Two-dimensional photonic quasicrystals by single beam computer-generated holography

Gianluigi Zito,[1,2,]* Bruno Piccirillo,[1,2] Enrico Santamato,[1,2] Antigone Marino,[1,3]
Volodymyr Tkachenko,[3] and Giancarlo Abbate[1,3,4]

[1] Dpt. of Physics, University Federico II of Naples, via Cinthia, 80126 Naples, Italy
[2]CNISM, Consorzio Nazionale Interuniversitario per le Scienze fisiche della Materia c/o Physics Dpt. University of Naples Federico II, via Cinthia, 80126 Naples, Italy
[3]CNR-INFM - Coherentia c/o Physics Dpt. University of Naples Federico II, via Cinthia, 80126 Naples, Italy
[4] CNR-INFM Licryl - Liquid Crystal Laboratory c/o Physics Dpt. University of Calabria 87036 Rende (CS), Italy
**Corresponding author:* gianluigi.zito@na.infn.it

Abstract: Recently important efforts have been dedicated to the realization of a new kind of photonic crystals, known as photonic quasicrystals, in which the lack of the translational symmetry is compensated by rotational symmetries not achievable by the conventional periodic crystals. Here we show a novel approach to their fabrication based on the use of a programmable Spatial Light Modulator encoding Computer-Generated Holograms. Using this single beam technique we fabricated Penrose-tiled structures possessing rotational symmetry up to 23-fold, and a two-dimensional Thue-Morse structure, which is an aperiodic structure not achievable by multiple beam holography.

OCIS codes: (090.1760) Computer holography; (160.5293) Photonic bandgap materials; (230.4000) Microstructure fabrication; (230.6120) Spatial light modulators.

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#91997 - $15.00 USD   Received 22 Jan 2008; revised 13 Mar 2008; accepted 14 Mar 2008; published 31 Mar 2008

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1. Introduction

Quasicrystals are structures exhibiting long-range aperiodic order and rotational symmetry [1-3]. Mesoscale quasicrystals may possess photonic bandgaps (PBG) [4-6] that are more isotropic than in conventional photonic and, hence, the PBG becomes more spherical leading to interesting properties of light transmission [7], wave guiding and localization [8], increasing the flexibility of these materials for many photonic applications. To construct two-dimensional (2D) or three-dimensional (3D) quasicrystals is a very difficult task, however, because conventional methods used in the micro-chip fabrication (such as semiconductor lithography, multi-photon adsorption, template-based techniques, etc.) and also special methods developed for making photonic crystals (such as self-assembling of colloidal micro-spheres or micro-fabrication) are inadequate. Previously used to realize periodic photonic crystals [9,10], holographic lithography was recently proposed and used to realize quasicrystals too at the mesoscale in polymer resins [11-13], Holographic Polymer Dispersed Liquid Crystals (H-PDLC) [14], and holographic plates [15]. The holographic lithography is based on the interference pattern of many coherent light beams, that are usually obtained by splitting a single laser beam by suitable grating [11,13], prism [15] or dielectric beam splitters [16], in a single or multiple exposure process [12]. Realizing a quasicrystal structure,

#91997 - $15.00 USD Received 22 Jan 2008; revised 13 Mar 2008; accepted 14 Mar 2008; published 31 Mar 2008

exhibiting *N*-fold symmetry like a 2D Penrose-tiled [14] structure, usually requires to control the amplitude and phase of *N* interfering laser beams, leading to several difficulties, which can limit this technique. Moreover, quasi-periodic structures may be envisaged that cannot be realized even in principle by multiple-beam interference. The Thue-Morse structure is an example of aperiodic pattern [17-20], defined by recursive sequences, that cannot be obtained by interference of coherent beams. One-dimensional Thue-Morse gratings are known to exhibit PGB with interesting omnidirectional reflectance [21-23]. 2D Thue-Morse gratings are defined mathematically by recursive procedure (see equation (3) below). Very recently, theoretical studies have shown the presence of PBG in a two-dimensional Thue-Morse structure [24]. But experimental realization of photonic structures exhibiting two-dimensional Thue-Morse arrangement has not been reported in the literature until now.

