Supporting Information

Two-dimensional closely-packed gold nanoislands: A platform for optical data storage and carbon dot generation

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1. Transmission spectra of Au films composed of closely-packed Au nanoislands

From the transmission electron microscope (TEM) images of the Au films obtained by using different sputtering times, it is noticed that the size of Au nanoislands becomes larger and the gap between them becomes smaller with increasing sputtering time (see Figure 1). The plasmonic coupling between Au nanoislands needs to be taken into account when Au nanoislands become closely packed. As a result, the resonant wavelength of the plasmonic hot spots, which is determined mainly by the gap between Au nanoislands, is shifted to a longer wavelength. In Figure S1, we show the transmission spectra measured for Au films deposited on SiO_2 substrates by using different sputtering times. For the Au film obtained at t = 10 s, a transmission valley induced mainly by the extinction of isolated Au nanoislands is found at ~600 nm. It becomes deeper when the sputtering time is increased to t = 15 s. For t = 20 s, one can see a large red shift and a further deepening of the transmission valley. For t = 25 s, the transmission valley is further shifted to ~800 nm. However, an increase of the transmission is found because of the reduced absorption of the Au film.

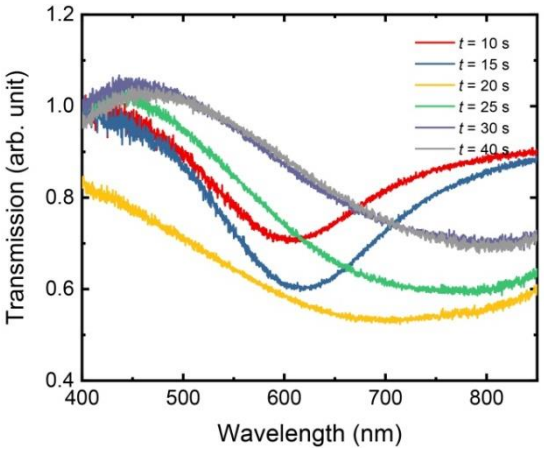

Figure S1. Transmission spectra measured for Au films deposited on SiO_2 substrates with different sputtering times.

2. Transmission spectra of Au films before and after the irradiation of femtosecond laser pulses

In order to gain a deep insight into the change of Au nanoislands induced by femtosecond laser pulses, we chose the Au film obtained with t = 15 s and examined the change of the transmission spectrum before and after the irradiation of femtosecond laser pulses with different fluences, as shown in Figure S2. It can be seen that the transmission at the laser wavelength (800 nm) was increased with increasing laser fluence, implying the elimination of the hot spots with resonant wavelengths at ~800 nm in the Au film. Accordingly, a blueshift of the transmission valley was observed.

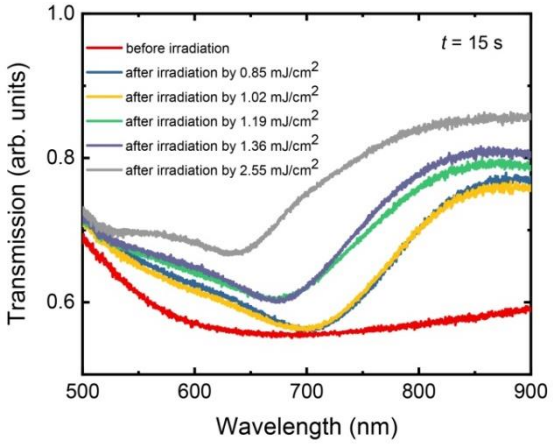

Figure S2. Transmission spectra measured for the Au film with t = 15 s before and after the irradiation of femtosecond laser pulses with different fluences.

3. Correlation coefficient and contrast of an extracted pattern

Basically, we can use correlation coefficient to characterize the quality (or error rate) of a pattern extracted by detecting the hot electron luminescence intensities of all the information units. The extracted pattern can be binarized by choosing appropriate threshold intensity, as shown in Figure S3.

