FOR KODAK PRODUCTS

Reversal Bleaching for Low Flare Light in Holograms

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It has been shown that the principal cause of flare light in a bleached hologram is self-interference of light from an extended object. In most bleaching processes the surface relief image and variation of refractive index of the bleached emulsion combine to enhance the self-interference pattern at low spatial frequencies and thus to enhance the flare light. This paper describes a reversal bleach process for Kodak spectroscopic plates, Type 649-F, such that the relief image tends to cancel the effects of the index variation for low spatial frequencies. This makes it possible to achieve high diffraction efficiencies and signal-to-noise ratios. Data are given.

Introduction

It is generally recognized that a good phase hologram should be able to diffract light much more efficiently than an amplitude hologram and, consequently, produce a much brighter image. A number of bleaching processes for photographic materials have been described in the literature,[1-8] but in general, they have received only limited acceptance. One of the primary difficulties encountered is that the image tends to be badly degraded by flare light. In some cases this can be so extreme that the image is essentially obscured. Among the processes that have been described, there are several that reduce the flare light, but most, if not all, of the silver halide processes tend to be complicated and time consuming. In this paper we will describe a process that is quite simple and reliable, but at the same time produces high-efficiency images that are fairly free of flare light.

Although it often has been assumed that flare light is the result simply of light scattering from the silver salts within the bleached emulsion, it is not difficult to show that for most bleach processes this is not true. For example, when a Kodak spectroscopic plate, Type 649-F, is exposed with ordinary white light, developed, fixed, and bleached in Kodak bleach bath R-10,[9] the resulting plate shows very little light scatter. On the other hand, a hologram processed similarly can show an excessive amount of flare light.

Upatnieks and Leonard[5] showed that the flare light results from the effects of self-interference of the light from an extended object, and they pointed out that the flare light becomes increasingly objectionable as the ratio of the light flux from the reference source to that from the object is decreased. Our own preliminary experiments showed that for holograms exposed with this ratio made very large, there was very little flare for most of the bleach baths we tried.

Because self-interference only can take place with an extended object, this type of flare light does not occur for holograms made by interfering the light from two point sources. A number of bleaching experiment results reported in the literature were obtained in this manner, and the data from such experiments can be quite misleading.

Theory

We can see the effects of an extended source in a phase-type holographic system by means of the following simplified first-order analysis. If we denote the complex amplitude of the reference beam as it falls on the plate by U_r and that of the object beam by $U_o(x)$, the exposure at the plate will be given by

$$E(x) = [|U_r|^2 + |U_o(x)|^2 + U_rU_o^*(x) + U_r^*U_o(x)]t, \quad (1)$$

where t is the exposure time. It is important to remember that the term $|U_0(x)|^2$ is not constant since it represents the laser speckle pattern formed from a diffuse object. If, as in a conventional hologram, the amplitude transmittance is linearly related to exposure, E, the term $|U_0(x)|^2$ causes the flare light around the reference beam upon reconstruction.

For a bleached hologram we can assume, as a first-order approximation, that the optical path through the bleached emulsion layer is proportional to the exposure. If we let B be the proportionality factor and $k = 2\pi/\lambda$, we can represent the complex amplitude transmittance in the form

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$$T_a(x) = \exp\{ikBt[|U_r|^2 + |U_o(x)|^2 + U_rU_o^*(x) + U_r^*U_o(x)]\}. \quad (2)$$

This also can be written as a product of four exponentials and each of these can be expanded in series form to give

$$T_a(x) = [1 + ikBt|U_r|^2 + \ldots][1 + ikBt|U_o(x)|^2 + \ldots] + [1 + ikBtU_rU_o^*(x) + \ldots][1 + ikBtU_r^*U_o(x) + \ldots].$$

Discarding the first bracket since it represents a constant phase retardation, we obtain to second order in kBt,

$$T_a(x) = 1 + ikBt\{|U_o(x)|^2 + U_rU_o^* + U_r^*U_o(x)\} - k^2B^2t^2|U_o(x)|^2\{U_rU_o^*(x) + U_r^*U_o(x) + |U_r|^2\}. \quad (3)$$

The first expression in the curly brackets is similar to that obtained for a linear system and these three terms respectively account for the flare around the reference beam, for the primary image, and for the conjugate image. The second expression in curly brackets also contains terms corresponding to the holographic images but, since it is multiplied by $|U_o(x)|^2$, this means that the flare light ordinarily appearing around the reference beam now also is combined with the images. For a Fraunhofer system the flare light would be convolved with the images.

