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Fabrication of a two-dimensional periodic microflower array by three interfered femtosecond laser pulses on Al:ZnO thin films

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Abstract. A two-dimensional periodic microflower array was fabricated on Al:ZnO thin films by the irradiation of three interfered 800 nm femtosecond laser beams. The petals of the microflowers are ~200 nm in size, and the intervals between the nanopetals are about 100 nm. These values are significantly smaller than the laser wavelength and its diffraction-limited scale. The evolution of microflowers with different laser fluences and irradiation times was analyzed. Theoretical analysis indicates that the interferential intensity and polarization distribution together account for the formation of microflowers with subwavelength petals. Flower periodicity with a period of microns was determined by the interferential intensity distribution, while nanopetal structures and their orientations were attributed to the interferential polarization distribution.

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1. **Introduction**

The interaction between ultra-short lasers and materials has become the focus of intensive research due to the ultra-high intensity and ultra-fast properties of ultra-short lasers [1, 2]. Many interesting phenomena have been found, which are different from the case of CW or pulse lasers with a long duration. Among them, short-periodic ripple-like nanostructures generated by a single femtosecond laser beam have been well reported on various materials, including semiconductors [3]–[10], metals [11]–[13] and dielectrics [14]–[18]. The periodic structures orient perpendicular to the direction of the laser electric field with periods much smaller than the laser wavelength. Various subwavelength structures such as nanogratings [12, 17] and nanoparticles [19] induced from one single femtosecond laser beam indicate that a femtosecond laser has great potential in the application of nanofabrication. It is a pity that the irradiation of a single femtosecond laser beam can only induce one-dimensional ripple-like patterns or particles, which are relatively simplex. On the other hand, semiconductor thin films have broad applications in various photonic and electric devices due to advantages of easy growth, capability of large area coating and low cost. However, investigations on laser-induced semiconductor nanostructures have been mainly conducted on semiconductor crystal surfaces [3]–[10]. In order to obtain a flexible application in nanotechnology, it is necessary to develop some more powerful techniques to fabricate more complex micro/nanopatterns, especially on thin solid films.

ZnO is an important semiconductor and attracts great interest for its potential in a variety of practical applications. It has a wide bandgap of 3.3 eV at room temperature [20], making it promising for UV photonic applications. A high exciton-binding energy of 60 meV [21] ensures stable excitonic emission even at room temperature. Al-doped ZnO film is a promising transparent conductive material for application in liquid crystal and plasma display panels and in optical devices such as solar cells and light emitting diodes [22]. Various interesting ZnO nanostructures, including nanobelts, nanobridges and nanonails, have been fabricated by thermal evaporation of oxide powders [23, 24]. Flower-like ZnO nanostructures have also been synthesized by a cetyltrimethylammonium bromide-assisted hydrothermal process [25]. In addition, laser-induced nanoripples, nanoparticles and nanosquare structures on the surface of ZnO crystals have been reported recently [8, 9, 26, 27].

Multi-laser-beam interference has been often used for the holographic fabrication of periodic microstructures [28]–[32]. The microstructures are usually of simple type such as microholes and micro-orbicular platforms [32]. Some work was done to obtain more complex microstructures and to investigate the fine structures of micropatterns down to subwavelength scale. Jia et al [33] reported that three interfered femtosecond laser beams could induce...
various complex periodic micro/nanostructures on the surface of a 6H-SiC crystal with certain laser polarization configurations. In this paper, we further demonstrate that two-dimensional, periodic microflower array structures are well fabricated by the irradiation of three interfered femtosecond laser pulses on Al:ZnO thin films. The present work shows the ability to grow complex microflower arrays by using the three-laser-beam interference technique on the surface of thin solid films. The petal sizes of the microflowers are only around 200 nm. In order to better understand the involved phenomenon, the evolution of microflower array structure is explored in depth with changes of laser fluence and irradiation time. Following a contrastive experiment conducted with single femtosecond laser beam irradiation, the interferential intensity and polarization distributions are calculated in order to explain the formation of microflower structures.

