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Diffractive optical devices produced by light-assisted trapping of nanoparticles

J.F. Muñoz-Martínez,[1,2,*] M. Jubera,[1] J. Matarrubia,[1] A. García-Cabañes,[1] F. Agulló-López,[1,3] M. Carrascosa[1]

[1]*Departamento de Física de Materiales, Universidad Autónoma de Madrid, Madrid 28049, Spain*
[2]*Departamento de Mecánica de Fluidos y Propulsión Aeroespacial, Universidad Politécnica de Madrid, Madrid 28040, Spain*
[3]*Centro de Microanálisis de Materiales (CMAM), Universidad Autónoma de Madrid, Madrid 28049, Spain*
**Corresponding author: juanf.munnoz@uam.es*

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One and two-dimensional diffractive optical devices have been fabricated by light assisted trapping and patterning of nanoparticles. The method is based on the dielectrophoretic forces appearing in the vicinity of a photovoltaic crystal, such as Fe:$LiNbO_3$, during or after illumination. By illumination with the appropriate light distribution, the nanoparticles are organized along patterns designed at will. One- and two-dimensional diffractive components have been achieved on X- and Z-cut Fe:$LiNbO_3$ crystals, with their polar axes parallel and perpendicular to the crystal surface, respectively. Diffraction gratings with periods down to around a few micrometers have been produced using metal (Al, Ag) nanoparticles with radii in the range of 70-100 nm. Moreover, several 2D devices, such as Fresnel zone plates, have been also produced showing the potential of the method. The diffractive particle patterns remain stable when light is removed. A method to transfer the diffractive patterns to other non-photovoltaic substrates, such as silica glass, has been also reported.

OCIS codes: *(230.4000) Microstructure fabrication; (050.1950) Diffraction gratings; (050.1965) Diffractive lenses; (050.1970) Diffractive optics; (160.5320) Photorefractive materials; (350.4855) Optical tweezers or optical manipulation*

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A method for massive parallel trapping and patterning of nanoparticles has been recently developed [1-3]. It is based on the high bulk photovoltaic (PV) effect of certain ferroelectric materials [4]. This effect can generate very intense light-induced electric fields (up to 10^5 V/cm at the surface of Fe-doped $LiNbO_3$ crystals) [4, 5]. Moreover, using an inhomogeneous illumination pattern an electric field distribution (the space charge field) is generated, which has been largely studied in the area of photorefractive (PR) non-linear optics [6]. This space charge field is here used to attract the nanoparticles to the crystal surface. The technique is often called PV tweezers since it allows micro- and nanoparticle manipulation [7, 8]. It has been shown to offer a great potential for applications in several areas [9-12] although reported data are still very scarce in the field of photonics.

The aim of the present work is to provide an actual demonstration of the application of the technique in the field of photonics. Specifically, in this letter we demonstrate the feasibility of the method to produce diffractive components. These optical devices [13, 14] play a key role in a variety of photonic devices. They include diffraction gratings, beam deflectors, tunable filters, Bragg reflectors, grating couplers, Fresnel zone plates and so on. There are different methods to produce those kinds of components [15], such as mechanical or irradiation (laser, electrons and ions) engraving of periodic structures and different deposition methods. More flexible methods include acoustic-wave modulation [16] and photorefractive-induce gratings [17].

The technique proposed here is a novel and simple fabrication method that allows achieving periodic nanoparticle patterns designed at will using the imaging capabilities of light. The method consists of three consecutive steps: 1) a space charge distribution is recorded in a photovoltaic Fe-doped $LiNbO_3$ sample by periodic illumination with a suitable wavelength [6, 7], 2) then, nanoparticles are approached to the surface of the illuminated sample and they are trapped and arranged on it by the dielectrophoretic forces associated to the PV field [7, 8], 3) the original space charge field is erased by heating (around 250ºC) [18], so it will not affect to the device performance. As a result, a diffractive pattern appears associated to the periodic distribution of particles that remains stable at least during months due to adhesion forces. No changes have been observed in the nanoparticle distribution as a result of the PV field erasure.

A number of grating devices, both 1D and 2D, have been fabricated to test the method and their optical performance has been characterized. Spherical metallic nanoparticles (Al, Ag), having average diameters of 70 and 100 nm respectively, have been used. Linear gratings with periods down to a few micrometers have been achieved. As to 2D devices, Fresnel lenses (strictly speaking, Fresnel zone plates) have been fabricated having tight focusing capabilities. Finally, the possibility to transfer the diffractive patterns to non-photorefractive substrates has been also investigated. This strongly enlarges the variety of applications of PV tweezers in photonics and other fields.

