Electronic Supplementary Information

Rewritable full-color computer-generated holograms based on color-selective diffractive optical components including phase-change materials

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1. Derivation of the mathematical expression for first-order diffraction efficiency of one-dimensional binary diffraction grating

Here, we derive the mathematical expression for the first-order diffraction efficiency of the proposed diffractive optical component. In general, the reflection-type one-dimensional binary grating can be illustrated as the schematic shown in Fig. S1.



Figure S1. Schematic of the one-dimensional binary diffraction grating.

Here, r_1 and r_2 represent the complex reflection coefficients of two types of pixels, whose magnitudes are less than or equal to 1. The pixel pitch and period of the grating correspond to Δp and $2\Delta p$, respectively. The magnitudes of the diffraction orders can be found by the plane-wave decomposition of the diffracted field, which can be obtained from the Fourier transform of the optical response u(x) of the grating, as follows:

$$FT\left[u\left(x\right)\right] = FT\left[\left\{\sum_{m}\delta\left(x-m\cdot2\Delta p\right)\right\}*\left\{r_{1}\cdot\operatorname{rect}\left(\frac{x-\Delta p/2}{\Delta p}\right)+r_{2}\cdot\operatorname{rect}\left(\frac{x-3\Delta p/2}{\Delta p}\right)\right\}\right]$$
$$= FT\left[\sum_{m}\delta\left(x-m\cdot2\Delta p\right)\right]\cdot FT\left[r_{1}\cdot\operatorname{rect}\left(\frac{x-\Delta p/2}{\Delta p}\right)+r_{2}\cdot\operatorname{rect}\left(\frac{x-3\Delta p/2}{\Delta p}\right)\right]$$
$$= \left\{\frac{1}{2\Delta p}\sum_{m}\delta\left(k-\frac{m\pi}{\Delta p}\right)\right\}\cdot\left\{\int_{0}^{\Delta p}r_{1}\exp\left(-ikx\right)dx+\int_{\Delta p}^{2\Delta p}r_{2}\exp\left(-ikx\right)dx\right\}$$
$$= \left\{\frac{1}{2\Delta p}\sum_{m}\delta\left(k-\frac{m\pi}{\Delta p}\right)\right\}\cdot\left[\Delta p\cdot\operatorname{sinc}\left(\frac{k\Delta p}{2}\right)\cdot\left\{r_{1}\cdot\exp\left(-ik\frac{\Delta p}{2}\right)+r_{2}\cdot\exp\left(-ik\frac{3\Delta p}{2}\right)\right\}\right],$$
(S1)

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where *m* is an integer representing the diffraction order, *k* denotes the spatial angular frequency along the *x*-axis, the sinc(*x*) function is defined as sinc(x) = sin(x) / x, and the rect(*x*) function is defined as

rect(x) =
$$\begin{cases} 1, & |x| < 1/2 \\ 1/2, & |x| = 1/2 \\ 0. & \text{otherwise} \end{cases}$$
 (S2)

The convolution theorem for Fourier transforms and Fourier series representations of the Dirac comb sequences are used in the derivation process. From Eq. (S1), we can find that the first-order diffraction efficiency $DE_{m=\pm 1}$ is

$$DE_{m=\pm 1} = \frac{1}{\pi^2} \left| r_1 - r_2 \right|^2.$$
(S3)

The spectral diffraction map shown in Fig. 1(b) is the plot of Eq. (S3) using the simulated complex amplitudes of the light reflected from the proposed structure, when the GST layer is in complete amorphous and crystalline states.

2. Numerically simulated spectral diffraction efficiency map of the reflection-type color-selective diffractive optical component comprising SiO₂/GST/SiO₂/TiW stacked layers



Figure S2. (a) Reflection-type color-selective diffractive optical component composed of $SiO_2/GST/SiO_2/TiW$ multilayer stacks, and (b) its spectral map of the first-order diffraction efficiency with respect to the thickness of the lower SiO_2 layer.