In this communication, we report the fabrication of several 2D Penrose and Thue-Morse quasicrystals in the mesoscale range, using a novel single-beam technique based on the spatial modulation of the optical beam by means of Spatial Light Modulator (SLM) and Computer-Generated Hologram (CGH). The realization of these structures clearly demonstrates the potential of the CGH-SLM technique. In fact, we were able to produce, with single-beam optical setup, 2D Penrose patterns of rotational symmetry as high as 23-fold, never reached before. Moreover, this technique allows to obtain, with no realignment of the optics nor mechanical motions, any design of quasiperiodic and aperiodic patterns that cannot be realized using *N*-beam interference, whatever large may be *N*, as for instance the 2D Thue-Morse pattern.

Holographic Polymer Dispersed Liquid Crystals (H-PDLCs) are materials that provide good mechanical and optical properties and can be switched by applying moderate external electric fields [16]. Our Penrose and Thue-Morse gratings were realized by induced photo-polymerization of liquid crystal-polymer composite. Nevertheless, the technique we employed is not limited to this kind of photosensitive materials.

2. Holographic patterning and fabrication

Computer-generated holography is a very attractive technique that permits to create almost any two-dimensional, and even three-dimensional, spatial distribution of the intensity of the optical beam, by controlling the phase profile of the laser beam impinging on a Diffractive Optical Element (DOE) [25], which is designed as a CGH. Usually, the DOE is previously fabricated and works as a static element to project in the Fourier plane an array of intensity spots [26]. In our work, we used a different approach implementing the DOE with a programmable liquid crystal SLM which can encode the CGH directly in its LC-display. This permitted us to exploit at best the potential of the CGH technique. While in the standard multi-beam holography is quite difficult to work with a large number of beams [27], with the CGH-SLM it is possible to get almost any design of the writing pattern encoding the desired spatial distribution in a single beam. Another drawback of the multiple-beam approach is the great difficulty in maintaining phase coherence of the interfering beams between subsequent exposures. Our CGH-SLM technique completely alleviates this issue, providing high reproducibility of the resulting structures. No complex optical setup is needed and mechanical noise problems are drastically reduced. Finally, real-time switching from one pattern to the other is easily accomplished without changing the optical alignment. The only limitations come out from the pixel resolution of the SLM and from the refresh rate of the computer graphic card. In our experiments we used a liquid crystal spatial light modulator HoloEye LC-R 3000. An intensity image of 256 grey levels with a maximum resolution of 1200×1920 pixels is addressed to the SLM via computer and corresponds to the optical phase profile added to the incident beam. The LC-display acts as a DOE modulating the irradiance spatial distribution in a given region of space, usually the focal plane of a lens. In a previous preliminary work, we realized 2D Penrose gratings by using an iterative algorithm to create the desired irradiance pattern in the Fourier plane of the CGH [28]. In the present work, we used a different approach for the beam shaping. Instead of computing the CGH suitable to work in the Fourier plane [25,29] we encoded the desired intensity pattern directly in a phase-

#91997 - $15.00 USD Received 22 Jan 2008; revised 13 Mar 2008; accepted 14 Mar 2008; published 31 Mar 2008

only CGH [30]. The desired irradiance profile is computed and then addressed to the SLM to create the phase hologram. Two relay lenses are then used to form the reduced image of the irradiance profile on the photosensitive sample. The lateral magnification of the imaging system determines the scale length of the pattern to be written. This method of "direct imaging" permits to obtain higher spatial resolution in writing the pattern. The irradiance profiles $I(\mathbf{r})$ associated to the Penrose structures were calculated simulating a multi-beam interference according to [11,15]

$$I(\mathbf{r}) = \sum_{m=1}^{N}\sum_{n=1}^{N} A_m A_n^* \exp[i(\mathbf{k}_m - \mathbf{k}_n)\cdot \mathbf{r} + i(\varphi_m - \varphi_n)], \tag{1}$$

where A_m, $\mathbf{k}_m$, φ_m, are the amplitudes, the wave vectors and the initial phases of the interfering beams, respectively. The number N of the interfering beams yields the order of the rotational symmetry of the quasicrystal pattern [27]. By adjusting the parameters in equation (1), different interference patterns can be obtained. The wave vectors of the interfering beams are given by [15]

