Fabrication of complex micro/nanopatterns on semiconductors by the multi-beam interference of femtosecond laser

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Abstract

Short periodic nanostructures with periods much smaller than the laser wavelength (1/10-1/2 λ) are fabricated on semiconductors after femtosecond laser irradiation. Laser polarization is an important factor to influence the formation of laser-induced nanostructures. Combining the fabrication of short periodic nanostructures induced by femtosecond laser with multi-beam interference, different types of complex micro/nanopatterns have been induced by the two-, three- and four-beam interferences of femtosecond laser with the modification of laser polarization combinations. The micro/nanopatterns are composed of two parts: two-dimensional long periodic micro-patterns and short periodic nano-patterns. Theoretical calculations of interferential polarization and intensity patterns for different polarization combinations explain well the experimental results. These researches have potential applications in the nanopatterning of semiconductors.

Keywords: femtosecond laser; complex micro/nanostructures; multi-beam interference; polarization; semiconductors

1. Introduction

Surface patterning of two-dimensional (2D) nanostructures has become increasingly important for applications in the fields of nanoelectronic, photonic crystals and biomedical devices. Holographic lithography (HL) is suitable for rapid fabrication of large-area periodic structures and becomes an important technology to fabricate various periodic...
patterns [1, 2]. Generally, one-, two- and three-dimensional patterns can be fabricated by interference of two beams, three beams and four (or more) beams, respectively. The number and spatial arrangement of interfering beams determine mainly intensity patterns. Laser polarization and phase also play important roles in interference patterns and are usually manipulated to improve the aspect ratio of periodic structures [3-6]. By adjusting the polarization and phase of laser beams, different microstructures, such as compound photonic crystals, magnetic metamaterials and plasmonic nanogap array, have been fabricated. Polarization combinations of the interfering laser beams are usually optimized for better interferential intensity patterns. However, there is little research concerning polarization patterns of the polarization-controlled interference.

Laser-induced periodic surface structures have been intensely studied in the last four decades. The periods were usually close to the laser wavelength, which was explained well by the interference between the incident laser and the surface scattered light [7-9]. Besides, nanostructures with periods much smaller than the laser wavelength (1/10-1/2 \( \lambda \)) are observed in semiconductors and dielectrics after femtosecond laser irradiation [10-17]. Laser polarization is an important factor for the formation of laser-induced nanostructures. In general, linearly polarized laser induces nanoripples, while circularly one induces nanoparticles. This is an attractive problem, and many explanations have been proposed to study the formation mechanism of the subwavelength periodic nanostructures [10-12].

In this paper, combining HL technology with femtosecond laser-induced nanostructures, we fabricated various types of complex micro/nanopatterns on ZnO surface by adjusting the laser polarization combinations of multi-beam interference. Theoretical calculations of laser intensity and polarization patterns accord well with experimental results. By using this method, diversity of surface nanopatterns fabricated by multi-beam interference can be greatly improved without any further change of the beam alignment. These micro/nanopatterns have the potential applications in optical nanofabrication and polarization diffraction elements [13, 14].

2. Experimental

The experiments were conducted on a commercial Ti: sapphire regenerative amplifier operating at 1 kHz repetition rate (Legend Elite, Coherent), which generated a linearly polarized laser at a wavelength of 800 nm with a pulse width of 50 fs and a maximum pulse energy of 3.5 mJ. A combination of a half-wave plate and a Glan polarizer was used to adjust the laser intensity and polarization. The laser beam was split with same intensity by the beam splitters. Each laser beam was focused by a lens with the focal length of 250 mm, and wave plates were used to adjust the polarization of each beam, respectively. Laser pulses of the laser beams arrived at the sample simultaneously, and the zero temporal delay point was determined by the signal of sum frequency via a \( \beta \)-barium borate (BBO) crystal.

