

Research Progress of Laser Shock Micro/Nano Forming Technology

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Keywords: Micro/nano forming, Laser shock, Metals, Micro/nano structure, Plastic deformation

Abstract: Metal micro/nano forming technology is broadly used in semiconductors, microelectronics, chips, MEMS, and other fields. Because the superplastic flow of metal materials requires an extremely high strain rate, it is still challenging to accurately and widely fabricate the high-quality micro/nano metal structures which are three-dimensional and complex. Laser shock micro/nano forming can trigger the superplastic flow of metal materials by laser-induced ultra-high pressure and conform to the geometric structure of micro/nano die with ultra-high strain rate, so as to realize the rapid formation of large and three-dimensional structure that is high-precision and complex. This paper focuses on the ultra-high strain mechanism of laser-induced material deformation and expounds on the laser-matter interaction. The forming methods of micro/nano structure metal surface prepared by laser shock and nanosecond structure surface covered in situ are described in detail. This paper also discusses the molding strategy of laser shock micro/nano forming and the prospects for its future development.

1. Introduction

Micro/nano forming refers to the plastic forming of parts or structures with micro/nano dimension [1-3], which is widely used in semiconductors, microelectronics, chips, MEMS, and other fields [4-6]. At present, lithography is the most widely used technology with high manufacturing precision and mature process. However, it is often limited by the processing of silicon-based materials and is mostly used for the manufacturing of planar micro/nano structures. It is still unsatisfactory in the manufacturing of complex three-dimensional micro/nano structures, the processing of extensible materials, and the complexity of systems [7-11].

Currently, nano-imprint lithography, microcontact imprint, micrometastasis casting, two-photon polymerization additive manufacturing, and other technologies have been applied to micro/nano forming manufacturing of three-dimensional complex structures, but they are often difficult to be applied to large-scale and high-precision micro/nano forming manufacturing of three-dimensional structures of metal materials [7, 9, 12-14]. Because the superplastic flow of metal materials requires an extremely high strain rate, it is still challenging to accurately and widely fabricate the high-quality micro/nano metal structures which are three-dimensional and complex [9, 15]. Although ultrafast laser removal and focusing ion beam can meet the requirements of precision and complex structures, the manufacturing efficiency and cost are too high, greatly hindering the application of this technology [16, 17]. Meanwhile, it's difficult to balance large-scale manufacturing and ultra-high precision resolution, which increases the difficulty of micro/nano forming of metal materials [18]. In 2014, Gao et al. developed a laser shock micro-nano forming method, which can realize high-precision micro/nano forming of large metal materials [19]. Its forming mechanism is that the ultra-high pressure induced by laser triggers the superplastic flow of metal materials and conforms to the geometric structure of micro/nano die with ultra-high strain rate, thus realizing the rapid formation of large and three-dimensional structure that is high-precision and complex.

Therefore, this paper will summarize the latest research progress on laser micro/nano forming, focusing on the ultra-high strain mechanism of laser-induced material deformation and expounding the laser-matter interaction. The forming methods of micro/nano structure metal surface prepared by laser shock and nanosecond structure surface covered in situ are described in detail. This paper also discusses the molding strategy of laser shock micro/nano forming and the prospects for its

future development.

2. Physical Mechanism of Material Forming Induced by Laser Shock

The interaction mechanism between laser and matter is determined by the optical properties of the laser, the physical and chemical properties of matter, and the external environment. With the laser irradiating on the surface of materials, due to the differences in physical and chemical properties of matters, different matters such as metals and nonmetals have various mechanisms with laser [20]. Metal materials realize laser-material energy coupling through the inverse bremsstrahlung effect. There are a large number of free electrons in the metal. Through electron-photon coupling, laser energy is converted into electron energy, and energy is transferred between electrons and metal lattice, thus completing the energy coupling between electrons and lattice before realizing the absorption of laser energy by materials [21-23].