3. Discussion on the reconstruction characteristics of spatially multiplexed CGHs

For CGHs having pixelated structures, the maximum diffraction angle, which is directly related to the viewing angle provided by the CGH, can be expressed as¹

$$\theta = \sin^{-1} \left(\frac{\lambda}{2\Delta p} \right), \tag{S4}$$

where λ is the wavelength of incident light and Δp denotes the pixel pitch. The key advantage of the spatial

multiplexing technique² used in our work is that the maximum horizontal diffraction angle can be maintained even though there are three kinds of diffraction areas periodically distributed to respectively diffract the three different wavelengths to reconstruct full-color holographic images. This is because the pitch of a cluster of pixels in each area is still Δp . When considering this, it seems that the performances of the CGHs are independent of the horizontal widths of the vertically striped diffraction areas. However, the width of each segmented diffraction area should be carefully determined because there is a horizontal blur in the reconstructed images, which can be a quality-degradation factor in holographic reconstructions.



Figure S3. (a) Simplified structure of the spatially multiplexed full-color CGH and (b) its equivalent model for monochromatic red light under the perfect color-selective diffraction condition.

This blurring effect in the reconstructed images can be explained using the model shown in Fig. S3(a), which is a simplified version of the spatially multiplexed full-color CGH, where Λ indicates the period of the RGBdiffractive stripes. If we assume that each of the RGB inputs can only be diffracted by its respective diffractive areas with no crosstalk, the periodically aligned vertical stripes of the diffractive regions can be regarded as a slit aperture array from the viewpoint of monochromatic light. Figure S3(b) illustrates the equivalent model when red light is incident on the CGH, in which the corresponding CGH pixels for red light are positioned in designated regions. In this structure, diffraction occurs not only by the CGH but also by the slit aperture array. Accordingly, additional diffraction orders are generated around each original diffraction order produced by the CGH pixel array. This is illustrated in Fig. S3(b) as the incident and reflected rays, where θ_i is the horizontal angle of incidence, $\theta_{r,(m,n)}$ indicates the horizontal angle of reflection, and *m* and *n* represent the diffraction orders associated with the diffraction gratings whose periods are Δp and Λ , respectively. The related grating equation can be formulated as

$$\theta_{r,(m,n)} = \sin^{-1} \left(m \frac{\lambda}{\Delta p} + n \frac{\lambda}{\Lambda} + \sin \theta_i \right).$$
(S5)

We verify this diffraction property through numerical simulations on the basis of the geometry shown in Fig. S3(b). The off-axis setup to generate a CGH is shown in Fig. S4(a). The size of the CGH is $8.192 \times 8.192 \text{ mm}^2$, the vertical off-axis angle is 7.6°, and the object is located at z = -0.1 m. The wavelength and pixel pitch are chosen as $\lambda = 530$ nm (green light) and $\Delta p = 2 \mu m$, respectively. The calculated CGH is masked with two different vertical slit aperture arrays of $\Lambda = 180$ and 60 μm . The parts of the masked CGHs and their holographic reconstructions at z = -0.1 m are shown in Fig. S4(c) and S4(d). For comparison, the corresponding result of the CGH with no masking is shown in Fig. S4(b), which shows a clear reconstruction of the object image. We can see from the results that masked CGHs generate horizontally blurred reconstructed images due to the additional higher-order diffractions from the slit aperture array structure, and the blur effect is more severe when the period of the slit aperture array is smaller. The degree of blur can be calculated using Eq. (S5). For example, for the case of $\Lambda = 60 \ \mu m$, the horizontal distance between the zero-order and first-order images is approximately 0.88 mm, which can be obtained by calculating (0.1 m) $\times \tan \theta_{r,(m,n)}$ with m = 0, n = 1, λ

= 530 nm, $\Delta p = 2 \mu m$, $\Lambda = 60 \mu m$, and $\sin \theta_i = 0$. This result coincides with that from the numerical simulation, as noted in the reconstruction result of Fig. S4(d). Although these extra diffraction orders can negatively influence the image quality, they are acceptable when the degree of blur is much smaller than the size of the reconstructed image. Our full-color CGH is designed in consideration of this characteristic so that the horizontal blur is hardly noticeable, as can be confirmed in the experimentally reconstructed images shown in Fig. 4.



