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Design of a patterned nanostructure array using a nanosecond pulsed laser

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For design the patterned nanostructure array (PNSA) on material surface using a nanosecond pulsed laser, we investigated the influence of phase shift between scattered lights on silicon (Si) substrate using 30-nm-wide gold lines (GLs) spacings. At a spacing of 5,871 nm, ten nanodot (ND) arrays were formed at intervals of 533 nm by nanosecond pulsed laser. The results show that the formation of the PNSA was affected by the resonance of scattered light. We conclude that ND arrays were formed with a spacing of $\Lambda = n\lambda$. And we have designed PNSA comprising two ND arrays on the substrate. The PNSA with dimensions of 1,600 nm $\times$ 1,600 nm was prepared using GLs. © 2018 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>). <https://doi.org/10.1063/1.5027060>

I. INTRODUCTION

Optomechanics focused on radiation pressure has attracted much attention in fields such as mechanical engineering. For example, it is well known that radiation pressure with high photon density can trap and accelerate the movement of micro-nanosize materials.^{1,2} Furthermore, micro-nanosize materials placed between the mirrors of a Fabry–Perot resonator can be moved by mechanical vibration to a stable position.^{3–6} Such examples suggest a causal relationship between light energy and mechanical vibration energy, which imply that the fluctuations and movements can be visualized as attraction and repulsion in these materials. It is now expected to increase precisely with the use of precision processing technology such as electron beam lithography (EBL).

Meanwhile, to makes changes to the local area of a material surface, the mechanism has been studied using nanosecond pulsed laser.^{7–13} The laser effects have been investigated so far because laser processing is advantageous for rapid production in air. The formation of nanodot (ND) arrays on silicon (Si) surfaces by means of a perpendicularly incident laser has been reported;⁷ the NDs, known as vidro-nanodots (VNDs), had an average diameter of 50 nm, were separated by ~ 110 nm, and had a periodicity of $\Lambda \approx 532$ nm. It was expected as the laser process for selectively fabricating more complex nanostructures.⁸ Although the technology for selectively manipulating VND arrays on Si surfaces by laser irradiation has the potential to trigger the development of novel functional devices, the comprehensive mechanism of light interference for the nanostructure array with a pulsed laser is not yet fully understood.^{7,8,14–16} To understand this mechanism, the formation of patterned nanostructure arrays (PNSAs) is investigated here using samples with separated gold lines (GLs) prepared on an Si substrate by high-resolution

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EBL. In this study, we design the fabrication of PNSAs using variously arranged and spaced GLs.

II. MATERIALS AND METHODS

A. Materials

The spatial patterns of GLs were fabricated on a Si substrate. The GLs were defined on Si substrates (10 mm × 10 mm) via high-resolution EBL conducted on a scanning electron microscope (ELS-770H; Elionix) working at a voltage of 125 or 130 kV. Conventional copolymer resist (ZEP-520a; Zeon) diluted with ZEP thinner (1:1) was spin-coated on the Si substrate (1,000 rpm for 10 s and 4,000 rpm for 90 s) and prebaked on a hot plate for 3 min at 180°C. The electron beam draw was conducted at a dose of $280 \mu\text{C cm}^{-2}$ and an electrical current of 1.2 nA. After the development (Figure 1), a 2-nm/30-nm chromium/gold bilayer was deposited by sputtering (MPS-4000; UL-VAC). Lift-off (resist removing) was then performed by immersion in an acetone solution for 2 min and in a dimethylformamide solution for 5 min in an ultrasonic bath. The GLs were 30 ± 10 nm wide and 30 ± 10 nm high, and samples were prepared with GL spacings from $1,100 \pm 30$ to $1,600 \pm 30$ nm and from $5,800 \pm 30$ to $6,200 \pm 30$ nm. A sample was also made with a fixed $1,600 \text{ nm} \times 1,600 \text{ nm}$ square GL pattern prepared on a Si substrate. Each sample was analyzed via scanning electron microscopy (SEM) (JSM-7001F; JEOL).

B. Methods

The sample surface was irradiated in air with an Nd:YAG pulsed laser (Inlite II; Continuum Co., Ltd.) at a wavelength of 532 nm, repetition frequency of 2 Hz, and pulse width of 5–7 ns at room temperature. The laser beam diameter was 6 mm, and irradiation was conducted normal to the surface at an average laser energy density of 1.24 kJ m^{-2} . The surface morphology of each specimen after 500 pulses of laser irradiation was observed via SEM.

