Color Reflection Holograms Recorded in a Panchromatic Ultrahigh-Resolution Single-Layer Silver Halide Emulsion

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A review of color holography is presented as an introduction. A new method of recording and processing high-quality color holograms in an ultrahigh-resolution silver halide emulsion has been developed. Color reflection holography presents no fundamental problems with regard to the geometry of the recording setup, but the final result is highly dependent on the recording material used and its processing. By the introduction of extremely high-resolution panchromatic emulsions it has become possible to obtain high-quality, large-format color reflection holograms. The use of three laser wavelengths on a single-layer emulsion in the recording process makes the holographic recording technique similar to the early Lippmann photography technique of the last century. That combination not only promotes good color rendition, but, additionally, because no dyes or pigments are used in the emulsion of the final hologram, high archival color stability can be predicted for the image. The recording procedure, employing a Denisyuk setup and three laser wavelengths, and the processing technique are described. The processing of such holograms is critical to obtain high diffraction efficiency and good color rendering. In particular, the prevention of emulsion shrinkage is extremely important. Color holograms up to 30 × 40 cm have been recorded.

Journal of Imaging Science and Technology 40: 134 – 146 (1996)

Introduction

Even 30 years after the appearance of the first monochromatic holograms the ability to record high-quality holograms in true colors is still very limited. Although various special techniques now allow for the production of holograms exhibiting several different colors, in most cases the colors displayed in these holograms are not the true, original colors of the holographed object. These holograms are often referred to as *pseudocolor* or *multicolor* holograms. Methods exist for creating colors that give an impression of a true color in the finished image, e.g., multiple recorded stereograms or rainbow holograms, although the recordings could well have been made from objects having completely different colors. They can also be made from multiple sets of color-separated photographs. By using the rainbow technique it has actually been possible to mass produce embossed holograms with either natural or artificial colors. It is also very common among artists to make multiple-exposure color reflection holograms using a single-wavelength laser, with the emulsion thickness changed between the recordings of special objects.

Sometimes lifelike holographic images have been referred to as *full color, natural color,* or *true color* holograms. The most logical name for these images that comes to mind in analogy with color photography, color movies, and color television would be *color holograms*. However, this term is sometimes objected to on the grounds that some colors of the objects we normally see are impossible to record holographically, because holograms can only *reproduce colors* of objects *created by the scattered laser light* (light consisting of various laser wavelengths). The colors we see are often the result of fluorescence, which cannot be recorded in a hologram. For example, some dyed and plastic objects achieve their bright, saturated colors by fluorescence. This limitation in color holography does not, however, seem to be very dramatic.

In this article, we present a reliable and rather simple method for recording large-format color holograms, based on a new holographic panchromatic, ultrahigh-resolution, single-layer silver halide emulsion. In addition to the description of our recording and processing technique for color holograms, we introduce the topic by presenting a review of the history and development of color holography.

To record high-quality color reflection holograms, extremely low scattering recording materials must be used, for example, ultrahigh-resolution silver halide emulsions.[1] This type of material has the advantage of higher sensitivity than photopolymer and dichromated gelatin (DCG), which are alternative materials for color holography. Ultrahigh-resolution silver halide emulsions for monochrome holography have been manufactured in Russia for many years, and currently these emulsions are panchromatically sensitized. In this paper we refer to the ultrahigh-resolution holographic emulsions (grain size 10 to 20 nm) as "Russian" emulsions, because they were first made in Russia (by both Protas and Kirillov).

Protas was able to slow down grain growth during emulsification by increasing the number of growth centers and introducing special growth inhibitors. The best Russian emulsion ever made is probably the one achieved by Kirillov. In this case, grain growth was hampered by the

Original manuscript received March 9, 1995. Revised January 12, 1996.

fact that in the emulsification process a highly diluted solution was used and the emulsion concentration was increased by applying the method of gradual freezing and thawing. A rather dilute solution of emulsion containing 0.5 – 1% gelatin was used. Just after the emulsion is mixed, it is poured into a beaker and frozen at a temperature between –10 and –20°C or even lower. The emulsion is kept in the beaker for 10 to 15 h. A rapid freezing method is also suggested. Here, a thin layer of emulsion is poured into an already chilled tray. The frozen emulsion is then chopped into small noodles and put on a grid for thawing. To speed up this process, the frozen emulsion noodles are showered with cold water (3–5°C). During this process, the emulsion is, at the same time, washed to remove unwanted salts. When the temperature of the emulsion increases (to about 25–30°C) the emulsion becomes liquid and undergoes a gentle ripening. The process of freezing and thawing can be repeated several times to increase the concentration of the emulsion. It has been verified that the silver content in the emulsion typically increases about 10 times during this repeated process. The silver content of the emulsion is about 2 – 2.5 g/L at emulsification, and after concentration 20 – 30 g/L. Sodium thiosulfate is added at a temperature of 30 to 32°C over 5 to 10 min. Then gold sensitization takes place for 5 min, as does optical sensitization. A 20% gelatin solution is added to the emulsion before it is coated on glass plates. This type of emulsion was used for the early PE-2 plates (now called *PFG-03),* which have grain diameters of about 10 nm. Ultrahigh-resolution emulsions have also been produced in other countries, such as Bulgaria and China, but it is only from Russia that such materials can be obtained today.

The highest resolution commercial holographic materials from Agfa, Ilford, and Kodak, which have grain diameters of 35 to 50 nm, are referred to here as "Western" materials.

Ultrahigh-resolution holographic emulsions are normally processed by solution physical development to create colloidal silver. In this way, high image resolution can be obtained.[2] Unfortunately, such processing also requires emulsions with ultrafine silver halide grains (about 20 nm). The most common procedure used for holograms of the Russian type is based on dilute emulsions processed in solution or semiphysical developers, e.g., the Russian GP-2 developer, in such a way that silver particles of an appropriate size are formed (colloidal silver). However, although this processing technique works extremely well for monochrome recordings, it is less suitable for color holograms. The colloidal silver accreted in the emulsion introduces a light-red or brownish color to the processed emulsion. This, in turn, affects color rendition and must therefore be avoided in holographic color recordings. By means of a special processing chemistry and processing baths it has been possible to obtain high-quality color holograms, as previously reported by Bjelkhagen and Vukičević.[3]

Lippmann Photography. At the end of the last century, Gabriel Lippmann was experimenting with color photography.[4–6] Although his photographic recording procedure, *Lippmann photography* (Fig. 1), shows similarities to holography, it was not very effective for color photography because the technique was complicated and the exposure times were too long for practical use. The difficulty in viewing the images was another contributing factor, in addition to the copying problem, that prevented Lippmann photography from becoming a practical photographic color recording method. Because of the demand for high resolving power, the material had rather low sensitivity. The emulsion coated on Lippmann plates was brought into contact with a highly reflecting surface, mercury, reflecting the light into the emulsion and then interfering with the light coming from the other side of the emulsion. The standing waves of the interfering light produced a very fine fringe pattern throughout the emulsion with a periodic spacing of $\lambda/(2n)$ that had to be recorded (λ is the wavelength of light in air and n is the refractive index of the emulsion). The color information was stored locally in this way. The larger the separation between the fringes, the longer the wavelength of the recorded part of image information.