If we assume the approximation in Eq. (3) to be valid, and consider only the coefficients of the terms representing the virtual image, we can see that

$$\text{signal/noise} = 1/(kBt\langle|U_o(x)|^2\rangle), \quad (4)$$

where the sharp brackets denote a spatial average. The total average exposure E_T is given by

$$E_T = |U_r|^2 + \langle|U_o(x)|^2\rangle, \quad (5)$$

and the beam balance ratio is given by

$$K = |U_r|^2/\langle|U_o(x)|^2\rangle. \quad (6)$$

Substituting Eqs.(5) and (6) into Eq.(4) gives

$$\text{signal/noise} = (1 + K)/(kBtE_T). \quad (7)$$

This shows that making the beam balance, K, very large should improve the signal-to-noise ratio, as was mentioned previously. However, since diffracted light flux is generally reduced by increasing the beam balance ratio, an undesirable consequence of using a large beam balance ratio to achieve good signal/noise is that the brightness of the holographic image is reduced. Clearly, it is desirable to be able to reduce the beam balance ratio to get good efficiency while somehow avoiding consequent reduction in signal/noise.

So far, we have assumed that the optical path length through the processed emulsion is related to the exposure by a constant, B, independent of spatial frequency. However, in considering a bleached photographic plate, we find that the optical path-length variations for the transmitted light result from two factors. The first is the variation of refractive index within the emulsion layer, caused by varying amounts of high-index silver salts imbedded within the gelatin.

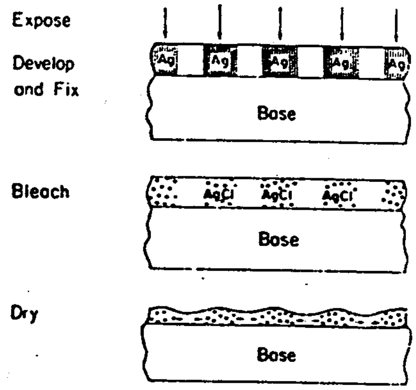

Fig. 1. Schematic of the emulsion cross section for the direct bleach process.

This can give the hologram a *thick* grating structure for high spatial frequencies which, as is well known, makes it possible in theory to approach 100% diffraction efficiency.[10]

The second factor is the variation of thickness of the emulsion layer, commonly termed the relief image. These generally conform to the developed photographic images, but it has been shown that they are strongest for spatial frequencies below about 100 cycles/mm, decreasing rapidly for higher frequencies.

Because the path length or phase variations result from a combination of these factors, the spatial frequency response of the system can be strongly frequency dependent. This means that in most practical cases the coefficient, B, is not a constant and to a crude approximation we can rewrite Eq.(2) in the form,

$$T_a(x) = \exp\{ikB_1t|U_o(x)|^2\}\exp\{ikB_2t[U_rU_o^*(x) + U_r^*U_o(x)]\}, \quad (8)$$

where B_1 is the coefficient for the lower spatial frequencies, characteristic of the flare-producing speckle pattern, and B_2 is the coefficient for the spatial frequency region of the holographic fringes. In such a case, Eq.(7) becomes

$$\text{signal/noise} = (1 + K)/(kB_1tE_T). \quad (9)$$

Hence, if we could keep B_2 constant while reducing B_1, the signal-to-noise ratio would improve with no loss in image brightness. One method of accomplishing this consists in immersing the emulsion surface in an index-matching liquid to cancel the effects of the relief image.[5,11] The use of such a liquid gate, however, can be rather cumbersome for many applications. Low-frequency suppression must undoubtedly occur for other low-noise processes that have been reported in the literature. This is notably the case for thermoplastic materials, as discussed by Urbach and Meier.[10]

Direct Bleaching Process

Most commonly used bleaching processes are what we can term *direct* bleaching processes, and these tend to enhance, rather than suppress, the low spatial frequencies. A typical process is sketched in Fig. 1. After the film or plate is developed and fixed, it is bleached in some solution that converts the metallic silver into a transparent, insoluble salt having a refractive index significantly higher than that of the gelatin. For example, Kodak bleach bath R-10 converts the silver into silver chloride.[9] (By modifying the *B* solution it is also possible to produce silver bromide or silver iodide.) When the emulsion is subsequently dried, a relief image is formed on the emulsion surface. This occurs because of the added bulk of the silver halide in the gelatin and because the gelatin is tanned or hardened by the reaction products formed when the developer converts the silver halide into metallic silver. The tanning tends to be fairly localized within the emulsion and, as a result, the emulsion pulls together, as shown by the arrows in the lower part of Fig. 1. Typical relief images for low-spatial-frequency line-pattern exposures can be several wavelengths high but they become much smaller for spatial frequencies above 100 cycles/mm. Smith[12] has discussed the formation of relief images in detail.

