Plasmonic Nanogap Arrays Fabricated via Moiré Holographic Lithography

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Metallic nanostructures with gap sizes in the order of tens of nanometers have gained a great interest in recent years due to their ability to create electromagnetic hotspots which can be exploited in sensing applications such as surface enhanced Raman scattering (SERS). Current top-down fabrication techniques for such structures include serial direct writing methods (e.g., e-beam lithography) and parallel-type multi-beam holographic lithography (HL) that both create a resist mask pattern for subsequent metal deposition and lift-off. While successful, the high cost and low throughput remains a drawback for such techniques. In this work, to alleviate such issues, we demonstrate the use of simple two-beam HL systems[1,2] (operating with a 325 nm HeCd laser) with the regulation of multiple exposures (i.e., superimposition of Moiré gratings) and the manipulation of the lithographic process conditions to produce nanogap arrays well below the diffraction limit of the irradiation source.

While a single exposure by two-beam HL results in 1D grating patterns, a second exposure with a substrate rotation is capable of producing complex lattices (Figure 1). While the multi-exposure with the fixed periodicity produces the structures of uniform lattice morphology (Figure 1a), the fabrication of periodic compound nano- and micro-scale features was enabled by superimposing the modulated periodicity between the multi-exposure steps (Figure 1b). Further, the lithographic process parameters, specifically exposure dose, were varied so as to control the geometry of the holographic compound nanostructures (Figure 2). Control of feature morphology via manipulation of the exposure dose is a common practice used in holographic lithography.[3] Lowering the exposure dose across both exposures in the case of patterning grating structures resulted in 3D hierarchical structures (Figure 2a→2b), whereas lowering the dose across all four exposures in making the square lattice patterns produced a quasi-periodic nanogap array with features separated by ~20 nm (Figure 2c→2d). The quasi-periodic nanogap array of resist then enabled us to produce plasmonic nanogap array of gold by lift-off process (Figure 3). The ~20 nm gap size of the gold nanostructures fabricated using this methodology is ~75% below the diffraction limit of conventional HL operating at the wavelength (325 nm).

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[1] I. Wathuthanthri, W. Mao, C.-H. Choi, ***Optics Letters*** 36, 1593-1595 (2011).
[2] W. Mao, I. Wathuthanthri, C.-H. Choi, ***Optics Letters*** 36, 3176-3178 (2011).
[3] I. Wathuthanthri, Y. Liu, K. Du, W. Xu, C.-H. Choi, ***Advanced Functional Materials***, DOI: 10.1002/adfm.201201814.

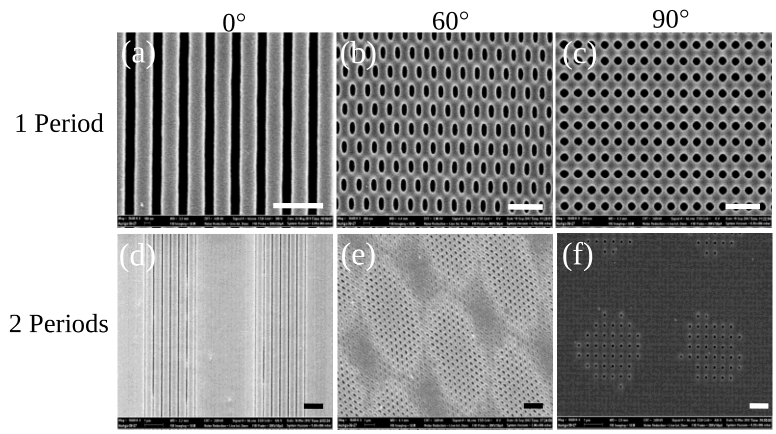

Figure 1: Moiré nanopatterns of various orders, made of resist by using holographic lithography. (a) After single exposure (500 nm period). (b-c) After second exposure (the same period of 500 nm) with substrate rotation of 60° and 90°, resulting in hexagonal and square lattice, respectively. (d) After double exposure with superimposed periods of 230 nm and 500 nm, resulting in compound nano/micro grating array. (e-f) With substrate rotation (60° and 90°) prior to 3rd and 4th exposure, resulting in microscale hexagonal and square lattice, respectively. Scale bar in each image represents 1 μm.

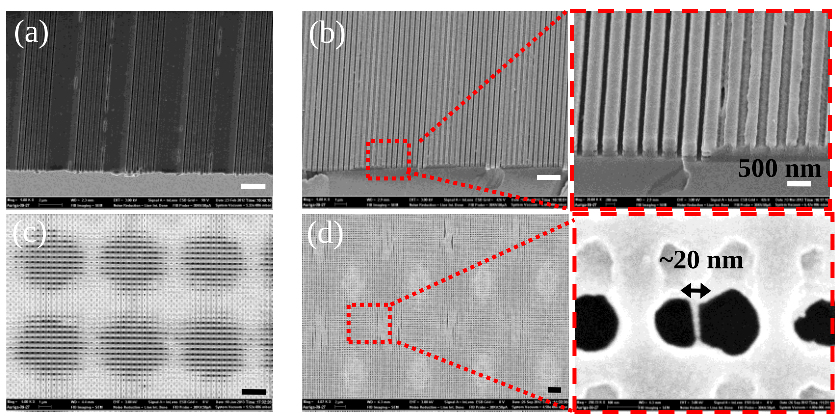

Figure 2: Effects of varying exposure dosage on compound structure morphology. (a) Grating structures made with regular dosage. (b) With lower dosage (33% less), resulting in hierarchical nanograting structures. (c) Square lattice structured made with regular dosage. (d) With lower dosage (65% less), resulting in the nanostructures with gap size of ~20 nm. Scale bar in (a-d) represents 2 μm.

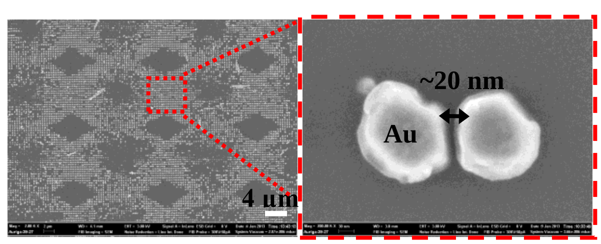

Figure 3: Quasi-periodic arrays of gold (Au) nanostructures made by lift-off process with the square-lattice compound nanostructures shown in Figure 2d. The higher magnification SEM image shows the gap size of ~20 nm between two adjacent Au nanostructures.