According to the pulse width, the pulsed laser can be divided into nanosecond laser, picosecond laser, and femtosecond laser. The effects of pulsed lasers with different time scales on metal materials are different. For long pulse laser [nanosecond laser], the main mechanism is the thermal effect. For short-pulse and ultrashort-pulse lasers, because the pulse width is equivalent to the electron-photon coupling time and electron-lattice coupling time scale, its main mechanism can be reflected by thermal effect and non-thermal effect. The influence of the external environment on the interaction mechanism between matter and laser is mainly realized through the ambient gas environment, absorption layer, and confinement layer. From the perspective of macroscopic physical phenomena, the response process of metal materials to laser can be divided into phase transition, heating, melting, gasification, and plasma production [24, 25]. Figure 1 shows the mechanism and application of laser-material interaction. According to the difference in material's response to laser, its non-thermal effect is the force effect of laser. Using the force effect of laser processing materials, materials can be strengthened, formed, cavitated bubbles, and cleaned. Using the thermal effect of laser processing materials, cutting, drilling, patterning, heat treatment, and surface cladding of materials can be realized.

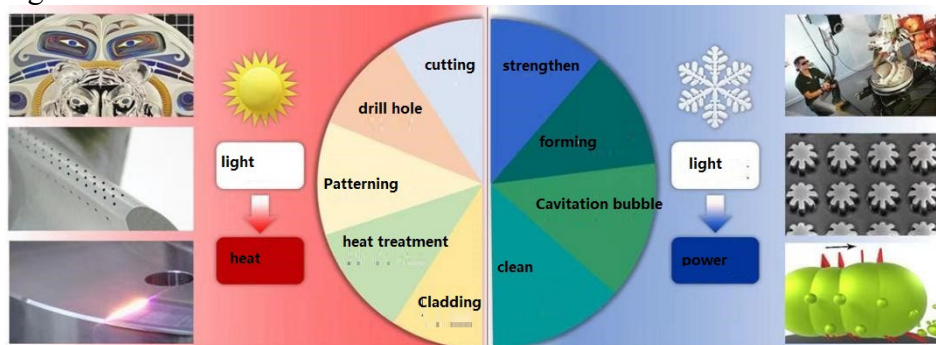


Fig.1 Applications and Mechanism of Laser-Matter Interaction

As for laser-induced materials to produce plasma, its effects can be summarized as plasma shielding effect, plasma force and heat effect, and plasma combustion effect [23, 26, 27]. Laser-induced plasma can realize the mechanical effect on materials through plasma expansion. Specifically, when the laser irradiates the material surface, the metal material absorbs the laser energy and produces high-temperature and high-pressure plasma clusters. The evolution of high-temperature and high-pressure plasma clusters will play an important role in the surrounding medium. Generally speaking, the method of using laser-induced plasma to produce mechanical effects on materials is called laser shock strengthening or forming technology. In laser shock strengthening or forming technology, the absorption layer and confinement layer are usually set. The absorption layer can be black tape, aluminum foil, and graphite coating. The confinement layer can be glass, deionized water, and flexible transparent materials [28-31]. Laser irradiates on the absorption layer, and the absorption layer material absorbs laser energy to produce high-temperature and high-pressure plasma mass. Under the restriction of the confinement layer, the

mechanical effect of plasma mainly acts on the metal target, thus realizing the strengthening and forming of the metal material [23, 32-34].

