Microfabrication of Au Film Using Optical Vortex Beam

Sho Kawagoe¹, Ryosuke Nakamura², Ryohei Tasaki¹, Hiroki Oshima¹, Mitsuhiro Higashihata¹,

Daisuke Nakamura¹, Takashige Omatsu²

 ¹ Graduate School of Information Science and Electrical Engineering, Kyushu University, 744 Motooka Nishi-ku, Fukuoka 819-0395 Japan
² Graduate School of Engineering, Chiba University, 1-33, Yayoicho, Inage-ku, Chiba-shi, Chiba, 263-8522 Japan E-mail: dnakamura@ees.kyushu-u.ac.jp

We fabricate Au microneedle structures by irradiating nanosecond optical vortex pulse, possessing orbital angular momentum (OAM), to Au thin film. Twisted microneedle associated with the handedness of optical vortex is formed. The partial liquid motion of the molten Au film and the OAM transfer effects play a role to establish a twisted Au microneedle. An Au microsphere on a twisted pillar is also achieved. Non-twisted structure is fabricated with picosecond optical vortex pulse irradiation due to unoptimized condition such as laser fluence, film thickness and unavoidable imperfections of the optical vortex.

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1. Introduction

Optical vortex, i.e. light possessing orbital angular momentum, $\hbar L$, (OAM) per a photon owing to its spiral wave front, has a central dark core arising from its phase singularity. It is widely applied in the fields, such as stimulated emission depletion (STED) [1], optical trapping of microparticles [2], and a mass optical communication system [3]. Light also carries spin angular momentum, $\hbar S$, (SAM) per photon, associated with a circular polarization, thus, it exhibits a total angular momentum, $\hbar J$, (TAM) per photon, defined as the sum of OAM and SAM. Recently, application of optical vortex to laser materials processing has been paid attention, and several studies have been conducted. For example, fabrication of twisted needle structures and influence of the angular momenta to shape of the fabricated structures by irradiating optical vortex to metal and semiconductor bulk targets, such as Si and Ta have been reported [4-8]. More recently, twisted nanoneedle formation from an Ag film irradiated by a vortex beam was reported [9]. In addition, twisted metal nanoneedle structure formation was demonstrated by a spiralshaped light beam without OAM owing to thermal diffusion induced OAM [10]. Optical vortex and spiral-shaped light beam can fabricate spiral structures with chirality. Such chiral twisted metal needles have a potential to provide chiral selectivity to nanoscale imaging systems [4], chemical reactions on plasmonic nanostructures [11], and planar metamaterials [12]. In particular, plasmonic metal structures such as Au and Ag are attractive for plasmonic applications in visible wavelength region [13, 14]. In this study, we focused on Au thin film and performed microfabrication by irradiating a single optical vortex pulse, and investigated the laser fluence dependence on the microfabrication.

2. Experiment

A schematic of experimental setup is shown in Fig.1. Au film was deposited on a silica glass substrate for this experiment. The thickness was measured to be 450 nm by an atomic force microscope (AFM). A Gaussian beam pulse from a nanosecond Nd:YAG laser (Spectra Physics, 532 nm,



Fig.1 Experimental schematic of irradiation to Au film by Nd:YAG laser.

15 ns) or a picosecond Nd: YAG laser (Japan Laser Corp, 532 nm, 20 ps) was converted into a right-/left-handed optical vortex beam pulse through a spiral phase plate (SPP) (topological charge l = +1 or -1), azimuthally divided into 16 parts providing $n\pi/8$ phase shift (*n* means an integer between 0 and 15) to 532 nm beam. Also, we used a quarter-wave plate (QWP) to give SAM to the optical vortex pulse. The optical vortex pulse was focused on the Au film by an objective lens. The target was translated from the focal plane along a *z*-axis by a defocused distance, *z*, as shown in Fig. 1. All experiments were performed in room temperature and atmospheric pressure. We observed irradiated spot by a scanning electron microscope (SEM).

3. Results

SEM images of fabricated structures by irradiation of a single right-handed optical vortex pulse (pulse width: 15 ns) to Au thin film (450 nm) focused by an objective lens (×40, NA = 0.6) are shown in Fig.2. The pulse energy was fixed to



Fig.2 SEM images of irradiation points around focal point. " $z = 0 \mu m$ " means the closest point to focal point of objective lens in the experiment.



Fig.3 SEM images of the fabricated structures on Au thin film with different orbital angular momentums of (a) l = +1 and (b) l = -1.

0.9 μ J at all experiments. The focused laser beam exhibited a diameter of approximately 6.2 μ m at the focal plane, and its fluence was then estimated to be ~3 J/cm². Within *z*=-20~+20 μ m, a micro-sized hole with a crown-like rim was created. At *z* = ±30 μ m, a broken bump-like structure with a sphere on its top was fabricated. The fabricated structure was shaped to be a needles at *z* = +40~50 μ m, and it was further transformed to be a pillar with a sphere on its top was fabricated at *z*= +60 μ m. The fabricated structures at *z*= +50~60 μ m were then twisted towards a clockwise direction. Also, note that the positive spherical aberration of the objective lens produces caustics at *z*=-40 μ m, forming a ring-shaped rim. An aplanatic objective lens should reduce such distortions of the defocused beam.

Figure 3 shows SEM images of structures fabricated on the Au thin film (450 nm) by the irradiation of a single right-/left-handed optical vortex pulse (pulse width: 15 ns, pulse energy: $0.9\pm0.1 \mu$ J). The defocused distance *z* was then fixed to be 50 µm. The insets in Fig. 3 show top views of the structures. A clockwise/counter-clockwise spiral structure was fabricated by the optical vortex pulse deposition, and its spiral direction was fully assigned by the handedness of the optical vortex (Fig. 3). These results indicate that the OAM of optical vortex pulse was transferred to the molten Au to establish twisted microstructure [7].

