Supplemental Document

Optics Letters

Plasmon-driven light harvesting in poly(vinyl alcohol) films for precise surface topography modulation: supplement

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Supplemental document

Section 1

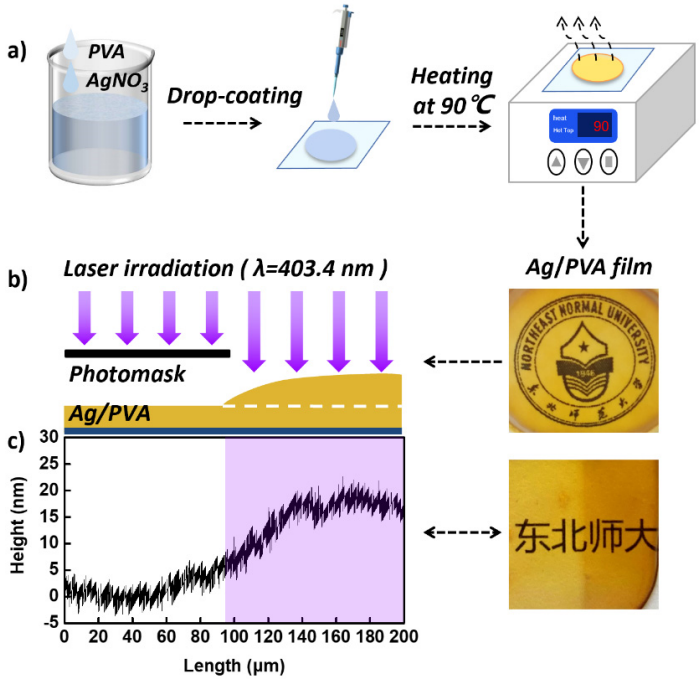

Fig. S1. (a) Fabrication process of Ag/PVA nanocomposite film. (b) Schematic of film surface deformation *via* laser irradiation with localized photomask (λ=403.4 nm, 50 mW/cm^2). (c) Modulated height for deformation on Ag/PVA film versus irradiation position.

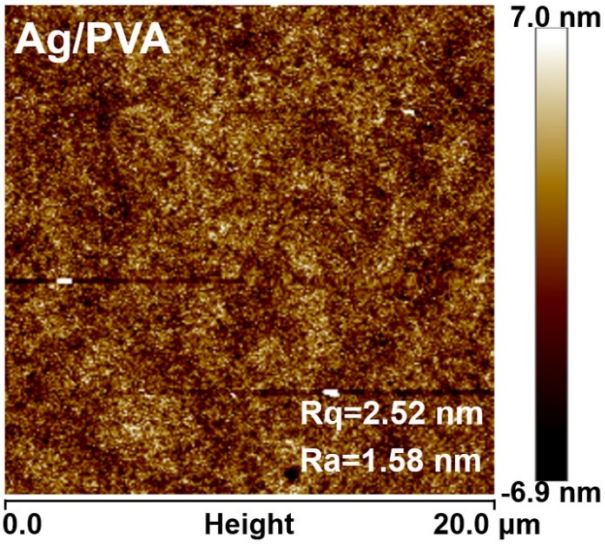

Fig. S2. AFM observation of surface of the obtained Ag/PVA film.

Section 2

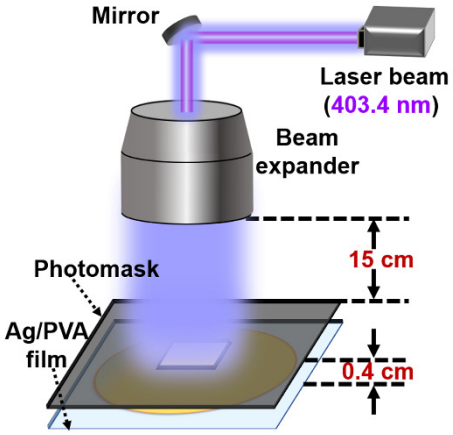

Fig. S3. Schematic of experimental device for photomask irradiation.

The specific steps of photomask irradiation are as followed: a blue-violet laser beam (403.4nm) passes through the beam expander, and the beam expander expands it into a parallel beam with a diameter of ~4 cm. And then the Ag/PVA film is irradiated through the self-made photomask that is an iron plate coated with black paint with 1.5 cm square hole in its center. In the non-hollowed out region, the transmittance of the blue-violet beam with power density of 50 mW/cm^2 is almost zero. Herein, to achieve good light-shielding effect and clear discoloration boundary, the short distance between the photomask and the Ag/PVA film is required, which is set as ~0.4 cm during the experiment.

