Study on Short-Pulsed Laser-Induced Periodic Surface Structures Assisted by Mechanical Processing

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論文内容要旨

Fine structures have been studied largely to provide materials with functionalities such as a reduction in friction, change of the wettability, the antireflection of light and an improvement of biocompatibility. Ultraprecision cutting, ultrasonic machining and photolithography have been widely used to create these functional surfaces, however, these traditional methods have many disadvantages such as complicated processing steps, environmental burden and difficulty processing large area and three dimensional shapes.

A laser is also a suitable method to fabricate fine structures, and an ultrashort-pulsed laser can fabricate fine periodic surface structures called as LIPSS (laser induced periodic surface structures) through self-organizing phenomena. An ultrashort-pulsed laser is a promising method for solving problems associated with traditional fabrication methods and providing functions by LIPSS.

However, this method still has such problems as low repeatability and difficulty controlling LIPSS since the principles and the phenomena of fabricating LIPSS have not been clarified completely. Moreover, a ultrashort-pulsed laser which unstably irradiates at high cost have mainly been used to fabricate LIPSS since it has been reported that the laser with shorter pulse duration than the collisional relaxation time (CRT) can fabricate LIPSS. Industry needs a laser with as long pulse duration as possible to stably fabricate LIPSS at low cost. Hence, in this dissertation, fabrication of LIPSS using a short-pulsed laser with 20 ps pulse duration was proposed and investigated to clarify principles and phenomena of fabricating LIPSS. In addition, the machining-assisted short-pulsed laser was proposed to investigate the effects of the surface geometry before laser irradiation on fabrication of LIPSS and to control LIPSS.

This thesis consists of seven chapters.

Chapter 1 gives the introduction of this dissertation. Various functionalities produced by fine structures on material surfaces and fabrication techniques of them are elucidated. A ultrashort-pulsed laser, capable of fabricating LIPSS in an efficient way, is stated as an effective method to fabricate fine structures. On the other hand, its issues concerning stability of laser irradiation, equipment cost, principles, phenomena and control of LIPSS are pointed out. In addition, principles and phenomena of fabricating LIPSS were stated, and the effects of laser irradiation conditions and environments on fabrication of LIPSS were studied. The interference between incident lights and scattered lights, surface plasmons or the parametric decay, one of them changes the electric field distribution and ionizes atoms inducing the Coulomb explosions periodically on the material surface. After the CRT, the heat distribution causes either the ablation or the inhibition of LIPSS, resulting in LIPSS. Dependency of the pitch length of LIPSS on the laser wavelength, the laser fluence and the incident angle is pointed out. Next, the pulse duration of a laser used in this study was decided to 20 ps, about the upper limit of CRT, via calculation of the CRT to clarify the principles and phenomena of fabricating LIPSS. Moreover, low-temperature environment experiment is introduced to fabricate sharp LIPSS easily and effectively due to extension of the CRT. Finally, the objectives and organization of this dissertation were drawn.

In Chapter 2, fabrication of LIPSS using the short-pulsed laser with 20 ps pulse duration was investigated on SUS304 surfaces to verify the effects of laser irradiation conditions and experimental environments on the fabrication of LIPSS. The fundamental experiments were conducted, that the 20-ps pulse laser was irradiated on a substrate, and LIPSS were fabricated at the specified conditions. The pitch length of LIPSS and the height of LIPSS slightly increased with increasing the energy density. Electromagnetic field analysis was performed using an FDTD simulation to investigate the effects of the electric field distribution on fabrication of LIPSS and to predict the effective conditions and the shapes of LIPSS. The simulation demonstrated that the decrease of the laser wavelength increased the electric field intensity at the bottom of grooves and decreased the pitch length of LIPSS, and the electric field intensity is large just at the bottom of grooves under a low temperature. Subsequently, Experiments were conducted to verify the effects of the laser irradiation conditions on fabrication of LIPSS. The pitch length of LIPSS was close to the laser wavelength, and increased with increase of the incident angle. In addition, LIPSS were fabricated sharply at 223 K, in contrast to those at 293 K and the pitch length of LIPSS depends on the laser wavelength regardless of the material temperature.

