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Recording of high efficiency volume Bragg gratings in a photopolymer using diffraction from very weak pre-recorded gratings

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Abstract: A two-step method of writing high diffraction efficiency holographic gratings in photopolymer using just one writing beam during the second step is discussed. The technique is based on the use of weak gratings (<1%) recorded during a first step which during the second step are illuminated with a single beam to form strong highly efficient holographic gratings. The weak grating provides a second interfering beam by diffracting a small portion of the illuminating light. Although effects such as beam pumping and two wave mixing are well known in dynamic re-writable recording media such as photorefractives, the ability to write with one beam, or ‘pump’ gratings is not usually observed in photopolymer materials. Other methods of recording with a single beam have been reported, especially for data storage, but they use compact optical systems, polarization effects or lenticular devices to produce the required second beam. In this method, the second beam is produced within the photopolymer medium. Earlier work by the authors reported increases in efficiency of weak photopolymer gratings illuminated by a single exposure beam, and more recently evidence was presented that diffraction at the weak pre-recorded grating was responsible for this effect. Here we demonstrate a strong dependence of the diffraction efficiency on grating thickness and show that the method allows the writing of high diffraction efficiency gratings in adverse conditions using a single beam. The possibility of angular multiplexing is also demonstrated. The technique may find application in writing information using pre-recorded weak holographic gratings and, potentially, in other data storage applications. In this paper it is shown that multiple new gratings can be written which are angularly separated and distinct from the pre-written grating and it is also possible to choose one grating from among a number of pre-written gratings and selectively amplify it without amplifying the others.

1 Introduction

1.1 Holographic recording

Holography has many well-known applications including display holography, security holograms, holographic optical elements, holographic data storage, holographic interferometry and sensing. In applications such as sensors, security holograms and display holograms, the hologram is often fabricated in an environment where vibration has been deliberately minimized and then either used by the end user in its finished form, or copied for mass production.

However, in some applications such as holographic data storage, certain types of holographic sensors [1], and the production of some security holograms it is necessary to record holograms in an environment which is difficult to control. Recording holograms and holographic data outside of vibration-controlled environments is a challenge. Away from the optical table, environmental noise and vibration will affect the fringe stability and severely reduce the contrast of the holographically recorded fringe pattern. In addition, design and cost considerations usually necessitate small, compact writing heads. The technique discussed here is one approach to avoiding the difficulties of recording holograms in such environments and involves recording the data gratings with a single beam, using diffraction at previously recorded weak gratings.

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This work reported here demonstrates a method for recording with a single beam that involves first ‘seeding’ the material with very low efficiency gratings and later selectively forming new gratings with a single beam using diffraction at these gratings. Despite the very low beam intensity ratio that is initially present, we show that high diffraction efficiencies of up to 80% can be achieved in gratings recorded.

1.2 Holographic recording with a single beam

It has long been known that the diffraction efficiency of recorded holograms and gratings can be amplified by various techniques. Chemical development processes are necessary to amplify the latent recorded pattern in materials such as silver halide and dichromated gelatin, and other materials require heating or other physical development.

Fully self-processing holographic materials are those which require no chemical, physical or optical processing in order to record the hologram, though there is usually a fixing step (e.g. photobleaching) to remove further photosensitivity [2]. The acrylamide-based photopolymer used here is one such fully self-processing material and for regular holographic recording with two beams, a refractive index modulation will be produced within a fraction of a second.

The optical method presented in this paper, however, doesn’t merely amplify, develop or strengthen the existing grating, but allows the writing of new gratings using just a single writing beam, angularly separated from the original grating. This is highly desirable for some applications, especially as the grating recording is achievable without the need for interferometric stability.

Most standard holographic recording is, of course, achieved by interfering two coherent recording beams. The interference between them creates the pattern that is recorded in the medium as a diffractive structure. However, due to the challenges of stability and size associated with split beams, various methods have been employed in order to achieve recording with a single beam.