$$\mathbf{k}_m = \frac{2\pi n}{\lambda}(\sin(\frac{2\pi m}{N})\sin\theta, \cos(\frac{2\pi m}{N})\sin\theta, \cos\theta), \tag{2}$$

where the $\mathbf{k}_m$, m=(1, …, N), are oriented at angle θ with respect to the longitudinal z-direction, and are equally distributed along the transverse (x, y)-plane; n is the average refractive index of the photosensitive mixture, and λ is the common wavelength of the beams. In our simulations θ ranges from 30° to 65°. We simulated (and realized) quasi-periodic patterns with 8-, 9-, 10-, 12-, 17-, 23-fold rotational symmetry.

In the one-dimensional case, a binary Thue-Morse sequence is constructed using the following rule: given an arbitrary sequence of two symbols, A=0 and B=1, say, a new sequence is formed by replacing each occurrence of A with the pair (A, B) and any occurrence of B with the pair (B, A) [17]. The one-dimensional quasi-periodic Thue-Morse grating is formed starting from the single element A. The one-dimensional Thue-Morse grating is generalized to the two-dimensional case by the recursive rule defined by

$$\mathbf{M}_{\mathrm{n+1}} = \begin{pmatrix} \mathbf{I}_{\mathrm{n,n}} - \mathbf{M}_{\mathrm{n,n}} & \mathbf{M}_{\mathrm{n,n}} \\ \mathbf{M}_{\mathrm{n,n}} & \mathbf{I}_{\mathrm{n,n}} - \mathbf{M}_{\mathrm{n,n}} \end{pmatrix}, \quad \mathbf{I}_{\mathrm{n,n}} = \begin{pmatrix} 1_{11} & \cdots & 1_{1\mathrm{n}} \\ \vdots & \ddots & \vdots \\ 1_{\mathrm{n}1} & \cdots & 1_{\mathrm{nn}} \end{pmatrix}, \tag{3}$$

where $\mathbf{I}_{n,n}$ is a 2^nx2^n matrix in which each element is equal to one, and the initial matrix, for n=1, is $\mathbf{M}_1$=(0). It is easily verified, in fact, that any row and any column of the $2^{(n+1)}$x$2^{(n+1)}$ symmetric matrix $\mathbf{M}_{n+1}$ is a Thue-Morse sequence of order n+1 generated from the first element of the row or column itself. The initial condition of the recurrence, defined by the starting matrix (a single element is a particular case), determines the symmetry and the properties of the 2D-Thue-Morse pattern affecting the optical properties of the resulting dielectric structure. In fact, given a binary square matrix of arbitrary size as the initial condition of Eq. (3), the resulting matrix of the iteration is a Thue-Morse 2D-pattern in which the unit cell corresponds to the matrix given in the initial condition. We expect that changing the unit cell from a scalar to an arbitrary square matrix would imply a modification of the Thue-Morse superstructure and, hence, of the photonic band gap properties of the quasicrystal. Typically ten or more iterations of Eq. (3) are needed in order to achieve the matrix size saturating the resolution of our SLM.

A homogenous mixture of photopolymer, liquid crystal, and photo-initiator is exposed to the required irradiance profile at the conjugate plane of the hologram. Photo-polymerization of the monomer occurs in the bright region of the writing pattern, initiating a diffusion process where the LC-molecules diffuse into the dark regions of the pattern. The intensity modulation is finally recorded as a refractive index modulation due to the phase separation of LC-rich regions and polymer-rich regions, and due to the refractive index mismatch between the

#91997 - $15.00 USD Received 22 Jan 2008; revised 13 Mar 2008; accepted 14 Mar 2008; published 31 Mar 2008

polymer an the liquid crystal. The modulation of the dielectric function gives rise to a photonic crystal with a quasi-periodic and/or aperiodic arrangement of the structure. We used the frequency doubled continuous wave Coherent Verdi at λ=532 nm for the optical curing of our samples during the holographic process. This was a single step process with time exposures between a few seconds and several minutes depending on the power of the single laser beam incident on the sample after the spatial modulation. The laser power ranged typically from 1 to 20 mW at the position of the sample. Our quasicrystal structures were obtained by curing, at room temperature, a pre-polymer/LC photosensitive mixture in the image plane of the CGH, where the desired reduced irradiance profile is reconstructed by means of two relay lenses with focal lengths ranging between 63 and 500 mm depending of the wanted lateral magnification. The spatial light modulator LC-R 3000 (for visible light) had a pixel pitch of 9.5 μm and we adopted a lateral magnification of 0.1-0.2 in this work.