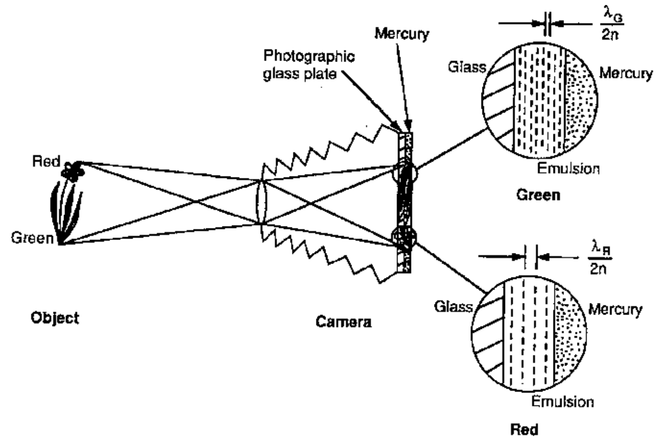

Figure 1. The principle of Lippmann photography.

When the developed photograph was viewed in white light, different parts of the recorded image produced different colors. This difference was due to the separation of the recorded fringes in the emulsion. The light was reflected from the fringes, creating colors corresponding to the original ones that had produced them during the recording. It was obvious that there was a high demand on the resolving power in order to record the fringes separated in the order of half the wavelength of the light. It was also clear that the processing of these plates was critically important, because the separation between the fringes could not be changed or it would create wrong colors. In addition, one had to find ways of obtaining high efficiency.

Gabriel Lippmann was awarded the Nobel prize for his invention in 1908. It was his idea of recording interference fringes throughout the depth of an emulsion that was used by Denisyuk in Russia when he introduced the technique of recording single-beam reflection holograms in the early 1960s.[7–9] Recent years have seen a revival of interest in Lippmann photography among scientists and holographers.[10–17] In particular, a recent paper by Fournier and Burnett[17] describes color rendition and archival properties of Lippmann photographs.

Color Recording in Holography. The first methods for recording color holograms were established in the early 1960s. Leith and Upatnieks proposed multicolor wavefront reconstruction in one of their early papers.[18] Mandel[19] pointed out that it might be possible to record color holograms in a more straightforward way, using a polychromatic laser and an off-axis setup. Lohmann[20] introduced polarization as an extension of the suggested technique. These first methods concerned mainly *transmission holograms* recorded with three different wavelengths from a laser or lasers, combined with different reference directions to avoid crosstalk. The color hologram was then reconstructed, using the original laser wavelengths from the corresponding reference directions. Color holograms of a reasonably high quality could be made this way, but the complicated and expensive reconstruction setup prevented this technique from becoming popular. The first transmission color hologram was made by Pennington and Lin.[21] The authors used the 15-μm-thick Kodak 649-F emulsion with a spectral bandwidth of about 10 nm. This narrow bandwidth eliminated, in principle, crosstalk between the two colors (633 and 488 nm) when reconstructed.

The Lippmann color technique is very interesting for color recordings using *reflection holograms.* Lin et al.[22] made the first color reflection hologram that could be reconstructed in white light. They recorded a reflection hologram of a color slide illuminated with two wavelengths (633 and 488 nm). The material used here was the Kodak 649-F plate, which was processed without fixing in order to avoid shrinkage. This technique, using reflection holography and the white-light reconstruction technique, seems to be the most promising one with regard to the actual *recording* of color holograms and will be discussed further later. However, the three-beam transmission technique might eventually become equally applicable if inexpensive multicolor semiconductor lasers appear on the market in the future.

Relatively few improvements in color holography were made during the sixties and seventies, although several papers on color holography were published.[23–41] During the following decade and until the present, several new, improved techniques have been introduced.[42–117] A review of various transmission and reflection techniques for color holography can be found in a publication by Hariharan.[51] Concerning reflection color holography, an extensive contribution was made by Hubel and Solymar.[91]

Color Reflection Holography. Color reflection holography presents no problems in terms of the geometry of the recording setup, but the final result is highly dependent on the recording material used and the processing techniques applied. The single-beam Denisyuk recording scheme has produced the best results so far. Color holograms have been recorded in single-layer silver halide emulsions,[24,25,31,34,83,84,88,90,100,115–117] or in two separate silver halide emulsions in a sandwich.[39,42,43,46,55,57,58,70,77,91] Pure dichromated gelatin emulsions,[41,79,97] or a DCG emulsion in combination with a silver halide emulsion in a sandwich[61,92,102] have also been used. Even three-layer emulsions for color holography have been proposed and manufactured.[86] Photopolymer recording materials for color holography[32,75,84,85,96,99,101,104,114] have been used experimentally as well.

At least four fundamental problems are associated with the recording of color reflection holograms in silver halide emulsions, which are the most convenient materials for large-format color holograms because of their high sensitivity:

- Scattering that occurs in the blue part of the spectrum found in Western silver halide emulsions makes them rather unsuitable for the recording of color holograms. Therefore, in the most successful color holograms, the blue part has so far always been recorded in dichromated gelatin.[41,61]
- Multiple exposures of a single emulsion reduce the diffraction efficiency of each individual recording.[54,118] The diffraction efficiency of a multiply exposed single-layer emulsion varies inversely as the square of the total number of recordings.
- During processing, emulsion shrinkage frequently occurs, causing a wavelength shift. White-light-illuminated reflection holograms normally show an increased bandwidth upon reconstruction, thus affecting the color rendition.[78]
- The fourth problem, related to some extent to the recording material itself, is the selection of appropriate laser wavelengths and their combination in order to obtain the best possible color rendition of the object.

Some of the problems mentioned above have been discussed in the paper by Lin and LoBianco.[29] Noguchi[34] tried to make a quantitative colorimetric comparison between the recorded color holograms and the colors of the test targets. He used the standard 649-F emulsion and four primary wavelengths for the recording. However, reproduction of the blue colors caused Noguchi some problems. Kubota and Ose[41] demonstrated that a good color reflection hologram could be recorded in a dichromated gelatin emulsion, which gives high efficiency and low-noise blue reconstruction. Hariharan[42] introduced the sandwich recording technique to improve image luminance as compared with that of the earlier triple-exposed 649-F emulsions. He used Agfa 8E75 emulsion for the red (633-nm) recording and the 8E56 emulsion for the green (515-nm) and blue (488-nm) recordings.

The sandwich technique has been further used by Sobolev and Serov,[43] Smaev et al.,[57] and Sainov et al.[58] for the recording of color holograms. The most successful sandwich recording technique has been demonstrated by Kubota,[61] who used a dichromated gelatin plate for the green (515-nm) and blue (488-nm) components, and an Agfa 8E75 plate for the red (633-nm) component of the image. Because the DCG plate is completely transparent in red light, the silver halide plate (containing the red image) is mounted behind the DCG plate in relation to the observer.

Hubel and Ward[70] have been using Ilford silver halide materials for the recording of color reflection holograms. The sandwich technique (SP 672T for blue and green and SP 673T for red) was used with recording illuminations at 458, 528, and 647 nm. Although Ilford blue/green material worked better with regard to scattering noise than Agfa 8E56 HD material, the holograms produced on Ilford materials suffer from the blue recording scattering noise. The color rendition, however, is good, and Hubel and Ward have achieved, so far, the best results with the Western commercial silver halide materials. Hubel[90] reported that he had successfully recorded a color hologram in a single-layer panchromatic emulsion specially prepared for him by Ilford.

