Laser nanostructuring by tailored free carrier generation in designer semiconductor metasurfaces

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Abstract: We demonstrate the formation of high aspect ratio nano-trenches in ultrafast laser-illuminated semiconductor meta-atoms enabled by photoinduced free-carrier generation inside localized hot spots.

1. Introduction

Laser machining and material processing using ultrashort pulses has been an industry standard in manufacturing and printing for decades [1]. While the diffraction limit often restricts the feature sizes realized with this family of techniques, several approaches were proposed to remove or reshape matter at the nanoscale [2-4]. Here, we demonstrate nanotrench formation in semiconductor microresonators [5] by breakdown plasma from femtosecond mid-infrared radiation. Trenches with widths from 50 nm to 500 nm and a depth of 600 nm were formed in silicon microparticles by femtosecond laser pulses with a carrier wavelength of 3.9 µm. Full-wave simulations of the field distribution in the resonators with photogenerated free carriers (FCs) reveals hot spot formation at the apex of the resonator notch that acts as a seed feature for nanotrench formation [6]. The highly nonlinear nature of photoinduced FC generation by mid-infrared radiation and enhanced local fields create conditions for localized phase explosion of silicon [7]. Using microresonator hot spots for subwavelength nanotrench formation can become a powerful tool to scale down the critical dimensions enabled by modern laser machining.

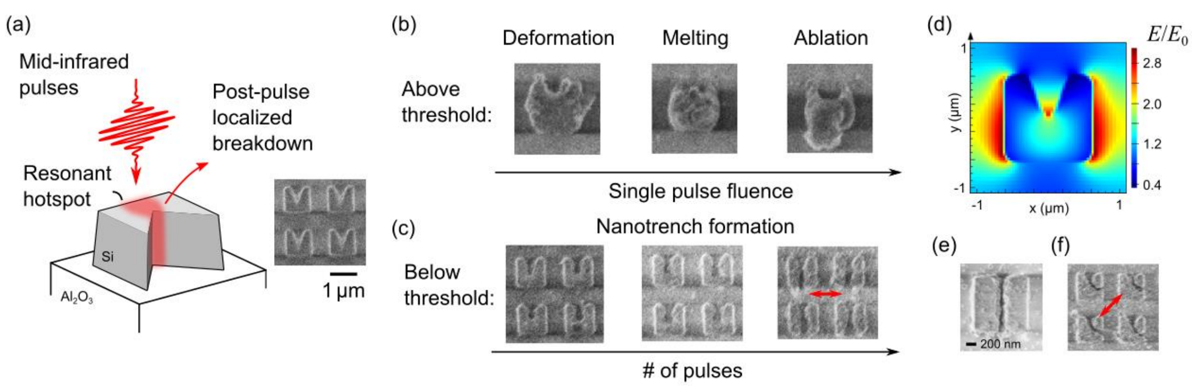

Fig. 1. Nanomachining with mid-infrared laser pulses using free-carrier generation in hot spots of pre-fabricated silicon resonators. (a) Schematic: free carriers are produced by resonant mid-infrared ($\lambda = 3.9$ µm) pulses inside a hot spot. (b) Above the **s**ingle-**pu**lse **d**amage (SPUD) threshold ($F > 0.15$ J/cm^2): resonator deformation due to the heat generated by the pulse. (c) Below the SPUD threshold (0.05 J/cm$^2 < F <$ 0.15 J/cm^2): ablative nanotrench formation due to localized phase explosion. (d) Strongly localized optical field distribution in a silicon resonator at $\lambda = 3.9$ µm. Plasma frequency: $\omega_p = 10^{15}\ \mathrm{s}^{-1}$. (e) Multi-pulse generation of long and deep trenches as narrow as ≈50 nm. (f) Polarization (red arrow) dependence of the trench profile.

2. Results and discussion

Resonator samples were fabricated from a silicon-on-sapphire wafer (University Wafer, scientific grade, 600 nm device layer). The pattern was defined in a PMMA resist layer (495K at 1600 rpm with 1 minute 90 °C and 12 minutes 170 °C bake) at 1000 µC/cm^2 (JEOL 9500FS with proximity effect correcxtion) and developed in MIBK:IPA solution (1:3). A 30-nm layer of Cr was thermally evaporated and lifted off in sonicated acetone for 22 minutes at room temperature. The pattern was transferred to the device layer by HBr reactive ion etch (Oxford

Instruments Cobra), and the remainders of the mask were removed in a wet Cr etch. A scanning electron microscope (SEM) image of the resulting resonators before nanomachining is shown in Fig.1(a).

