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Research paper

Laser Technology of Optical Meta-Materials

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Abstract

The basic methods of laser technology used to obtain 2D and 3D meta-materials are considered. The main attention is paid to the methods of forming submicron and nanometer structures using the two-photon polymerization effect and various types of laser interference lithography. It is shown that these technological methods, being planar and scalable, are promising for use in industrial optical production due to their potentially high productivity and low cost compared to many other methods of nanotechnology, in particular, with electronic lithography and processing with focused ion beams.

Keyword: *technology, production, meta-materials, development, samples.*

1.Introduction

At present, meta-materials are being intensively studied - composite formations that are absent in nature and possess unusual properties or “superproperties” with respect to the starting materials [1-3]. V.G. Veselago in 1967 considered theoretically [5].

In the high-frequency part of the terahertz (optical) waveband, the sizes of meta-atoms have submicron / nanometer values. This required the use of nanotechnology methods for the implementation of optical meta-materials, and only with the development of these methods in the last decade has it become possible to obtain samples of optical meta-materials.

This article is a review of laser technologies that can be applied to the manufacture of optical meta-materials.

2.Research Methods

Laser technologies mastered in the last century were not designed for processing submicron elements, i.e., elements with dimensions of the order of wavelength and sub-wavelengths below the diffraction limit. This either does not allow the use of known laser technology, or greatly narrows their scope. At present, new methods have been implemented that allow solving many production problems. Of these, two-photon polymerization for direct laser formation of 2D and 3D structures and laser lithography should be mentioned.

Laser photochemistry is used to form various miniature and micro-sized objects [8] using two-photon absorption (2 FP). This process is possible at a high radiation density, which is achieved only at the focal point, due to which the process is highly localized. On the path of radiation to the point of focus and further absorption is absent due to the threshold effect on the energy of absorbed quanta and the nonlinear nature of the process. The speed of photoreaction at 2FP is proportional to the square of the photon flux density, and at 1FP - just the flux density. Accordingly, the spatial distribution of photopolymerization at 2FP will be quadratic compared to the distribution at 1FP. Therefore, for example, at a radiation wavelength of 7 = 790 nm, the transverse size of the polymerization zone will be almost the same as the size of the light spot, limited by diffraction at a wavelength of λ = 395 nm. The spatial resolution of the 2FP method is limited by the diffusion of radicals and the growth of polymer chains outside the illuminated region, but this effect is less pronounced than in the 1FP method, due to polymerization only in the focus area with high radiation density. Outside this region, the photon flux density is low, the 2FP process is not possible, and photons of long-wave radiation with low energy are not separately absorbed. As a result, when the focus point is moved in a photosensitive medium, point-wise formation of a 3D volume object is possible. After the exposure procedure is completed, the unpolymerized starting material is washed out with a solvent, and the desired object remains on the substrate, for example, a photonic crystal lattice in the form of a firewood or micro-spiral lattice.

Femtosecond laser radiation on a titanium-doped sapphire laser (wavelength 2 ~ 780 nm, pulse duration and frequency ~ 100 fs and 80 MHz, respectively, the average radiation power is tens to hundreds of milliwatts) is usually used to implement the 2FP method. Sum of the energies of the two absorbed quanta of this radiation corresponds to the energy of the quanta of the ultraviolet range, while the threshold and nonlinear nature of the absorption process, as stated above, provides a resolution below the diffraction limit.