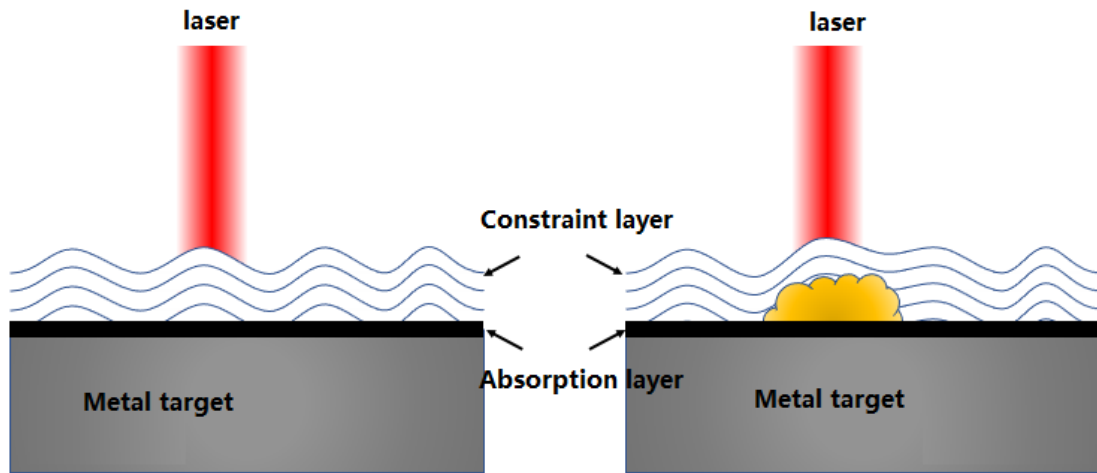


Fig.2 Schematic of Force Effect by Laser-Induced Plasma

3. Functional Surface Forming

By virtue of the force effect of laser shock, micro/nano forming of large metal material can be realized [19]. Micro/nano structured surfaces often have unique optical, mechanical, and biological properties. The mechanism of laser-induced plasma micro-nano forming, in particular the functional surface forming, lies in the metal's ultra-high strain rate as its forming flow characteristics [35-37]. Generally speaking, functional surface forming can be divided into two types, that is, micro/nano structured metal surfaces and in-situ covered nano-structured surfaces.

3.1 Micro/Nano Structured Metal Surface

Micro/nano structured metal surfaces are widely used in plasma, optics, biological detection, and other fields. Because of the fluidity of metal itself, it is a great challenge to form micro/nano complex structural surfaces with high precision and large area. Laser shock imprinting can realize large micro/nano forming of metal materials at a low cost with high precision. Huang Gao et al. proposed the direct forming of three-dimensional micro/nano structured metal surfaces at room temperature, air, and atmospheric pressure. This work achieved a minimum machining accuracy of 10 nm, which is even less than the lattice size of metal materials. At the same time, this surface is smooth and high in structural uniformity without mechanical damage and chemical pollution [35]. This study further extends the laser shock imprinting method to the large-area preparation of micro-grooves, pyramids, fishing net protrusions, micro-gears, and other structures, creating a new method for rapid and high-quality forming of micro/nano structured metal surfaces.

Yaowu Hu and others deposited gold thin films on flexible aluminum foil and prepared large plasma nanostructures by laser-induced direct imprinting. This method can realize direct and high-efficiency forming of cross-scale micro/nano arrays with accuracy from several microns to 15 nm. Such gold nanostructures have a local light field enhancement effect and can be applied to low concentration molecular detection and other fields [38]. Shigeru Tanaka et al. of Kumamoto University used underwater shock waves for high-quality imprinting on the thin metal foil as an alternative to laser shock imprinting. The underwater explosion of high explosives will produce shock waves with a duration significantly longer than that of LSI, which provides a pressure relief depth of 90% of the groove depth in the mold [39]. Underwater shock waves can be used for large-area imprinting, and the thickness of processed aluminum foil can be increased. The researchers said that this will be especially beneficial to future applications in the fields of plasma and super surface. Liangliang Wang et al. of Guangzhou University of Technology proposed a method of manufacturing microgrooves by laser-induced cavitation impact forming via the

mechanical effect of laser-induced plasma, realizing the forming and manufacturing of microgrooves on copper film. Taking advantage of this process, the team realized the controllable forming of microgrooves of 14-50 microns. Meanwhile, with the increase in laser shock times, the hardness of the formed surface more than doubled [40].

It should be pointed out that in order to realize laser shock forming of specific three-dimensional micro/nano metal structures, specific dies must be designed and processed, undoubtedly reducing the adaptability and application prospect of laser shock forming. To lift this restriction, Shengyu Jin et al. directly used the soft optical disk as the substrate, and realized the forming and manufacturing of various complex structures by skillfully designing the deflection angle between the soft optical disk and the target surface during multiple impacts, instead of directly forming and copying the mold [41]. The team further used biomaterials as substrates to prepare the metal with a complex micro/nano structure surface, which realized the direct replication of dehydrated bamboo leaves, iris leaves, azalea leaves, and other structures, successfully applied to nano friction generators as a new way to manufacture biologically inspired micro/nano scale patterns that are scalable and large on metal surfaces [1]. Yaowu Hu et al. adjusted the superplastic flow of metal nanoparticles by controlling the laser energy and produced ultrafine tunable line gaps at a scale lower than 10 nanometers. From the perspective of molecular dynamics analysis, metal nanostructures are changed from crystalline metals to liquid-like metals, which expanded rapidly but never merged [25].

