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Ultrafast dynamics of femtosecond laser-induced high spatial frequency periodic structures on silicon surfaces

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Numerical simulation of local enhancement of light field

When the groove depth of LSFL is as shallow as 15 nm, it is difficult for the light field to locally increase on the ripple ridges[S1]. However, the split phenomenon of the LSFL ripples, with narrow and deep features, followed by the evolution of uniform HSFL, was clearly observed in ultrafast imaging. Based on experimental observations, it is suggested that sub-surface nano-cavities exist in the middle of the ridge during the LSFL preparation on silicon.

As shown in Fig. S1, an air ellipse is placed in the subsurface layer of the LSFL ridge to represent the nanocavity. The distance from the air-silicon interface is 5 nm, and the radii in the x and y directions are r_x and r_y, respectively. To observe the transient response of the structural surface to the light field during femtosecond laser irradiation, we numerically simulated the transient process during femtosecond laser irradiation in steps of 0.1 fs. A plane wave with central wavelength of 800 nm, pulse width of 50 fs, polarization direction parallel to the silicon surface and parallel to the paper, enters the surface vertically from top to bottom. The amplitude of the incident laser electric field is set to 1 for comparison of the light field under different conditions. Since only the electric field distribution at the groove and ridge is of interest, we set the simulation area to one cycle to improve grid accuracy and reduce computational time.

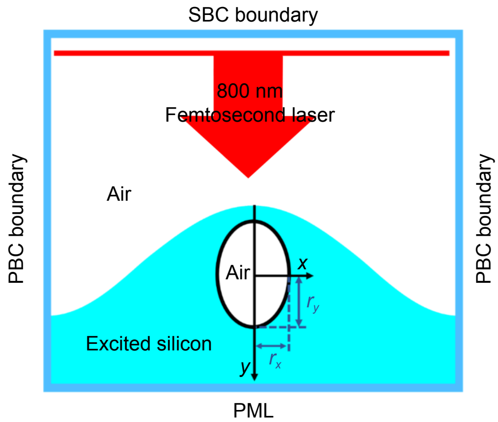

Fig. S1 | Schematic diagram of LSFL nanostructure with a nanocavity and the incident light.

The numerical calculations in a volume of 760 × 1200 nm^2 were performed with COMSOL Multiphysics software. It contains two layers: air layer (800 nm) and excited state Si (400 nm thick). The carrier density on the silicon surface is 8.04×10^{21}/cm^3 when irradiated with a femtosecond laser of 0.82 J/cm^2, corresponding to a dielectric constant of −8.79+8.65i[S2]. There is a LSFL structure with a period of 760 nm and a depth of 14 nm on the surface, which comes from the measured results of SEM and confocal optical microscopy (COM). The scattering boundary (SBC) was set at the upper face, and perfectly matched layer (PML) at the bottom face of the cube. The boundary perpendicular to the silicon surface is set as a periodic boundary condition (PBC)[S3,S4].

As shown in Fig. S2, the simulation results indicate that femtosecond laser irradiation of the silicon surface containing nano-cavities generates a strong localization near the white dashed line on the surface, particularly around the cavity. The nano-cavities are modeled as circular with a diameter of 10 nm. If there are no nanocavity, the light is mainly localized in the grooves, which is 1.23 times the light intensity at the ridge. This is similar to the results reported in the literature[S1], and cannot explain the narrow and deep slit formed at the ridge of the LSFL after single pulse laser irradiation. However, when a nanocavity is set at the LSFL ridge, laser field greatly localized there, and peak intensity increases to 3.78 near the nano-cavity, much higher than 3.25, the intensity at the valley.