Laser Wavelengths for Color Holograms. The problem of choosing optimal primary laser wavelengths for color holography is illustrated in the 1931 CIE (Commission Internationale de l'Eclairage) chromaticity diagram (Fig. 2) and the 1976 CIE version (Fig. 3) with indication of suitable laser wavelengths for color holograms. These diagrams are useful for predicting the colors that can be matched by additive mixing of a set of primary laser wavelengths. After the three primary spectral colors have been selected, a triangle is made by joining the three points corresponding to the spectral colors of the diagram. The colors within the area covered by the triangle correspond to all the colors that can be produced by appropriate mixing of the chosen spectral colors. It may seem that the main goal of choosing the recording wavelengths for color holograms would be to cover as large an area of the chromaticity diagram as possible. However, many other considerations must be taken into account when choosing the wavelengths for color holograms. One important question is whether *three* wavelengths are really sufficient for color holography.

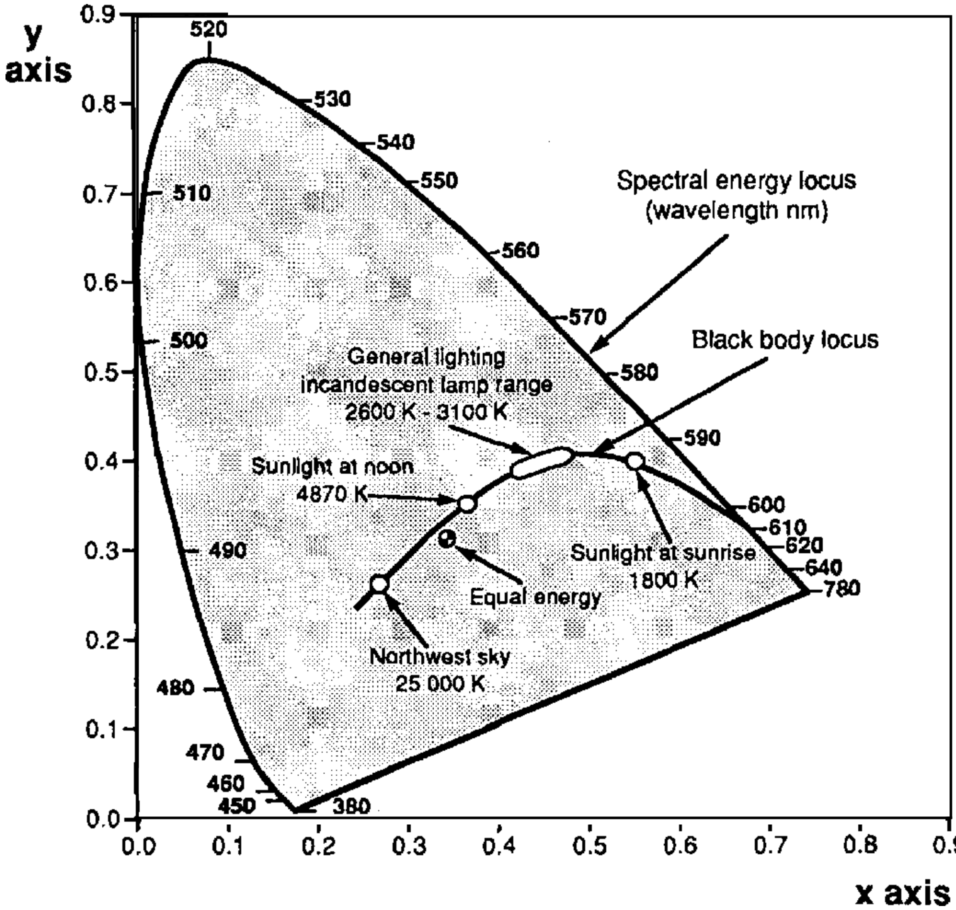

Figure 2. The 1931 CIE chromaticity diagram, illustrating the chromaticities of various light sources.

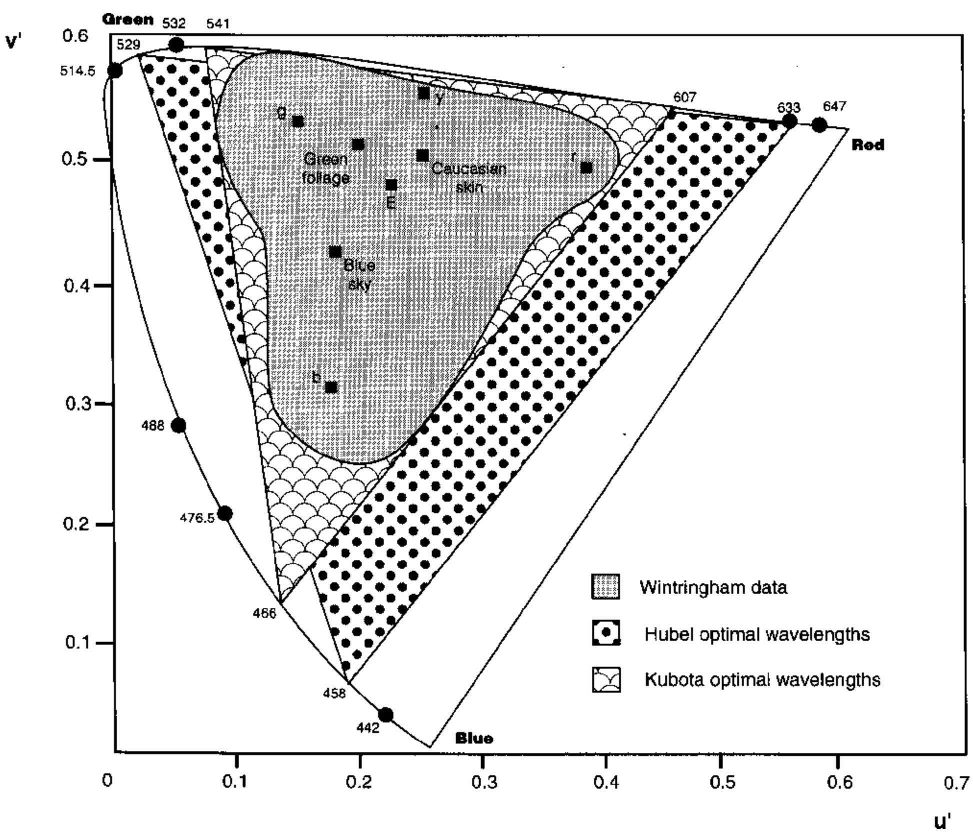

Figure 3. The 1976 CIE uniform scales chromaticity diagram shows the gamut of surface colors and positions of common laser wavelengths. Hubel's and Kubota's three optimal color-recording laser wavelengths are also indicated.

Jeong and Wesly[83,84] gave a heuristic definition of a color hologram, which relates directly to the colors obtained through the laser recording process: “A hologram is said to have true color, if it recreates an image which has the same combination of wavelengths and their relative intensities as those laser wavelengths detected from the object during recording.”