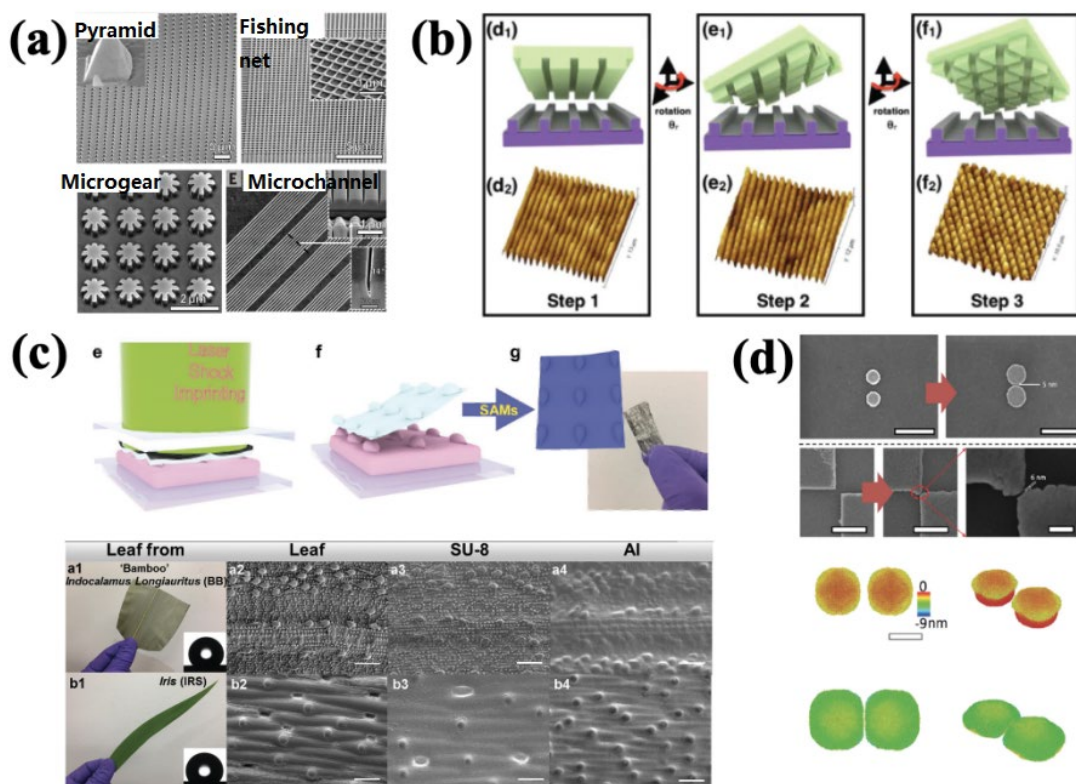


Fig.3 Surface Forming of Micro/Nano-Structured Metal Surfaces.

(a) Laser shock surface forming of Ag nanopyramids, Ag fish nets [line width 30 nm, spacing 60 nm], an array of nanogears imprinted on cold-rolled aluminum foil, and an array of triangular V-grooves [width 100 nm, depth 500 nm].

(b) Schematic illustration of surface forming using a DVD mold.

(c) Schematic of surface forming with leaves and the corresponding results.

(d) SEM images of metal nanostructure arrays with ultrafine maps, the FEI simulation results showing the changes of gap width with different deformations of metallic particles.