Early work by Kukhtarev *et al.* [3], demonstrated holographic recording using only one input beam in a photorefractive BaTiO₃ crystal using the photogalvanic coupling between orthogonal birefringent modes. Naruse *et al* [4] multiplexed ten gratings into a Fe-doped LiNbO₃ crystal with a single recording beam by using the crystal edge in a wavefront-splitting arrangement. Mitsuhashi and Obara [5], using a similar approach, reported successful demonstration of a compact holographic memory sys-

tem using a single beam geometry in Fe-doped LiNbO₃. At that time they estimated a maximum total capacity of 23 Gbit based on a beam diameter of 5mm in a system using 635nm light and a 10mm crystal using both angular and spatial multiplexing.

Yau, *et al.* [6] have proposed another method that uses a single object beam to record images in a photorefractive LiNbO₃ crystal which allows imaging through a dynamically varying medium. The photorefractive fanning effect creates the reference beam. The advantage is that when rapidly changing distortions occur on a shorter time scale than the recording time, the image of the static object is recorded with minimal distortion

Chiang *et al.* propose a method that uses a single object beam to record multiple images in a medium without the need for a reference wave using a lenticular array [7]. This is demonstrated by recording four holograms in a 30 × 30 × 1 mm³ Fe-doped LiNbO₃ crystal with a single exposure.

More recent work by Kukhtarev, develops a dynamic version of his early single beam recording system, again in photorefractive crystals demonstrating dynamic holographic interferometry [8] and also holographic amplification of weak images without phase distortions [9].

In the commercial arena, Optware, a Japanese based company, developed a new method of holographic storage called collinear holography. Instead of separate signal and reference beams to create the interference pattern, Optware are using a collinear approach by aligning the two laser beams into a single beam of coaxial light to create data fringes. This approach significantly simplifies the recording set-up [10]. Optware released a prototype of this recording system operating at a wavelength of 532 nm with an overall storage capacity of 200 GB on a recording medium with diameter of 120 mm (HVD Pro Series 1000). They also released a credit card sized layer with a storage capacity of 30 GB and demonstrated a recording system with a storage capacity of 1 TB with a data transfer rate of 128 MB/s.

1.3 Single beam recording in acrylamide-based photopolymer

An example of the single beam recording method described here was first reported by the authors in 1998 [11]. After recording weak diffraction gratings in the photopolymer, gratings were illuminated on Bragg, with just one of the recording beams. For gratings with initial diffraction efficiencies ranging from less than 1% to 64%, a further increase in diffraction efficiency was observed dur-

ing the single beam exposure. For example, a grating with 7.5% efficiency was observed to increase to 60% efficiency over several minutes of single beam exposure. It was suggested at the time that the increase may be caused by either uniform polymerization of unreacted monomer in the grating, as had been observed in other photopolymers [12], or diffraction from the recorded grating. More recent work [13] demonstrated that the grating strength only increased significantly under single beam illumination when the single writing beam was incident close to the Bragg angle of the pre-recorded grating.

In this paper this method of writing holographic gratings using weak pre-recorded gratings is explored further, because of its potential to allow the use of just one beam at the data-writing stage. New gratings, angularly separated from the pre-written grating are written using a single beam and the dependence on grating thickness is demonstrated. We demonstrate that this approach allows the writing of high diffraction efficiency gratings in unstable conditions due to the fact that the second beam is generated from within the photopolymer layer, in a manner similar to beam pumping in photorefractive crystals. We demonstrate that angular multiplexing is also possible, allowing one grating to be amplified without amplifying the other pre-recorded gratings.

2 Experimental

2.1 Holographic recording

A two step process was used: (1) recording the weak ‘seed’ gratings with two recording beams (2) exposure of the seed grating to one beam. There was a short delay between the two steps during which both beams were blocked.