For the direct bleach, the variations of optical path length from the relief image and those from the index variations within the emulsion are additive, and consequently the emulsion acts as a low-pass spatial filter. Because the speckle pattern is confined predominantly to the lower spatial frequencies, it becomes greatly enhanced in the direct bleaching process and excessive flare light is produced upon reconstruction.

Table I. Kodak Special Developer SD-48[a]

Solution A	
Water	750 ml
Sodium sulfite, desiccated	8 g
Pyrocatechol[b]	40 g
Sodium sulfate, desiccated	100 g
Water to make	1 liter
Solution B	
Water	750 ml
Sodium hydroxide	20 g
Sodium sulfate, desiccated	100 g
Water to make	1 liter

[a] *Caution:* Use rubber gloves or take special precaution to keep hands from coming in contact with the developer.

[b] This is available as Eastman organic chemical P604. It is a hazardous solid material and breathing of dust or vapor can be harmful. Avoid getting pyrocatechol on the skin, in eyes, or on clothing.

Table II. Processing Procedure

Step	Time	Temperature °F	Temperature °C
1. Develop (Kodak special developer, SD-48)	5 min	75	24
2. Stop bath (Kodak stop bath SB-1)	15 sec	70–80	21–27
3. Rinse, running water	1 min	70–80	21–27
4. Bleach (Kodak bleach bath R-9)	3 min	70–80	21–27
5. Wash, running water	5 min	70–80	21–27
6. Stain removal (Kodak stain remover S-13, Solution A)	1 min	70–80	21–27
7. Clear (Kodak stain remover S-13, Solution B)	1 min	70–80	21–27
8. Wash, running water	5–10 min	70–80	21–27
9. Dry[a]			

[a] As is the case with other processes, drying is critical. For uniform drying, it is suggested that the plate be rinsed in a solution of methanol diluted 1:1 with water, then washed twice in isopropyl alcohol.

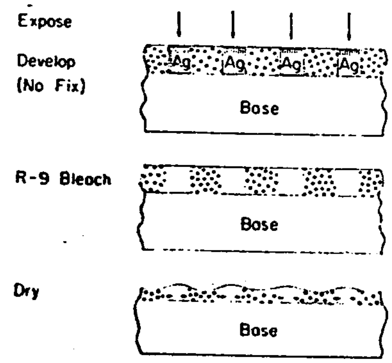

Fig. 2. Schematic of the emulsion cross section for the reversal bleach process.

Reversal Bleaching Process

As a solution to these problems, we have devised a bleaching system wherein the relief-image path-length variation is kept to a minimum and may even counteract the index variation. This greatly reduces the over-all system response for low spatial frequencies and drastically reduces the flare light in the holographic image.

The essential steps of our process are sketched in Fig. 2. The exposed emulsion is developed but not fixed, so that silver halide remains in the unexposed regions. The plate then is bleached in Kodak bleach bath R-9, which converts the metallic silver to a soluble salt and removes it from the emulsion. Because the remaining silver halide appears reversed from what it was for the R-10 process, we refer to this as a *reversal bleach*. Chang and George have also reported good results for a somewhat different reversal bleach process.[13]

Maximum tanning occurs in regions of maximum development, and there will be a tendency for a relief image to be formed as shown at the bottom of Fig. 2. However, the bulk of the silver halide tends to counteract this effect and, for commonly used developers, the bulking appears to be the stronger effect. In order to produce sufficient tanning, we found it necessary to formulate a new developer, Kodak special developer SD-48, whose formula is given in Table I.

