![frontiers]

Supplementary Material of "Ultrafast laser direct-writing of self-organized microstructures in Ge-Sb-S chalcogenide glass"

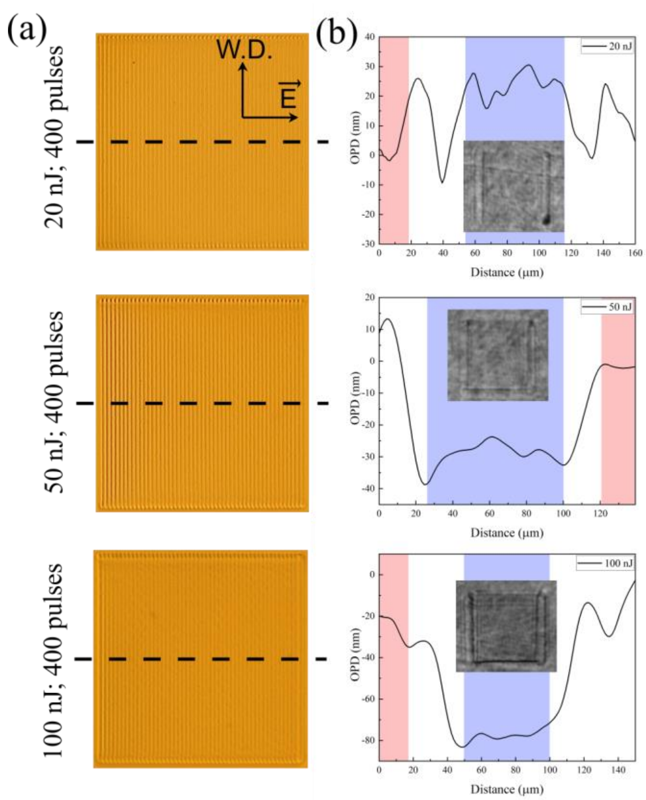
Gözden TORUN[1*], Anupama Yadav[2,+], Kathleen A. RICHARDSON[2], Yves BELLOUARD[1]

[1] Galatea Lab, IEM/STI, Ecole Polytechnique Fédérale de Lausanne (EPFL), Neuchâtel, Switzerland

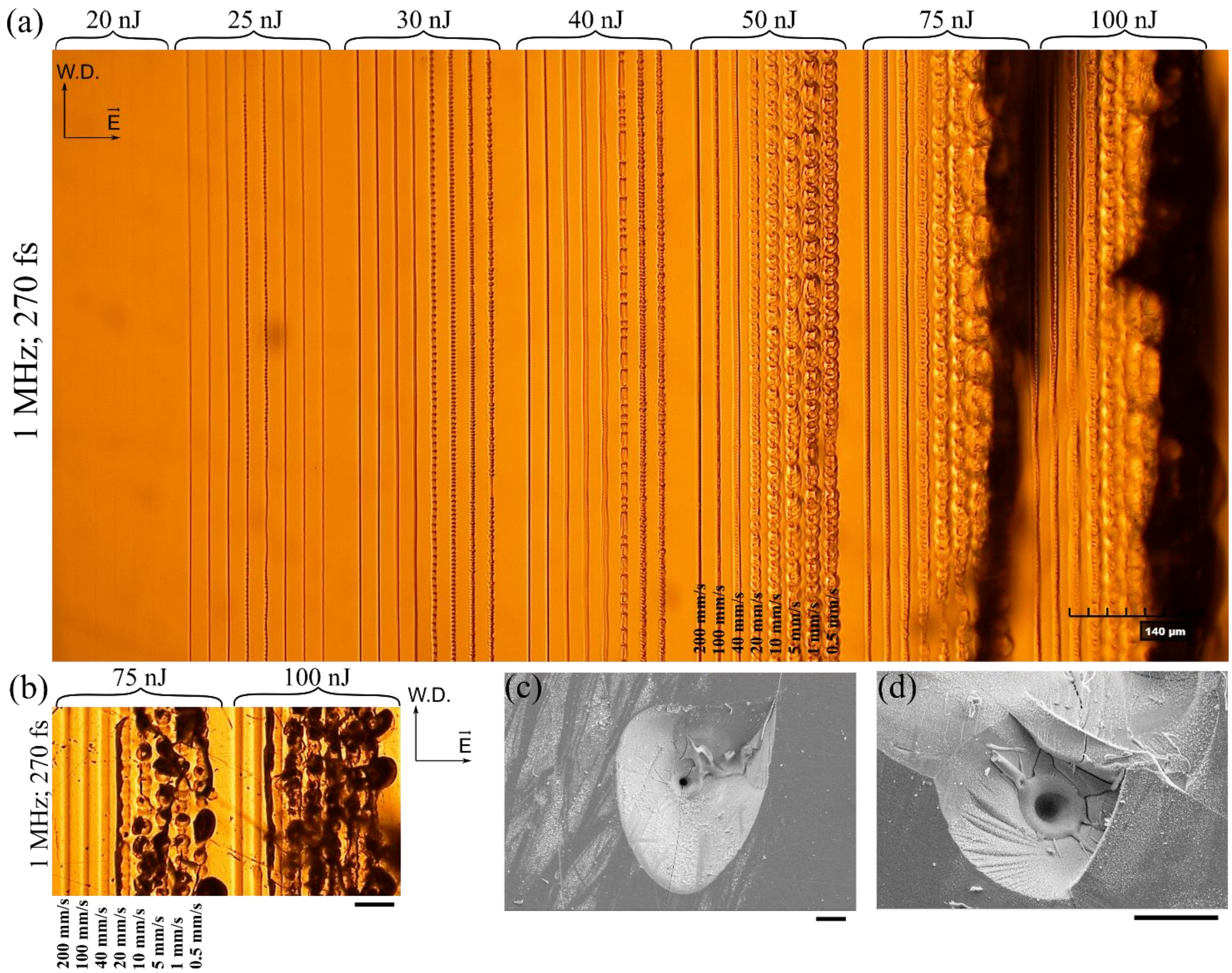
[2] CREOL and College of Optics and Photonics, University of Central Florida, Orlando, USA