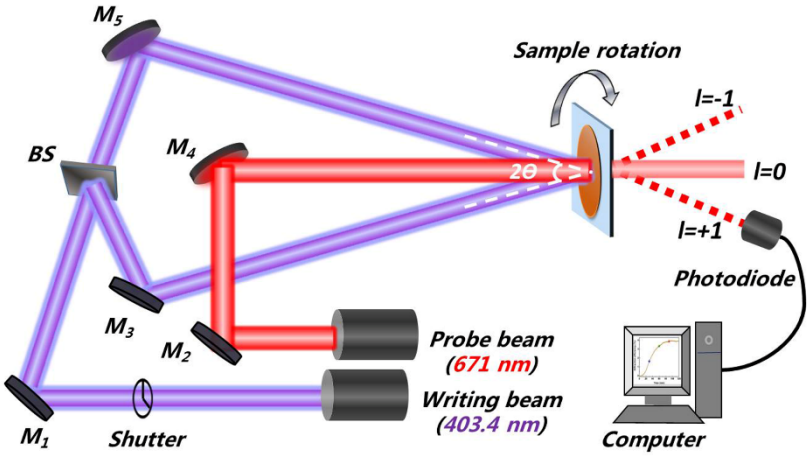

Fig. S4. Schematic of optical setup for inscribing SRGs & CSRGs.

A laser beam passed through a beam splitter and then was divided into transmitted and reflected lights. The two *s*-polarized and coherent 403.4 nm beams with an irradiance of 5 mW were aligned to interfere on the plasmonic polymer, forming a spatially modulated periodic structure. The grating period (Λ) can be determined according to the equation: $\Lambda = \lambda/(2\sin\theta)$, where λ is the wavelength of the writing beam and θ is the half-angle between the two writing beams. The Ag/PVA film was mounted on the sample holder, which can be rotated freely at any angle in the plane perpendicular to the beam propagation direction. At the same time, the grating growth kinetics was probed by a red laser beam (671 nm, *s*-polarized, Changchun New industries Optoelectronics Tech. Co. Ltd.) that was incident normally on the film, and the first-order diffraction signal was temporally monitored by a photodiode.

Section 3

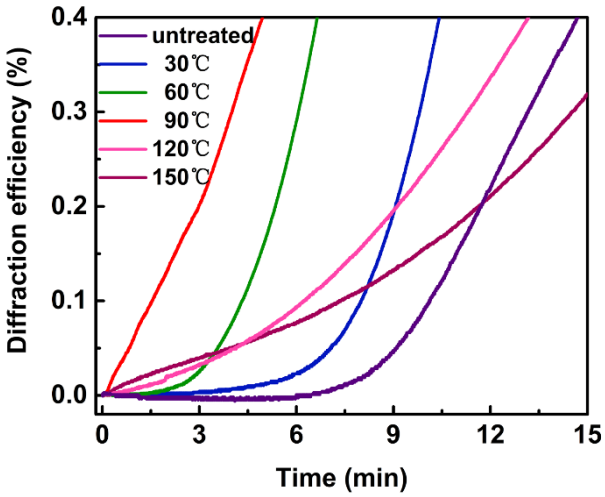

Fig. S5. First-order diffraction kinetics of the thermally pretreated and untreated samples for the recording period of 0~15 min.