Obviously, single-beam laser formation of micro / nano-objects is a long process, besides being sensitive to mechanical and thermal effects. Therefore, this approach is not optimal and does not provide great advantages over electron and ion beam methods in large-scale production. However, this problem was solved by applying a system of a large number of micro-lenses, creating a corresponding number of focus points [9], or diffraction gratings and holographic techniques [10]. As a result, it is possible to simultaneously form several hundred identical objects on a relatively large area (~ 2 cm2) [9]. We note that electron-beam

lithography provides for obtaining samples of meta-materials only on an area of up to 10 μm2.
Most often, two-photon absorption is used for the manufacture of objects from polymeric materials by their polymerization from the liquid phase. However, these materials have a low refractive index n. A higher value of n can be obtained by applying a photopolymerizable material containing Ti atoms in its composition. After the 2FP procedure with drawing the specified structures and the sol-gel process at a relatively low temperature of 400 ° C, a refractive index of more than 2 was obtained due to the presence of the TiO2 phase in the material of the structures [11].
Let us give an example of the technology of making 3D metal micro / nanostructures [12, 9]:

- applying a hydrophobic coating on a glass substrate;
- applying a liquid modified with special additives photo-polymerizable material on a substrate;
- laser formation of structures by two-photon absorption using a micro-lens system;
- removal of an uncured substance with a solvent (acetone);
- surface treatment with SnCl2 solution to improve metal deposition and its adhesion;
- chemical deposition of Ag from AgNO3 solution on hydrophilic surfaces (sediment thickness - 50-100 nm, grain size is governed by the deposition conditions and amounts to tens of nanometers);
- washing the product.

This technology provides for obtaining 3D configurations of meta-material elementary cells with a lateral resolution of ~ 100 nm. In addition to chemical precipitation of a metal from a solution of its salt, low-temperature chemical vapor deposition (CVD) can be used [13, 14]. It is necessary to achieve a reduction in the roughness of the polymer surface layer. In [15], a roughness of 4–11 nm or 1 / 50–1 / 100 of λ was achieved in the visible or near IR range with a lateral resolution of 100 nm. Lasers are capable of effectively generating short-wave UV radiation of the required intensity (~ 200 mW / cm2), while excimer inert-gas lasers turned out to be the most suitable [17, 18]. The era of laser photolithography has begun. KrF lasers emitting at λ = 248 nm (resolution 250–110 nm) were the first to use submicron technology, but to obtain element sizes less than 100 nm, most modern photolithographic systems began to use radiation at a 193 nm wave generated by an ArF laser (resolution 100-80 nm). In the next 5–10 years, it is predicted that the industry will use extreme UV (EUV) photolithography using radiation with λ ~ 13.5 nm (resolution 10–4 nm). To obtain such radiation, it is planned to use a strongly radiating plasma of a cold Xe gas, generated using a laser (Nd: YAG, λ = 1064 nm) [17].
Interference lithography is a formless (maskless) method of irradiating a photoresist with two or more coherent laser beams, which form a standing wave as a result of overlaying and interference in the form of a pattern of elements of a given micro or nano structure [19–21]. The coherence of the rays ensures the formation of a stable interference pattern. The method is applicable for obtaining structures with a periodic or quasi-periodic pattern, i.e. meta-materials. The big advantage of interference lithography is the simultaneous, parallel formation of a large number of identical elements / cells over a large surface area of the substrate, which significantly increases the productivity of the process, reduces its cost, and, importantly, reduces sensitivity to mechanical stress (vibration) and thermal size care. It is also possible to form not only flat 2D structures, but also 3D structures.

3.Analysis of Results

Figure 1 shows the scheme of using the interference of two coherent laser beams for the manufacture of a strip grid of photoresist. To calculate the lattice period, we present the complex amplitudes of the waves in the form [19]:

$$E_1 = A\exp\left[-ik\left(z\sin\Theta - x\cos\Theta\right)\right],$$
$$E_2 = RA\exp\left[-ik\left(-z\sin\Theta - x\cos\Theta\right) - i\varphi\right]$$

Where x is the axis of coordinate perpendicular to the plane of the pattern, z is the horizontal axis in the plane of the pattern, R is the ratio of the amplitudes of the waves, φ is the phase difference of the waves. Hence the intensity of light on the surface of the photoresist is equal to:

$$\left|E_1 + E_2\right|^2_{x=0} + \left|A\right|^2\left[1 + R + 2R\cos 2\left(kz\sin\Theta - \varphi\right)\right].$$

It is easy to see that the intensity is modulated along the z coordinate along the photoresist surface (Fig. 1) with a period, where n is the refractive index of the photoresist (n ~ 1.6), λ is the wavelength of the laser beam in vacuum. The maximum modulation amplitude is obtained at equal wave amplitudes (when). After processing (development and hardening), the photoresist forms strips in areas with a maximum amplitude of the standing wave.