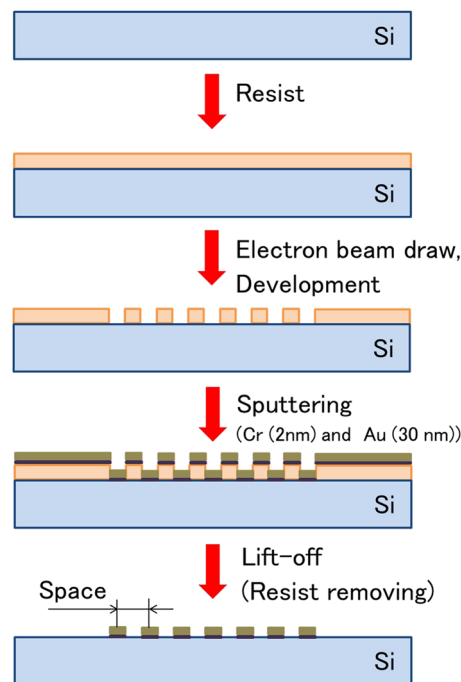


FIG. 1. Working fabrication of surface nanostructure on silicon (Si). Conventional copolymer resist diluted with ZEP thinner is spin-coated on the Si substrate and prebaked. Electron beam lithography is conducted at an adjusted dose and electrical current. A bilayer of gold (2 nm) and chromium (30 nm) is deposited by sputtering. Lift-off is performed by immersion in acetone and dimethylformamide solutions in an ultrasonic bath, and the line-and-space nanostructure is prepared on the substrate.

III. RESULTS AND DISCUSSION

We fabricated patterned ND arrays between the GLs using the same laser conditions, namely 500-pulse laser shots at an average energy density of 1.24 kJm^{-2} . Figure 2 shows SEM images of the surface morphology observed before and after laser irradiation. Figure 2(a) shows the various GL spacings before laser irradiation. Figures 2(b) and 2(c) show the surface of the local area after laser irradiation; an incomplete ND array was observed periodically at spacings of 1,096 and 1,200 nm. The GL spacing was more than twice the laser wavelength of 532 nm. The GLs clearly melted and spread after the laser irradiation; we assume that they were alloyed by the laser irradiation.⁹

At the GL spacing of 1,280 nm, the NDs were scattered between the GLs as shown in Fig. 2(d), and two incomplete ND arrays appeared between the 1,393-nm GLs, as shown in Fig. 2(e). Figure 2(f) shows two complete ND arrays formed with a spacing of 526 nm. They are clearly aligned at sample of the space between the GLs is close to an integral multiple of the wavelength because the 1,587-nm spacing is almost three times that of the incident laser wavelength. We consider that the spacing of the scatterers affects the formation of PNSA.

The discussion is supported by Fig. 3. Figure 3(a) shows the fluctuation of a nanostructure on the Si surface at $\Lambda \neq n\lambda$. From this result, we assume that the radiation pressure of the scattered light during the nanosecond laser pulses causes the attraction and repulsion that moves the scatterers on the Si substrate in cases of light scattering. While the scatterers attracted each other in the region of one incomplete ND array, the scatterers repulsed each other in the region of two incomplete ND arrays. Although the force created by radiation pressure that contributes to this process is very small, it would be sufficient to assist the liquid state at the nanoscale. As demonstration, the liquefied gold such as dewetting^{9,10,17} by thermal effect of the laser irradiation was stirred on surface. However, a complete ND array is observed in Fig. 3(b) for a GL spacing of $\Lambda \approx n\lambda$. These results support the assumption that this formation can be attributed to the resonance of scattered light between the GLs, as shown in Fig. 3(c). Figure 3(d) shows the tilted ND array at a GL spacing of 1,580 nm.

