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Fabrication of an ordered nanodot array by thermal dewetting on a patterned substrate

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Abstract

A new process to fabricate an ordered gold nanodot array by thermal dewetting on a patterned substrate is proposed in this paper. Its feasibility was demonstrated through the experiments. The process comprises three steps; fabrication of a square nano-grooves grid on a substrate by nano-plastic forming, deposition of a thin gold film on the patterned substrate, and annealing the metal coated substrate to form an ordered nanodot array. The effects of the thickness of the deposited metal film on the morphology of nanodot array are studied. It is found that a nanodot array of good uniformity and regularity is obtained by optimizing the gold film thickness. It is verified that patterning of groove grid on a substrate is effective to improve the uniformity and regularity of the nanodot array in the thermal dewetting process.

Keywords: Nano-plastic forming; Metal nanodot array; Patterned substrate

Metallic nanodot arrays are attracting interests due to their unique optical property known as the localized surface plasmon resonance (LSPR). Utilizing LSPR, many studies have reported broad applications of metallic nanodots arrays such as in plasmonic solar cells [1], plasmonic sensing devices [2], and SERS substrates [3]. Nanodot array can be fabricated by the electron beam lithography (EBL) [4]. However, EBL is disadvantageous in terms of facility cost, stringency of process conditions, and complicate processes. In order to overcome these disadvantages of EBL, many researchers proposed self-organization processes for fabrication of nanodot arrays, such as thermal dewetting method [5]. However, many of these self-organization methods have drawbacks in controllability of dot morphology, i.e. uniformity of dot size and regularity of dot alignment.

The authors proposed new self-organization processes by thermal dewetting method to overcome the drawbacks of controllability of dot morphology; one is thermal dewetting of a patterned metal layer deposited on a flat substrate [6], the other is thermal dewetting of a metal layer deposited on a patterned substrate. In this paper, the latter process is focused on. Objective of this paper is to study effects of deposited film thickness on morphology of dots in order to improve controllability of dot morphology by thermal dewetting on the patterned substrates.

1. Experimental method and conditions

Figure 1 shows the proposed process to fabricate a nanodot array by using a patterned substrate. It consists of three steps; (1) fabrication of a square nano-grooves grid on a substrate by nano-plastic forming (NPF), (2) deposition of a thin metal layer on the patterned substrate, (3) annealing the metal coated substrate to form an ordered nanodot array.

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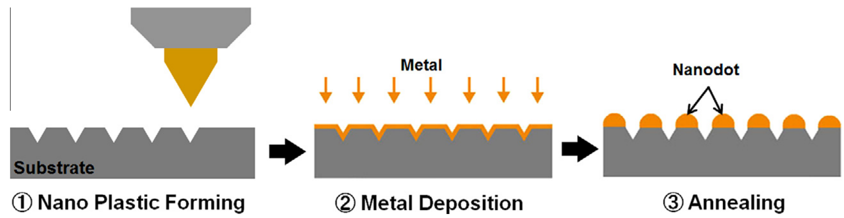

Figure 1. Schematic illustration of proposed fabrication process of an ordered nanodot array.

A quartz slide glass made by Saito Optics Co., Ltd. was used as a substrate material. The thickness of the quartz slide glass was 1 mm. The quartz slide glass was cut into plates of 10 mm × 10 mm in size. These quartz plates were cleaned by acetone using ultrasonic bath for 10 min.

Then, a groove grid pattern was fabricated on a quartz plate by the nano-plastic forming (NPF) method. Detail of the NPF technique is presented in reference [6]. In this paper, the indentation load was 1.0 N, and the distance between the grooves was 100 nm. Depth of the grooves was about 5 nm.

After fabricating groove grid on a quartz plate, the plate was again cleaned by ultrasonic acetone bath for 10 min.

Then, the patterned quartz plate was used as a substrate of gold deposition by DC sputter coating. The sputter gas was Argon, and gas pressure was 15 Pa. The sputtering current was kept to 5 mA during sputter deposition. In the experiment, the thickness of gold deposited film was controlled from 8 to 16 nm by adjusting the sputtering time. The gold deposited substrates were subjected to annealing under air atmosphere in an electrical furnace. Annealing temperature was 1000 °C, and annealing time was 30 min. After annealing, the specimens were taken out of the furnace and cooled down quickly in the air. The metal nanodots on the patterned substrate were characterized by a field-emission scanning electron microscope (FE-SEM).

2. Results and discussion

Figure 2(a–c) show the FE-SEM micrographs of the square grid patterns of nano-grooves fabricated on the quartz glass substrates by NPF, and deposited with gold film for different thicknesses: (a) 8 nm, (b) 12 nm, (c) 16 nm. It was confirmed that the square grid patterns were successfully fabricated on the quartz glass substrates. Figure 2(e–f) show the FE-SEM micrographs of nanodot arrays aggregated on the corresponding patterned quartz glass substrates in Figure 2(a–c) by thermal dewetting. Thicknesses of gold film were: (a, e) 8 nm, (b, f) 12 nm, (c, g) 16 nm.

In Figure 2(e), when the gold film was thin, the gold film aggregated into multiple small dots both on the grid square and the grooves. Dot size is not uniform, and the regularity of the nanodot array is poor. In Figure 2(f), when the thickness is 12 nm, the gold film mainly aggregated on the grid square, and formed one dot on each grid square. Therefore, good uniformity and regularity of nanodot array were achieved. The dots were aligned on a grid pattern, and the dot distance coincided with the distance between grooves preformed on the substrate. In Figure 2(g), when the gold film thickness is 16 nm, the uniformity and regularity of the nanodot array become worse because some dots merged with the adjacent dots, forming large dots randomly. On the other hand, Figure 2(h) shows nanodots aggregated on a quartz substrate without grid patterning (a flat substrate as in Figure 2 (d)), for comparison. Their dot sizes are not uniform, and their alignment is random. It was found that the patterning of grooves grid pattern on the substrate is effective to improve the uniformity and regularity of nanodot array when gold film thickness is proper.

Figure 3 shows the variation of the mean diameter of the nanodots and the relative alignment error (RAE) against the thickness of gold film. The mean diameter was defined as the average of diameters of each equivalent circle, whose area is equal to that of metal dot. The area of metal dot was calculated from the FE-SEM micrographs using a graphic analysis system (ImageJ software).

The vertical bars on the graph of the mean diameter indicate the standard deviations of the diameter. The RAE represents the degree of irregularity of a nanodot array. It is defined as the mean square root of deviation from the ideal position, i.e. the square grid pattern [6]. It is found that the mean diameter increases with the increase of gold film thickness. Meanwhile, the standard deviation of the dot diameter decreases with the increase of the gold film thickness when the thickness is less than 12 nm. The standard deviation becomes minimum at 12 nm, and it increases with the increase of the film thickness when the film is thicker than 12 nm. The variation of RAE is similar to that of standard deviation of dot diameter. RAE becomes minimum when gold film thickness is 12 nm. This implies that uniformity and regularity of the nanodot array becomes the best when the gold film thickness is 12 nm.

Figure 4 illustrates three types of dot aggregation patterns observed in the experiments. As shown in Figure 4(a), when the gold film is very thin, through thermal dewetting process, the gold film is well separated by the debossed grooves grid pattern since the ratio between the depth of debossed grooves on gold film and the thickness of gold film is fairly large. It is known that thermal dewetting of a thin film is caused by the nucleation of holes or voids which reach the substrate surface [7]. Groove at the grain boundary triple junction progressively penetrates through