In order to fabricate the diffractive patterns, the substrate is illuminated with the appropriate light pattern to generate the evanescent PV fields on the surface. Fe:$LiNbO_3$ samples with total iron concentrations of 1000 ppm are used as substrates for the experiments. They have been cut with the polar axis parallel (parallel configuration) or normal (perpendicular configuration) to the surface. The oxidation/reduction ratio $[Fe^{2+}]/[Fe^{3+}]$ for the iron doping is estimated to be around 0.1, that leads to PV fields as high as 10^4 -10^5 V/cm.

For 1D diffraction gratings we apply a sinusoidal illumination obtained by two beam interference on a X-cut Fe:$LiNbO_3$ substrate [8, 19], using the parallel photorefractive configuration with the *K*-vector along the ferroelectric *c*-axis. For 2D gratings, we use a two-dimensional distribution of light intensity generated by a spatial light modulator (HOLOEYE, LC-R 1080). This light distribution is projected on a Z-cut Fe:$LiNbO_3$ substrate (perpendicular configuration) that allows an accurate reproduction of the 2D light pattern [11, 20]. Light comes from a frequency doubled Nd:YAG, operating at 532 nm with typical intensities in the range 1-100 mW/cm^2. Although Z-cut crystals could be also used for 1D patterning, we have used X-cut substrates because there are larger experimental experience and theoretical background for this cut. After illumination, the nanoparticles are deposited by immersing the substrate in a hexane suspension containing them. We have used mostly metal nanoparticles of Al ($d \sim$ 70 nm) and Ag ($d \sim$ 100 nm). After the pattern fabrication the initial bulk photorefractive charge grating is erased by thermal heating [18] during about 10 min at $T \sim 250$ºC to avoid its competition with the particle grating. The deposition and light-assisted patterning procedures are described in more detail in [8, 19] and [20] for 1D and 2D, respectively. Finally, it is worthwhile remarking that the method can be considered reconfigurable in the sense that the same substrate can be reused by simply removing the particles and repeating the fabrication process. In fact, we have often used the same substrate for several experiments without decreasing the quality of the patterns.

The simplest case to test the new method is to fabricate 1D diffraction gratings that appear schematically illustrated in Fig. 1. They can be obtained by illuminating a Fe:$LiNbO_3$ crystal surface (X-cut) with a sinusoidal light intensity profile, $I=I_0[1+m\cos(kx)]$, the x coordinate being along the ferroelectric *c*-axis. It is convenient to use a high light contrast, i.e. $m \sim 1$, in order to enhance the PV fields and generate a well-defined particle pattern [19]. The periodicity of the final particle patterns coincide with that of the light. The grating period, designated as Λ, is the sum $\Lambda = a + b$, a being the width of the strips formed by the aggregation of nanoparticles in the PV field and b is the inter-strip spacing corresponding to a particle-free Fe:$LiNbO_3$ surface.

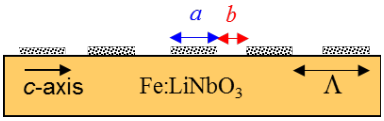

Fig. 1. Schematic diagram for the 1D diffractive grating.

Fig. 2(a) shows an example of a good quality diffractive pattern. The whole pattern extends over an area of about 1 mm^2 and it was obtained with a sinusoidal light pattern of $\Lambda = 20$ μm. It was fabricated using spherical Al nanoparticles of diameter $d = 70$ nm. A smaller region of the pattern with higher magnification is shown in Fig. 2(b) where the uniformity of fringes, having a width of $a \sim 8$ μm, is clearly appreciated. The regions between fringes (width $b \sim 12$ μm) are nearly free of particles as illustrated in the two micro-photographs of Figs. 2(a) and 2(b). The corresponding light diffraction profiles in both reflection and transmission configurations have been determined under plane wave illumination with 4 mW light power using a He-Ne laser emitting at $\lambda = 633$ nm. The result in the reflection configuration using a probe beam at ~10º from the normal of the sample surface is given in Fig. 2(c). It shows seven diffraction orders at angles $\theta_n = n\lambda/\Lambda$ that perfectly correlate with those expected for the spatial period, $\Lambda =$ 20 μm. By using particles of sufficiently small diameter (10-50 nm) grating periods down to around 4 μm can be achieved as previously reported [19]. As an example the diffraction pattern for a grating with $\Lambda = 8.5$ μm is also shown in Fig. 2(d).

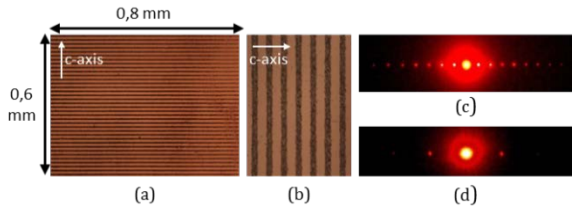

Fig. 2. (a) Photographic image of a grating device with Al nanoparticles ($d = 70$ nm) and $\Lambda = 20$ μm. (b) Local magnified image of a region of the pattern in (a). (c) Diffraction intensity diagram generated by reflection in (a). (d) Reflection diffraction diagram of a grating device fabricated with Ag nanoparticles ($d = 100$ nm) with a smaller period $\Lambda = 8.5$ μm.

