Supplementary Material

Controllable generation of large-scale highly-regular gratings on Si films

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Numerical simulation of interfacial electric field in vicinity of silica nanoparticles

We perform the numerical computations by using FDTD-based method to investigate the interfacial electric field distribution when the incident light reacts with the nanoparticles. Here, we assume the nanoparticles are silica, which are oxidation product of the a-Si after absorbing the heat from the femtosecond laser. The simulation model consists of three layers: the bottom layer is substrate (Al_2O_3), the middle layer is a T = 200 nm thick a-Si, and the silica nanoparticles are embedded in the top layer. The simulation region is a cuboid of $P_x \times P_y \times P_z = 4\times4\times4$ μm dimensions and perfectly match layer (64 layers) in $x-y$ plane, $x-z$ plane and $y-z$ plane is adopted. The incident light is a monochrome plane wave at 1030nm and polarizes along x direction. A frequency-domain field and power monitor sets to investigate the interfacial electrical field at Si surface. A mesh is set to thoroughly encompass the region where nanoparticles are located, and the maximum mesh step in x and y

directions are equal to 20 nm while that in *z* direction is 10nm. The simulation time is 1000 fs and we assume the simulation converges when the auto shutoff level is below 1.0×10^{-5} and the simulation process is no more than 90%. We employ an ellipsoid (maximum cross section with the diameter of d = 500 nm in *x*-*y* plane and the third radius of 100 nm in *z*-axis) to represent the silica nanoparticle and it is set to be located at the center of the simulation region. Because we assume the oxidation happens along both directions at the interface of amorphous silicon and air, half of the ellipsoid is exposed to the air surroundings while the other part is embedded into the amorphous silicon. In such situation, we set the mesh order of the ellipsoid to be 1 while that of the amorphous silicon to be 2 so the ellipsoid will take priority over amorphous silicon within the overlap region. After simulating the monoparticle, we obtain the electrical filed distribution within the interface of air and the amorphous silicon. We then add other ellipsoids with smaller dimension size (maximum cross section with the diameter of 200 nm and the third radius of 50 nm) around the first ellipsoid to investigate the coupling of field induced by different nanoparticles.

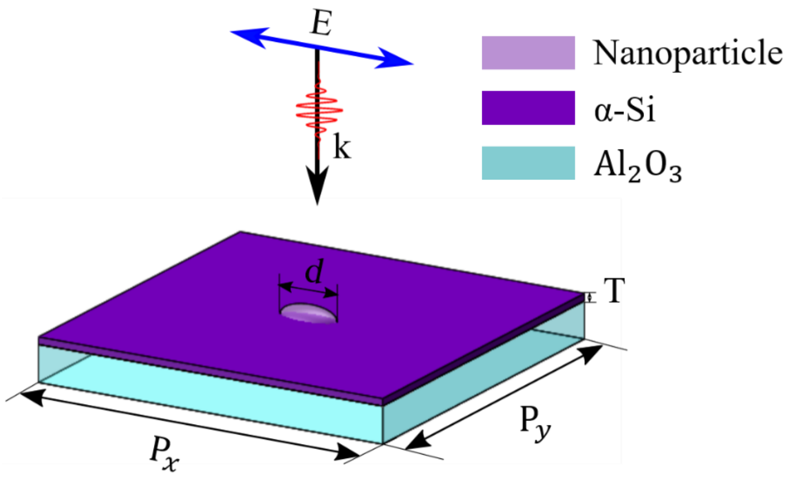

Figure S1. Scheme of the numerical simulation model.

LIPSS on a 200 nm thick a-Si film by a circularly polarized laser

Figure S2(a, b) depict the SEM images of the nanostructures that are produced by a circularly polarized femtosecond laser. The applied laser fluence is the same as that for the linearly polarized laser. The circular polarization can be visualized as rotating linear polarization (Fig. S2c). Therefore, a seeding nanoparticle will grow towards every direction. As a result, we do not obtain periodic ripples. Instead, a porous surface is observed.

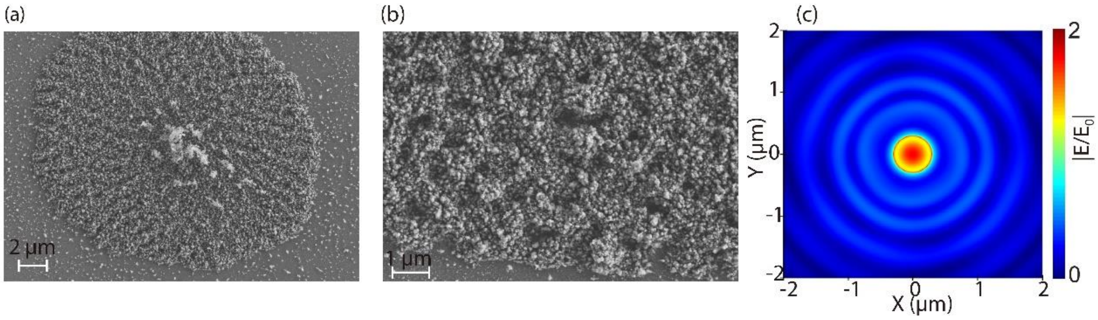

Figure S2. Overview (a) and high resolution (b) SEM of surface structuring by circularly polarized femtosecond laser. (c) Numerical simulation of interfacial electric field on 200 nm thick a-Si film with an embedded silica nanoparticle when irradiating by a circularly polarized 1030 nm light source.