Chapter 3 describes the effects of material characteristics (the refractive index, the extinction coefficient and the reflectance) and the crystal structures on fabrication of LIPSS. The FDTD simulations were performed under the configuration of various material properties to verify the effect of material characteristics, especially the refractive index, the extinction coefficient and the reflectance, on fabrication of LIPSS. SUS304, Ti and Ni-P were conducted as the substrate. The results demonstrate that the low LIPSS with the long pitch length are fabricated on the Ni-P surface, and the pitch length of LIPSS of SUS304 and Ti will be same but their height of SUS304 will be greater than that of the Ti surface because it has the lowest refractive index, extending the wavelength of incident lights. Subsequently, experiments were conducted with different materials to verify the effects of material characteristics on fabrication of LIPSS. The work materials were SUS304, Ti and Ni-P. LIPSS were fabricated on all materials perpendicular to the direction of the laser polarization. The pitch length of LIPSS on the Ni-P surface was calculated to be about 0.85 times as long as the laser wavelength of 1064 nm, while that of LIPSS on the SUS304 and Ti surfaces were about 0.75 times as long as the laser wavelength. The height of

LIPSS decreased in the order of SUS304, Ti and Ni-P, according with the analytical results. Next, the effects of the crystal structure on fabrication of LIPPS was investigated with short-pulsed laser irradiation at low energy density on SUS304 surfaces. The surface topographies considered as crystal grain boundaries were seen on irradiated areas and an electron backscattered diffraction (EBSD) pattern analysis was performed to confirm the appearance caused by the crystal structure and the appearing structures after the irradiation of the short-pulsed laser depend on the crystal structure. Moreover, the fabricated LIPSS varied with crystal orientations. Additionally, the three-dimensional topographies were analyzed to verify the difference in expansion on each crystal orientation and the geometry of LIPSS depended on the atom arrangement which varies with the crystal orientation.

In Chapter 4, the short-pulsed laser assisted by the magnetic abrasive finishing (MAF), capable of creating straight nanogrooves, was proposed to investigate the effects of the initial surface geometry on fabricating LIPSS and to fabricate straight and high-aspect-ratio LIPSS since LIPSS follow depressions and debris. In order to investigate the effects of nanogroove geometry on the electric field distribution by laser irradiation and predict LIPSS on the MAFed surface, an FDTD simulation was performed and influence of the groove pitch on the electric field is larger than that of groove depth, in the case of nanogrooves, causing large ablation and removal of surfaces. Subsequently, experiments were conducted to fabricate straight LIPSS with high aspect ratio and to investigate the effects of the groove geometry on fabrication of LIPSS. Straight nanogrooves with various depths and different pitches were fabricated on all surfaces by using MAF. A short-pulsed laser was irradiated on them to fabricate straight and high-aspect-ratio LIPSS and to investigate the effects of the nanogroove geometry on the fabricated LIPSS. The direction of laser polarization was perpendicular to the direction of MAF grooves. Straight LIPSS were fabricated on all MAFed surfaces with various scratching depths, whose direction was perpendicular to the laser polarization. . The aspect ratio of LIPSS on the Ni-P and SUS304 surfaces fabricated using the MAF-assisted short-pulsed laser are approximately 0.2–0.25 and 0.4–0.7, respectively. From analytical and experimental results, the surface geometry before laser irradiation as the key factor is pointed out to control LIPSS since grooves facilitate the induction and the propagation of the surface plasma waves.