3.2 In-Situ Coating of Nanostructured Surface

In-situ coating of nano-structure surface refers to the structure [34, 35] that covers

two-dimensional materials in situ on the surface with nano-structure, so that it has both the geometric characteristics of metal nano-structure and the special properties of two-dimensional materials. Because of their special structural characteristics, two-dimensional materials have been widely used in materials science, electronics, physics, mechanics, biology, and chemical engineering [42-44]. Nanostructured surfaces have been proven to significantly improve the performance of nano-optical devices, imaging, sensing, energy conversion, and material control. The preparation of in-situ coated nanostructure surfaces harmonizes their unique physical and chemical characteristics with potential application prospects in the new thermal, electrical, and optical micro-nano generation [45-47].

Graphene is a typical two-dimensional material, which has special physical and chemical properties [37, 48, 49]. Gary J. Cheng et al. of Purdue University developed a graphene-nano structure [50] in 2015, where graphene was coated on nanoparticles, nanorods, and nano double pyramid structures without gaps by laser shock pressure, forming a special composite heterostructure of the graphene-coated nanostructure. The author further verified that this kind of structure can realize high-precision detection of trace elements by using the local plasma enhancement effect. Using graphene-coated gold nanospheres, gold nanorods, and gold nanopyramids as surface-enhanced Raman scattering activation substrates, it's verified that these surfaces have significant biosensing ability [34]. At the same time, it is verified that adjusting the laser power can change the band gap of graphene through laser shock imprinting so that its energy band gap reaches 2.1 eV. Besides, the graphene single-layer structure is pressed into the groove-shaped mold by using the force effect induced by laser, and the permanent deformation of this kind of heterostructure is realized, which provides a new processing method for flexibly adjusting the optical, magnetic, and electronic characteristics of graphene materials [51].

Complex two-dimensional materials and nano-structure surface composite structures can be further applied to the field of optical detection. Graphene/PbS-quantum dot/graphene hybrid photodetector is very important for industrial and scientific detection [52]. Traditional graphene transfer methods can only cover graphene on three-dimensional micro-nano structures, but can not achieve perfect seamless wrapping, which brings great challenges to improving the performance of photodetectors. By using the method of laser shock imprinting, the original grid space contact is promoted to seamless tight wrapping. The light response rate, light response gain, and accuracy of this kind of sandwich structure are much higher than those of the original structure [53]. Therefore, compared with the original method, laser-induced shock micro/nano forming can realize ultra-precision deformation of two-dimensional materials at a very small scale and closely fit with the surface of three-dimensional micro/nano structure without gaps, thus providing potential technical means for significantly improving the performance of various functional devices.

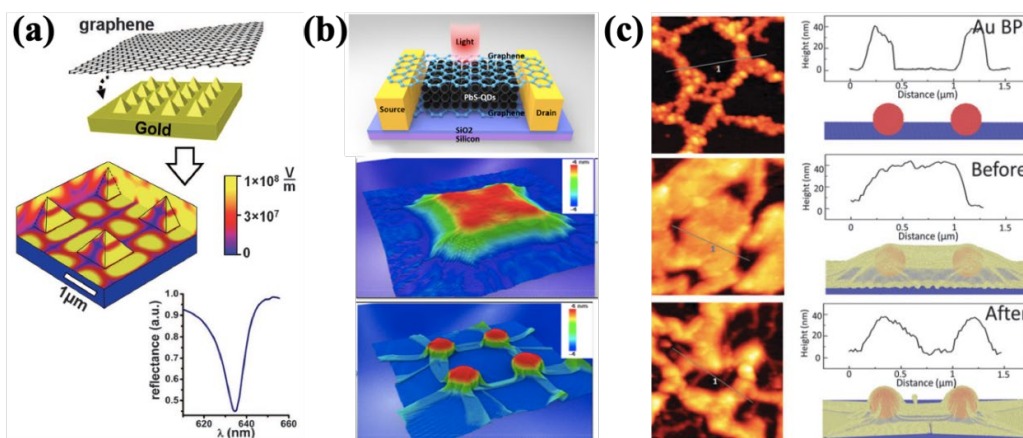


Fig. 4. In-situ Surface Forming of Hetero-nanostructures with Covered Layers. [a] Illustration of the graphene-covered Au nanopyramid structures and the field enhancement around the tip. [b] Hybrid graphene/PbS-QDs/graphene photodetectors and the detailed hetero-nanostructures with covered layers. [c] Images of hetero-nanostructures before and after laser shock forming, the corresponding sectional profiles, and presentative molecular dynamics simulations.