Figure S4. (a) Calculation scheme for the CGHs based on the model shown in Fig. S3(b). (b) Calculated binary CGH without masking and the holographic reconstruction at z = -0.1 m. (c) and (d) show the corresponding results of CGHs masked with slit aperture array structures when $\Lambda = 180$ and 60 µm, respectively.

Figure S5 depicts the horizontal viewing angle provided by the full-color CGH and observed images from three different viewpoints. In these kinds of multi-wavelength color CGHs, it is reasonable to define the viewing angle as twice the maximum diffraction angle of the light having the shortest wavelength among the used wavelengths and can be calculated using Eq. (S4) as

$$\theta_{\rm VA} = 2\sin^{-1} \left(\frac{\lambda_{\rm B}}{2\Delta p} \right)$$

= $2\sin^{-1} \left(\frac{473 \text{ nm}}{2 \cdot 2 \,\mu\text{m}} \right)$ (S6)
= 13.6°,

where λ_B indicates the wavelength of the blue light (473 nm). Thus, depending on the viewing angle, we can determine the size of the observable area at a specific longitudinal distance from the CGH plane. In particular, when there are more than two focal planes in the reconstruction images, the transverse shifts of the comprising images differ according to the observation angles, as illustrated in Fig. S5(b).



Figure S5. Illustrations of (a) horizontal viewing angles supported by the CGH and (b) observed images from three different perspectives.



4. Determination of the laser pulse conditions for writing and erasing operations

Figure S6. Experiments to find appropriate conditions for writing and erasing operations in the proposed multilayer structures having (a) red-diffractive, (b) green-diffractive, and (c) blue-diffractive characteristics. In the three sub-figures, left and right results correspond to writing (95%) and erasing (0%) overlap conditions, and the energy densities are indicated on the relevant areas.

Figure S6 shows the experimental investigations that were performed in order to determine appropriate energy density and overlap conditions of laser pulses for writing and erasing operations. Since the SiO₂ layer of the proposed optical component has three different thicknesses, three kinds of flatly deposited ITO/GST/ITO/SiO₂/TiW multilayer samples were prepared, with common thicknesses for ITO/GST/ITO (20/7/30 nm) and TiW (210 nm) layers, but different SiO₂ thicknesses of 295 nm (red-diffractive), 415 nm (green-diffractive), and 145 nm (blue-diffractive). The left-hand figures in the three sub-figures show the results for different energy densities with the common overlap condition of 95%, in which the target area receives 20 consecutive pulses. We can see that, for the three samples, energy density from 25 to 40 mJ/cm² can be used for the writing operation, that is, a state transition from amorphous to crystalline. The broad energy density range viable for crystallization is due to fact that the GST layer can be transformed into the completely crystallized state as an aggregate result of partial crystallizations, attained by a sufficient number of low-energy optical stimuli. Contrarily, the laser pulses with energy density of 45 mJ/cm² keeps the GST layer in the amorphous state, and those with higher energy density cause the target area to be damaged. The right-hand figures in the three sub-figures show the results for different energy densities with the common overlap condition of 0%, in which the target area receives only one pulse. Here, for clarity, part of the target area was pre-crystallized. The results demonstrate that the proper energy density for the erasing operation lies between 50 and 55 mJ/cm², the same for all three samples. It turns out that the overlap and energy density conditions for the three samples are almost the same, regardless of the SiO₂ layer thickness. From the results, we selected the (95%, 40 mJ/cm²) and (10%, 55 mJ/cm²) conditions for writing and erasing in our full-color diffractive optical component, respectively. Note that for the erasing of the full-color CGH, the overlap condition was increased to 10% to assure no area was excluded from the laser exposure.

References

- 1. J. W. Goodman, Introduction to Fourier Optics, Robert & Company Publisher, Englewood, 2005.
- 2. Y. Tsuchiyama and K. Matsushima, Opt. Express, 2017, 25, 2016–2030.