The correlation coefficient between the binarized pattern and the original one is defined as follows:

$$C = \frac{\sum_m \sum_n (A_{mn} - \bar{A})(B_{mn} - \bar{B})}{\sqrt{\left[(A_{mn} - \bar{A})^2\right]\left[(B_{mn} - \bar{B})^2\right]}}, \quad \bar{A} = \frac{\sum_m \sum_n A_{mn}}{m \times n}, \quad \bar{B} = \frac{\sum_m \sum_n B_{mn}}{m \times n}.$$

Here, A_{mn} and B_{mn} represent the intensities of individual information units (m, n) while $\overline{A}$ and $\overline{B}$ denote the averaged intensities of all the information units in the extracted and original patterns, respectively.

For the extracted pattern shown in Figure S3a, the distribution of the luminescence intensities of all the information units is shown in Figure S3d where one can see two Gaussian distributions with different average intensities (I_1 and I_2, $I_1 > I_2$,), corresponding to the information units without (I_1) and with (I_2) the irradiation of femtosecond laser pulses. A threshold intensity I_{th} in between I_1 and I_2 is used to discriminate the two types of information units and to do binarization. The correlation coefficient can be deduced after the binarization of the extracted pattern. In generally, the contrast of the extracted pattern is defined as $R = (I_1-I_2)/(I_1+I_2)$. It influences significantly the correlation coefficient. As far as the recording energy is concerned, the correlation coefficient and contrast of the pattern have to be specified.

