Non-volatile dynamically switchable color display via chalcogenide stepwise cavity resonators

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In Fig. S1(a), we schematically present the experimental setup of non-contact microsphere femtosecond laser irradiation. A microsphere is used to focus the high repetition rate fs laser (Mira 900 of Coherent, Inc., 800 nm, 76 MHz) via the objective lens (10×, 0.26 NA). The laser fluence can be tuned by an isolator and half-wave plate. A lens holder can fix the soda-lime glass microsphere with a radius of ~27 μm (SLGMS, Cospheric) and is lined up to a microscope system. The device is placed on a three-dimensional nano-stage with a maximum speed of 5 mm/s, a minimum moving accuracy of 10 nm, and a traveling range of 20 mm (FS-3200P-WE2 series, OptoSigma). An in-house code is programmed to automatically move the nano-stage. The working distance is experimentally observed from top and side views using two charge-coupled devices and a long working distance objective lens. The side view of the experimental setup is presented in the zoom-in picture. The focal length is ~35 μm, which includes a working distance of ~8 μm and a microsphere radius of ~27 μm. This focal length is about 44 times larger than the laser wavelength of \( \lambda = 0.8 \) μm. Thus, the fs laser irradiation integrated with the microsphere operates in an optical far field, where a significant ablation depth can be obtained\(^{1-4}\). Note that, near-field fs laser writing mainly demands a smooth target surface because of its short working distance. As the sub-50 nm ablation relied upon near field effect is caused by the generation of evanescent waves and the ablated depth is less than 10 nm\(^{45}\), Thus, the far-field fs laser fabrication has the advantage of a longer working distance over the near-field. By lifting up the microsphere, random surface patterning can be realized using the programming movement of the nano-stage. To explore the possibility of printing with a sub-diffraction resolution, nano-line arrays with different gaps and profiles are patterned on the Sb\(_2\)S\(_3\) film surface. At the scanning speed of 100 μm/s and laser fluence of 0.42 mJ/cm\(^2\), the Sb\(_2\)S\(_3\) film is ablated obviously and a sub-50 nm nano-line is created. Figure S1(b) shows the SEM images of the sub-50 nm line structures realized on the 30 nm thick Sb\(_2\)S\(_3\) film, where we have written a series of irregular nano-lines at the linewidth of sub-50 nm, and the spaces from 100 to 400 nm are achieved. The creation of these nano-lines indicates the microsphere femtosecond laser irradiation is able to realize desirable nanostructures and make high-performance optical devices.

**Fig. S1** (a) Scheme of non-contact microsphere fs-laser setup. Inset: side view of microsphere focusing of fs-laser beam. (b) Various types of super-resolution nano-lines are formed by microsphere fs laser irradiation on 30 nm-thick Sb\(_2\)S\(_3\) layers residing on the Si substrate.
References


Fig. S2 | (a) Optical microscope image and (b) corresponding cross-sectional profile of the nano-structures created on Sb$_2$S$_3$ thin film at a laser power of 0.12~0.22 mW.

Fig. S3 | The photo images of the R-CR strip (ii) in Fig. 1(c) at 270 °C for the various durations of (a) 2 min, (b) 10 min, and (c) 15 min. Scale bar is 100 μm.

Fig. S4 | The reversible color variation of the strip (ii) in Fig. 1(c) between red (MQ-AM) and cyan (R-CR). Scale bar is 100 μm.