2.1.1 Two beam recording:

The first step was to record a grating, usually with a diffraction efficiency of approximately 1%, in the photopolymer medium. A standard holographic grating recording arrangement was used, with two coherent interfering beams (532nm) using beam splitters and mirrors, as shown in figure 1. The arrangement is for unslanted gratings. A He-Ne beam (633nm) was used to monitor the diffraction efficiency throughout the initial recording and subsequent illumination. This was possible because, in the formulation used, the photopolymer is not sensitive to red light. A short exposure time (around one second) was used for the

initial recording in order to keep the efficiency of the initial grating low. The spatial frequency was controlled by adjusting the angle between the two interfering beams and is 500 lines/mm in the work reported here.

2.1.2 Single beam recording

The next step was to illuminate the grating, on Bragg, with a single beam and observe the change in diffraction efficiency. The simplest way to do this was to use an additional shutter to block one of the two recording beams. Usually, a short interval with no writing beam illumination was allowed after the initial grating recording in order to allow the system to record any spontaneous change that may be occurring in the absence of illumination. Exposure times and writing beam illumination were controlled using Uniblitz electronic shutters. The photopolymer grating was mounted on a rotation stage so that angles of illumination could also be varied.

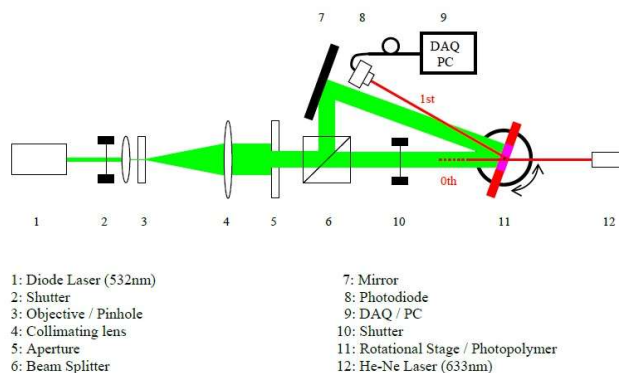


Figure 1: Experimental set-up

2.2 Photopolymer solutions

Photopolymer was prepared from stock solution of 10% polyvinylalcohol binder, acrylamide and bisacrylamide monomers, a green-sensitizing dye and triethanolamine, according to the standard method for this photopolymer described elsewhere [14]. Where different thickness layers were required the volume deposited on the substrate was adjusted appropriately.

3 Results

Figure 2 shows the diffraction efficiency changing during a typical exposure starting with a standard two-beam holographic recording of 2 seconds followed by a 25 second delay, during which there is no illumination. Then, at 27 seconds, illumination with one of the writing beams commences. The diffraction efficiency increase obtained during the single beam exposure is significant and diffraction efficiency of the final grating much higher than at the point when single beam exposure commences. In this case the grating spatial frequency is 500 lines/mm and the layer thickness is 135 μm . Each exposing beam has an intensity of 2.5 mW/cm^2 . Figure 2 is typical of the diffraction efficiency increase observed upon exposure to a single on-Bragg recording beam. No dependence was found on the delay time between the two beam and single beam exposure and weak gratings could be enhanced by single beam exposure after 12 weeks, as long as the photopolymer was still sensitive.

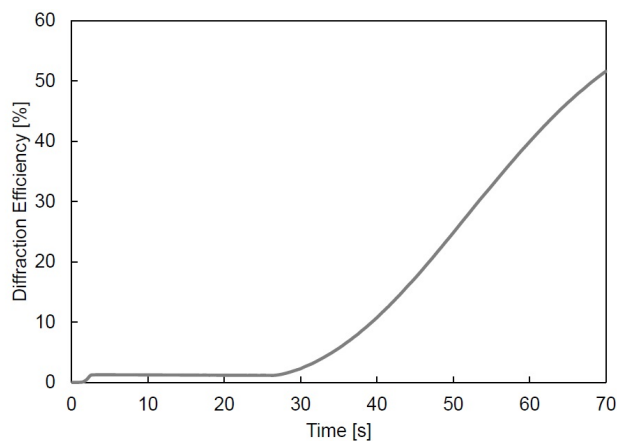


Figure 2: Diffraction efficiency versus exposure time. A standard two-beam holographic recording of 2 seconds is followed by a 25 second delay (no illumination) and then illumination with just one of the writing beams. Exposure intensity is 2.5 mW/cm^2 in each beam

The increase in diffraction efficiency consistently observed under single beam illumination in this photopolymer is due to a new grating formed by the interference between the single writing beam and the 1st order beam generated by diffraction at the pre-recorded weak grating (see discussion below).