As it is well known from basic diffraction theory, the diffraction intensity profile results from the convolution of the Airy pattern from a single reflecting or transmitting strip-like diffracting element, and the interference comb function of the grating. The analysis of the intensities of the diffraction orders for the various fabricated gratings is consistent with identifying the uncovered Fe:$LiNbO_3$ stripes between the nanoparticle fringes (width b in Fig. 1) with the diffracting elements, even in reflection configuration. In other words, the metal particles strips act as incoherent light scatters that do not contribute to the diffraction order intensities. This analysis can be clearly illustrated in Fig. 3 using the data corresponding to an Al grating fabricated on purpose with the inter-fringe spacing ($b = 4$ μm) rather different from the nanoparticle fringe width ($a = 11$ μm). The diffracted light intensity profiles for both, 4 μm (continuous line) and 11 μm (dashed line) diffracting widths, have been calculated and plotted together with the measured relative intensity of the first two orders for reflection and transmission diffraction. As it is clearly appreciated the intensity data fit much better with a diffractive width of 4 μm which corresponds to the uncovered Fe:$LiNbO_3$ strips (b in Fig. 1).

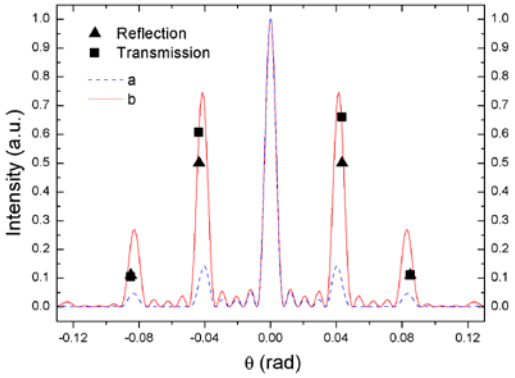

Fig. 3. Calculated diffraction intensity profile of a linear grating with period $\Lambda = 15$ μm and diffractive width $b = 4$ μm (solid red line) or $a =$ 11 μm (dashed blue line). The experimental intensity data of the first and second orders for reflection (triangles) and transmission (squares) diffraction are indicated.

As a further step in our work we have moved to the fabrication of a variety of 2D diffractive optical components. To this end we have taken advantage of the recently reported [11, 20] new perpendicular configuration that uses Z-cut crystals illuminated with 2D light patterns. This configuration is a necessary requirement to guarantee isotropic particle trapping and patterning over the crystal surface and so, good quality patterns.

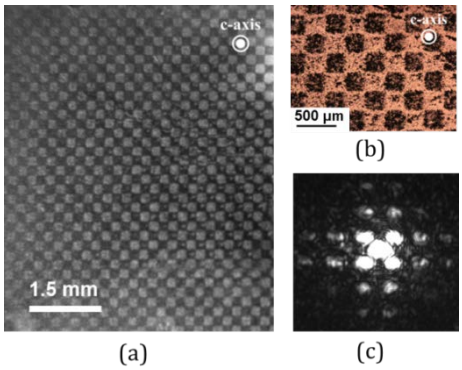

Fig. 4. (a) Photographic image of a 2D pattern of Al nanoparticles. (b) Amplified microscope image showing a local region of the pattern. Note that particles are white in (a) and black in (b). (c) Diffraction diagram of the 2D pattern.

In Fig. 4 we show the case of illumination with a mosaic of squares. An overall image of the sample is presented in Fig. 4(a) showing that a large area of the sample has been uniformly patterned. A detail with larger magnification is shown in Fig. 4(b) and the diffraction diagram in Fig. 4(c). However, a most common and investigated 2D diffraction device is the so-called Fresnel zone plate. The binary plate is constituted by alternative opaque and transmitting ring zones whose radii obey the relation, $R_N = (Nf\lambda)^{1/2}$, N being an integer number, λ the light wavelength and f the element focal length. Focal lengths of around 0.1-1 m can be routinely achieved. A typical device made up by depositing Al nanoparticles of d = 70 nm is displayed in Fig. 5(a). The focal length of this component should be f = 39.5 cm for λ = 633 nm. The focalization properties of the pattern have been tested by measuring the diffracted light intensity profiles obtained along planes parallel to the lens at different distances from it. The results are shown in Figs. 5(b) and 5(c). A sharp focalization of the beam is clearly observed at a distance of 39 cm that matches with the predicted focal length.