Section 4

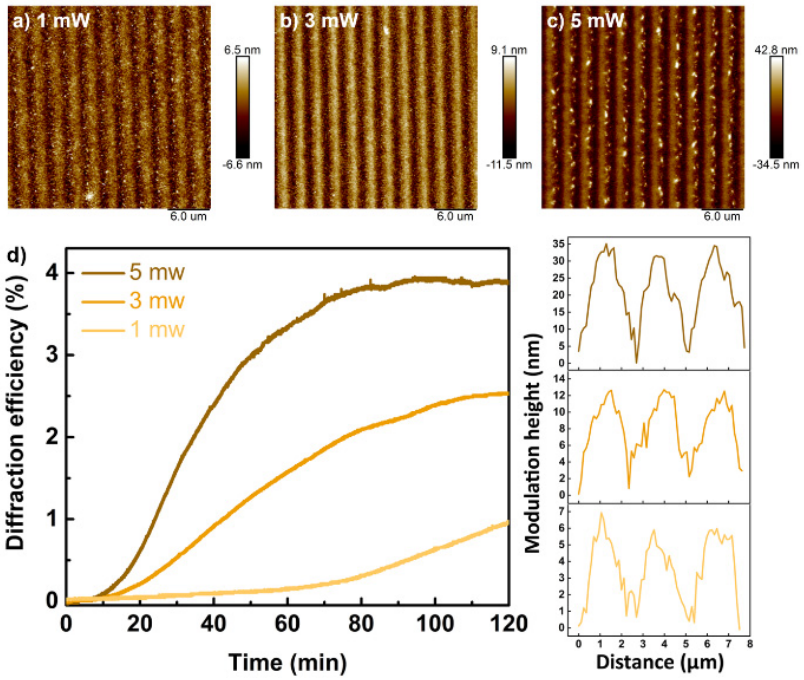

Fig. S6. AFM images of surface of Ag/PVA films under different irradiation powers of (a) 1 mW, (b) 3 mW, and (c) 5 mW. (d) Diffractive kinetics of *in-situ* recording with different powers of 1 mW, 3 mW, and 5 mW. The corresponding surface modulation heights are shown on the right side.

Section 5

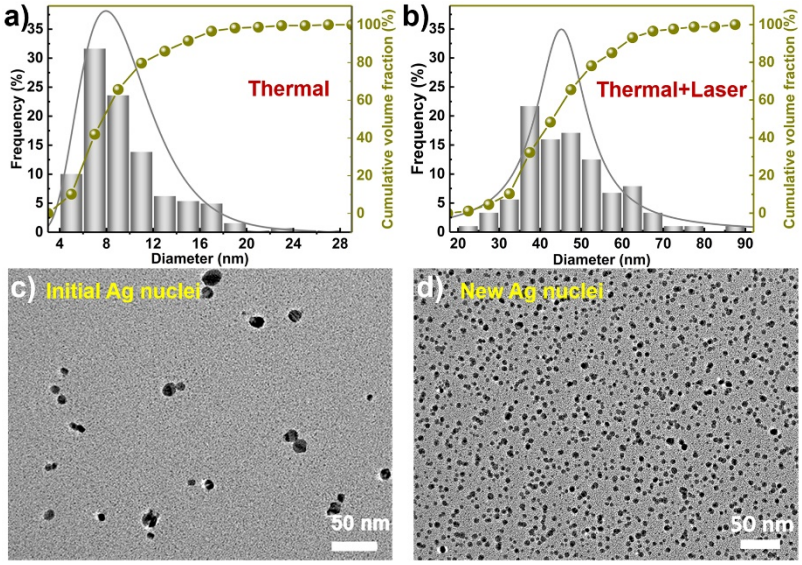

Fig. S7. (a) Histogram of Ag particle size distribution generated by thermal reduction at the initial stage. (b) Histogram of Ag particle size distribution for the growth of initial Ag nuclei after the 403.4 nm laser irradiation. (c) TEM image of initial Ag nuclei generated by pre-thermal treatment. (d) TEM image of new Ag nuclei generated by laser irradiation.

Section 6

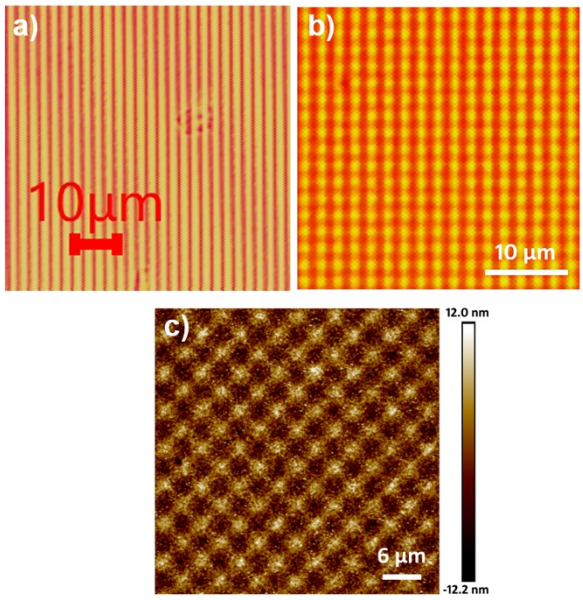

Fig. S8. Optical micrographs of (a) SRGs and (b) CSRGs. (c) AFM image of CSRGs in Ag/PVA films.