Schematic demonstration of optical localization-induced nonlinear competition (O-LINC) mechanism

As shown by the SEM image in Fig. S3(a), prior to the formation of periodic ripples, we observe a high amount of laser-produced subwavelength nanoparticles. Each nanoparticle behaves as a dipolar scatter. The interference between the incident/reflected and scattered waves results in periodic pattern in vicinity of the nanoparticles. Each nanoparticle will grow simultaneously due to the local field enhancement at the poles of the nanoparticles. However, their growing speed is related to their initial size. As confirmed by the numerical simulation of interfacial electric field distribution in Fig. S3(b-f), the bigger nanoparticle acquires a stronger scattering field and thus exists a higher field enhancement. This is also confirmed by the SEM image in Fig. S3(a). The higher field enhancement results in a faster growing speed and bigger size, forming a positively nonlinear feedback, as schematically illustrated in Fig. S3(g). We refer this nonlinear feedback as an optical localization-induced nonlinear competition (O-LINC) mechanism, which is capable of choosing a single nanoparticle as an effective seed to initiate a regular nanograting.

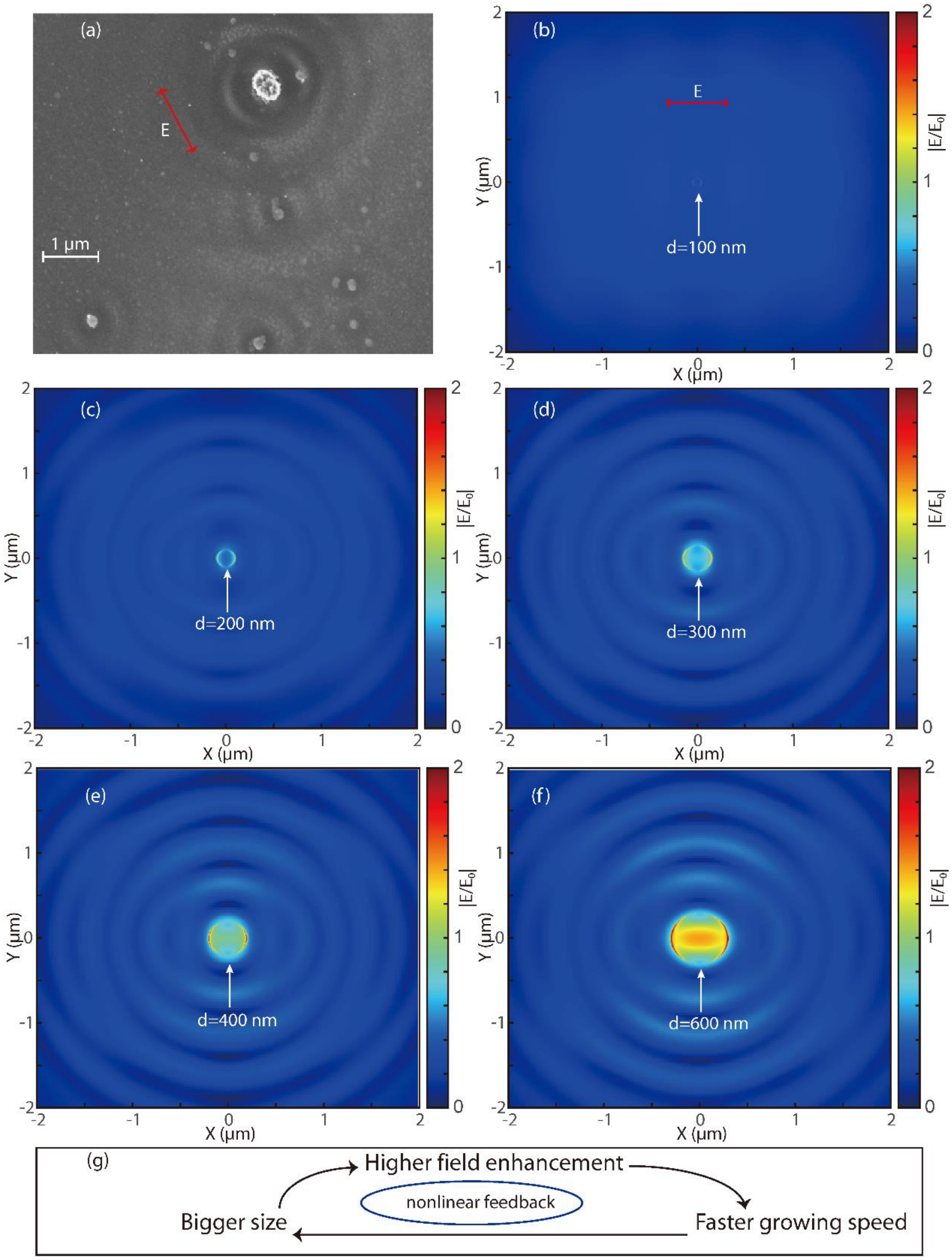

Figure S3. (a) SEM image of laser-produced nanoparticles and interfering patterns on a 200 nm thick a-Si film. Red arrow indicates the laser polarization direction. (b-f) Numerical simulation of electric field distribution on 200 nm thick a-Si film surface with an embedded ellipsoidal SiO_2 nanoparticles. The diameter of these particles in $x-y$ plane set to 100, 200, 300, 400 and 600 nm, respectively. The diameter in the third axis is fixed to 200 nm. (g) Scheme of the nonlinear feedback mechanism.