3.1 Single beam recording with Bragg mismatch

The need for near-Bragg matching of the single writing beam rules out any bulk photochemical effect as the cause of the diffraction efficiency increase and supports the idea that diffraction is the main contributor. In order to study this further, and also assess the potential for multiplexing, the angle of incidence of the single writing beam was varied around the Bragg angle of the pre-recorded grating. The results are shown in Figure 3. The original seed grating diffraction efficiency was close to 1% in each case and the photopolymer thickness was 200 μm . The spatial frequency of the seed grating was 500 lines/mm. It can be seen from Figure 3 that there is an optimum angle, close to the Bragg angle of the pre-recorded grating, that maximizes the strength of the grating recorded with the single beam writing process. As the illuminating beam is moved further away from the optimal angle the final diffraction efficiency is reduced under the same exposure conditions. This is probably due to the reduced coupling between the single writing beam and the seed grating. In this example the optimal angle is about 0.5 degrees from the Bragg angle for the original grating. The asymmetry of the sidelobes is also a consistent feature. Both of these are thought to be due to fringe bending during formation of the grating under single beam exposure as discussed below.

This work demonstrates that the grating strength only increases significantly under single beam illumination when the single writing beam is incident close to the Bragg angle of the pre-recorded grating.

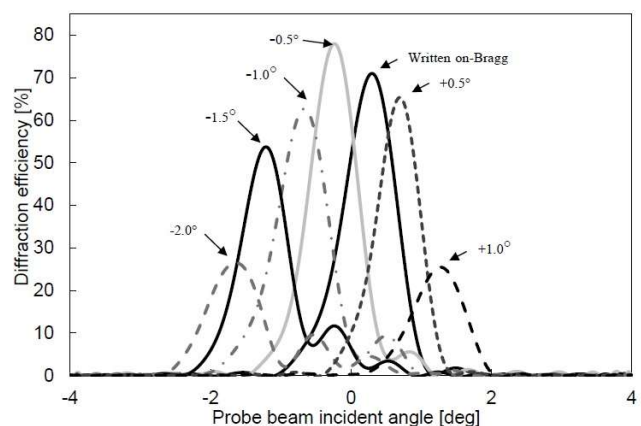


Figure 3: Bragg curves (the variation of diffraction efficiency with reading beam angle of incidence) for a series of gratings formed using the single beam process using different angles of incidence of the single writing beam. The layer thickness is 200 μm . The arrows indicate the offset (in degrees) from the Bragg angle of the seed grating (0°).

As was discussed in [13] the angular position of the Bragg peak for the gratings recorded in this way is linearly dependent on the angle of incidence of the single writing beam. This shows that the formation of a new grating formed by diffraction is responsible for the observed increases.

We propose that the weak diffracted beam interferes with the undiffracted beam to produce a low-contrast interference pattern which is immediately recorded in the material. If the new grating is in phase with the original grating more light will then be diffracted into the weaker beam, reducing the beam ratio and increasing the contrast in the interference pattern, in turn producing an even stronger diffracted beam. In this way, quite weak gratings can rapidly ‘seed’ the growth of relatively high diffraction efficiency gratings. This growth of a new grating is analogous to the energy transfer between the strong beam and the weak beam in ‘beam pumping’ in photorefractive crystals, except that the refractive index modulation created is permanent.