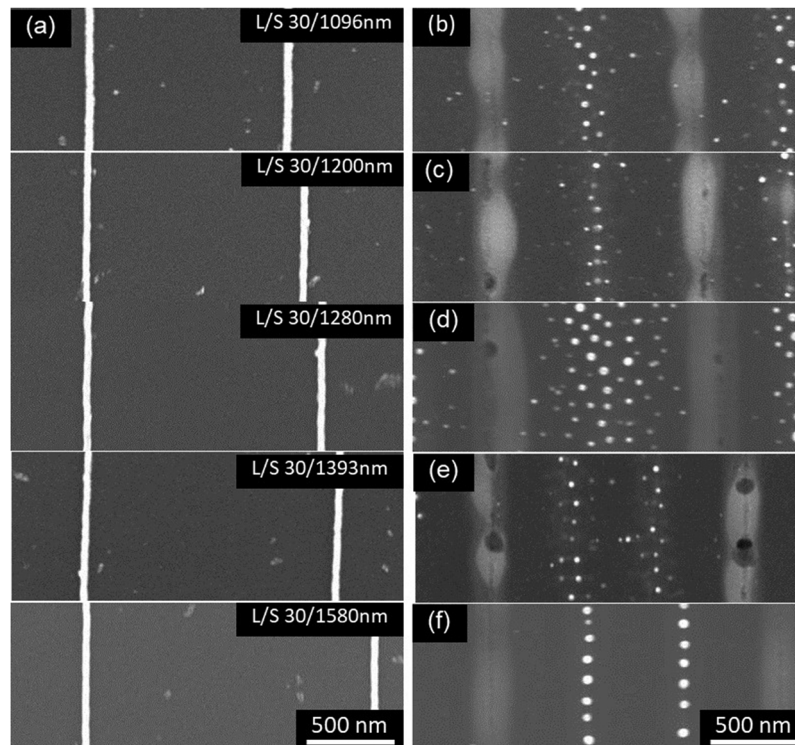


FIG. 2. Scanning electron microscope (SEM) images of surface nanostructure on Si substrate before (a) and after (b–f) irradiation by 500 pulses, with ordered lines and spacings of roughly 1,100–1,600 nm: (b) 1096 nm; (c) 1,200 nm; (d) 1,280 nm; (e) 1,393 nm; (f) 1,580 nm.

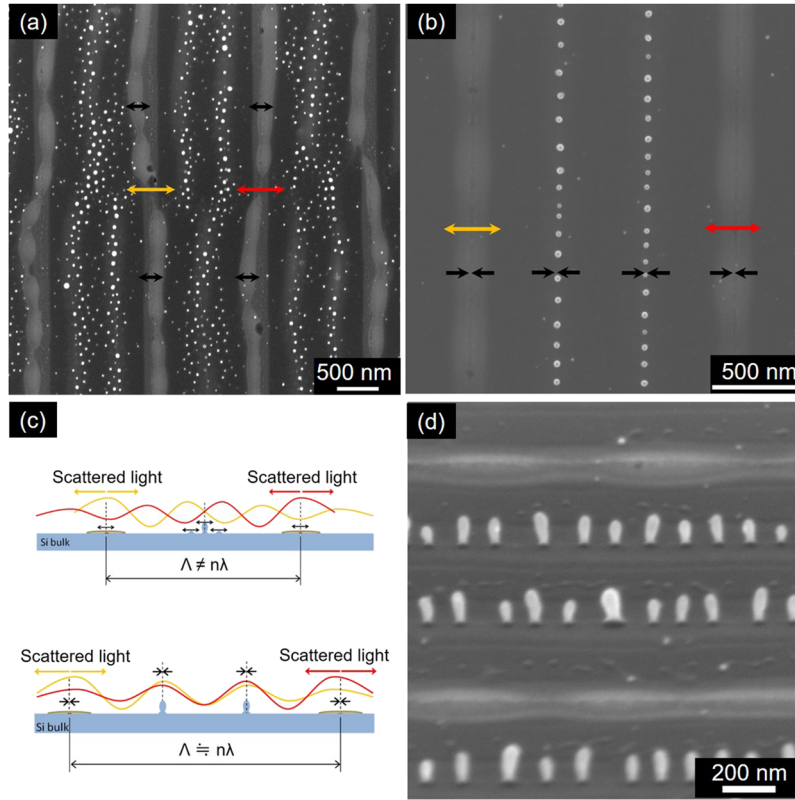


FIG. 3. SEM images and illustrations of nanostructure on Si substrate at spacings of (a) 1,280 nm and (b) 1,580 nm. (c) Schematic of scattered light on Si substrate. (d) Tilted sample of 1,580 nm.

The patterned ND arrays are formed in a VND shape,⁸ and the alloy structure presented is that of an Au/Si composite at a local area of $\sim 200 \mu\text{m}^2$.⁹

To understand the phenomenon of complete and accurate ND array formation, we also investigated the GL spacings from 5,793 to 6,207 nm to discuss the GL spacing. Figure 4 shows the SEM images of the surface morphology analyzed at GL spacings from 5,700 to 6,200 nm before