4. Molding Strategy of Laser Shock Micro/Nano Forming

The core of laser shock micro/nano forming of large metal materials lies in the force effect of laser-induced plasma cluster, and the prefabricated die directly determines the structure and properties of materials after plastic deformation. In addition, when the feature size of the preset die is equal to the grain size of the material itself, the fluidity of the material itself and the boundary effect with the interface will directly affect the surface forming quality of micro/nano structures, and even determine the machining scale limit of the forming technology. At present, laser shock micro/nano forming can be divided into three types according to the difference of preset dies, that is, structural rubbing forming, micro-nano particle forming, and structural bionic forming.

The die used for structural rubbing is usually a spatial complement [54] of the three-dimensional shape of the target surface. This kind of mold generally uses silicon as material [35], which can make the mold not damaged after more than 100 laser shock tests. There are two reasons: a) ultra-thin Al₂O₃ layer [5 ~ 10 nm] is deposited on the silicon nano-mold by atomic layer deposition to improve its strength and wear resistance, and dry lubricant also helps to reduce the wear of the nano-mold; b) the impact load applied in this study ranges from 1 to 1.8 GPa, which is sufficient to deform the selected material within 5 to 10 ns, but not enough to damage the Si nano-mold. The shapes of nano-molds include nano-strips, nano-rods, V-shaped wedges, fishing nets, and nano-pyramids, with different shapes meeting the needs of different application scenarios. The die used in structural rubbing forming takes a long time to manufacture, and the target structure and die structure often correspond one to one, which limits its extended application. Therefore, it is a feasible process to form complex structures by plastic deformation accumulation by using existing simple micro/nano structures. The flexible disc has a regular groove structure, which makes it a potential application mold of laser shock imprint technology. Using this as a mold, the fabrication of a graded metal nano-photon structure at a nano-scale is realized by a plurality of laser shock impressions [41]. Optical discs composed of grooves at a micrometer scale to the nanometer scale, such as optical discs, digital versatile discs, and Blu-ray discs, can be manufactured at low cost with excellent geometric accuracy and mechanical reliability [39]. It is found that when applied in multiple laser shock imprinting processes, each imprinting is controlled by mold shape, laser power density, and rotation angle of sequential laser shock imprinting. Therefore, the final metal structure is controlled by the mold shape, laser intensity, and rotation angle, thus forming a hierarchical nanostructure.

The die used in the bionic forming of structure is a natural structure in nature. All kinds of blade surfaces have the characteristics of high-density micro/nano structure and low surface energy, which is a natural excellent super hydrophobic surface. Therefore, rubbing its high-density micro/nano structure on natural blades, supplemented by subsequent surface treatment, can realize the replication of natural blade structure and function. For example, dehydrated bamboo leaves, iris leaves, azaleas, and the like can be used as a mold [1] for the bionic forming of structures. In the process of structure bionic forming, the natural blade structure needs to be rubbed with PDMS firstly, and the geometric shape of the prepared PDMS master mold is complementary to the bonding properties of the original blade. Then, the surface of the PDMS master mold is immersed in the light curing material, and the light curing master mold is obtained by exposure treatment. The geometry of the light-cured master mold is the same as the bonding properties of the original leaves. Finally, the required metal foil is covered on the photo-curing master mold, and the geometric structure on the natural blade is copied by laser shock imprinting. In addition, the surface wettability is further adjusted by coating silanes with different surface energies, such as perfluorooctane trichlorosilane, octyl trichlorosilane, etc. By combining high strain rate metal deformation with 3D soft mold, laser shock imprinting uniformly transfers layered microstructures from natural leaves to metal surfaces, which solves the challenges of scalability, resolution, and layered complexity in the preparation of micro-nano structures.