The resonators were irradiated by laser pulses from a homebuilt $KNbO_3$/KTA 3-crystal/3-pass optical parametric oscillator pumped by a homebuilt 80-fs Ti:Sapphire chirped pulse amplification system. For the experiments, the 200-fs-long mid-infrared (idler) beam was fixed at $\lambda = 3.9$ μm, pulse energy of up to 5 μJ and focused down an area of ≈2500 μm^2, resulting in a fluence of up to 0.2 J/cm^2. A controllable number of pulses (from 1 to 1,000) was sent to the resonator array, with each pulse's power characterized by a calibrated pick-off arm. Above the **s**ingle-**pu**lse **d**amage (SPUD) threshold of $F > 0.15$ J/cm^2 (Fig. 1(b)), the whole resonator deforms due to the heat generated by the pulse, with deformation and melting happening at lower fluences and ablation and resonator detachment occurring at higher fluences. Below the SPUD threshold but above the multi-pulse damage of the resonators, 0.05 J/cm^2 $< F <$ 0.15 J/cm^2 (Fig. 1(c)), a nanotrench is formed by nanoablation due to hot-spot-induced localized phase explosion.

The characteristic speed of nanotrench formation depends on the fluence of the pulses and is on the scale of 30 nm/pulse, which is consistent with previous findings in flat mirror coatings [6]. Nanotrenches formed through the entire silicon layer (600 nm) vary in width, with the highest fluences forming wider trenches. The overall range of the observed trench widths is 50 nm to 500 nm. Examples of very narrow trenches and trenches formed using a train of diagonally polarized (red arrow) pulses are shown in Fig. 1(e) and (f), respectively. The main mechanism of nanotrench formation relies on FC generation by localized laser field hot spots, which funnel the energy of the incoming mid-infrared pulse to a small region of the resonator and induce photoinduced breakdown. As shown previously [7], under mid-infrared pulse illumination, silicon undergoes phase explosion at fluences in the vicinity of those used in our experiments. By pumping the resonators below the single-pulse damage, we enable localized phase nanoexplosions that chip away silicon in the places of the most localized field. Fig. 1(d) shows a calculated electric field profile for a resonator that includes an electron-hole plasma with a plasma frequency of $\omega_p = 10^{15}$ s^{-1}. A hot spot can be seen at the apex of the triangular notch of the resonator, with fields penetrating silicon with a local enhancement field factor of $E/E_0 \approx 2$. Even a moderate enhancement can have dramatic impact over the process of free-carrier generation in strong mid-infrared field. Estimations of the total rate of the direct inter-band electron transitions by the Keldysh formula [8] (Γ-point, two-band approximation) show more than two orders of magnitude enhancement of FC generation at $\lambda = 3.9$ μm in silicon, which serves a strong indication of highly inhomogeneous plasma formation and explains localized nanoablation.

3. Conclusions

We have shown high aspect ratio nanomachining in silicon-based microresonators by mid-infrared femtosecond laser pulses. Features with subwavelength (down to ≈ λ/80) dimensions have been engraved by pulse trains below the single-pulse damage threshold of the resonator due to localized hot spots in the resonator coupled with the highly nonlinear nature of free carrier generation in silicon by strong mid-infrared radiation. The established approach to nanomachining has the potential to remedy the existing limitations of photolithography and to be applied in novel light-based machining techniques.

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References

[1] Perry, M. D. et al., "Ultrashort-pulse laser machining of dielectric materials," *Journal of Applied Physics* **85**, 6803 (1999).
[2] Joglekar, A. P. et al., "Optics at critical intensity: Applications to nanomorphing," *Proceedings of National Academy of Science* **101**, 5856 (2004).
[3] Gattass, R. R., Mazur, E., "Femtosecond laser micromachining in transparent materials," *Nature Photonics* **2**, 219 (2008).
[4] Makarov, S. V. et al., "Light-Induced Tuning and Reconfiguration of Nanophotonic Structures" *Laser & Photonics Reviews* **11**, 1700108 (2017).
[5] Kuznetsov, A. et al. "Optically resonant dielectric nanostructures," *Science* **354**, aag2472 (2016).
[6] Rasedujjaman, Md., Gallais, L., "Polarization dependent laser damage growth of optical coatings at the sub-picosecond regime," *Optics Express* **26**, 24444 (2018).
[7] Werner, K. et al., "Single-Shot Multi-Stage Damage and Ablation of Silicon by Femtosecond Mid-infrared Laser Pulses," *Scientific Reports* **9**, 19993 (2019).
[8] Keldysh, L., "Ionization in Field of a Strong Electromagnetic Wave," *Soviet Physics JETP* **20**, 1307 (1965).