Direct laser printing of chiral plasmonic nanojets by vortex beams

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Direct laser printing of chiral plasmonic nanojets by vortex beams

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Abstract. Donut-shaped laser radiation, carrying orbital angular momentum, namely optical vortex, was recently shown to provide vectorial mass transfer, twisting transiently molten material and producing chiral micro-scale structures on surfaces of different bulk materials upon their resolidification. In this paper, we show that nanosecond laser vortices can produce chiral nanoneedles (nanojets) of variable size on thin films of such plasmonic materials, as silver and gold films, covering thermally insulating substrates. These results suggest optical interference of the incident and reflected laser beams as a source of complex surface intensity distributions in metal films, possessing spiral components and driving both center-symmetric and spiral thermocapillary melt flows to yield in frozen nanoneedles with their pre-determined spiral nanocarving.

1. Introduction
Chirality is a specific inherent feature, typically occurring in natural living organisms at almost all length scales from snails and sea shells to chiral molecules and DNA. Artificially designed nanoscale chiral-shaped structures, mimicking their natural analogues owing to unique ways of interaction with optical radiation, possess remarkable properties as circular dichroism, enhancement of nonlinear signals, highly directional emission and photoactivity. Meanwhile, modern methods of fabrication such chiral structures are rather expensive and small-scale (low-throughput) or limit the number of the available materials as well as the achievable size of the produced structures, that triggers, in its turn, search for alternative technologies. Recently, another alternative pioneering approach, employing optical radiation with specially designed intensity-, polarization- or phase states for fast fabrication of chiral structures, was demonstrated. More importantly, vortex laser pulses, carrying simultaneously orbital (OAM) and spin (SAM) angular momenta, were demonstrated to twist transiently molten metal, producing chiral-shaped micron-size needles [1]. Based on their experiments, the authors revealed that mass transfer and handedness of the produced surface structures are associated with corresponding phase helicity (or OAM), while the polarization helicity (or SAM) accelerates/decelerates the movement of the molten material.

In this work, we explore formation of twisted nanojets under direct translative ablation of Ag films of variable thickness, using nanosecond (ns) laser vortex pulses generated from circularly polarized pulses by an S-waveplate. We suggest an alternative explanation of the chiral nanoneedle formation on the Ag film surface based on a combination of straight and twisted thermocapillary flows of the molten material, which are triggered by the characteristic spiral-like intensity distribution, originating
from the interference of the incident donut-shape vortex beam and its replica reflected from transiently evolving molten metal surface.

2. Experimental details
A vortex beam, carrying OAM $l = \pm 1$ (per photon, in units of ħ), was generated by transferring second-harmonic radiation from a Nd:YAG laser system (Brio, Quantel: central wavelength – 532 nm, pulse duration – 4 ns, repetition rate – 20 Hz, maximal pulse energy – 50 mJ) through a Glan-Taylor polarizer and a quarter-waveplate (Fig. 1), to produce a circularly-polarized beam, and finally through a commercially available radial polarization converter (S-waveplate, Altechna). Then, generated ns-laser vortex pulses were focused onto the sample surface (Fig. 1(A)) by means of different microscope objectives (Xirox: NA = 0.45, 50x; Nikon Plan Fluor: NA = 0.3, 15x and NA = 0.65, 50x), yielding in donut-shape intensity distributions (Fig. 1(B)) with the outer optical diameter $D_{vortex}$ of ~2.05 µm, ~2.7 µm and ~3.9 µm for NA = 0.65, 0.45 and 0.3, respectively.

![Image](image_url)  
**Fig. 1.** (A) Schematic of the experimental setup for nanostructuring with the nanosecond vortex beams. (B) Vortex-beam intensity distributions measured in the focal plane of the microscope objectives with NA = 0.3 (top), NA = 0.45 (middle) and NA = 0.65 (bottom).

3. Results and discussion
Series of energy-resolved SEM images (Figs. 2(A)-2(D)) illustrates the formation and evolution of the twisted nanojets produced under single-pulse irradiation of the 500-nm thick Ag film, covering the silica glass substrate, with ns-laser vortex pulse focused at NA = 0.3. Besides the evident twisted shape of the produced nanojets with their clockwise rotational symmetry (the sign of vortex helicity), expectedly coinciding with the vortex pulse handedness (Figs. 1(C)-1(E)), their formation process appears to be very similar to that for common nanojets fabricated under single-pulse ablation of noble (semi-noble) metal films with Gaussian-shape (zero-OAM) ns-laser pulses.