Micro/nano particle forming avoids the difficulties of mold manufacturing and selection fundamentally, which realizes the direct forming of micro/nano particles through superplastic deformation of micro/nano particles. Pulsed nanosecond laser irradiates transparent confinement

layer, sacrificial layer, and momentum transfer layer. Under laser irradiation, the sacrificial layer [graphite] ionizes instantaneously and the expanded plasma is confined by a transparent confinement layer, which induces instantaneous impact pressure of tens of GPa. A 4 μ m Al thin film is used as a momentum transfer layer to transfer impact pressure to metal nanostructures. Superplastic deformation of metal nanostructures leads to the rapid expansion of metal nanostructures and the narrowing of metal gaps. The impact pressure of tens of GPa in nanoseconds is much higher than the flow stress of metal and its nano-counterparts, thus generating superplastic flow in metal nano structures. The difference between the response of metal nanostructures under laser shock and their mechanical properties can be analyzed by size effect and non-equilibrium plastic flow under ultra-high strain rate laser shock. When metal nanostructures are supported by rigid substrates, the influence of height is more obvious than that of transverse dimensions. The friction force from the substrate hinders the lateral material flow at a high strain rate. Under non-equilibrium conditions, compared with large particles, small particles can flow freely without restriction.

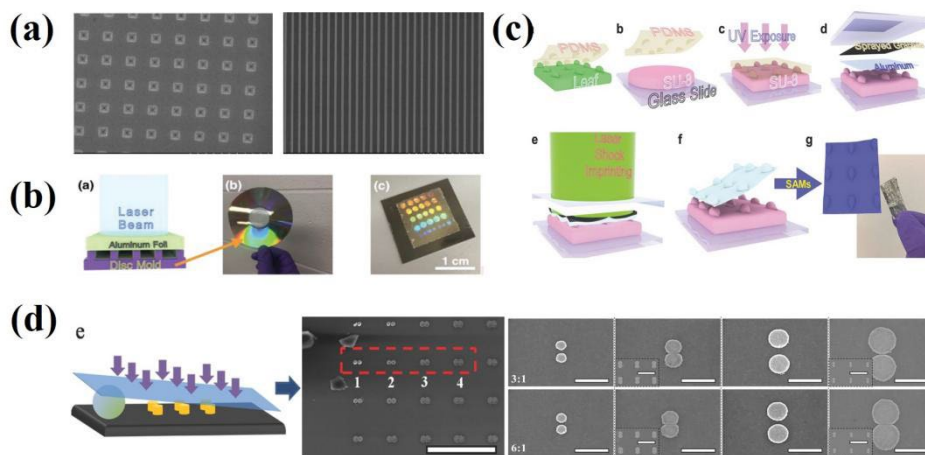


Fig.5 Strategy of Mold Design in Laser Shock Forming. [a] Rubbing Forming. [B] Multi-Path Rubbing Forming. [C] Biomimetic Forming. [d] Micro-to-Nano Partial Forming.

5. Outlook

Using the force effect of laser shock, micro/nano forming of large area metal materials can be realized. Micro/nano structure surfaces often have unique optical, mechanical, and biological properties. The mechanism of laser-induced plasma micro-nano forming, in particular the functional surface forming, lies in the metal's ultra-high strain rate as its forming flow characteristics. Generally speaking, the functional surface forming can be divided into two types, including micro/nano structured metal surfaces and in-situ covered nano-structured surfaces. Besides, prefabricated dies directly determine the structure and properties of materials after plastic deformation. When the feature size of the preset die is equal to the grain size of the material itself, the fluidity of the material itself and the boundary effect with the interface will directly affect the surface forming quality of micro/nano structures, and even determine the machining scale limit of the forming technology. Currently, laser shock micro/nano forming can be divided into three types according to the difference of preset dies, that is, structural rubbing forming, micro/nano particle forming, and structural bionic forming.

However, although laser shock micro/nano forming has solved many problems in forming large and complex three-dimensional structures with high precision on metal surfaces, there are still big key technical problems and scientific challenges as follows.

1) Stability of prefabricated die. Prefabricated die directly determines the structure and performance of the deformed material, and its surface quality will gradually deteriorate with the increase of press-in and demoulding times of metal materials.

2) Forming of micro/nano structure with a three-dimensional curved surface. At present, laser

shock forming is mostly used in the plane. How to realize complex structure forming on three-dimensional curved surfaces is still an obvious technical challenge.

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