The sample was a commercially available c-cut ZnO (0001) single crystal with size of 10×10×1 mm\(^3\). The two surfaces were optically polished with a roughness less than 10 nm. The sample was mounted on a \( XYZ \) translation stage controlled by a computer. After laser irradiation, the ablation areas were observed with scanning electron microscope (SEM).

3. Results and discussions

3.1. Complex micro/nanopatterns fabricated by two-beam interference of femtosecond laser

When the two-beam interference of femtosecond laser was performed, the laser beam was split into two beams with the same intensity and polarization by a beam splitter. Figure 1(a) shows the sketch of two-beam interference. The two beams denoted by two red lines propagate in \( xz \) plane and are focused on the same spot of ZnO simultaneously. The cross angle (2\( \theta \)) is set as 13.9°. Figure 1(b) shows the calculated intensity distribution of two-beam interference in \( xy \) plane with a grating period of \( \lambda/2\sin\theta \), where the intensity of each beam was normalized. Figures 1(c) and 1(d) show the 2D periodic surface structures fabricated by the interference of two femtosecond laser beams. The long periodic grating is determined by the interferential intensity pattern (see Fig. 1(b)). Besides, there are short periodic nanoripples with a period of 200 nm embedding in the long periodic gratings (see Fig. 1(d)). The nanoripples were induced by the femtosecond laser pulses, and the orientation was perpendicular to the laser
polarization. The long periodic micro-grating and the short periodic nanoripples made up the complex periodic micro/nanopatterns.

Fig. 1. (a) Sketch of two-beam interference; (b) calculated intensity pattern of two-beam interference in xy plane; (c) ablated spot irradiated by the femtosecond laser pulses; (d) the magnified SEM image of the black box in (c). The inset in (d) indicates the laser polarization combination.

3.2. Complex micro/nanopatterns fabricated by three-beam interference of femtosecond laser

Figure 2(a) shows the sketch of three-beam interference, and the cross angle between any two beams was about 14.8°. Figures 2(b-d) show the experimental results for three types of polarization combinations shown in the insets of Figs. 2(b), 2(c) and 2(d), respectively.
The two-dimensional spots with a period of 3.6 µm formed hexagonal long-periodic micropatterns. Meanwhile, we surprisingly found periodic nanostructures on the micropatterns. Figures 2(b) shows the SEM image of ablation area for parallel polarization of the three laser beams. The short period of nanoripples was about 200 nm with orientation perpendicular to the laser polarization. In Fig. 2(c), the circularly distributed nanoripples formed around each bulgy micro-spot. Besides, there were some nanoparticles formed on the six dots around a bulgy spot. The period of nanoripples and the diameter of nanoparticles were about 200 nm. We changed the laser polarizations of the three laser beams by 90°, and obtained different complex nanostructures shown in Fig. 2(d). The nanoripples were radially orientated on each bulgy spot, which formed another type of complex micro/nanopatterns.

### 3.3. Complex micro/nanopatterns fabricated by four-beam interference of femtosecond laser

Figure 3(a) shows the sketch of four-beam interference, and the convergence angle between any laser beam and z axis was about 10.6°. Figures 3(b-d) show the experimental results for three types of polarization combinations shown in the insets of Fig. 3(b), 3(c) and 3(d), respectively. In Fig. 3(b), the square micropatterns were determined by the intensity pattern of the four-beam interference. The period of the micropattern was 3 µm, which was in accordance with the calculated value of 3.08 µm. Additionally, there were several nanoripples with a period of 200 nm embedded in each spot. These nanoripples were perpendicular to the laser polarization.

We adjusted the polarization combination as that shown in the inset of Fig. 3(c), and observed, surprisingly, very different micro/nanopatterns. The spindly micro-spots were covered with two types of nanoripples, which were referred to as the vertical-oriented ripples (VOR) and the horizontal-oriented ripples (HOR). The cross-linked VOR and HOR nanopatterns structured the binary arrays of the complex micro/nanostructures.

