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

Kuan Liu^{1†}, Zhenyuan Lin³, Bing Han¹, Minghui Hong^{2*} and Tun Cao^{1†*}

¹School of Optoelectronic Engineering and Instrumentation Science, Dalian University of Technology, Dalian 116024, China; ²Pen-Tung Sah Institute of Micro-Nano Science and Technology, Xiamen University, Xiamen 361102, China; ³Department of Electrical and Computer Engineering, National University of Singapore, 4 Engineering Drive 3, Singapore 117576, Singapore.

 $^{\dagger}\textsc{These}$ authors contributed equally to this work.

*Correspondence: MH Hong, E-mail: elehmh@xmu.edu.cn; T Cao, E-mail: caotun1806@dlut.edu.cn

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Liu K et al. Opto-Electron Adv 7, 230033 (2024)

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 $\sim 27 \mu m$ (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 μ m. This focal length is about 44 times larger than the laser wavelength of $\lambda = 0.8 \mu$ m. Thus, the fs laser irradiation integrated with the microsphere operates in an optical far field, where a significant ablation depth can be ob tained^{S1-S3}. 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^{\$4,55}. 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_2S_3 film surface. At the scanning speed of 100 μ m/s and laser fluence of 0.42 mJ/cm², the Sb₂S₃ 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 Sb₂S₃ 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 Sb₂S₃ layers residing on the Si substrate.

Liu K et al. Opto-Electron Adv 7, 230033 (2024)

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Fig. S2 | (a) Optical microscope image and (b) corresponding cross-sectional profile of the nano-structures created on Sb₂S₃ 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.





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