The main energy-resolved steps include (i) accumulation of the molten material at the spatial center of the beam through thermocapillary forces and melt flows, which rapidly thin the peripheral part, (ii) formation of the liquid nanojet, which undergoes a Rayleigh-Plateau hydrodynamic instability, resulting in appearance and ejection of the molten droplets and, finally, (iii) formation of the through hole by breaking the significantly thinned area, surrounding the nanojet, at increased pulse energy. The tipped twisted nanojets are expectedly fabricated for typical pulse energies sufficient to trigger the ejection of all molten droplets from the resolidifying tip. Under such condition, the produced twisted nanojet can be characterized by very fine tips with the typical averaged curvature radius $R_{tip} \sim 12 \pm 6$ nm, as it was shown by the high-resolution SEM imaging (see inset in Fig. 2(C)).
Fig. 2. (A-D) Side-view (view angle of 40°) and normal-view (in false color) SEM image pairs of twisted Ag nanojets produced under single-pulse ablation of 500-nm thick Ag film with the vortex pulses focused at NA = 0.3. Similar side-view (view angle of 40°) SEM images showing twisted nanojets fabricated on the surface of the same Ag film with single vortices focused with microscope objective at NA = 0.45 (E-H) and NA = 0.65 (I-L). The pulse energy in each image row increases from left to right. Insets in figures (C) and (L) show the magnified view of the nanojet tip indicating the typical curvature radii smaller than 12 nm.

Similarity of our present observations and the previous extensive experience in fabrication of thin-film nanojets, using short- and ultrashort laser pulses, provokes us to compare formation of the non- and chiral nanoneedles. For Ag films studied in this paper, both Gaussian- and donut-shaped beams produce nanojets, having non- and chiral shapes, respectively. In particular, this indicates that for almost similar general mechanism, underlying the formation of the nanojets on the noble-metal film surface and attributed to the temperature-gradient driven thermocapillary flow of the molten material, strong rotational movement appears under vortex-pulse irradiation, twisting molten material in the direction, coinciding with the vortex helicity sign. The helical thermocapillary melt flows within the evolving nanojet and surrounding microbump area possibly can originate from the corresponding characteristic spiral-like (or more complex) intensity distribution. For the noble-metal films considered in this study, the origin of such spiral-shaped intensity distribution and corresponding temperature
profile on the metal film surface can be explained in terms of “pure” optical processes as interference of the incident donut-shaped vortex beam with the zero-OAM beam, having a spherical (or more complex) wavefront. It is clear that appearance of such perturbed reflected wave, providing the desired “spiral” component of the corresponding surface intensity distribution, can be associated with the transient nanosecond evolution of the metal film surface, which can be triggered via its heating, melting and thermal expansion by the incident donut-shaped laser pulse. Scalar diffraction theory calculations show that structure of a vortex beam reflected from a metal film can be perturbed during its reflection from a film, containing multiple submicroscale inhomogeneities with average size more than 20 nm. In this case, interference of the incident vortex beam and the reflected speckle-modulated zero-OAM one produces the spiral-like intensity distribution on the metal film surface, which handedness coincides with that one of the incident vortex beam (Fig. 3). To the contrary, similar calculation performed for a smooth metal mirror shows a common annular-shape interference pattern, originating from two counter-propagating vortex beams, retaining with opposite helicity and the similar OAM (this intensity distribution is equal to that one presented in left-most image of the Fig. 3). As any substantial surface evolution of the initially smooth metal film starts after electron-phonon relaxation (typical timescale - few picoseconds for both noble metal film and Si), the “heating spiral” as well as the helical shape of the nanojets are expected to be missing for ultra-fast energy deposition with the pulse durations shorter, than this characteristic time.

Fig. 3. Intensity distributions of the incident focused first-order vortex beam (left), distorted beam reflected from the rough metal surface (middle) and their interference pattern (right). The white arrows in the right-most image indicate the direction of the intensity gradient, which leads to the appearance of a “thermal spiral”.

4. Conclusions
To conclude, donut-shaped nanosecond vortex pulses generated from circularly polarized ones by an S-waveplate, were found to produce twisted nanojets under single-pulse translative ablation of Ag films. We suggest an alternative explanation of the chiral nanoneedle formation on the silver film based on a combination of straight and twisted thermocapillary flows of the molten material, which are triggered by the characteristic spiral-like intensity distribution originating from interference of the incident donut-shape vortex beam and its replica reflected from the transiently evolving molten metal surface.

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References
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