[+] Present address: Department of Physics, Sri Aurobindo College, University of Delhi, New Delhi, India

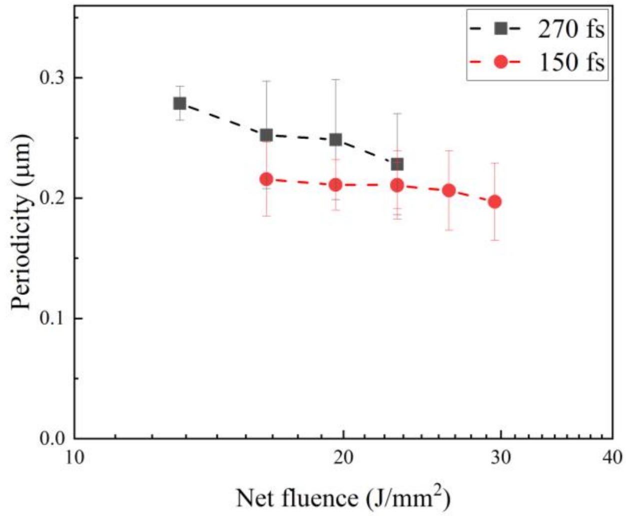
*** Correspondence:**
Gözden Torun
gozden.torun@epfl.ch



Supplementary Figure 1: (a) Transmission optical microscope images of 1x1 mm^2 laser-affected areas obtained at a pulse rate of 100 kHz with 400 overlapping pulses. (b) Optical path difference across the laser modified area (insets: digital holographic microscope images of the area). The average optical path differences are calculated averaged from the red and blue areas. Red areas are considered as pristine material, while blue areas are considered laser-modified zones.



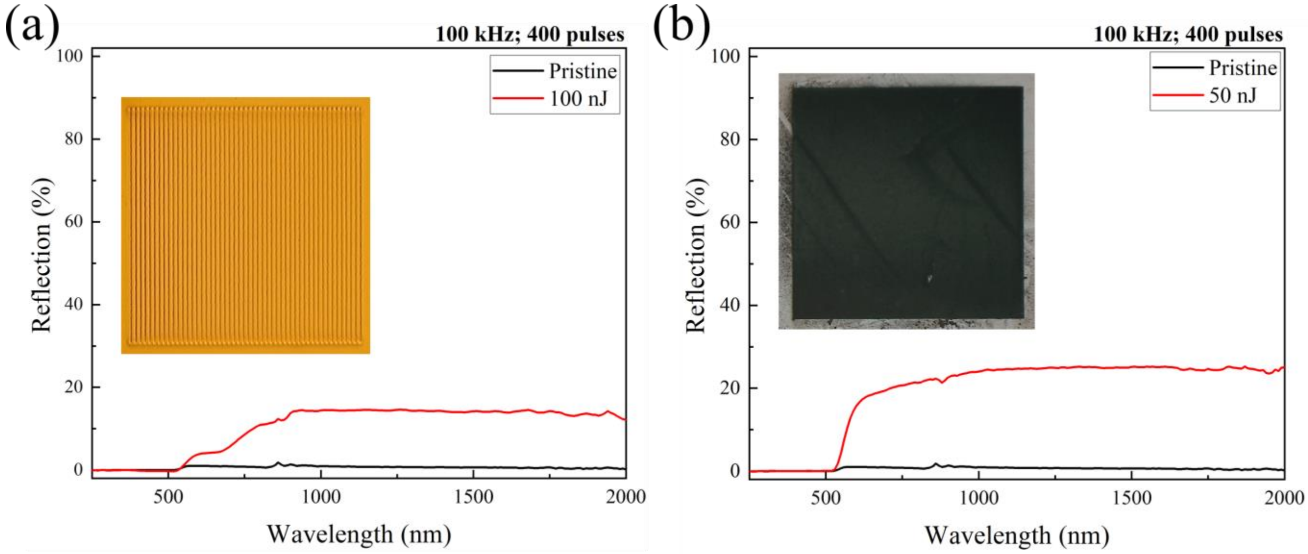
Supplementary Figure 2: (a-b) Transmission optical microscope images of laser inscribed lines in $Ge_{23}Sb_7S_{70}$ glass volume at 1 MHz (thermal cumulative regime) with various writing speeds and pulse energies. (c-d)SE images of the top surface of laser inscribed lines at 75 nJ with 4000 pulses. Note that the focal spot was located at 20 µm below the surface.