The processing procedure for the reversal bleach is simple and straightforward, as shown in Table II. It is basically no more complex than an ordinary black

Table III. Bleach and Stain Remover Formulas

KODAK BLEACH BATH R-9	
Distilled water	1.0 liter
Potassium dichromate	9.5 g
Sulfuric acid (concentrated)*	12.0 ml
KODAK STAIN REMOVER S-13	
Solution A	
Water	750 ml
Potassium permanganate	2.5 g
Sulfuric acid (concentrated)*	8.0 ml
Water to make	1.0 liter
Solution B	
Water	750 ml
Sodium bisulfite	10 g
Water to make	1 liter

* *Caution:* Always add the sulfuric acid to the solution slowly, stirring constantly, and never the solution to the acid; otherwise the solution may boil and spatter the acid on the hands or face, causing serious burns.

and white development. Table III gives formulas for Kodak bleach bath R-9 and for Kodak stain remover S-13 solutions. The latter remove brown stain that appears after development. These solutions also make the final plate much less subject to darkening from printout.

Developer Formulation

The roles of the various constituents of a developer are well known and have been described in detail in the literature.[14,15] Our developer is unusual in that it uses pyrocatechol (also known as catechol) as a developing agent. Used at high pH, this is a highly active developer with a strong tanning action.

The sodium sulfite acts as a preservative but it is used here in only a small amount since it counteracts the tanning action. Consequently (and unfortunately), the developer tends to oxidize (or turn black) fairly rapidly in the presence of air, but this occurs only when the A and B solutions are mixed together; the individual solutions are relatively stable, and are mixed only just before use.

Because of the rapid oxidation we have found it advantageous to develop the plates in a specially constructed tank that will accommodate a standard plate holder and that requires a minimum of the developing solution. Our model has a built-in gas distributor for nitrogen-burst processing. This consists simply of a section of polystyrene tubing with No. 80 holes drilled into it. Such a procedure also makes it easier to avoid contact of the developer with the skin.

The sodium sulfate serves to prevent excessive swelling of the gelatin during development, but otherwise has little effect on the development. Potassium bromide is present in many developers to suppress the fog level, among other things, and our original formulation contained 15 g of KBr per liter of developer. However it was found that diffraction efficiency for 649-F plates was improved as much as three to six times by eliminating it. This was attributed to the fact that it acts as a silver halide solvent and causes solution physical development to occur. We found that any such solvent (such as sodium thyocyanate) caused an immediate reduction of diffraction efficiency for reversal processing.

Experimental Details

To evaluate the effectiveness of the solutions and procedures, measurements were made with an instrument described earlier,[16] which scans a one-dimensional pattern and records the optical path variation (OPV) of the transmitted light. Kodak spectroscopic plates, Type 649-F, were exposed in a camera used for MTF evaluation of photographic materials.[17] By using special test objects we could make sinusoidal line exposures with a modulation of approximately 25% at the image plane of the lens.

Figure 3 shows the OPV for a direct bleach process plotted as a function of exposure time for spatial frequencies of 10, 25, 100 and 200 cycles/mm. The plate was developed for 5 min in Kodak HRP developer at 75°F, (24°C), fixed, and bleached in R-10 bleach bath. The data in this figure can be compared directly with those in Fig. 4, which shows similar data for our reversal bleach process. The development in the latter case was 5 min in SD-48 at 75°F (24°C). Clearly the OPV for low spatial frequencies is very much reduced by using the reversal bleach process. The exposure required for a conventional unbleached holo-

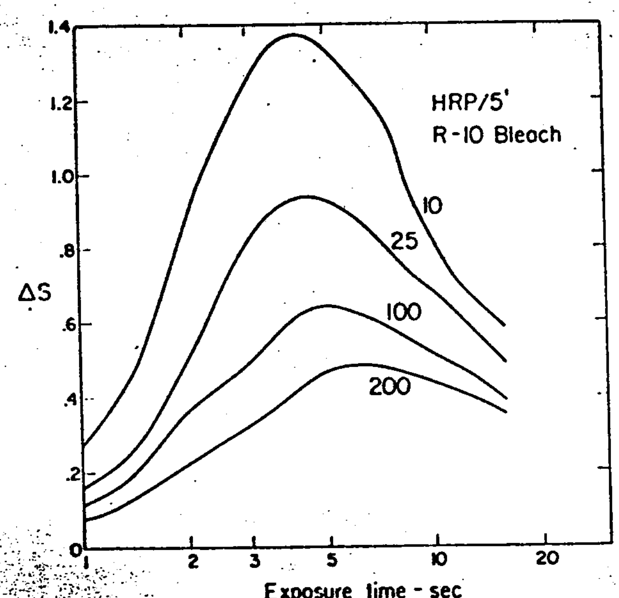

Fig. 3. Optical path variation, ΔS, measured in wavelengths and plotted as a function of relative exposure for sinusoidal patterns of 10 cycles/mm, 25 cycles/mm, 100 cycles/mm, and 200 cycles/mm. Kodak 649-F plate was processed by direct bleaching.

