Tuning chirality of laser-printed plasmonic nanoneedles via tailored spiral-shape pulses

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Abstract. Chiral mass transfer on the surface of plasmonic-active metals appeared upon their ablation with vortex laser pulses was recently found to be driven by a helical-shape temperature and corresponding surface tension gradients rather than optical angular momentum transfer from the incident beam. Here, we demonstrate that by properly designing and tailoring the spiral-shape intensity pattern used for direct single-pulse laser ablation, the chirality of produced nanoneedles can be controlled in a wide range of parameter. Such optimization of the laser intensity pattern governing the helical movement of the transiently molten metal allows to produce nanostructures with controlled chirality suited for various nanophotonics and biosensing applications.

1. Introduction

Optical vortex (OV) [1], a singular optical beam characterized by a helical wavefront (or an orbital angular momentum, OAM, $\ell$) and a circular polarization (or a spin angular momentum, SAM), has refreshed research interest in the area of laser-material processing [2]. Such interest can be generally attributed to the specific character of the OV-matter interaction. Particularly, an OV laser pulses were demonstrated to “twist” irradiated matter producing a helical relief on the surfaces of various materials as transition metals [3,4], semiconductors [5] and azo-polymers [6]. For transition metals, the appearance of such helical-shape nanorelief was initially described in term of the direct OAM transfer from the OV pulse to a transient material melt [3]. Meanwhile, recent experiments provided alternative explanation of this phenomenon based on spiral intensity distribution emerging as a result of constructive interference of the incident donut-shape OV beam and its reflected replica [4]. Further studies performed for plasmonic-active metal films ablated with specially designed spiral-shape intensity patterns having both zero OAM and zero SAM confirmed the suggestion made [7]. Additionally, it was qualitatively demonstrated that the chirality of the produced nanoneedles is affected by a number of spirals in the designed intensity distribution.

In this study, we designed and fabricated several diffraction optical elements (DOEs) generated various intensity patterns having variable number of intensity spirals. The chirality of the obtained nanoneedles defined as a number of turns per height unit was found to depend on the spiral intensity pattern.
2. Experiment
The spiral intensity patterns were obtained with the various binary spiral axicon with the phase given by:
\[
\tau(r, \varphi) = \begin{cases} 
0, & \cos(k\alpha_0 r + m\varphi) \geq 0, \\
\pi, & \cos(k\alpha_0 r + m\varphi) < 0,
\end{cases}
\]
where \((r, \varphi)\) are the polar coordinates, \(\alpha_0 = \lambda/d\) is the so-called numerical aperture of the axicon with the period \(d\); \(m\) is the topological charge of the OV, \(k = 2\pi/\lambda\) is the laser wavenumber at the wavelength \(\lambda\) (Figure 1a). The transmission function of the binary spiral axicon can be represented as a superposition of transmission functions of the scattering and collecting spiral axicons with the opposite topological charges. In this case, the total OAM of the generated focal-plane intensity distribution will be zero.

![Figure 1](image)

**Figure 1.** a) Designed phase pattern of the binary spiral axicon (white-color area denotes the phase of \(\pi\) rad, while the black one - 0 rad). b) Focal-plane intensity profile generated with the designed DOE. c) Experimental setup for laser nanotexturing. The inset explains the definition of the nanoneedle chirality. d-f) SEM images of the twisted nanoneedle printed with the double-spiral beam (Figure 1b) on the surface of a 500-nm thick Ag film at NA=0.42 and different incident fluences.

Single-pulse printing of twisted nanoneedles with the zero-OAM spiral intensity pattern was performed with a microfabrication setup (Figure 1c), using second-harmonic pulses of a Q-switched Nd:YAG nanosecond laser. The Gaussian laser beam was directed through a beam collimator and the transmissive DOE to generate a beam with a spiral-shape intensity pattern. This pattern was projected onto a sample surface by microscope objective with numerical aperture of 0.42. We used thick silver films with a thickness of 500 nm, which were deposited onto silica glass substrates, using e-beam evaporation. In the process of nanotexturing, the samples were mounted onto a three-dimensional motorized translation stage with a movement resolution of 150 nm. All structures were produced under single-pulse irradiation.

3. Results and discussions
SEM images in Figure 1(d-f) illustrate the surface modification produced at laser fluence slightly above the ablation threshold of the irradiated 500-nm thick film as well as a chiral nanoneedle formed.
at higher fluence of 2.2 J/cm². As seen, the produced structures are twisted in the clockwise direction, coinciding with the chirality of the double-spiral intensity pattern used (Figure 1b). Upon increasing the applied fluence, the geometric shape of the nanoneedle typically evolves as follows [7]. The formation of the protrusion starts in the center of the irradiated area via accumulation of the molten material. At further increase of the fluence, the height of this protrusion gradually grows. For a certain height, the liquid helical nanoneedle becomes hydrodynamically unstable decaying via ejection of the droplets. Taking into account this evolution, we further analyze the nanoneedles with the maximal height (aspect ratio), which can be observed at moderate fluences according to our previous studies. For such high-aspect ratio needles, we found direct correlation of their helical shape and the number of spirals in the designed intensity pattern used for single-pulse ablation.

![Figure 2](image.jpg)

**Figure 2.** a,d,g) Focal-plane intensity distributions of the DOE-generated beams designed to have 4, 6, and 8 spirals. (b-i) Top-view and side-view SEM images of the twisted nanoneedles produced on the surface of the 500-nm Ag film using the generated spiral-shape beams.

For these experiments, we have designed and produced several DOEs generating beams with a lateral intensity distribution having various number of spirals (Figure 2a,d,g). Presented SEM image clearly illustrate that the chirality of the needles produced with multi-spiral intensity pattern significantly increase being compared to the needles produced by simple double-spiral pattern (see Fig.1c,e,f). To further quantitatively analyze the obtained results, we classified the chirality of the high-aspect-ratio nanoneedles as a number of spiral turns normalized on the structure height (see inset in Figure 1c). Analyzing the geometry of the produced structures from this point of view, the chirality was found to increase gradually with the number of intensity spirals reaching its maximum value of 2.4 μm⁻¹ for six-spiral intensity pattern (Figure 3).

It is worth noting that using 8-spirals pattern results in decrease of the nanoneedle’s chirality. Apparently, it is due to the fact that on the target surface, the material doesn’t resolve spirals
individually and they become overlapped owing to thermal blurring of the intensity distribution governed by strong lateral thermal conductivity in the silver film irradiated by the laser pulse.

Figure 3. Chirality of the laser-printed nanoneedles versus the number of the spirals in the designed intensity pattern. The statistics for each intensity pattern having different number of intensity spirals is accumulated over 50 twisted nanoneedles produced under similar experimental conditions.

4. Conclusions

Herein, we showed that the chirality of the laser-printed nanoneedles can be tailored by precisely tuning the spiral-shape lateral intensity distribution used for single-pulse laser printing. Specifically, the chirality was found to increase gradually with the number of intensity spirals, reaching maximum value of 2.4 μm⁻¹ for six-spiral intensity pattern. Further increase of spiral number in the intensity pattern decreases the chirality of the laser-printed nanoneedles, apparently owing to thermal blurring of the intensity distribution governed by strong lateral thermal conductivity in the silver film irradiated by the laser beam. Presented flexible and high-performing approach is promising for making functional nanotextures with controllable chirality.

Acknowledgments

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