Supplement Information for "*In-situ* real-time monitoring of ultrafast laser processing using wide-field high-resolution snapshot compressive microscopy"

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In-situ monitoring of self-organized nanostructures with 1kHz repetition rate

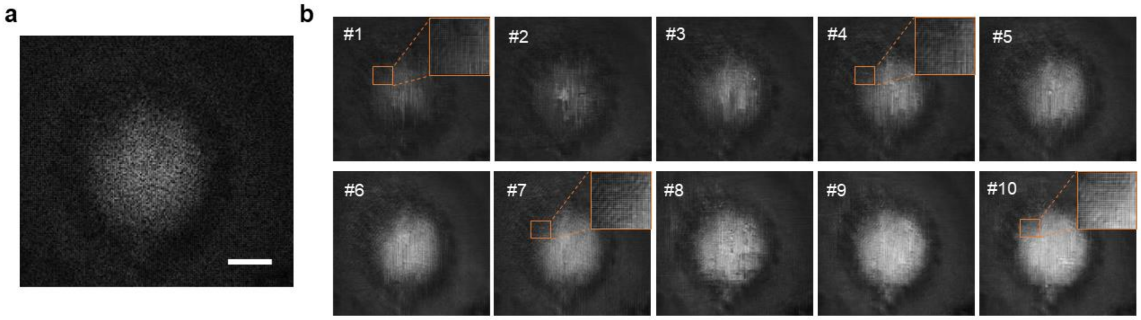

Figure. S1. ***In-situ* and real-time monitoring of the growth of self-organized periodic nanostructures with a laser repetition rate of 1kHz.** Experiments are conducted in high-resolution path. Compressed ratio and exposure time are set to be 10 and 20ms respectively. (a) Compressed measurement. (b) Reconstructed frames. Scale bar in a is 10um.

To visualize the ultrafast laser-matter interactions, a progressive way is to reduce the laser repetition rate to the reconstructed frame rate, allowing each reconstructed frame to distinguish the difference between laser pulses. In light of this, we decreased the repetition rate of the femtosecond laser to 1 kHz and employed our proposed snapshot compressive imaging to monitor the growth of nanogratings. The exposure time and compression ratio were set to 20ms and 10, respectively, resulting in an imaging speed of 500 fps. Consequently, each reconstructed frame corresponds to the interaction of 2 laser pulses with the sample. The reconstructed results is shown in Fig. S1, where we observed a continuous amplification of the signal over time, and the growth of nanogratings was dynamically visualized as the experiment progressed. The reconstruction images showcase undesired artifacts which is primary due to the low signal-to-ratio of the compressed measurement. By lowering the laser repetition rate, lower laser power is injected into the sample surface, leading to low signal from the surface that can be detected. Note that we opted for an imaging speed of 500 fps instead of 1000 fps, as increasing the speed to 1000 fps would adversely impact the quality of the reconstructed signal.

In-situ and real-time monitoring of the laser printing process

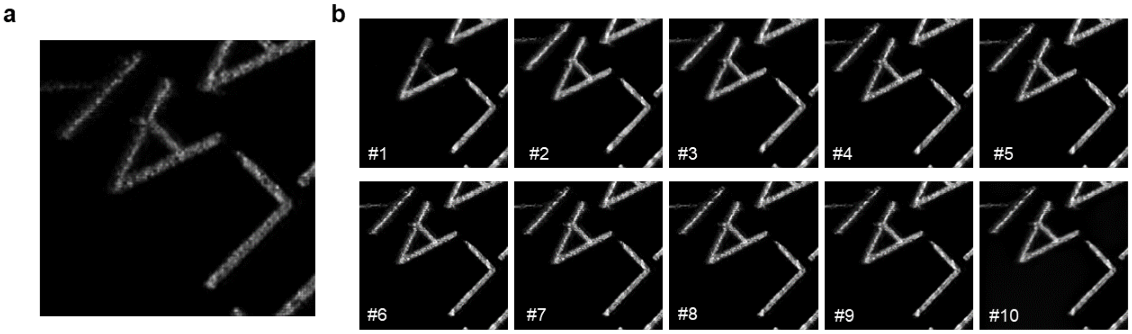

Figure. S2. *In-situ* and real-time monitoring of the laser printing process in wide FOV path. Compressed ratio and exposure time are set to be 10 and 20ms respectively. (a) Compressed measurement. (b) Reconstructed frames.

Our method can also be used to monitor the laser printing process. An interesting application would be customizing desired patterns by laser printing. Specifically, we utilized a resonant scanner to laser-print the designed pattern and used our system to monitor the printing process across a wide field of view. As shown in Fig. S2, we successfully monitored the generation of letter “K”, demonstrating the high-speed capture of printing process. Please note that the experiments is performed in wide FOV path.