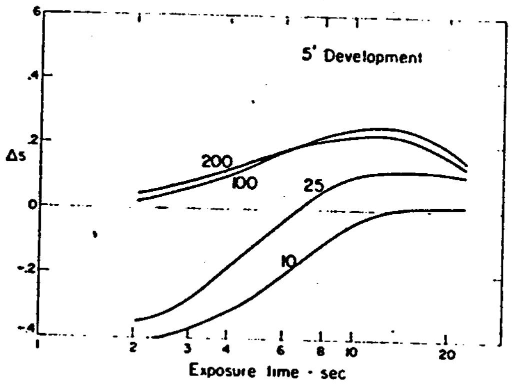

Fig. 4. Data similar to those in Fig. 3 but for the reversal bleach process given in Table I.

gram is equivalent to about 2 sec on the exposure scale, with a 5-min development in HRP.

In further experiments, measurements were made of holograms processed with the reversal bleach. The holographic object was somewhat similar to that used by Upatnieks and Leonard.[5] It consisted of an illuminated ground glass approximately 25 mm square with a round opaque area about 6 mm in diameter in the center. Both the object and the reference beam pinhole were placed about 380 mm from the plate. The angle separating the beams was such that it gave a mean spatial frequency of about 1000 cycles/mm on the hologram. A He–Ne laser was used for all the holographic experiments.

Diffraction efficiency was measured by the following procedure: we first determined the beam balance ratio by placing a diffuser at the plane of the hologram plate and measuring the light from the reference and object beams with a photomultiplier placed directly behind it. Next, we removed this diffuser and imaged the object diffuser with an auxiliary lens onto a photomultiplier and measured the light flux. We then placed the hologram in place, blocked off the object, and measured the flux from the holographic image. For the last two measurements the aperture of the photomultiplier was displaced from the black spot in the center of the image. The ratio of the hologram to the object flux was then divided by the beam balance ratio to give the diffraction efficiency.[18,19] Efficiency so measured is in terms of the incident flux. This method makes it possible to make accurate measurements for most object configurations and circumvents problems that ordinarily arise because of nonuniformities of angular response or of the detecting photosensitive surface.

The system noise was determined by measuring the light with the black spot in the holographic image falling onto the photomultiplier.

Holograms were made with our reversal bleach process for a series of beam balance ratios. An exposure series was made for each ratio and a typical plot of the diffraction efficiency and signal-to-noise data is shown in Fig. 5. Each such plot showed a rather broad exposure latitude and a signal-to-noise ratio that improved with increased exposure.

The values of maximum efficiency for each of the plots are shown by the lower solid curve in Fig. 6 as function of the beam balance ratio, K. The values of signal-to-noise corresponding to these efficiency values are plotted as the dashed curve. For increased exposure the curves would have shown slightly lower efficiency but improved signal-to-noise.

A similar series of holograms was made with plates that had been presoaked in distilled water for 10 min and then dried. This represented an attempt to relieve strains in the emulsion as originally coated

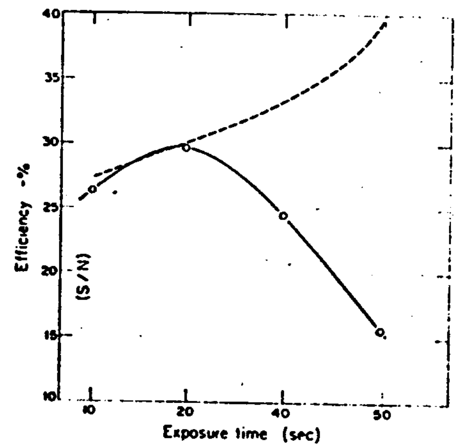

Fig. 5. Typical plot of diffraction efficiency and signal-to-noise ratio as a function of exposure.