A potential difficulty with the above explanation is the phase mismatch between the ‘seed’ grating and any grating created by diffraction at the ‘seed’ grating. This is due to the fact that there will be a phase difference ($\pi/2$) between the incident beam and the beam diffracted by the phase grating, which would cause any new grating to be out of phase with the original. Beam pumping in photorefractives, which is also initiated by diffraction at a weak grating, occurs only because the recorded grating in photorefractive crystals is laterally displaced with respect to the interference fringes that create it.

Unlike photorefractive crystals, photopolymers are usually considered to produce gratings that are not laterally shifted from the interference pattern that creates them. Thus we might not expect the interference pattern created by the incident beam and its diffracted beam to produce a grating that is in phase with the one that created it. However, such shifts and non-linear recording profiles have been observed in holographic recording materials such as acrylamide photopolymers [15], nanoparticle doped photopolymers [16] and silverhalide emulsions [17]. Murciano *et al.* [18] showed that effects such as beam bending and two wave mixing, have been observed even with very small phase shifts in similar materials. They analyzed the origin and effects of fringe bending and Bragg detuning in holographic gratings recorded in rigid media such as photopolymerizable inorganic silica glass materials and proposed that they occurred as a result of the non-sinusoidal nature of the recorded pattern. Using an acrylamide-based photopolymer-doped sol gel, Murciano *et al.* observed two-wave mixing during two-beam record-

ing and used a two-wave mixing model to explain the asymmetry and angular shift (fringe bending) observed in their angular selectivity (Bragg) curves. The reconstruction model used by Murciano *et al.* to analyse the gratings used coupled wave theory taking into account two wave mixing occurring during recording. Fringe bending gets larger as thickness and refractive index modulation increase, and of course, depends greatly on the initial beam ratio. Good agreement was obtained between experimental and theoretical results of simulations with a shift of just 2.6° between the recorded grating and the light pattern, demonstrating that very small shifts can cause such effects.

It seems likely that the results described above would have a similar origin. i.e., the non-sinusoidal refractive index profile of the recorded grating provides enough of a phase shift to allow coupling from the strong beam to the weak diffracted beam during single beam recording. It should also be borne in mind that the beam ratio is very large (typically 99:1) at the start of the single beam recording step, so small amounts of energy transfer from the strong beam will have a significant effect, and also the fringe period is very much larger than the wavelength for these low spatial frequency gratings.

In the case of these acrylamide-based photopolymers, gratings are recorded via photopolymerization and diffusion. Either of these processes can dominate depending on the recording intensity, spatial frequency and photopolymer formulation [19] and non-sinusoidal grating profiles are also common especially at low spatial frequencies [20]. It has been observed that the process of enhancing diffraction efficiency reported here is stronger at lower spatial frequencies.

3.2 Single beam recording with different layer thickness

Experiments with different layer thicknesses ranging from 60 to 240 μm showed that the enhancement of the seed grating occurs much more in thicker layers. Figure 4 shows the diffraction efficiency increasing under single beam illumination from an initial diffraction efficiency of approximately 1% for thicknesses of 60, 130, 190 and 240 μm . These curves are each obtained in the same way as that in Figure 2 but the delay between the two exposures was 110 seconds in these experiments. The gratings were exposed to a single beam at the Bragg angle for 150 seconds. The spatial frequency is 500 lines/mm and each exposing beam has an intensity of 2.5 mW/cm^2 . It is observed that the increase in efficiency when recording with a single beam is much more pronounced in thicker layers and

practically non-existent in the layer with thickness 60 μm .