Resolution comparison between high-resolution path and wide-field path

We provided a quantitative indicator result concerning the line-profiles of resolution target. We exploited a long-working-distance objective lens with 0.42 NA for imaging (Thorlabs - MY20X-804 20X Mitutoyo Plan

Apochromat Objective, 436 - 656 nm, 0.42 NA, 20 mm WD). Using the theory of Rayleigh Criterion, the Rayleigh resolution is given by $r_R = 0.61\lambda/NA$, where λ is the wavelength of the light. Assuming that the wavelength of a white-light LED is centered at 550nm, the theoretical resolution is then measured to be 0.8 μm, which is equivalent to the measured resolution in Fig. S3. The line pairs in the nineth element of high-resolution target (USAF 1951 Glass Slide Resolution Targets, Edmund Optics, the smallest line-width is 775nm) are observed with fine details in high-resolution path whilst the resolution target in wide-field path is barely seen. Hence, we infer that the imaging system has the capability to achieve its theoretical resolution.

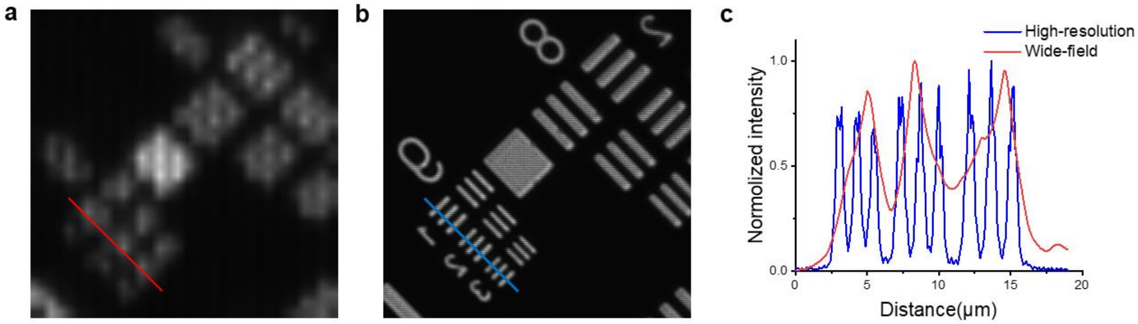

Figure. S3. Comparison of lateral resolution between high-resolution path and wide-field path. (a) Resolution target captured by high-resolution path. (b) Resolution target captured by wide-field path. Note that the image in (b) is interpolated into the same image size for comparison.

Choice of compression ratio

Higher compressed ratio means faster imaging speed. For example, for camera running at 50fps with compressed ratio of 10, this corresponds to an imaging speed of 500fps, but is compromised with degraded reconstruction. We further conduct experiment to verify this point as follows, with simulation results. We test the simulation data (specifically hummingbird data used in [1]) with different compressed ratio B=8, 16, 24, 32, 40, 48. And the reconstruction results are shown in Fig. S4. The peak-signal-to-noise-ratio (PSNR) is used as the performance indicators of reconstruction quality. The PSNR value decreases as compressed ratio increase. When compressed ratio surpasses 20, all algorithms gradually move toward stability.

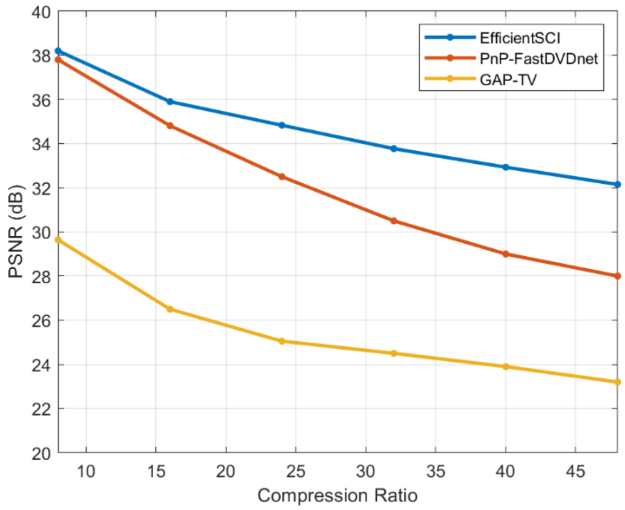

Figure. S4. Reconstruction quality (PSNR in dB, higher is better) of different reconstruction algorithms, with varying compression ratio from 8 to 48.

On top of the above analysis, we choose compressed ratio to be 10 or 20 in our experiments. However, it is common to all that, for PSNR over 30dB, the human eye can hardly discern differences in image quality. Therefore, if there is a need for higher speed imaging in the experimental scenario, higher compressed ratio over 20 can still be adopted.

Reference

1. Wang, L. S, Cao, M. & Yuan, X. Efficientsci: Densely connected network with space-time factorization for large-scale video snapshot compressive imaging. In IEEE/CVF Conference on Computer Vision and Pattern Recognition (CVPR), 18477-18486 (2023).