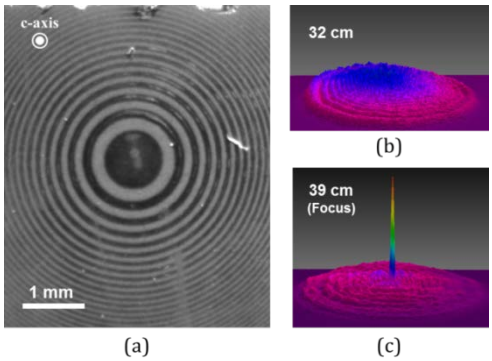

Fig. 5. (a) Photography of a Fresnel lens fabricated with Al nanoparticles (d = 70 nm). (b, c) Intensity profile of the diffracted light along a transversal direction at two distances from the crystal surface acting as zone plate: 32 cm and 39 cm (focus).

So far, nanoparticle patterns in this and previous works have been always obtained and kept on the PV substrate. However, for practical purposes, it is very convenient to be able to use any kind of substrate, particularly silica glass or polymers, as support of the patterns. Specifically, for our diffractive devices, this avoids possible absorption effects from the doped substrate or the contribution of competing photorefractive gratings. The latter can be generated during the operation of the device when using PR-sensitive wavelengths. Hence, we have tested a simple method to transfer the diffractive patterns from the Fe:$LiNbO_3$ surface to other substrates using a thermal release tape (supplied by Graphene Supermarket, Graphene Laboratories Inc.). First of all, the PV field generated in the active crystal substrate (Fe:$LiNbO_3$) is erased by heating it to 230 ºC. As already mentioned, the patterns remain stable due to the adhesion forces. Next, the tape is stuck to the substrate surface and peeled off, with the particles attached to it. No particles remain on the original substrate. Then, the tape is firmly stuck to the new substrate, such as silica glass. Finally, the tape comes off very easily after a smooth heating process at 100-200 ºC during a few minutes. This way the diffractive pattern has been transferred onto the new surface. The stability of the transferred pattern is as good as it was before the transference process. The best results have been obtained heating during 3-4 minutes at 200 ºC and applying slight pressure at the same time in the last step of the process. A careful inspection of the particle patterns images before and after the transference allows to make an estimation of the fraction of transferred particles, that is in the range of 40-50 %. Figs. 6(a) and 6(b) show the comparison between the microscopic images of a Fresnel pattern with aluminum particles on Fe:$LiNbO_3$ and the one after transferring it to a glass substrate. It can be observed that the transferred replica faithfully reproduces the original pattern. Moreover, the diffractive pattern for the transferred nanoparticle pattern at the Fresnel lens focus (Fig. 6(c)) shows again a sharp peak at the distance predicted by theory.

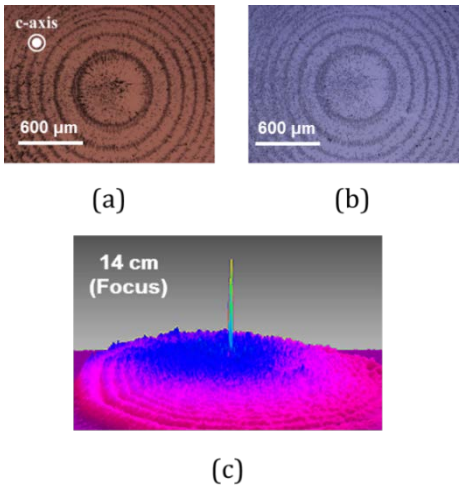

Fig. 6. (a) Microscopic image of a Fresnel pattern of Al nanoparticles on the $LiNbO_3$. (b) Pattern transferred to a glass substrate. (c) 2D intensity profile at the focus of the transferred Fresnel pattern.

In summary we have proposed and demonstrated the application of PV tweezers to the fabrication of 1D and 2D diffractive components, a key element for photonic devices. This is one of the first demonstrations of the technological applications of this nanoparticle patterning technique. The most relevant feature of the novel proposed method is its flexibility to easily obtain diffractive patterns at will, with

rather simple and low cost instrumentation. Recycling capabilities of the PV substrates are also a valuable plus. Moreover, the possibility to transfer the pattern to other active or passive substrates represents a remarkable advance either, for a flexible fabrication of diffractive devices, or for other future applications of PV tweezers. Applications of the obtained diffractive patterns may include beam steering, or integrated optical components. As examples, one could mention the use of these structures as optical components such as couplers or un-couplers in optical waveguides, or as tracks for photonic integrated circuits. In fact, the use of optical waveguides as substrates for PV trapping has been already reported [10].

The results of this work are a good example of the capabilities of this emergent nanoparticle manipulation technique. In fact, the further development of the method is expected to lead to new applications in different fields (nano-photonics, bio-photonics, nano-materials, etc.). For instance, using smaller nanoparticles active devices that take advantage of plasmonic effects are envisaged.

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