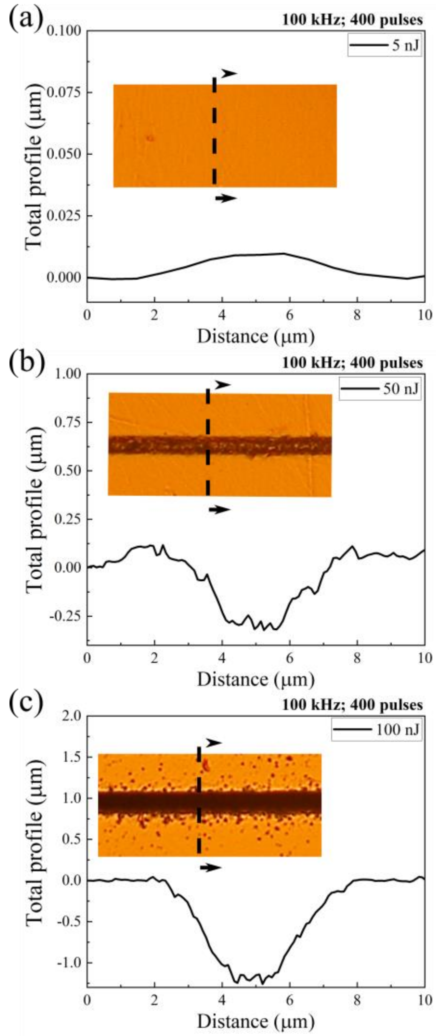
Supplementary Figure 3: Volume nanogratings periodicity of $Ge_{23}Sb_7S_{70}$ as a function of laser net fluence for a pulse duration of 150 and 270 fs emitted at a rate of 100 kHz.

Table S1: Literature survey of periodicity of surface self-organized structures in chalcogenide glass [29-34].

Glass composition	*Pulse duration (fs)*	*Wavelength (nm)*	*Periodicity in nm (in parenthesis: number of overlapping pulses)*
As_2S_3 [29]	150	800	700 (2-5) and 250 (>10)
As_2S_3 [30]	150	800	740 to 780 (for overlapping pulses of 65%-80%) and 300 (higher overlapping or secondary scan)
As_2S_3 [31]	100	800	(2) to 180 (10)
As_2S_3 [33]	34	806	180 on top of 720 (10)
As_2S_3 [34]	150	3000	599 (1000)
As_2Se_3 [34]	150	3000	583(1000)
$Ge_{25}Ga_1As_9S_{65}$ [32]	34	806	720 (50)
$Ge_{23}Sb_7S_{70}$ [This study]	150	1030	210 (5) and 170 (400)
$Ge_{23}Sb_7S_{70}$ [This study]	270	1030	330 (1), 250 (5) and 170 (400)



Supplementary Figure 4: Reflection spectra of laser-affected zones (a) in the volume and (b) on the surface. The inset shows a reflective optical micrograph of a single plane (1x1 mm^2) formed with parallel lines, themselves made of overlapping pulses with 100 nJ pulse energy. Note that the peaks around 800 nm are measurement artifacts due to the change of detector types from visible to NIR measuring spectral ranges.



Supplementary Figure 5: Surface profile obtained for three different surface regimes, firing 270 fs pulses at 100 kHz. The inset images show transmission optical microscope images of the surface, corresponding to (a) homogeneous modifications at 10nJ, (b) self-organized surface structures at 50 nJ, and (c) ablation patterns at 100 nJ, all of them achieved with 400 pulses overlapping.