2. Experimental

The sample was Al-doped ZnO thin film with a thickness of 500 nm deposited on fused silica substrate by dc reactive magnetron sputtering with a target of 98 wt.% ZnO and 2 wt.% Al₂O₃. A generatively amplified Ti:sapphire laser was used in our experiments with pulse duration of 50 fs, a central wavelength of 800 nm, a pulse energy of 3.5 mJ and a pulse repetition rate of 1 kHz. The laser beam went through a half-wave plate and a Glan-Laser polarizer to obtain variable pulse intensity. Then it was split into three beams with equal pulse intensity by two beam splitters. By adjusting the optical path difference via mechanical delay lines, the three beams finally focused simultaneously on the sample at the same point by three lenses. Three half-wave plates were introduced to rotate the polarizations of the three beams. The geometric configuration of the three laser beams is shown in figure 1. The left and right parts of figure 1 are a side-view and front-view of configurations corresponding to the sample, respectively. The three beams were incident symmetrically on the sample with the same obliquity. The electric fields of the three laser beams crossed over each other at an angle of 60° in a plane parallel to the sample surface, as shown in the right part of figure 1.

3. Results and discussion

Figures 2(a) and (b) show the scanning electron microscope (SEM) images of the ablation area under the irradiation of three interfered femtosecond laser pulses on Al:ZnO thin films.
A regular two-dimensional periodic microflower array structure is clearly shown in figure 2(b), which presents the magnified SEM image in the white square marked in figure 2(a). The laser fluence and irradiation time are 0.14 J cm\(^{-2}\) and 0.1 s, respectively. The microflowers have sizes of \(\sim 2.5 \mu m\), and array two-dimensionally with a period of 3.6 \(\mu m\). The petals of the microflower are spokewise with sizes of \(\sim 200 \text{ nm}\), and the intervals between the nanopetals are about 100 nm. The petal size and petals interval are significantly smaller than the diffraction-limited scale of a single laser beam with a wavelength of 800 nm.

Figure 3 shows the evolution of the microflower periodic array with changes in laser fluence and irradiation time. In figures 3(a)–(c), the irradiation fluence is fixed at 0.083 J cm\(^{-2}\) for a single laser beam with different irradiation times of 0.5, 2.5 and 10 s, respectively. For 0.5 s irradiation time or 500 shot numbers, the microflower array is regular with a period of 3.6 \(\mu m\), flower sizes of 3.5 \(\mu m\) and petal sizes of around 200 nm. On increasing the irradiation time to 2.5 s, some particles with sizes of \(\sim 100 \text{ nm}\) are clearly seen in the central part of the regular triangle enclosed by three neighboring microflowers. The petals extend toward the flower center and become partly ablated. For 10 s irradiation, the petals extend more to the center, get mostly ablated and become illegible. In figures 3(d)–(f), the irradiation time is fixed at 0.1 s with changing laser fluences of 0.083, 0.14 and 0.22 J cm\(^{-2}\), respectively. For a laser fluence of 0.083 J cm\(^{-2}\), the microflower array is regular with a period of 3.6 \(\mu m\), flower sizes of around 3.5 \(\mu m\) and petal sizes of around 200 nm. When the fluence increased to 0.14 J cm\(^{-2}\), the microflowers become separate with the whole size reducing. Further increasing the laser fluence to 0.22 J cm\(^{-2}\), the microflowers gradually disappeared. The appropriate laser fluence and irradiation time are requisite in order to obtain a regular microflower array with certain shape.

As is well reported, subwavelength patterns can be induced under single femtosecond laser irradiation. In order to analyze the underlying mechanism involved in three interfered femtosecond laser beam experiments, we also conducted the experiment with single femtosecond laser beam irradiation. Figure 4(a) shows the experimental results under single linearly polarized femtosecond laser beam irradiation. The laser fluence and irradiation time are 0.42 J cm\(^{-2}\) and 2.5 s, respectively. Ripple-like structures with sizes of \(\sim 200 \text{ nm}\) are formed in a period of 250 nm. The ripple direction is perpendicular to the electric field direction.
Figure 3. Microflower periodic array with different laser fluences and irradiation times. (a)–(c) Different irradiation times with a laser fluence of 0.083 J cm$^{-2}$; (d)–(f) different laser fluences with an irradiation time of 0.1 s.

When irradiated by a circularly polarized femtosecond laser beam (laser fluence of 0.50 J cm$^{-2}$, irradiation time of 2.5 s), nanoparticles with sizes of around 100 nm are induced regularly, as shown in figure 4(b). Compared with the experimental conditions in figure 3(b), the irradiation time in figure 4 is the same, whereas the laser fluences in figures 4(a) and (b) are 5 and 6 times larger than that of the single laser beam in figure 3(b), respectively. However, the laser fluence in figure 4 is close to the strongest intensity point for three interfered laser beams, which is 4.5 times that of the single laser beam, as will be calculated in the next part. The ripple and particle sizes in figures 4(a) and (b) are close to those in figure 3(b). Several models were developed to explain the formation mechanism of these laser-induced nanostructures, such as interference [17], Coulomb explosion [19] and self-organization [34]. Recently, Shen et al [3] and Juodkazis et al [35] proposed that the refractive index of the interaction region played an important role. The exact model for the formation of nanoripples is still a matter of debate. In this paper, we will not explore what accounts exactly for the formation of nanoripples. What we are interested in is the relation between the mechanisms for the subwavelength petal structure from three interfered laser beams and for one single laser-induced nanostructure.
In figures 5(a) and (b), we calculated the distribution of three-beam-interfered laser intensity and polarization. The three laser beams were assumed to be plane wave and expressed as the Jones vector for different polarizations,