Figure S3 shows the electric field distribution along the x-axis and y-axis when placing different nanocavities below the surface. Near the circular nano-cavity with a radius of 5 nm, the width of the x-directional region where the local electric field intensity exceeds 4.0 is only 8.24 nm. However, the maximum local electric field intensity in the y-direction reaches 5.08, with a depth exceeding 13.3 nm for intensity greater than 4.0. This indicates that the nano-cavity is more likely to extend in the y-direction during the laser irradiation. Therefore, keeping the x-direction radius r_x at 5 nm, when the y-direction radius increases to 10 nm, the maximum local electric field intensity in the y-direction reaches 6.16, with a depth exceeding 24.8 nm. When the y-direction radius is further increased to 15 nm, the maximum local

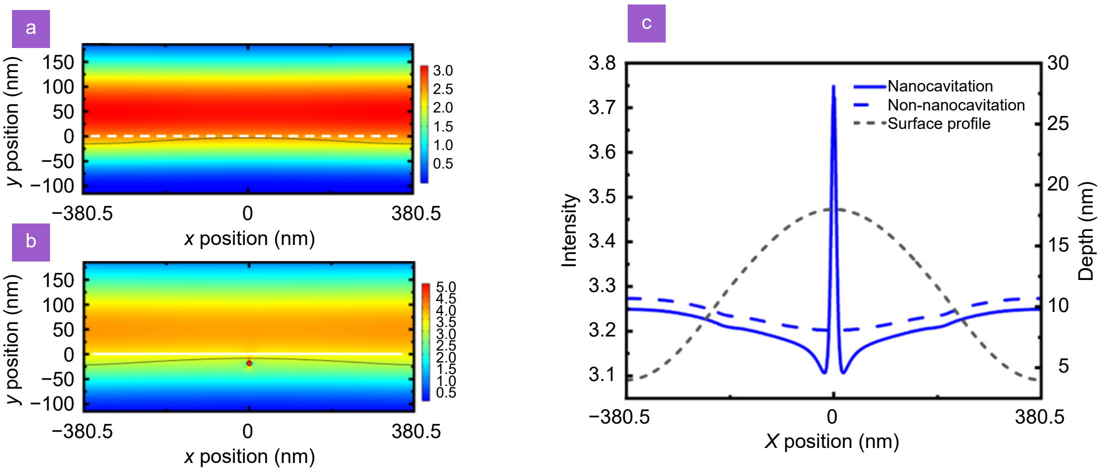

Fig. S2 | (**a**, **b**) Two-dimensional distribution of the laser electric field without and with nano-cavities. (**c**) The blue solid and dashed curves indicate the light intensity distribution at the white dashed line on the silicon surface with and without nano-cavites, respectively. The black dashed line is the surface profile.