Hubel and Solymar[91] gave a more quantitative and exact definition: “A holographic technique is said to reproduce ‘true’ colors if the average vector length of a standard set of colored surfaces is less than 0.015 chromaticity coordinate units, and the gamut area obtained by these surfaces is within 40% of the reference gamut. Average vector length and gamut area should both be computed using a suitable white light standard reference illuminant.”

The wavelength selection problem for color holography has been treated by Buimistryuk and Dmitriev,[45] Bazargan,[59,93] Kubota and Nishimura,[80,81] Kubota,[92] Hubel,[90] Hubel and Solymar,[91] and Peercy and Hesselink.[107,108]

Bazargan’s[59,93] discussion is based on Wintringham’s gamut of surface colors.[119] Surface colors refer to colors of natural and man-made objects. Normally, these colors are of low saturation. Such colors are also found in most of the objects considered for display color holography. Pointer[120] has extended the gamut of surface colors to include some of the highly saturated fabric dyes that were introduced after Wintringham’s publication. So far, the recording wavelengths for color holography have usually been located at 476.5, 514.5, and 632.8 nm. These wavelengths cover the Wintringham data sufficiently well. A factor to bear in mind when working with silver halide materials is that a slightly longer blue wavelength might give higher quality holograms because of reduced Rayleigh scattering during the recording. However, if we disregard the scattering problem, Hubel[90] and Hubel and Solymar[91] have made an important observation, explaining that, for example, only half of the relative efficiency is needed if a short, 450-nm wavelength is used as the blue primary (as compared with the 480-nm wavelength) to obtain good white balance with the other primary wavelengths. This observation is very important for multiple exposures of holographic single-layer silver halide emulsions. Using a short blue wavelength, the diffraction efficiency of this recording can be rather low, compared with that of the red and green recordings, and still produce a good color hologram. What is needed here is a low-scattering recording material that will make it possible to use the optimal short blue wavelength.

Another important factor to consider is the reflectivity of the object at primary spectral wavelengths. Thornton[121] has shown that the reflectivity of an object at three wavelength bands, peaked at 450, 540, and 610 nm, has an important bearing on color reconstruction. These wavelengths can also be considered optimal for the recording of color holograms and are in good agreement with investigations performed in Russia.[45] Using these wavelengths, a better rendition of the yellow part of the spectrum is obtained. The popular set of wavelengths (476.5, 514.5 and 632.8 nm), although covering the surface colors defined by Wintringham, often causes a distortion in the yellow colors so that they are reconstructed as brown or pink.[49]

The luminosity of the color image is affected by the drop in luminous efficiency at very short or very long recording wavelengths. This important factor has been pointed out by Bazargan[49] and should be remembered when choosing the wavelengths. This is also one of the reasons why trying to cover the largest possible triangle may not be the best procedure. Hubel[68] does not agree with this view and has argued that a triangle larger than is normally used is necessary to compensate for the *color desaturation* (color shifting toward white) that takes place when reconstructing color reflection holograms in white light. Hubel and Ward[70] used the 528-nm wavelength (instead of the 514.5-nm wavelength of the argon laser) combined with 458 and 647 nm, which was the main reason why their holograms gave good yellow rendition. Hubel[90] and Hubel and Solymar[91] continued the investigation of the optimal wavelengths, both theoretically and experimentally. According to their color rendering analysis, these wavelengths are 464, 527, and 606 nm for the sandwich silver halide recording technique. If the calculations are performed to maximize the gamut area instead, the following combination of wavelengths is obtained: 456, 532, and 624 nm. Hubel suggests that 458, 529, and 633 nm is the optimal wavelength combination for practical color holography recordings.

Bazargan[59,93] found the ideal wavelengths to be 450, 540, and 610 nm. Kubota and Nishimura[80,81] approached the wavelength problem from a slightly different angle. These authors calculated the optimal trio of wavelengths based on the reconstructing light source of 3400 K, a 6-μm-thick emulsion with a refractive index of 1.63 and an angle of 30° between the object and the reference beam. Kubota and Nishimura obtained the following wavelengths: 466.0, 540.9, and 606.6 nm. These wavelengths have accurately reproduced the colors of the Macbeth Color Checker chart[122] in a hologram, according to the paper.

Peercy and Hesselink[107,108] discussed wavelength selection by investigating the sampling nature of the holographic process. During the recording of a color hologram, the chosen wavelengths sample the surface-reflectance functions of the object. This sampling on color perception can be investigated by the tristimulus value of points in the reconstructed hologram, which is mathematically equivalent to integral approximations for the tristimulus integrals. They used both Gaussian quadrature and Riemann summation for the approximation of the tristimulus integrals. In the first case they found the wavelengths to be 437, 547, and 665 nm. In the second case the wavelengths are 475, 550, and 625 nm. According to Peercy and Hesselink, the sampling approach indicates that three monochromatic sources are almost always insufficient to

TABLE I. Wavelengths from CW Lasers

Wavelength (nm)	Laser type	Single line power (mW)
442	Helium cadmium	<100
458	Argon ion	<500
468	Krypton ion	<250
476	Krypton ion	<500
477	Argon ion	<500
488	Argon ion	<2000
497	Argon ion	<500
502	Argon ion	<400
514	Argon ion	<5000
521	Krypton ion	<100
529	Argon ion	<600
531	Krypton ion	<250
532	Nd:YAG (frequency doubled)	<400
543	Green neon	<10
568	Krypton ion	<100
633	Helium neon	<80
647	Krypton ion	<2000

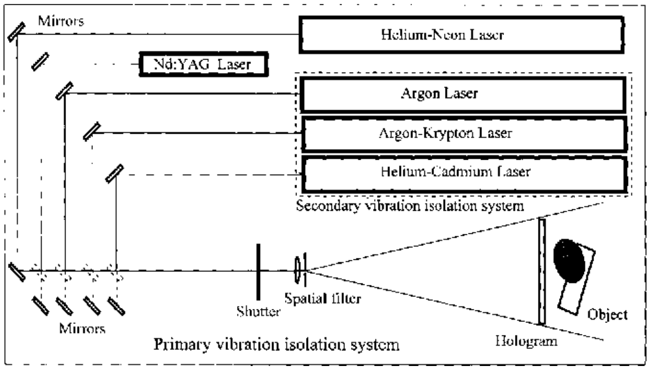

Figure 4. A setup for the recording of color holograms (Lippmann holograms). The most promising recording technique for color reflection holograms is the single-beam Denisyuk setup, which can provide color holograms with a very large field of view and full parallax.

preserve all of the object's spectral information accurately. They claim that four or five laser wavelengths may be required.

Only further experiments will establish how many wavelengths are necessary and which combination is the best for practical purposes. Another factor that may influence the choice of the recording wavelengths is the availability of wavelengths in different cw lasers currently in use in holographic recordings: argon ion, krypton ion, diode-pumped frequency-doubled Nd:YAG, helium–neon, and helium–cadmium lasers (Table I).

Lippmann Holography. In the early days of holography, the term *Lippmann hologram* was used with reference to white-light-viewable monochrome reflection holograms recorded with a single laser wavelength. We suggest that the term *Lippmann hologram* should be used only for a color reflection hologram. The method of recording any type of color reflection holograms (Denisyuk or image-plane) can therefore be referred to as Lippmann holography in analogy with Lippmann photography.