In Chapter 5, ultraprecision cutting prior to short-pulsed laser irradiation was proposed to investigate the effects of the groove geometry on fabricating LIPSS and to fabricate micro/nanostructures with microgrooves and LIPSS. The FDTD simulation was conducted to investigate the effects of straight microgrooves on the electric field distribution and intensity by laser irradiation, causing fabrication of LIPSS. Ni-P, an amorphous material, was used. Compared with the effects of the pitch length of the microgrooves on the electric field, high intensity was also detected on the surface with narrow-pitch microgrooves, causing ablation and large removal of material. Subsequently, experiments were conducted to fabricate straight LIPSS with microgrooves and to investigate the effects of the surface shape (straight microgrooves) on the fabricated LIPSS. Straight microgrooves with different pitches and depths were created on the flattened surface of an Ni-P substrate by using ultraprecision cutting, and the processed surface was then irradiated by a short-pulsed laser. Straight LIPSS were fabricated on all Ni-P surfaces compared to LIPSS on the flat surface, whose direction was perpendicular to the laser polarization. The pitch length of LIPSS independent of the surface shapes after ultraprecision cutting in both cases. The height of LIPSS on the processed surfaces is larger than that on the flattened surface, and increased slightly with shortening grooves pitch of the processed surface due to intensification of surface plasma waves. Microgrooves are effective to expand the aspect ratio of LIPSS. Finally, micro/nanostructures were successfully fabricated using the short-pulsed laser assisted by ultraprecision cutting and ultraprecision cutting prior to irradiation with a short-pulsed laser is effective to fabricate micro/nanostructures.

In Chapter 6, the effects of the groove angle on fabrication of LIPSS by laser irradiation on the surface with microgrooves changing the angle of laser polarization were investigated to fabricate LIPSS in complex structures. First, in order to investigate how groove-polarization angle makes the direction of LIPSS follow grooves or the laser polarization, a short-pulsed laser was irradiated on the surface with microgrooves changing the direction of the laser polarization. The direction of LIPSS is attributed to the direction of the grooves when $\varphi = 45^\circ$–90° since microgrooves favorably induces the surface plasma waves. The newly proposed fabrication method of complex LIPSS uses a short-pulsed laser assisted by ultraprecision cutting were tried to fabricate complex LIPSS. The created zigzag and crosshatch microgrooved surfaces were passed through short-pulsed laser irradiations to fabricate complex LIPSS. As the results, the surface shape before laser irradiation is the important factor for fabrication and control of LIPSS, and a short-pulsed laser assisted by machining can control LIPSS and fabricate complex structures.

In Chapter 7, the general conclusions of this research were given out.

In this dissertation, a short-pulsed laser was proposed to clarify principles and phenomena of fabricating LIPSS and to fabricate LIPSS effectively. The short-pulsed laser capable of fabricating LIPSS and the geometry of LIPSS depending on laser irradiation conditions, material characteristics and crystal structures were stated. Moreover, the machining-assisted short-pulsed laser irradiation was proposed to investigate the effects of the initial surface geometry on fabrication of LIPSS and to control LIPSS. The simulations and experiments demonstrated the initial surface geometry as the key factor for control of LIPSS and the effectiveness of machining-assisted short-pulsed laser to fabricate straight and high aspect ratio LIPSS. Finally, micro/nanostructures and complex fine structures with zigzag and crosshatch shapes can be fabricated using the machining-assisted short-pulsed laser.

The results of this dissertation will let to the development of laser processing and largely spread the machining-assisted short-pulsed laser among the industry for fabrication of various functional interfaces.