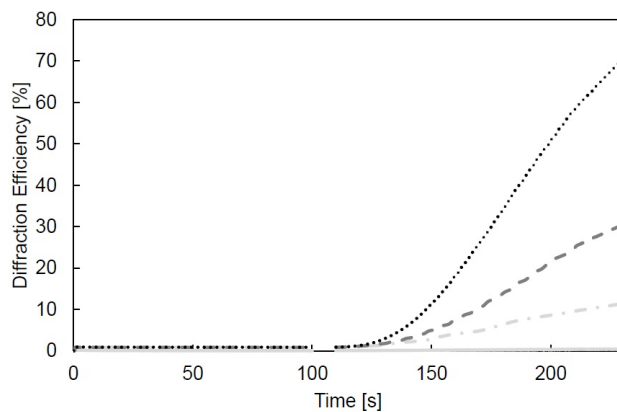


Figure 4: Growth in diffraction efficiency, under single beam illumination, of ‘seed’ gratings in samples prepared with different thicknesses. The grating spatial frequency is 500 lines/mm and the layer thicknesses are approximately 60 μm (solid), 130 μm (dot-line), 190 μm (dashed line) and 240 μm (dotted line). Initial diffraction efficiency is approximately 1% for the two-beam grating.

The behavior at a range of angles was investigated for three thicknesses, including the 200 μm sample in Figure 3. In each case graphs similar to fig 3 were obtained with the Bragg angle for the final grating changing with the angle of incidence of the single writing beam. In Figure 5, the location of each peak is plotted as a function of the Bragg mismatch of the writing beam for the three layer thicknesses. The slope is close to 1 in each case but the intercept is different for each thickness. This is consistent with beam bending as discussed above.

3.3 Recording in mechanically unstable environments

One of the biggest challenges in holographic recording is the great sensitivity to relative movement in the optical recording system. Any change in the optical path difference between the two recording beams due to movement of the optical components can shift the interference pattern and ‘blur’ the recorded fringe pattern. If the movement causes the interference pattern to move by half a fringe period for half the exposure time, the hologram will be erased.

In a regular two-beam recording the diffraction efficiency increases as the recording progresses but sudden mechanical disturbances, at times shown by the arrows in Figure 6(a), causes the grating growth to stop until the sys-

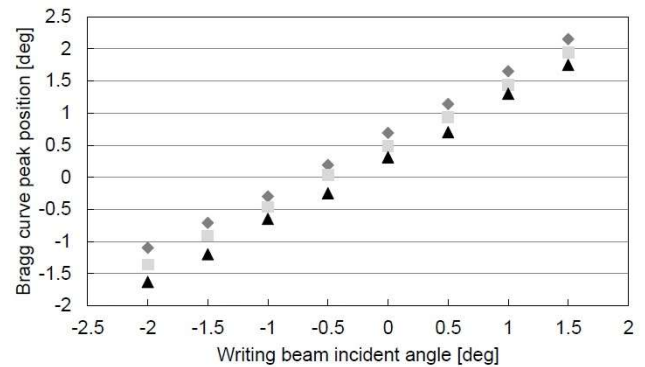


Figure 5: The angular position of the Bragg peak of gratings formed by the single beam process, as a function of the angle of the writing beam. 0° on the x axis corresponds to the angle of the beam used in writing the ‘seed’ grating. The grating spatial frequency is 500 lines/mm and the layer thicknesses are 60 μm (diamond), 135 μm (square), 200 μm (triangle). Initial diffraction efficiency is approx. 1% for the two-beam grating.

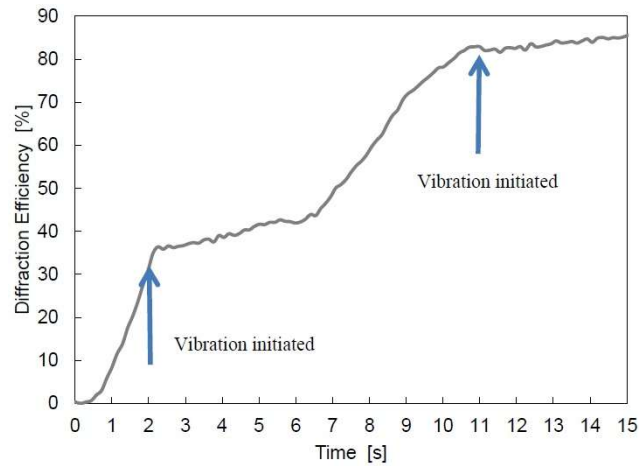
tem stabilizes and greatly reduces the final diffraction efficiency. However, if a single beam recording is subject to the same mechanical disturbance, as in Figure 6(b), no change in the grating growth is observed

These results demonstrate that single beam recording works well in unstable conditions.