Furthermore, by changing the laser polarization geometry, we can manipulate the cross-linked micro/nanopatterns. In Fig. 3(d), the polarizations of laser beams A, B, and C were rotated by 90°, and the beam D was horizontally polarized. Each spot exhibited hyperbolic nanopatterns with the straight nanoripples in the middle and the incurved nanoripples on the edge. Additionally, the relative ablation depths of the cross-linked VOR and HOR micro/nanopatterns also changed. The HOR micro/nanopatterns were ablated slightly comparing with the VOR ones.
3.4. Theoretical explanations

In order to understand the micro/nanopatterns described above, we calculated the interference patterns of the laser intensity and the polarization according to the theory of optical interference. The laser beams were supposed as plane wave, and expressed as Jones vector for different polarizations. The electric field of laser beams were normalized and the initial phase \( \varphi \) were set as zero. We conducted vector synthesis of the electric field of the laser beams for different position on the interference area. Consequently, we can calculate the distribution of compound electric field, and obtain the patterns of polarization and intensity in the interfering area.

Figure 4 shows the experimental and theoretical results for the polarization combination shown in the inset of Fig. 2(c). Figure 4(a) shows SEM image of a lattice of the complex periodic micro/nanopatterns. Figures 4(b), 4(c) and 4(d) are the theoretical calculation results for the same polarization combination as that of Fig. 4(a). Figure 4(b) is the intensity pattern, which is in accordance with the hexagonal long periodic bulgy spots. The polarization is described by the polarization ellipticity (Fig. 4(c)) and the polarization orientation (Fig. 4(d)). Figure 4(c) shows the distribution of polarization ellipticity, where 0 represents the linearly polarization and 1, the circular one. The light field on the six dots around a bulgy micro-spot are nearly circularly polarized and nanoparticles formed on the corresponding positions (see the white circle parts of Fig. 4(a)). The light polarization is linear on the three symmetrical axes and elliptical elsewhere. The linearly polarized laser pulses can induce nanoripples, while elliptical ones induce short nanoripples (see Fig. 4(a)). The orientation of the nanoripples is usually perpendicular to the polarization or the major axis of ellipse. We calculate the polarization orientation denoted by the angle \( \alpha \) between the major axis of ellipse and \( x \) axis. The angles \( \alpha \) on the three symmetrical axes are 30°, 90° and 150°, respectively. The embedded nanoripples are all orthogonal to the light polarization (see Fig. 4(a)). Besides, the angle \( \alpha \) rotates by 360° round a bulgy micro-spot, so the ring-like nanoripples form there. Theoretical and experimental results indicate clearly that long periodic micropatterns are determined by interferential intensity distribution and nanopatterns depend on the interferential polarization patterns. Theoretical calculations for the other polarization combinations of three- and four-beam interference are also conducted, and the results also explain well the corresponding complex periodic micro/nanopatterns. Therefore, different patterns of intensity and polarization are produced according to the corresponding polarization combinations due to vector superposition principle and the directions femtosecond laser-induced nanoripples are always perpendicular to the laser polarization, which result in the various micro/nanopatterns.
Fig. 4. (a) SEM image of a lattice of complex micro/nanopatterns fabricated by three-beam interference of femtosecond laser. (b), (c) and (d) theoretical results of the patterns of light intensity, polarization ellipticity and polarization orientation, respectively.

4. Conclusion

In summary, the complex micro/nanopatterns were obtained by the two-, three-, and four-beam interference of 800nm femtosecond laser. The micro/nanopatterns included the two-dimensional long periodic micro-spots and the short periodic nanoripples and nanoparticles. Theoretical calculations indicated that the long periodic micropatterns were determined by the interferential intensity distribution and short periodic nanopatterns depended on the interferential polarization pattern. By adjusting the polarization of laser beams, various types of complex micro/nanostructures can be fabricated, which have potential applications in the field of nanofabrication by femtosecond laser.

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References