A typical reflection color recording setup is illustrated in Fig. 4. The different laser beams necessary for the exposure of the object pass through the same beam expander and spatial filter. A single-beam Denisyuk arrangement is used, i.e., the object is illuminated through a holographic plate. The light reflected from the object constitutes the object beam of the hologram. The reference beam is formed by the expanded laser light used to illuminate the holographic plate as well as the object itself. Each one of the three primary laser wavelengths forms its individual interference pattern in the emulsion, which is recorded during the exposure. Thus, three holographic images (a red, a green, and a blue image) are superimposed in the emulsion.

The Denisyuk setup is the most demanding one with regard to the material's resolving power and scattering. Only materials with the lowest possible scattering in the blue part of the spectrum can be considered. Traditionally, only dichromated gelatin and photopolymers have low blue scattering, which is why they have been regarded as most suitable for color holography. Western silver halide materials for holography have never been intended for color holography and only ultrahigh-resolution silver halide materials can be used in this area. Earlier experiments with Russian emulsions for color holography have been reported by Watson[74] and Ross and Watson.[100] Sainov[48] described the use of Bulgarian materials for color holograms.

Concerning color work on Western commercial materials, the work by Hubel and Ward[70] shows progress. Here holographic silver halide materials from Ilford were used. In a later work, Hubel and Ward[77] compared different silver halide materials produced by Western companies as potential recording materials for color holography.

Hubel[78] has treated the problem of emulsion shrinkage and the resulting wavelength shift, as well as the desaturation problems that make holographic color reproduction difficult. He introduced a simple model to simulate bandwidth variations between the recording phase and the white-light reconstruction. The white-light reconstruction of a color hologram shows a decreased signal-to-noise ratio and an increased bandwidth, compared with the wavelengths used at the recordings. Hubel shows that his theoretical model fits in with his experimental results and concludes that desaturation is caused primarily by noise but partly by the increased bandwidth. The model shows that desaturation is so large that it remains as one of the main problems in the reproduction of color holographic images in silver halide materials.

Experimental

Color Hologram Recording. After the initial experiments, a special recording setup was arranged in order to produce large-format color holograms (Fig. 4.) It is also used for the investigation and development of color holography in general. For regular recording of color holograms with laser wavelengths of 477, 532, and 633 nm, the blue light is obtained from an etalon-equipped mixed gas laser (Kr–Ar–ion laser), the green light from a cw diode-pumped frequency-doubled Nd:YAG laser, and the red light from a HeNe laser in which an etalon has also been installed.

To record holograms with deep scenes, long coherence is absolutely required from all wavelengths. Furthermore, severe problems in mode hopping were experienced, early in our experiment, in the HeNe laser due to the lack of temperature control in its etalon. The usual way to ascertain single-mode operation of a laser is to monitor its output with a scanning Fabry–Perot interferometer. This instrument requires a specified set of mirrors for specific wavelengths. To be certain that each wavelength is operating in a single axial mode at a constant power output during each exposure, a novel scheme was devised, as shown in Fig. 4. The outputs from each of the three lasers are directed by two sets of mirrors so that each beam can be diverged by a single spatial filter. The alignment process requires that the 633-nm light from the HeNe laser be adjusted first. The 532-nm output from the Nd:YAG is then aligned, followed by the 477-nm light from the argon–krypton laser. In making the hologram, blue light is exposed first, followed by the green and then the red, by sequentially removing the respective mirrors as shown.

To monitor constantly both the output and the temporal coherence of all the light, a glass wedge is placed before the shutter, at a small angle of incidence. Thus each of the two surfaces on the wedge reflects about 5 to 6% of the incident light. One of these reflected beams is used for power monitoring; the other is reflected by a mirror into a classical Michelson interferometer. The two mirrors of the interferometer are adjusted to have an optical path difference of approximately 1 m. The recombined beam at the beamsplitter is directed to a first-surfaced concave mirror, which directs the spread-out interference pattern onto a screen. A video camera relays the interference pattern to a television monitor situated near the site of the hologram being exposed. Exposures are made while observing the contrast of the interference pattern, as well as the power output of whichever wavelength is being used. This monitoring system allowed us to pinpoint the chief cause for mode hopping in the argon–krypton laser as mechanical vibration due to the moving cooling water. This problem was solved by mounting this laser on an independent vibration-isolation system.

Previously, the 488-nm wavelength was selected when recording our first color holograms, as reported earlier.[3] This choice was based on a desire to obtain the highest possible signal-to-noise ratio, considering scattering during blue recording. However, for blue there are several possible wavelengths: 442-nm from the HeCd laser and 458, 476, or 488 nm obtained from the argon–ion laser. However, recent experiments with the 442-nm wavelength from the HeCd laser have shown equally high-quality color recordings. Employing the 442-nm wavelength, better color rendering, as expected, was obtained in the deep blue, violet, and purple parts of the spectrum.

In our first investigation,[3] we found that the green wavelength, 532 nm, obtained from a cw frequency-doubled diode-pumped Nd:YAG laser was suitable as the green primary wavelength. As to the red wavelength, there is a choice between the 633 nm of the HeNe and the 647 nm of the krypton-ion laser.

A specially designed test object including the 1931 CIE chromaticity diagram, a rainbow ribbon cable, pure yellow dots, and a cloisonné elephant was used for the experiments and is shown in Fig. 5. To simulate any color filtering effect caused by the dyes in the holographic emulsion, the test object illuminated with a halogen spotlight was photographed (using Kodak Royal Gold film) through an unexposed holographic plate positioned in the same way as the plates were for recording the Denisyuk color holograms.

The holographic plates (PFG-03C) are produced by the Slavich photographic company outside Moscow.[123] The sizes used in our laboratory range from 5 × 5 cm up to 30 × 40 cm. Slavich can coat 60 × 80-cm glass plates, which represent the largest format. Because there are great variations from batch to batch of this material it is rather difficult to make a more detailed investigation of the emulsion itself at this time. However, the silver halide grain size is the most important parameter of this material and is the main reason for the quality of the holographic images obtained. Some characteristics of the Slavich material are presented in Table II. A recent investigation of this new color material was presented by Markov.[117]

The actual sensitivity values for blue, green, and red were found experimentally. The exposure of the color holograms was performed *sequentially,* starting with the blue recording. Concerning the decrease in diffraction efficiency of the reconstructed images caused by multiple exposures, the publication by Johnson et al.[124] can be considered. Here the degree of decrease in diffraction efficiency is attributed to the time delay between the exposures and is directly related to the lifetime of the individual silver atoms in the latent-image prespecks. Because of the dissociation of single silver atoms, the first exposure produces the strongest effect. Although subsequent exposures reinforce the signal of the previously recorded images, these exposures also increase the noise in later recordings. The blue, which is the most critical recording, is therefore selected to be the first exposure of the emulsion. The second is the green and the last one is the red exposure. The second and third exposures are extended according to the formula given by Johnson et al.[124]

TABLE II. Characteristics of the Slavich Color Emulsion

Silver halide material	PFG-03C
Emulsion thickness	7 µm
Grain size	12–20 nm
Resolution	~10,000 lp/mm
Blue sensitivity	~1.0–1.5 × 10^{-3} J/cm^2
Green sensitivity	~1.2–1.6 × 10^{-3} J/cm^2
Red sensitivity	~0.8–1.2 × 10^{-3} J/cm^2
Color sensitivity peaked at 633 nm and 530 nm	

For example, if the blue exposure is 1, then the green is calculated to be 1.3 and the red 1.7, assuming equal sensitivity of the recording material for all three wavelengths. However, these ratios have been corrected to compensate for variations in the spectral sensitivity of our present emulsion. The corrected factors are blue, 1; green, 0.9; and red, 1.2. The recorded color hologram is considered satisfactory when color rendering in the reconstructed image of the chromaticity diagram is acceptable within the limits of the chosen wavelengths.