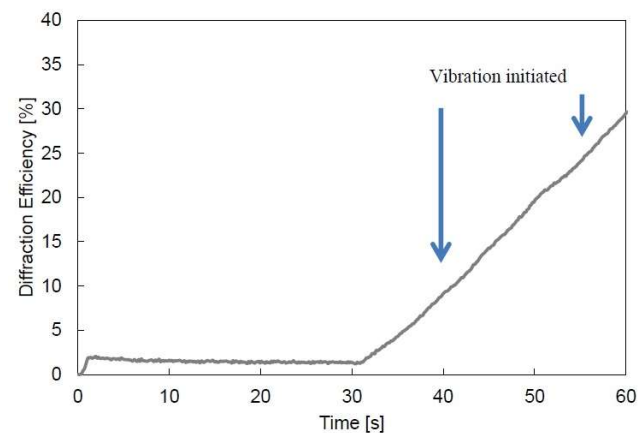
3.4 Angular multiplexing of data using a one beam system

In some applications it may be necessary to obtain a diffraction efficiency increase in one grating selected from a number of gratings angularly multiplexed in the same region of the photopolymer layer, without affecting its neighbors. The selectivity demonstrated in the above data (Figure 3) implies that this should be possible.

In order to investigate if it would be possible to selectively boost one grating from a series, a layer of photopolymer 135 μm in thickness was used and 5 gratings of equal strength were angularly multiplexed into it. One of the gratings was then illuminated at its Bragg angle in order to increase its efficiency. For separations up to 1.8 degrees illumination caused a significant increase in the diffraction efficiency of gratings on either side of the intended grating, Figure 7 shows the result for gratings 2° apart. Although there are only five seed gratings in these examples, the principle is demonstrated that one of a set of angularly multiplexed gratings, separated by 2 degrees, can be enhanced by illuminating it at the appropriate angle with a single beam of light. Working with higher spatial



(a)



(b)

Figure 6: Comparison between a regular two-beam recording and the single-beam recording in mechanically unstable environment. The growth curves for both recording schemes in a disturbed environment are shown in figure 6(a) and 6(b). Vibration was initiated at 2 and 11 seconds during the two beam recording in 6(a) and at approx. 40 and 55 seconds during the one beam recording in 6(b).

frequency and thickness would be likely to allow smaller angular separation between neighboring gratings.

4 Conclusion

A method of writing holographic gratings based upon the use of weak pre-recorded gratings is discussed. We demonstrate that this approach allows the writing of high diffraction efficiency gratings in unstable conditions due to the fact that the second beam is generated within the photopolymer layer. The dependence on grating thickness was investigated and it was shown that the diffraction efficiency enhancement increases with grating thickness. The

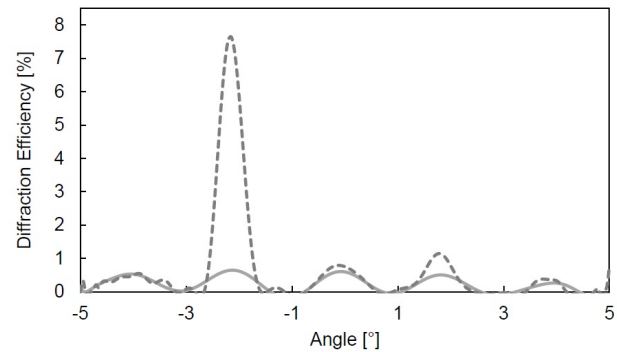


Figure 7: Variation of diffraction efficiency with reading beam angle of incidence for a series of gratings, from which one individual grating has been enhanced by illuminating with a single on-Bragg beam of light. The angular separation between neighbouring gratings is 2° .

potential for angular multiplexing is shown by illuminating a single grating from among five multiplexed weak gratings increasing its efficiency eight-fold with negligible effect on the other gratings.

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