Figures 4(a) and (b) show the fabricated structures by irradiation of a single right-handed optical vortex pulse (l =

+1) around 0.9 μ J. The inset in Fig.4(b) shows the top view of the fabricated structure. The clockwise spiral structure was formed during pinch-off of the molten Au film. A counter-clockwise spiral structure was also provided by the single left-handed optical vortex pulse (l = -1) as shown in Figs. 4(c) and (d).



Fig.4 SEM images irradiated by optical vortex pulse (l = +1) at (a) 0.9 µJ and (b) 1.0 µJ. Similarly, by optical vortex pulse (l = -1) at (c) 0.9 µJ and (d) 1.0 µJ.

Next, a picosecond OAM light beam was focused by an objective lens (×10, NA = 0.26) to be approximately 6.9 μ m (diameter) annular spot on the Au film with a thickness of 450 nm. Its pulse energy was then measured to be 0.35 μ J. Figure 5 shows the SEM images of fabricated structures with a single optical vortex pulse (l = 1). In Fig.5(a), a deformed needle with a small sphere on its top was created on a deformed bump surrounded by short nanoneedles. Such structure was interestingly reproduced, as shown in Fig. 5(b). A



Fig.5 SEM images of fabricated structures by irradiation of optical vortex (20 ps, $l = +1, 0.35 \mu$ J) to Au film with a thickness of 450 nm at (a) a certain spot and (b) another spot.

previous report mentioned that SAM reinforces the helical structure [6]. We performed the experiments by employing the circular polarized optical vortex pulse (l = +1, s = +1), however, no fine needle structures were fabricated. To create fine twisted needles by picosecond optical vortex pulses, further investigations should be needed.

4. Discussion

We discuss a mechanism of formation of Au twisted microneedle by optical vortex pulse. Wang et al. have experimentally and theoretically investigated the formation of structured matters, such as a bump and a nanojet with a spherical-shape droplet on its top, on the Au thin film by irradiating a femtosecond Gaussian pulse [15,16]. The Au film gains a separation velocity from substrate owing to laser-induced vapor pressure, and it is deformed. This film deformation is very sensitive to a heat affected zone determined by the laser fluence and the pulse duration. In fact, Morning et al. have reported that the deposition of a nanosecond Gaussian laser pulse also enables us to form a bump/jet structure even on a thick (650 nm) Au film [17]. This indicates that after melting and equilibration the Au film gains the separation velocity to form bump and jet-like structures. Thus, our experimental results can be explained by a following mechanism. Figure 6 shows a schematic of the mechanism. (i) The optical vortex-irradiated Au film is melted, while the film at the dark core of the irradiated optical vortex might be still solid-phase. After that, (ii) the film at the dark core is also melted due to excess of injected laser energy and



Fig.6 Schematic of twisted microstructure formation by optical vortex pulse irradiation to Au thin film: (a) laser-irradiated Au film changed into liquid-state, and vapor pressure and/or thermal stress of the liquid metal can be the driving force for partial liquid motion and formation of a microneedle and a droplet by surface tension, where OAM is transferred to the molten metal, producing orbital motion to twist the microneedle, and the twisted microneedle formed. (b) The droplet is pinched off at slight higher laser fluence. (c) When holes appear on the surface of the bump during fabrication, the bump partly remains.

thermal diffusion in of Au film (320 Wm⁻¹K⁻¹). Subsequently, (iii) vapor pressure and/or thermal stress of the molten Au acts as a mass-transport (partial liquid motion) driving force to collect the molten Au within the dark core. Also, OAM was then transferred to molten Au film. (iv) A droplet, a spiky microneedle, and bump are formed by surface tension. (v) The superfluous droplet is pinched off to establish a twisted microneedle with a microsphere on the bump, as shown in Fig. 6(a). The laser fluence of ~ 1.3 J/cm² in the experiment was comparable to that in previous reports by using ns laser pulse [17]. The droplet on the microneedle is pinched off when the laser fluence is slightly high, as shown in Fig. 6(b). The pinching off of the Au droplet strongly depends on the irradiated laser fluence as shown in Figs.4(b) and 4(d). Also, note that imperfections, for instance, slight distortion of the donut-shape, of irradiated optical vortex may induce the rupture of the bump to prevent the OAM transfer effects, as shown in Fig. 6(c).

Picosecond optical vortex irradiation also allows the formation of microneedle, indicating the partial liquid motion of the molten Au occurs after the laser irradiation. However, unaviodable imperfections of picosecond optical vortex with high peak intensity should significantly distort the structured matters. In fact, the optical vortex should carry undesired higher-order radial and azimuthal modes arising from the misalignment of the SPP. The non-uniform thermal diffusion effects, governed by many physical parameters, such as nonuniformity of the film, spallation of the surface layer, [18] etc., should further contribute to the distortion of structured matters [19]. Further investigation concerning ps pulse optical vortex processing is required to make clear the mechanism.

5. Summary

The several microstructures were fabricated by a single optical vortex pulse irradiation onto Au thin film. With ns pulse irradiation, twisted microneedle and micro sphere on the microneedle have been successfully fabricated. Twisted direction of structures was fully assigned by the handedness of the irradiated optical vortex. The mechanism of formation of the twisted microneedle is based on the partial liquid motion and OAM transfer effects. Fine structures might be achieved by ps optical vortex irradiation by further optimization of experimental parameters, such as the film thickness, irradiated laser fluence, etc. These metal microstructures are expected to be applied to plasmonic devices.

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