The temperature and relative humidity (RH) in the laboratory must be kept stable. A temperature of 20°C and 50% RH should be maintained in the laboratory. If the recording takes place in a warm room with, e.g., high humidity, the emulsion absorbs moisture and will increase in thickness. After processing, emulsion shrinkage will affect the image colors. The unexposed emulsion is very sensitive to humidity changes. During processing the emulsion is hardened, and when the hologram is finished it is much less sensitive to variations in humidity and temperature.

Processing of Color Holograms. The processing of the plates is critical. The emulsion is rather soft, and it is important to harden the emulsion *before* the development and bleaching take place. Emulsion shrinkage and other emulsion distortions caused by the active solutions used for the processing must be avoided. The following bath is used for this first processing step:

Distilled water	750	mL
Formaldehyde 37% (Formalin)	10	mL(10.2 g)
Potassium bromide	2	g
Sodium carbonate (anhydrous)	5	g
Distilled water to make	1	L

The time in this solution is 6 min.

The developer used is the holographic CWC2 developer:

Distilled water	750	mL
Catechol	10	g
Ascorbic acid	5	g
Sodium sulfite (anhydrous)	5	g
Urea	50	g
Sodium carbonate (anhydrous)	30	g
Distilled water to make	1	L

The developing time is 3 min at 20°C.

The catechol-based CW-C2 developer[125] has become one of the most successful developers for the processing of the cw-laser-exposed monochrome reflection holograms. The use of urea serves to increase the developer's penetration into the emulsion, which is important for uniform development of the recorded layers within the emulsion depth. Catechol also has a tanning effect on the emulsion, but with low staining effect compared with pyrogallol. Therefore, this developer can be considered suitable for processing color holograms.

The bleach bath used to convert the developed silver hologram into a phase hologram is very critical. The bleach must create an almost stain-free clear emulsion in order not to affect the color image. In addition, no emulsion shrinkage can be accepted, because it would change the colors of the image. A special rehalogenating bleach for holography used here is based on the idea of mixing a bleach by using an oxidation process between persulfate and a common developing agent, e.g., ascorbic acid, amidol, metol, and hydroquinone. New rehalogenating bleach baths for holography were previously introduced by Bjelkhagen et al.[126] These baths have very good performance in terms of high efficiency and low noise, and some of them introduce no emulsion shrinkage.

These bleaches have been named PBU (Phillips–Bjelkhagen Ultimate) bleaches followed by the name of the developing agent on which they are based. The PBU-amidol bleach, which was selected for the color processing, is mixed as follows:

Distilled water	750	mL
Cupric bromide	1	g
Potassium persulfate	10	g
Citric acid	50	g
Potassium bromide	20	g
Distilled water to make	1	L

After these chemicals have been mixed, we add 1 g amidol [$(NH_2)_2C_6H_3OH \cdot 2HCl$, 2,4-diaminophenol dihydrochlo-ride]. The bleach can be used within a few minutes of being mixed. The developing agent amidol must be allowed to oxidize enough. Dilute one part stock solution with two parts distilled water for use. The bleaching time is normally about 5 min. The process must continue until the plate is completely clear. After the bleach step is finished, the plate is washed for at least 10 min and then soaked in water to which 20 mL/L glacial acetic acid has been added. This is done to prevent printout of the finished hologram. Washing and drying must be performed so that no shrinkage occurs. The best way to dry the plates is to let them slowly dry in air at room temperature. Warm air will introduce emulsion shrinkage. The processing steps are summarized in Table III.

Table III. Color Holography Processing Steps

1. Tan in formaldehyde solution	6 min
2. Rinse	5 s
3. Develop in the CW-C2 developer	3 min
4. Wash	5 min
5. Bleach in the PBU-amidol bleach	~5 min
6. Wash	10 min
7. Soak in acetic acid bath	1 min
8. Short rinse	1 min
9. Wash in distilled water with wetting agent added	1 min
10. Air dry the holograms	

Results

Color prints of some of our recorded color holograms are presented in Figs. 6 through 10. The photographs of the reconstructed color holograms were recorded using a halogen spotlight positioned at the correct distance from the

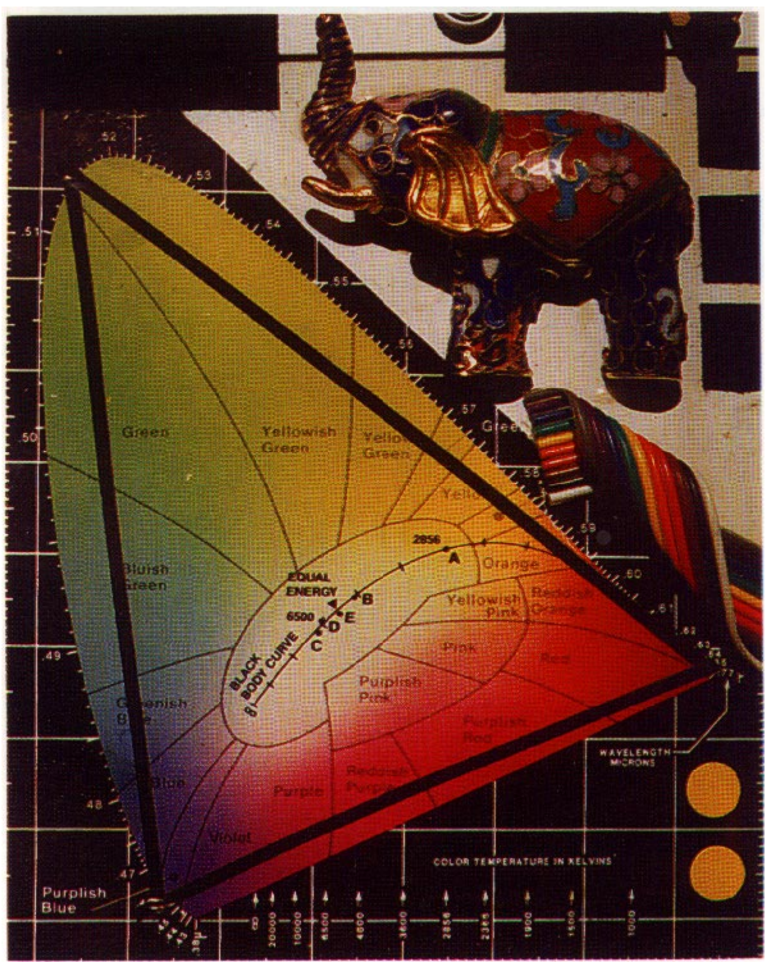

Figure 5. Photographed color test object including the 1931 CIE chromaticity diagram, a rainbow ribbon cable, pure yellow dots, and a cloisonné elephant.

hologram and illuminating the hologram at the correct angle according to the recording geometry. The photograph of the holographic image of the test object in Fig. 6 must be compared with the photograph of the test object in Fig. 5. However, by direct comparison between the photograph of the test object and the object itself, the colors appear slightly different to the eye. In particular, green is too weak in the photograph. The photographic prints were made with a manual filtering process necessary for better photographic color rendering. Nevertheless, there are visible differences between the holograms and photographic prints of holograms. However, because we are concerned with display holograms, here the human eye is the best detector for a qualitative evaluation of the image, which can be directly compared with the object itself under the same illumination.