論文審査結果の要旨

近年，様々な製品において省エネルギ化や高機能化に対する需要が高まっており，材料表面に微細構造を創成し機能性を付与する研究が数多く行われている。従来，機能性表面作製技術としてフォトリゾグラフィなどが挙げられるが，加工能率が低く，より高能率の加工技術が要求される。そこで，高い加工能率を有する超短パルスレーザによる微細周期構造創成技術が注目されている。しかし，加工原理・現象が明らかになっておらず，構造の制御が困難である。さらにパルス幅が短くなるほど，コストが高く，レーザ照射が不安定となり，長期的な安定性に欠けるといった問題が存在する。

本研究では構造創成において限界と考えられるパルス幅 20 ps の短パルスレーザを用いて原理・現象の解明，構造の制御を狙っている。電磁場解析および照射実験により検討を行い，加工メカニズムを明らかにし，構造の創成現象の単純化を行っている。また，材料により創成される構造が異なる現象に関して，3 種の金属を用いて，解析および実験により検証を行い，特定の材料物性により形状が異なること，さらに結晶構造によって構造の形状・方向が異なることを明らかにしている。さらに，産業への大幅な普及にあたり，材料や結晶構造に関係なく構造を制御することが要求される。そこで，世界初となる機械加工援用短パルスレーザによる表面微細周期構造の創成・制御を提案している。その結果，構造の直進性・アスペクト比の向上，マイクロ・ナノ複合構造の創成，さらに今まで創成が困難であった複雑な微細構造の創成に成功している。本論文はこれらの研究成果をまとめたもので，全編 7 章からなる。

第 1 章は序論であり，本研究の背景，加工原理，目的および構成を述べている。

第 2 章では，短パルスレーザによる表面微細周期構造の創成プロセス，照射条件の構造形状への影響について，電磁場解析および照射実験により検討を行っている。その結果，パルス幅 20 ps の短パルスレーザにより構造創成が可能であり，材料表面に形成される微小な凸が構造創成のトリガーとなること，構造の形状が照射条件に依存することを明らかにしている。これはレーザによる表面微細周期構造創成の産業への大幅な普及に向けた重要な成果である。

第 3 章では，構造創成に対する材料の影響について SUS304，Ti，NiP を用いて，解析および実験により検討を行っている。その結果，構造の形状は消衰係数，屈折率，イオン化エネルギにより決定すること，さらに結晶構造により構造の形状が異なることを明らかにしている。加工現象の単純化は，材料および照射条件の選定の指標となる重要な成果である。

第 4 章では，磁気粘性流体研磨援用短パルスレーザによる表面微細周期構造の制御を提案し，解析および実験により検討を行っている。その結果，レーザ照射前の表面粗さ，表面形状が構造の創成・制御において重要であること，新加工法により直進性およびアスペクト比の向上を実現している。これは加工原理・現象の解明に対して重要な知見である。

第 5 章では，超精密切削援用短パルスレーザによる表面微細周期構造の制御，マイクロ・ナノ複合構造の創成を提案し，解析および実験により検討を行っている。その結果，直進性・アスペクト比の向上，マイクロ・ナノ複合構造の創成を実現している。これはこれまで困難であった構造の制御を可能とする重要な成果である。

第 6 章では，超精密切削援用短パルスレーザによる複雑微細構造の創成を提案し，実験的に検討を行っている。その結果，これまでレーザにより創成されたことのないジグザグおよびクロスハッチの構造の創成に成功している。これは新たな微細構造，より高付加価値の機能性表面の創成を可能とする重要な成果である。

第 7 章は結論である。

以上要するに本論文は，加工現象を単純化し，構造の制御を可能とする新たな技術を提案するものであり，従来よりも長いパルス幅のレーザにより構造創成を可能とし，要求される構造の形状に対する材料および照射条件の選定指標を与えるものである。さらにレーザ照射前の表面形状を制御することで，構造を制御することを可能としている。これは工学的に高い意義を有する。また，本研究で得られたレーザによる微細周期構造の創成メカニズム，結晶構造による影響，レーザ照射前の表面粗さ・表面形状による影響に関する知見は，これまでほとんど研究が行われておらず，加工原理・現象の解明に対して有益な知見であり，学術的にも意義深く，機械機能創成およびナノ精度加工学の発展に寄与するところが少なくない。

よって，本論文は博士(工学)の学位論文として合格と認める。