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Laser interference lithography using spray/spin photoresist development method for consistent periodic nanostructures

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ABSTRACT

Generally, a simple immersion method for development of photoresist (PR) has been used to fabricate nanostructures by interference lithography (IL). However, the immersion method has the disadvantage that fabrication is inconsistent, especially for large-area periodic structures. Herein, we introduce the spray/spin PR development (SSPRD) method to fabricate periodic nanostructures using IL. By quantitative analysis and comparison, we characterized the effectiveness of the SSPRD method to develop PR. In our experiments the SSPRD method produced reliable uniform nanostructures, whereas the immersion method showed very poor consistency. In the SSPRD, rotation speed was very important: if it was too low the development speed differed between edges and center; if the rotation speed was too high it caused a distortion of nanostructures by unstable local flow induced by spraying and rotation So, to reduce this distortion, we adopted the puddle developing process; as a result the uniformity and repeatability of developed nanostructures were improved. These results demonstrate that the SSPRD method can be useful for fabrication of consistent periodic nanostructures.

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1. Introduction

Periodic nanostructures have numerous applications such as photonic crystals [1–3], cell culture plates [4–7], solar cells [8] and sensors [9–12]. Interference lithography (IL) is one of the powerful fabrication methods for periodic nanostructures without photomasks for different designs. Recently, IL has also been used to fabricate 3D nanostructures or coplanar multishape nanostructures [13,14]. However, IL has the disadvantages that the light source and optical components are expensive, and that it cannot easily be used for large-area fabrication; many efforts have been made to overcome these disadvantages [7,14–27]. Wathuthanthri et al. (2011) developed a two degree-of-freedom Lloyd-mirror interferometer for large-area application of IL, and fabricated periodic patterned nanostructures on a 4-inch silicon (Si) wafer [24]. Byun and Kim (2010) successfully developed cost-effective nanostructures by IL using an AlInGaN semiconductor laser which is relatively inexpensive (<US\$ 15,000) and has a very long coherent length $(\sim 20 \text{ m})$ for nanostructure formation on a large area [26]. Korre et al. (2010) also reported a compact, low-cost IL system [15]. These efforts have increased the feasibility of using IL to fabricate periodic nanostructures for real devices. However, for applications in devices to be successful, these nanostructures must be uniform in size and repeatable [28,29]. More recently, some researchers have reported about the uniformity of nanostructures, but no quantitative measurements have been presented [18,24,27,30].

Lithography is a process of patterning a photoresist (PR) by exposing and developing it; both steps can cause the pattern to be inconsistent. O'Reilly and Smith presented a model to explain the variation of exposure characteristics in a Lloyd's mirror IL system for uniformity relation [28,31], but research about the development process was not fully conducted. Also, the process of PR development can be influenced by differences in experimental conditions, and by human error, whereas the uniformity of the exposure process is mainly determined by the laser source and the optical components of the device used. For this reason, obtaining a consistent development process has been considered as a technical problem that is strongly influenced by human skill. However, this issue should be solved for the next step of IL; i.e., real device application [29].

For development of PR in IL, a specimen with an exposed PR layer is briefly immersed in developer solution. This 'immersion' method is very simple, but does not provide consistent results. Some agitation is needed for a complete development of exposed PR, but local nonuniform flow of the developer during immersion can degrade the uniformity and repeatability of the developed PR



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nanostructures. This problem can be crucial in large-area PR development due to the difficulty of applying the developer uniformly to the whole specimen. Industrially, the spray/spin PR development (SSPRD) method has been used, because it has three major advantages over the immersion method: (1) it reduces the amount of developer solution needed, (2) it produces nano-structures with good uniformity even in large-area fabrication, and (3) it has good repeatability. The second and third advantages are also important to when developing periodic nanostructures for real devices, so the SSPRD method can be an effective tool to improve nanostructure consistency. Here, we introduce and precisely analyze the periodic nanostructures obtained using the SSPRD method for large-area applications with consistent results using a Lloyd's mirror IL system.

2. Experiment

2.1. Importance of PR development process in IL

In the general photolithography process, a PR layer on a substrate (Fig. 1a) is exposed to ultraviolet (UV) light source through a photomask (Fig. 1b), and the exposed area of the PR layer (unblocked area by the photomask, a dark color in Fig. 1b) from UV light is clearly distinguished with the unexposed area (blocked area by the photomask, a light color in Fig. 1b). However, in IL, two coherent light beams interfere to generate a sinusoidal light intensity profile, not a square-wave profile (graph in Fig. 1c). The exposed and unexposed areas of the PR layer (a dark color and a light color in Fig. 1c, respectively) are not clearly separated. When the exposed PR is dissolved by a developer solution, an overdeveloped profile frequently occurs (Fig. 1d). In spite of a highly non-linear dissolution rate of PR as a function of UV intensity, the PR development process can be crucial in IL because of absence of the photomask, especially when fabricating nano-scale structures.



Fig. 2. Schematic drawing of experiment set-up for laser interference lithography.

2.2. Experimental setup

A 30-mW AlInGaN semiconductor laser with a 405-nm wavelength in single longitudinal mode (BCL-030-405-S, CrystaLaser), and a Lloyd's mirror interferometer with a mirror fixed perpendicularly to the specimen were used (Fig. 2). The beam from the laser is expanded and noise in the beam is removed by passing it through a spatial filter. Two beams reach the PR on the substrate, one directly and one after reflection from a mirror; due to differences in phase, they generate an interference pattern. The beam had an expansion length of 2.2 m (i.e., distance from a laser to a substrate), and a diameter > 20 cm when it reached the specimen holder. We used 2 cm \times 2 cm Si specimens to ensure that they were uniformly exposed in the center of the Gaussian beam.

2.3. Fabrication process

The Si substrate was cleaned for 15 min with a 4:1 (v:v) mixture of sulfuric acid (H₂SO₄) and hydrogen peroxide (H₂O₂), then immersed in buffered oxide etchant for few seconds to remove the native SiO₂ layer, then dehydrated on a hot plate at 150 °C for 10 min. After the substrate preparation, a 10:1 (v:v) mixture of propylene glycol monomethylether acetate (PGMEA) and



Fig. 1. Schematic drawings to illustrate the difference of development process between conventional photolithography and interference lithography. (a) prepared specimen, (b) photolithography using a photomask, (c) interference lithography without any photomask, and (d) difference of development degrees.

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