In Fig. 6, the recording wavelengths were 488, 532, and 633 nm. The main difference in color rendering is that the photograph of the object shows colors with higher saturation than the reconstructed colors of the holographic images. The colors are slightly "washed out" in the holograms, an effect caused by the already explained white-light reconstruction desaturation combined with some low-level scattering noise. Color rendering is otherwise good. In Figs. 7 and 8, two 10×12-cm color holograms are featured. The first one is a recorded ceramic mask in different colors, mounted on a blue background, and the second one is a color hologram of a polychrome vase.

Color prints of two recent large-format color holograms are presented in Figs. 9 and 10. They were recorded with 477-, 532-, and 633-nm wavelengths. In Fig. 9, a 20×25-cm hologram of a model car is shown. Figure 10 shows a 30×40-cm color hologram of a toy truck.

Discussion and Conclusion

The recording of large-format color holograms in single-layer silver halide emulsions has been demonstrated. In spite of the common opinion that silver halide materials are inferior to grainless recording materials, we found that the performance of the ultrahigh-resolution emulsions was very good. Given the proper choice of the three recording laser lines, good color rendering can be achieved on such materials. For practical color holography, we have found that the following laser wavelengths can be recommended: blue, 477 nm; green, 532 nm; and red, 633 nm. However, depending on the emulsion's grain-size variation from batch to batch, the 488-nm blue wavelength may produce a higher contrast color hologram. The processing of such holograms requires special attention to avoid shrinkage or other emulsion distortions. The diffraction efficiency of color holograms produced this way is about 25 to 30% but can be increased. Although good color rendition can be obtained, problems connected with color desaturation still remain to be solved. The development process can be further improved to avoid the desaturation caused by nonuniform development. Other limitations concerning the recording of colors in a hologram include the fact that

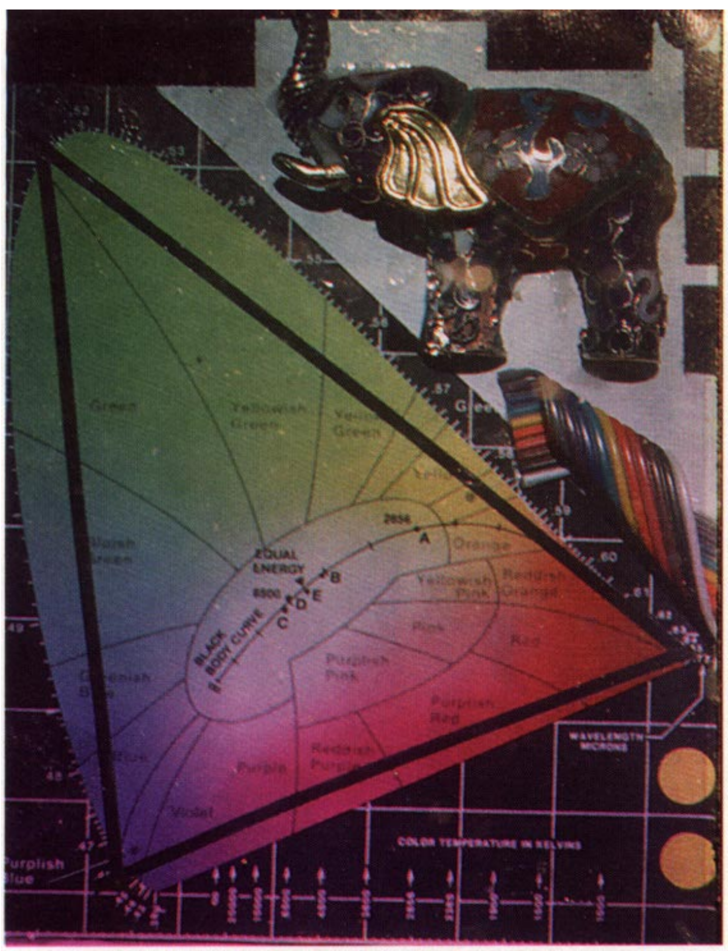

Figure 6. A 10×12-cm color hologram of the test object. The hologram was recorded with 488-, 532-, and 633-nm laser wavelengths. Note the rainbow ribbon cable, the pure yellow dots, the full range of colors, and a balanced white in the center of the CIE color test target.

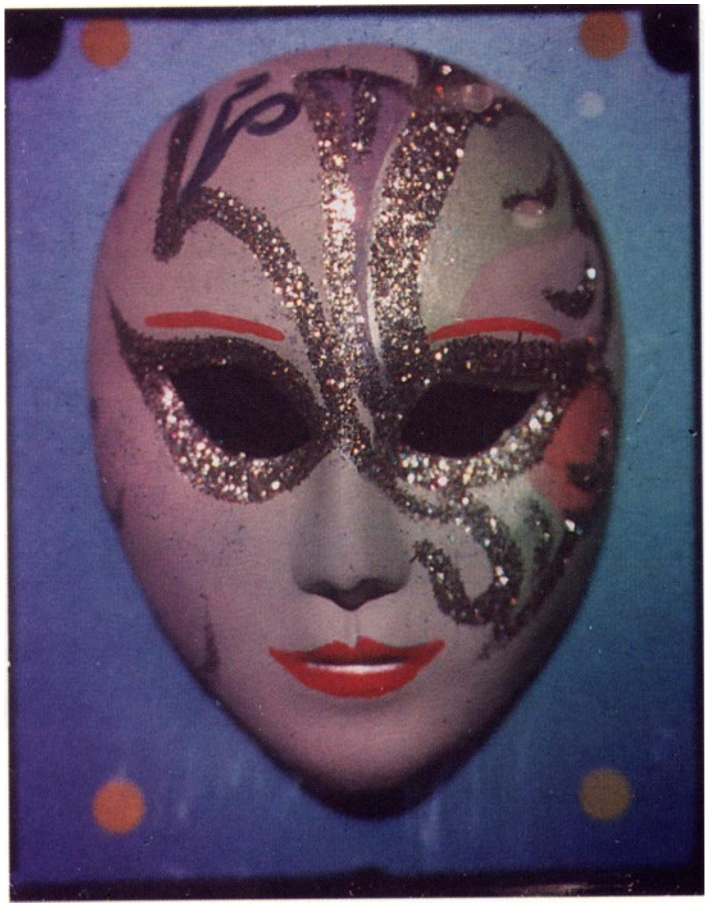

Figure 7. A 10 × 12-cm color hologram of a ceramic mask in different colors, mounted on a blue background. The hologram was recorded in the same way as the test object.

Figure 8. A 10 × 12-cm color hologram of a polychrome vase. The hologram was recorded with 477-, 532-, and 633-nm (blue, green, and red) laser wavelengths.