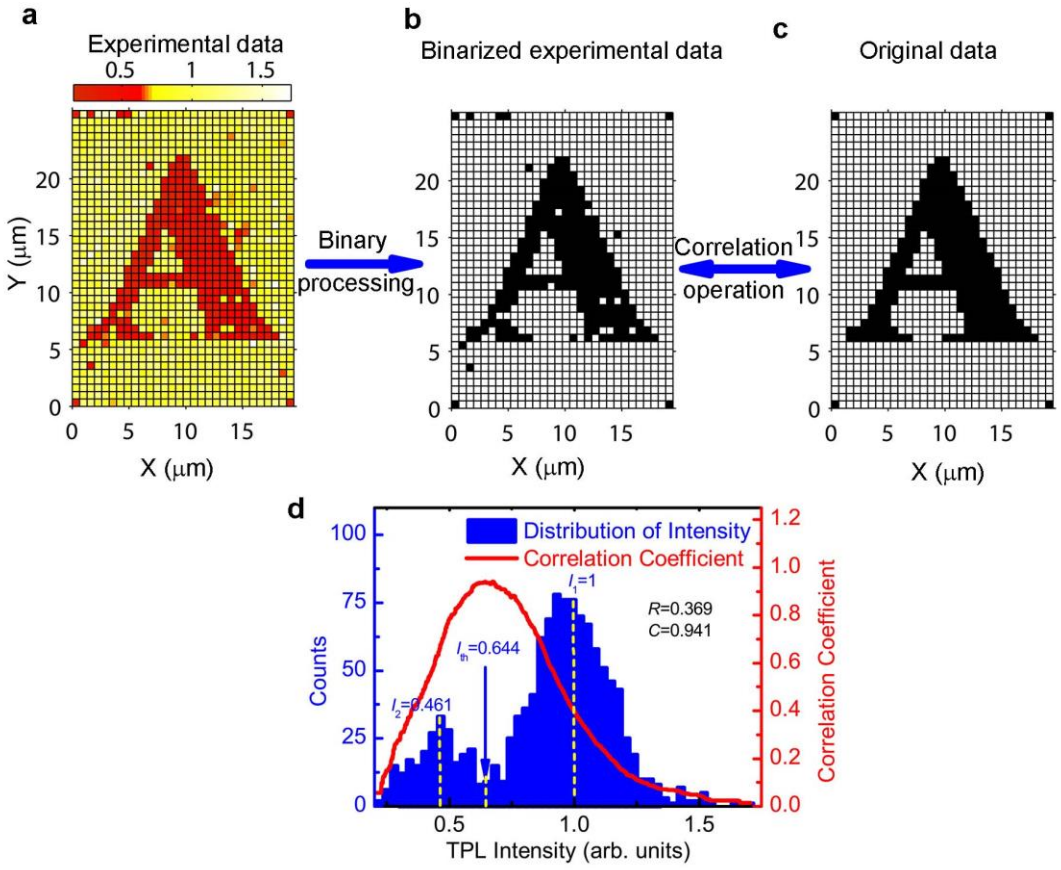

Figure S3. Procedure used to derive the correlation coefficient and contrast of an extracted pattern. a) The pattern obtained by detecting the hot electron luminescence intensities of all the information units. b) The binarized pattern obtained by choosing appropriate threshold intensity. c) The original pattern used for data recording. d) The distribution of the luminescence intensities of all the information units in the extracted pattern. Here, I_1 = 1 and I_2 = 0.461 are the averaged luminescence intensities for the information units without and with the irradiation of femtosecond laser pulses and I_{th} is the threshold intensity used to discriminate the two types of information units (or used for extracting the binarized

pattern). The calculated correlation coefficient (C = 0.941) and contrast (R = 0.369) for the extracted pattern are also provided.

4. Comparison of two optical data storage media

As discussed above, the quality of optical data storage can be evaluated by using the correlation coefficient and contrast calculated for an extracted pattern. In Figure S4a, we present the pattern extracted by detecting the hot electron luminescence of closely-packed Au nanoislands. The laser fluences used for data recording and readout were 0.84 and 0.17 mJ/cm^2, respectively. Based on the binarized pattern shown in Figure S4b, the correlation coefficient and contrast of the extracted pattern are calculated to be 0.93 and 0.42, as shown in Figure S4c. For comparison, we also evaluated the correlation coefficient and contrast of the pattern recorded in a polymer film doped with high-density Au nanorods, as shown in Figure S5a. From the binarized pattern (see Figure S5b), the correlation coefficient and contrast of the pattern are calculated to be 0.92 and 0.29, as shown in Figure S5c. It indicates that the Au film composed of closely-packed Au nanoislands is much better than the polymer film heavily doped with Au nanorods in optical data storage.