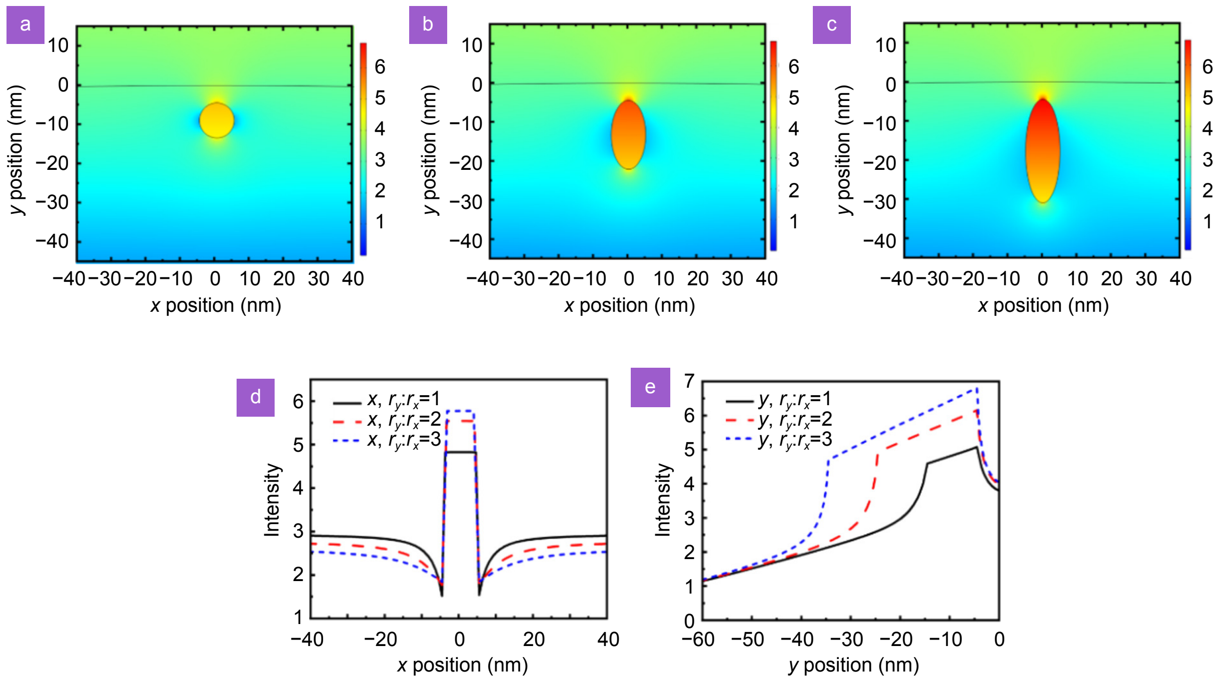

Fig. S3 | (**a**–**c**) 2D electric field distribution around the nanocavity with r_x:r_y=1,2,3, respectively. (**d-e**) Electric field intensity distribution along the *x*-axis and *y*-axis, respectively.

electric field intensity in the *y*-direction reaches 6.81, with a depth exceeding 35.4 nm, while the width of the *x*-directional region decreases to 7.13 nm. This indicates that the nano-cavity extends in the *y*-direction and forms a deep and narrow nano-plane with a positive feedback effect as the nano-cavity grows and the electric field localization increases.

During the fabrication of LSFL by multi pulse irradiation, there are transient nano grooves on the ridges. After solidification, the molten material will fill and submerge these transient nano grooves. The filled material exists in a partially amorphous or polycrystalline state with a lot of defect states. Compared to single-crystal silicon, the re-solidified material exhibited higher absorption rate. During laser irradiation, nanoscale plasmas in higher excited states are formed rapidly.

Figure 11 shows that the defect size at the ridge is generally 10–60 nm. Therefore, a trapezoidal region with an upper width of 30 nm, a lower width of 10 nm, and a height of 20 nm represents the nanoplasma of the filled molten material. The dielectric constant of nanoplasma is set as −8.79+8.65i, and silicon is in the ground state. The optical field and boundary parameters are the same as the simulation of Fig. S1.

As shown in Fig. S4, the simulation results show that the light field is strongly localized in the nanoplasma. Compared to other areas in the ridge, the light intensity is enhanced by more than 3.04 times.

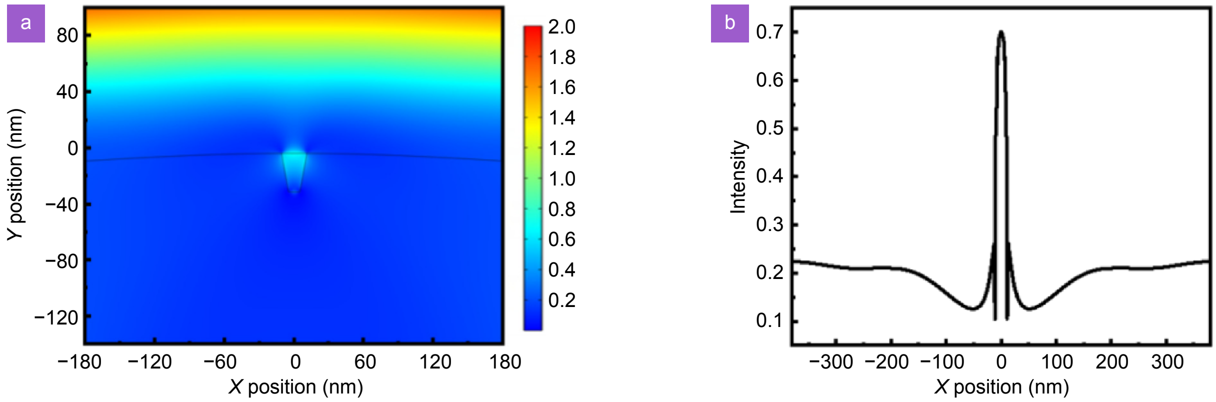

Fig. S4 | (**a**) 2D electric field distribution around the nano plasma. (**b**) Electric field intensity distribution along the *x*-axis.

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