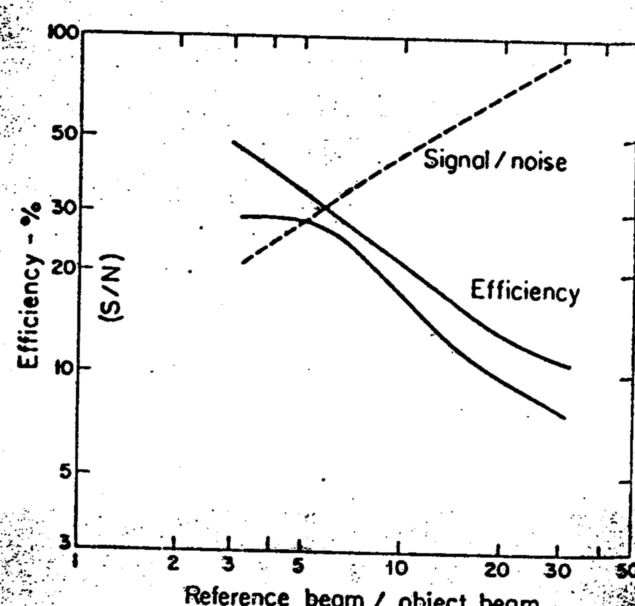

Fig. 6. Diffraction efficiency and signal-to-noise ratio for 649-F plates, plotted as a function of the beam balance ratio, K. The lower solid curve shows the diffraction efficiency obtained with unsoaked plates, and the upper curve shows the data obtained with plates that had been presoaked in distilled water and dried.

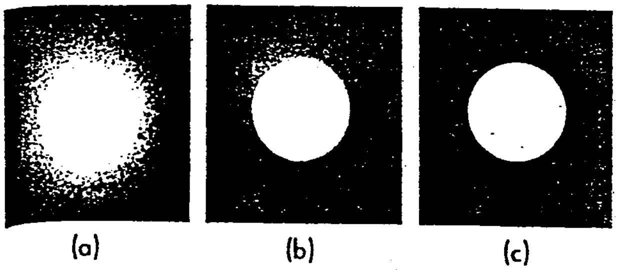

Fig. 7. Photographs of the holographic image of a diffusely illuminated disk: excessive flare light occurs when the plate is bleached in R-10 as shown at (a); this is reduced when the emulsion surface is immersed in an index-matching liquid (b); and almost eliminated with the reversal bleach process (c).

Measurements for these plates are shown by the upper solid curve in Fig. 6. The signal-to-noise data for the presoaked plates were so close to the previous data that a single curve is shown in the figure. In general the presoaking increased the diffraction efficiency by about 10%. For a beam balance ratio of 3.2:1, these plates gave a measured diffraction efficiency of approximately 40% with a signal-to-noise ratio of about 17. While the data show a distinctly improved efficiency resulting from presoaking, the improvement is hardly sufficient to make a noticeable difference when the holograms are viewed.

Figure 7 shows three photographs made of holographic images of a simple disk diffuser. Photograph (a) is the image from a hologram that had been developed for 5 min in HRP developer, fixed, and then simply bleached in R-10, washed, and dried. Photograph (b) is an image from the same hologram but with the emulsion surface immersed in a liquid with refractive index of 1.54. Photograph (c) is the image from a hologram processed with the reversal bleach process described above. The holograms were made with a beam balance ratio of about 6:1. All the photographs were exposed, processed, and printed identically. Diffraction efficiencies for these three cases were approximately the same. It is interesting to note that the flare reduction with the reversal bleach process is greater than can be accounted for simply by the elimination of the surface relief with a liquid gate. Similar holograms were made by using other published bleach processes, but none showed as little flare light as the reversal bleach process, for comparable diffraction efficiencies.

In a further direct visual comparison between the primary holographic image formed by a bleached hologram with that formed by a conventional unbleached hologram (developed 5 min in HRP developer and fixed), we found that when a 0.9 neutral density was placed over the bleached hologram the images appeared essentially identical. This showed that the image from the bleached hologram was about eight times as bright as that of the other while the signal-to-noise ratios were comparable. Both holograms were made with a beam balance ratio of about 5:1.

Finally, it should be noted that, whereas the principles for reducing flare light are general, the experiments were performed with only one photographic material, Kodak spectroscopic plates, Type 649-F. It has not been determined how (or if) the developer should be modified for best results with other photographic materials.

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