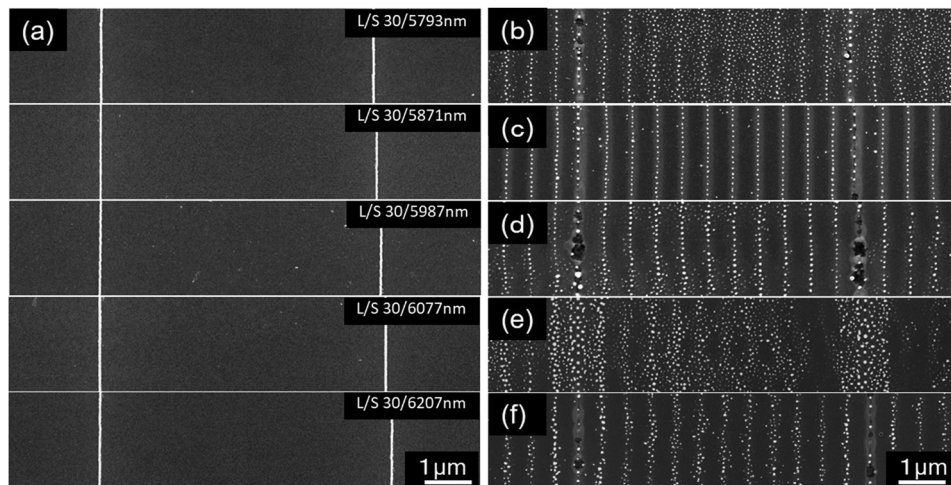


FIG. 4. SEM images of surface nanostructure on the Si substrate before (a) and after (b–f) irradiation with ordered lines and spacings of roughly 5,800–6,200 nm: (b) 5793 nm, (c) 5871 nm; (d) 5,987 nm; (e) 6,077 nm; (f) 6,207 nm.

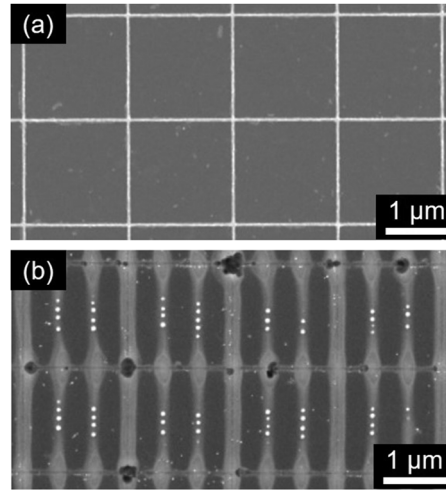


FIG. 5. SEM images of surface nanostructure on the Si substrate before (a) and after (b) irradiation in a $1,600 \times 1,600$ nm square with 500 pulses.

and after 500 pulses of laser irradiation at an average energy density of 1.24 kJ m^{-2} . An incomplete ND array was formed with the NDs scattered between the 5793-nm GLs, as shown in Fig. 4(b). A complete ND array was formed at a GL spacing of 5,871 nm, as shown in Fig. 4(c). The periodic spacing ($\Lambda = 533$ nm) of the complete ND array was nearly equal to the incident laser wavelength ($\lambda = 532$ nm). As the spacing was widened, the NDs gradually scattered as shown in Figs. 4(d) and 4(e). At a GL spacing of 6,077 nm, the surface structure fluctuated and its position was not fixed, as shown in Fig. 4(e). Although it was incomplete, the 12th ND arrays appeared at a GL spacing of 6,207 nm as shown in Fig. 4(f). The interference of scattered light affected the creation of the patterned nanostructures even at spacings of 10 or more times the wavelength, and the nanostructure present on the surface remained stationary (stable position) or moved because of attraction and repulsion due to the radiation pressure.

On the surface, the GLs on the Si substrate were broken into NDs by the thermal effect of the laser beam.¹⁸ The molten gold was pushed under the influence of the radiation pressure due to the scattered light and was patterned by phase matching between the scatterers. And the position error occurred because the phases of the scattered light are shifted from each other. This implies that the phase shift is related to this formation.

Finally, we used nanosecond pulsed laser irradiation to design a PNSA on the Si surface. Figure 5 shows the surface morphology observed in the $1,600 \text{ nm} \times 1,600 \text{ nm}$ square GL patterns before and after 500 pulses of laser irradiation at an average energy density of 1.24 kJ m^{-2} . It was fabricated two complete ND arrays which was founded in the $1,600 \times 1,600$ nm square GL patterns, as shown in Fig. 5(b); however, the PNSA was not formed in some local spots. Future work is required to understand the manner in which the accuracy of formation can be improved.

IV. CONCLUSIONS

We have demonstrated that the resonance of scattered light is assisted by preparing GLs with spacings from $1,100 \pm 30$ to $1,600 \pm 30$ nm and from $5,800 \pm 30$ to $6,200 \pm 30$ nm. From the results, it is concluded that the formation succeeded at spacings in excess of ten times the wavelength $\lambda = 532$ nm and that ND arrays were formed with a spacing of $\Lambda = n\lambda$.

And we have succeeded in fabricating a PNSA that was designed in the $1,600 \text{ nm} \times 1,600 \text{ nm}$ square GL pattern at a local area on the Si substrate. This result indicates that a laser beam may be a good tool for directly patterning a functional material at the nanoscale and microscale. By considering this resonance phenomenon, a complete ND array structure can be formed with high precision in a short time. We expect that the method could be expanded as a unique process of nanosecond pulsed

laser irradiation and high-resolution EBL in the field of nanotechnology. This laser process could be useful for design the PNSA on material surface.

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