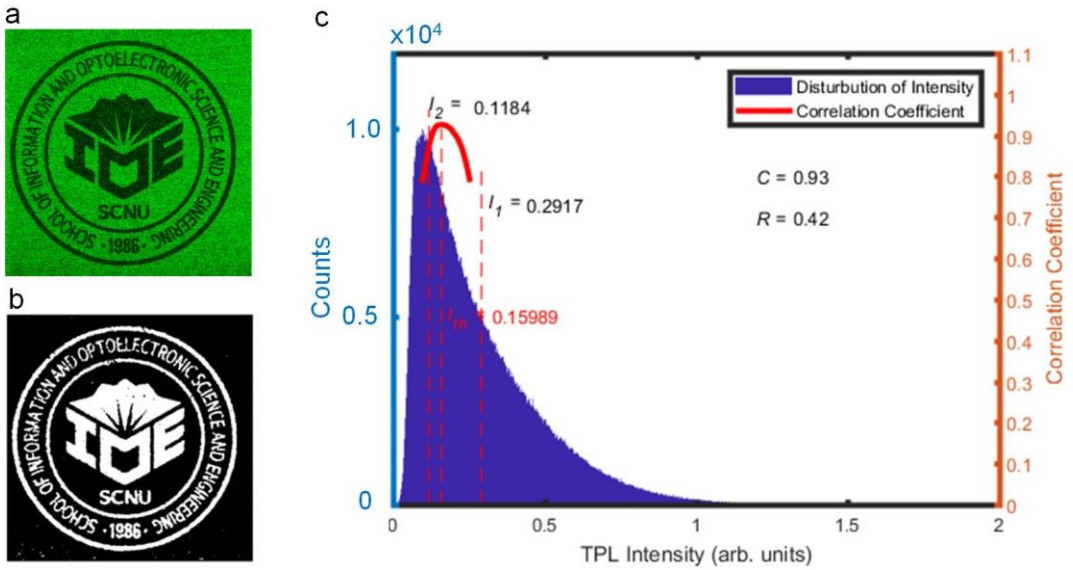

Figure S4. a) Pattern recorded in an Au film composed of closely packed Au nanoislands by detecting hot electron luminescence. b) Binarized pattern of the extracted pattern shown in a). c) Correlation coefficient and contrast of the extracted pattern shown in a).

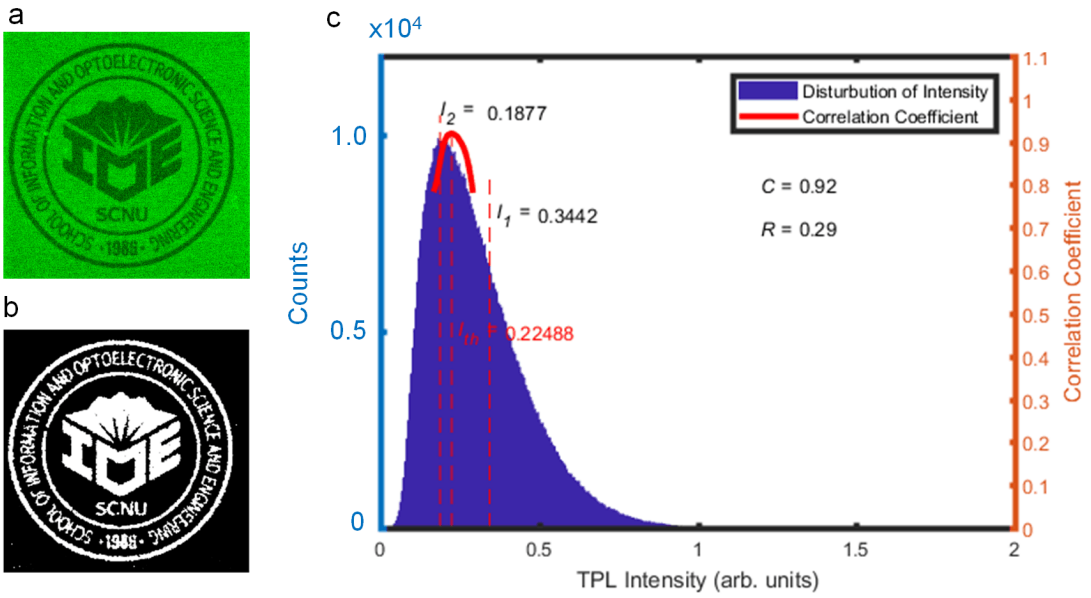

Figure S5. a) Pattern recorded in a polymer film doped with Au nanorods by detecting hot electron luminescence. b) Binarized pattern of the extracted pattern shown in a). c) Correlation coefficient and contrast of the extracted pattern shown in a).

5. Polarization multiplexing with three polarization channels

In Fig. 3, we have demonstrated that optical data storage with negligible cross-talk can be realized in the Au film composed closely-packed Au nanoislands by using femtosecond laser pulses with cross polarizations. In order to find out the maximum polarization channels available in the Au film, we also examined the optical data storage in three polarization channels separated by 60°, as shown in Figure S6. It is noticed that high-quality data storage with correlation coefficients larger than 0.99 and contrasts larger than 0.54 can be achieved in the three polarization channels. It implies that the Au film composed of closely-packed Au nanoislands is comparable to the polymer film doped with Au nanorods in polarization multiplexing.