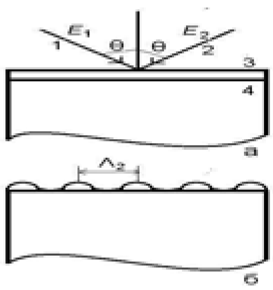

Figure 1. The imposition of two laser beams (a) and strips of photoresist after its processing (b). 1, 2 — coherent laser beams, 3 — photoresist layer, 4 — substrate.

The imposition of three rays provides the formation of intensity maxima at the nodes of a triangular lattice. If the azimuthal (horizontal) angles between the beams are 120°, the minimum distance between the maxima is determined by the relation [22].

$$\Lambda_3 = (2/3)\lambda/2n\sin\Theta$$

The overlap of four rays at azimuth angles between them of 90° leads to the formation of a square lattice with a period [22].

$$\Lambda_4 = \left(\sqrt{2}/2\right)\lambda/2n\sin\Theta$$

Instead of overlapping three rays, double exposure can be applied using two rays. To do this, after performing the first exposure, the second is immediately performed with the substrate rotated 90° around an axis perpendicular to the surface. The result is a square lattice, as shown in Figure 2.

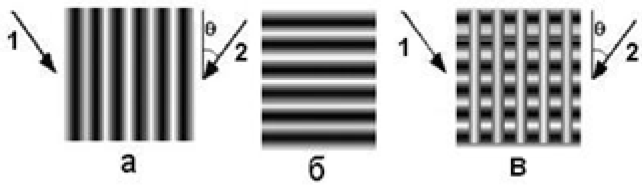

Figure 2. Obtaining a square 2D grating from a photoresist by double exposure [23]: a - 1D grating after the first exposure, b - rotation of the substrate by 90 в, in - 2D grating after the second exposure. 1, 2 — coherent laser beams incident on the substrate at an angle.

Accordingly, to obtain a triangular lattice, it is necessary to perform two rotations of the substrate by 120°. The above formulas for Λ2, Λ3, and Λ4 show that it is necessary to use short-wave UV lasers for the manufacture of submicron and nanometer structures. Therefore, in laboratory experiments on interference lithography, a He-Cd laser (λ = 441 nm and 325 nm) or an Ar laser (λ = 458 nm) is often used. The above formulas also indicate the possibility of reducing the grating period by increasing the refractive index of the medium in which the interference occurs. Practically, this is achieved by placing on the surface where interference occurs in a transparent immersion liquid or a transparent solid body in the form of a prism, as shown in Figure 3. In the latter case, the period of a strip grid obtained from a photoresist is defined by the above formula for Λ2, where n –is the refractive index of the prism. The value of θ is determined from the law of Snell. Such a device makes it possible to obtain Λ2 about 100 nm with a He-Cd laser at a wavelength of 325 nm [24]. It is obvious that interference photolithography in the presented embodiments allows to obtain only flat 2D structures. To obtain 3D structures of meta-materials, it is possible to carry out vertical overlapping of 2D structures with appropriate alignment of adjacent layers, but it is less laborious to combine interference laser lithography with two-photon polymerization; the technique of this approach is described in [25].

4.Conclusion

The basic laser technologies of meta-materials are considered. They make it possible to obtain structures with submicron and nanometer sizes of elements and to overcome the diffraction limit [26] and have several advantages over other beam methods of manufacturing nanostructures, primarily, before electronic lithography and processing with a focused ion beam. Among them, there are higher performance and lower cost and the ability to obtain structures on substrates of a large area. It is very important that the considered technologies are planar and scalable, which allows them to be integrated into the existing production of optical-electronic devices and materials.
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