\[ E_m(r) = E_{0m} \cdot \exp[i(\omega t + k_m r_m + \phi_{0m})] \begin{pmatrix} \cos \theta_m \\ \sin \theta_m \end{pmatrix} \quad (m = 1, 2, 3), \]

where \( \theta_m \) is the angle between the laser polarization and the x-axis. The three beams were normalized, and the initial phase \( \phi_{0m} \) was set as zero. A vector synthesis of the three beams, \( E(r) = \sum_m E_m(r) \), was conducted for different positions within the interference area. The distribution of the compound electric field leads to patterns of polarization and intensity \( I(r) = |E(r)|^2 \). Figure 5(a) shows the calculated result for the intensity distribution in an area of 10 \( \times \) 10 \( \mu \)m. The hexagonal area enclosed by the dashed line represents the experimental part as shown in the inset of figure 5(a), which is cut from figure 3(b) with a laser fluence of 0.08 J cm\(^{-2} \) and an irradiation time of 2.5 s. The position with theoretically stronger intensity becomes more ablated. The intensity distribution determines the elementary contour and induces microflower periodicity. Figure 5(b) is the theoretical polarization distribution.
with the same laser conditions as those in figure 5(a). The polarization is described by the polarization ellipticity, where 0 represents the linear polarization and 1 represents the circular one. Lines with arrows, circles and ellipses are guides to the eye to denote the electric field oscillating type in its corresponding position. The white parts with circular polarization denoted in figure 5(b) correspond to the vertexes of the dashed-line-enclosed hexagon in figure 5(a). The light polarization is linear on the three symmetrical axes and elliptical elsewhere. The linearly polarized laser pulses can induce nanoripples, whereas the elliptical ones induce short nanoripples [36]. The orientation of the nanoripples is usually perpendicular to the polarization or the major axis of the ellipse, leading to radially arranged nanopetals, which is analogous to periodic nanoripples with single laser irradiation as shown in figure 4(a). With appropriate laser fluence and irradiation time, nanoparticles related to circularly polarized laser irradiation can be clearly seen in the central part of the regular triangle enclosed by three neighboring microflowers in figure 3(b), which corresponds to the vertexes of the hexagon in figures 5(a) and (b). The above analyses indicate that the formation of microflowers with subwavelength petals is attributed to both interferential intensity and polarization distribution. There is an analogy between the nanopetals for the three interfered laser beams and the nanoripples for a single laser beam. The difference is that three-beam interference induces two-dimensional periodicity, and leads to more complex micropatterns embedded with fine nanostructures. Since the irradiation of a single femtosecond laser beam could induce subwavelength ripple-like nanostructures on various materials, including semiconductors [3]–[10], metals [11]–[13] and dielectrics [14]–[18], the three-beam interference technique is promising in the fabrication of complex micropatterns, including microflowers on various materials under appropriate excitation fluence, laser wavelength, polarization combination and irradiation time.

4. Conclusion

In conclusion, we have fabricated a two-dimensional periodic microflower array structure on Al:ZnO thin films by using three interfered femtosecond laser pulses. The petals of the microflower are spokewise with sizes of \( \sim 200 \text{ nm} \), and the intervals between the nanopetals are about 100 nm. The petal size and petals interval are significantly smaller than the diffraction-limited scale of a single laser beam with a wavelength of 800 nm. The evolution of micropatterns with different laser fluence and irradiation time is analyzed. The appropriate laser fluence and irradiation time are requisite in order to obtain a regular microflower array with a certain shape. Theoretical calculations show that flower periodicity with a period of microns is determined by the interferential intensity distribution, while petal nanostructures and their orientations are attributed to the interferential polarization distribution, which is analogous to nanoripples induced by a single femtosecond laser. Compared with single laser irradiation, three interfered femtosecond laser beams can induce a more complex structure, which have great potential applications in the field of nanofabrication.

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