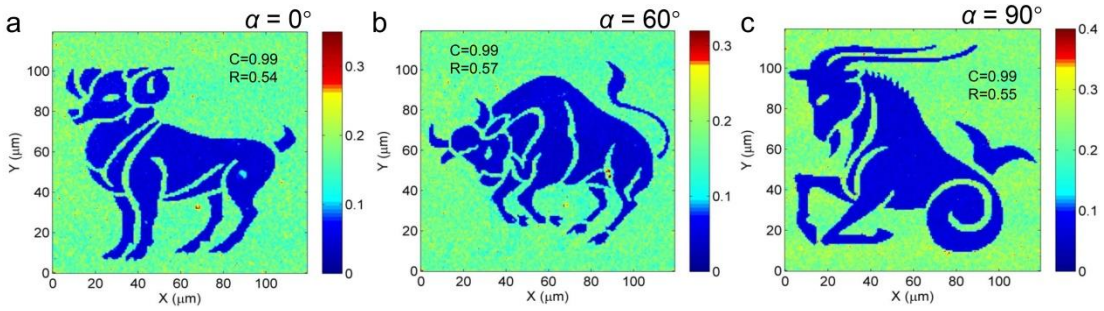

Figure S6. Patterns recorded and extracted in the Au film composed of closely-packed Au nanoislands (t = 25 s) by using linearly polarized femtosecond laser light with polarization angles of a) 0°, b) 60°, and c) 120°.

6. Wavelength multiplexing of the Au film in optical data storage

We have demonstrated the polarization multiplexing in optical data storage by using Au films composed of closely packed Au nanoislands (see Fig. 3 in the main text). It is interesting to explore the possibility for realizing wavelength multiplexing. In our experiments, we first recorded a pattern (school logo) in an Au film by using 800-nm femtosecond laser pulses with a fluence of 0.67 mJ/cm^2. Then, we tried to retrieve the pattern by using femtosecond laser pulses with a smaller fluence (0.17 mJ/cm^2) and with different wavelengths, as shown in Figure S7. It was found that the pattern became almost invisible when the laser wavelength was shifted to 780 nm, implying the existence of another wavelength channel at this wavelength. Surprisingly, the pattern extracted by using a longer wavelength of 820 nm possesses a larger contrast than that obtained at 800 nm. This behavior indicates that additional wavelength channels are not available in the long-wavelength side.

In order to find out the physical origin for this phenomenon, we measured the transmission spectra of an Au film after the irradiation of femtosecond laser pulses with different fluences (see Figure S2). With increasing laser fluence, a blueshift of the transmission valley as well as an increase of the transmission was observed. As a result, a large change in the transmission appeared at long wavelengths, which is responsible for the cross-talk observed at a longer wavelength.

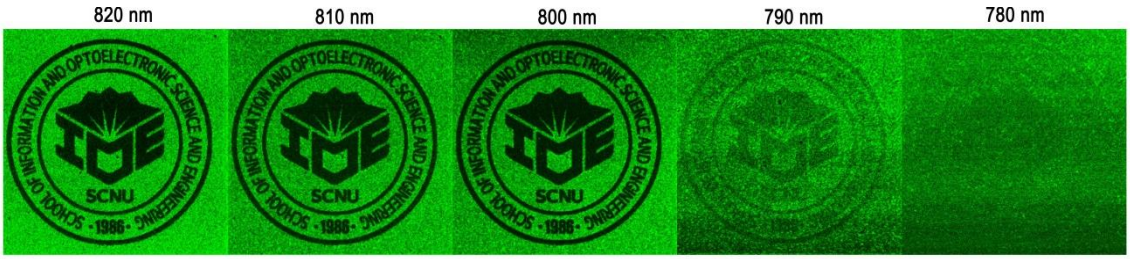

Figure S7. Pattern recorded by using 800-nm femtosecond laser pulses (0.67 mJ/cm^2) and extracted by using femtosecond laser pulses with different wavelengths (0.17 mJ/cm^2).