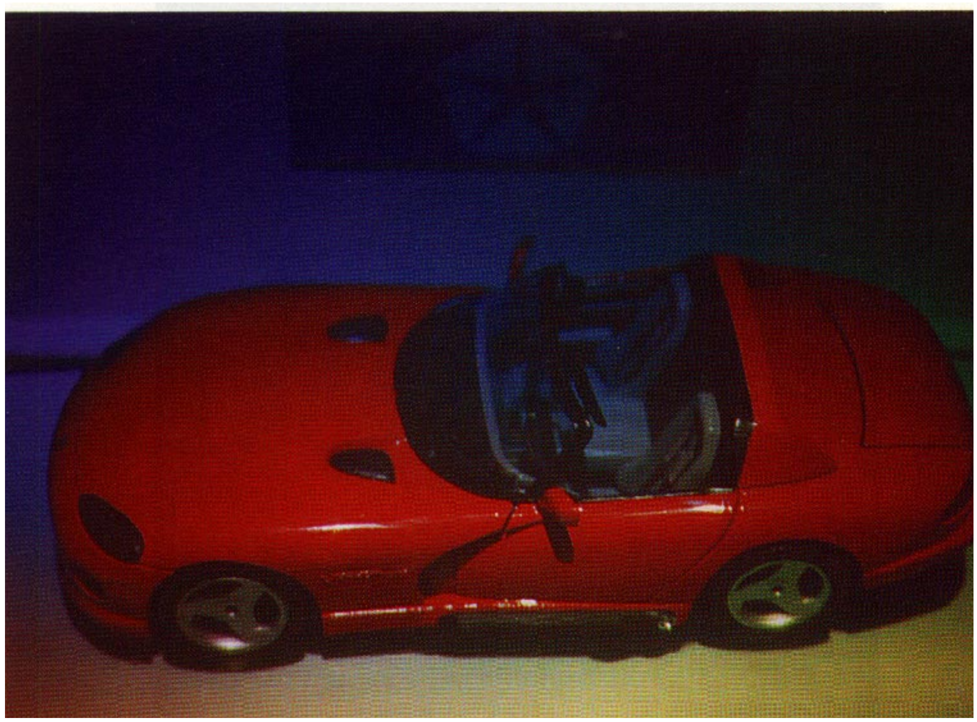

Figure 9. A 20 × 25-cm color hologram of a model car. The hologram was recorded with 477-, 532-, and 633-nm (blue, green, and red) laser wavelengths.

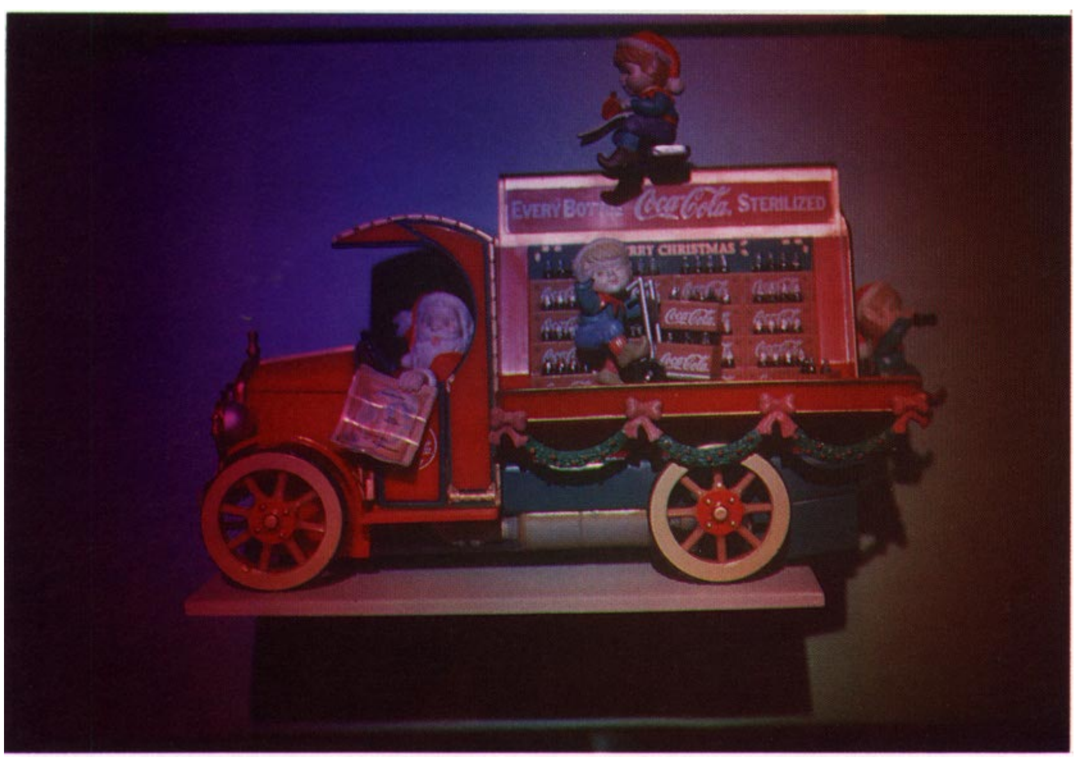

Figure 10. A 30 × 40-cm color hologram of a toy truck. The hologram was recorded with 477-, 532-, and 633-nm (blue, green, and red) laser wavelengths.

some of the colors we see are the result of fluorescence, which cannot be recorded in a hologram. At the present time, the research on processing color reflection holograms recorded on ultrahigh-resolution materials is still in progress. We are working on techniques that could completely eliminate shrinkage, as well as increase diffraction efficiency, by considering silver halide sensitized gelatin (SHSG) processing, a method for converting the silver halide hologram into a dichromated gelatin-type hologram. For best results SHSG processing requires emulsions with very fine silver halide grains in order to produce low-noise recordings.

In addition, more accurate color rendering analysis of our holographic recordings is planned, using, e.g., the Macbeth Color Checker chart and colorimetric evaluation, as well as scattering noise measurements.

An alternative to silver halide material for color holograms is the holographic photopolymer from E. I. du Pont de Nemours & Co. (OmniDex®). The monochrome materials are commercially available, but the panchromatic materials are still in the development phase. Eventually, photopolymer film can become a very suitable recording material for mass replication by contact-copying color holograms and color HOEs from silver halide masters.

Acknowledgments. The initial work was performed during two of the authors' (Bjelkhagen and Vukičević) visiting professorships at the University of Münster, Germany. The authors wish to thank M. Hoke, director of the Institute of Experimental Audiology, and G. von Bally, the head of the Biophysics Laboratory, for supporting the project. The authors thank G. Sobolev, MIREA, Moscow, for providing us with the first experimental PFG color emulsion. The color recording setup was arranged at ENSPS, Université Louis Pasteur, Strasbourg, France, supported by P. Meyrueis. Adlas GmbH is acknowledged for providing us with the cw frequency-doubled Nd:YAG laser.

The most recent work was performed at Lake Forest College by Bjelkhagen and Jeong. Coherent, Inc., is acknowledged for providing us with a cw diode-pumped frequency-doubled Nd:YAG laser (DPSS 532). A helium–cadmium laser by Kimmon (IK Series) was provided by ACI Systems, Inc.; and ATx Telecom Systems, Inc. (formerly Amoco Laser Company) provided a 50-mW frequency-doubled Nd:YAG laser, for which the authors are grateful.

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