffraction efficiencies after the steady state has been reached. It can be seen that the increment of the weak grating evidently reduces the difference between the consecutive gratings. Therefore, an uniform diffraction efficiency can be obtained by using a simple identical exposure method. Furthermore, we emphasize that the gratings must be recorded in the linear region of the sample and the dark enhancement can evidently improve the homogeneity. When the sample is close to saturation after the exposure, the effect of the dark amplification will decrease. Therefore, the multiplexing experimental procedures can be optimized as follows. Firstly, a few gratings are recorded in the linear region. After the dark enhancement, the same gratings are recorded again. Then, the procedures are repeated in the same spot for recording more gratings until the sample is close to saturation. It implies that the uniform diffraction efficiency in the multiplexing can be achieved by utilizing the above procedures. In this way, the complicated calculation technique and the complicated experimental procedure of the schedule exposure can be avoided.

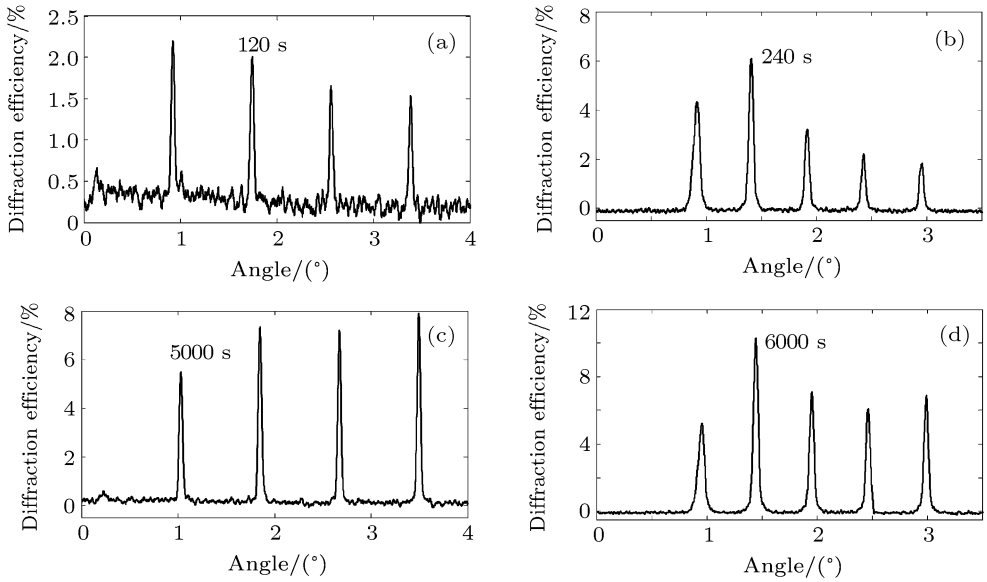



**Fig. 8.** Homogeneity improvement of diffraction efficiency in multiplexing using dark enhancement. The identical exposure is $3.4\times10^3$ mJ/cm$^2$ and the time interval is 2 min. Panels (a) and (b) show the response at the initial time after the exposure, panels (c) and (d) indicate the saturation response after the steady state has been reached.

## 5. Conclusion

The dark diffusional enhancement of the optimized holographic multiplexed grating in the PQ-PMMA photopolymer is investigated. The crucial experimental conditions including the preillumination and the time interval are optimized for improving the holographic characteristics, and the corresponding values are found to be $3.4\times10^3$ mJ/cm$^2$ and 2 min, respectively. When a series of holographic gratings are recorded at the same spot by rotating the sample, the obvious dark amplification of the multiplexed grating is observed. The experimental results indicate that the dark enhancement can improve the response region, enhance the effective storage capacity and prevent saturation of the material. The nearly the same FWHM of the angle response during the dark diffusion indicates that the storage density does not change by the enhancement. The dark reaction and the PQ diffusion are two significant factors contributing to the dark enhancement in the multiplexing. The dark reaction is responsible for the enhancement of the grating in a short period after the illumination has been turned off. The PQ diffusion is the primary reason for

the long-term dark amplification of the multiplexed grating. Furthermore, we introduce a simple experimental procedure to improve the homogeneity of the diffraction efficiency. In the linear response region, a few gratings are first recorded in the sample. After the dark enhancement, the same gratings are recorded again and then the procedures are repeated until the sample is close to saturation. The dependence of the PQ concentration on the dark increment of the multiplexing grating is obtained quantitatively by analysing the photochemical kinetics, which provides a significant basis for improving the homogeneity in the multiplexed gratings using quantitative calculations and experimental measurements.

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