We used a starting solution of the monomer dipentaerythrol-hydroxyl-penta-acrylate DPHPA (60.0% w/w), the cross-linking stabilizer monomer N-vinylpyrrolidinone (9.2% w/w), the liquid crystal BLO38 by Merck (30.0% w/w), and a mixture of the photoinitiator Rose Bengal (0.3% w/w) and the co-initiator N-phenylglycine (0.5% w/w) [28,31,32]. The polymer has a refractive index n_p=1.530, whereas the LC BL038 has an ordinary refractive index n_o=1.527 and an extraordinary refractive index n_e=1.799. The average refractive index of the mixture is estimated to be $n \sim 1.57$. The thickness of the sample cell varied from about 5 to 30 μm, whereas the cured area had a linear dimension ranging from ~ 4 to 20 mm. The diffraction patterns in Figs. 1 and 2 were obtained with a He-Ne laser at λ=633 nm in the back focal plane of a lens with a focal length of 300 mm. The non-diffracted beam was cut by a screen. The observed diffraction patterns are in a very good agreement with the calculated (FT) ones, as shown in Figs. 1 and 2.

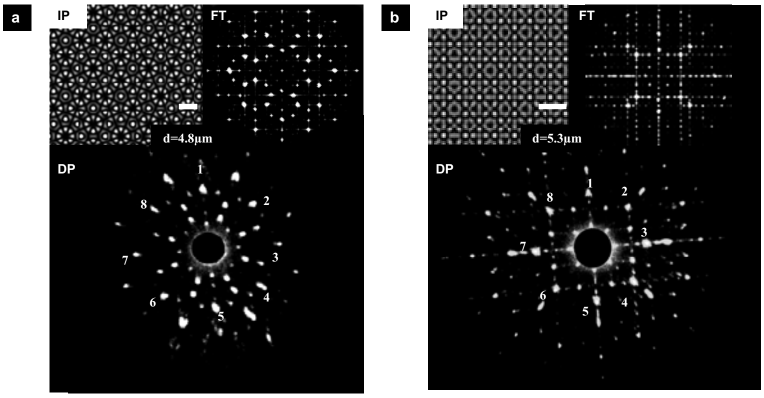

Fig. 1. (a) Penrose-tiled quasicrystal structure with 8-fold rotational symmetry and phase set 1 ($\varphi_i = 0$, for $i = \{1,\ldots,8\}$); (b) Penrose-tiled structure with 8-fold symmetry and phase set 2 ($\varphi_2 = \varphi_4 = \varphi_6 = \varphi_8 = \pi/2+\varphi$, where φ represents the phase of the remaining beams). (a)-(b) Top left inset: calculated irradiance profile (IP), top right inset: 2D Fourier transform (FT) of the irradiance profile, bottom inset: observed diffraction pattern (DP); d estimates the self-similarity cell size of the structures.

In Figs. 1(a)-1(b) and 2(a)-2(f) three insets are shown. Each figure, in fact, consists of: the calculated Irradiance Profile (IP) sent to our SLM and then imaged at the sample position; the calculated 2D Fourier Transform (FT) of this irradiance pattern; and the experimental Diffraction Pattern (DP) produced by the written structure. Examples of quasiperiodic structures achievable varying the initial phases of the interfering beams in the numerical calculation of the irradiance profile are presented in Figs. 1(a) and 1(b) for a structure with 8-fold rotational symmetry. In Fig. 1(a) all the beams possess the same initial phase, say, phase set 1; in Fig. 1(b), instead, the initial phases of the eight beams are $\varphi_2 = \varphi_4 = \varphi_6 = \varphi_8 = \pi/2+\varphi$,

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say, phase set 2, where φ represents the phase of the remaining beams [15]. Figures from 2(a) to 2(e) refer to 2D Penrose-tiled structures having 9-, 10-, 12-, 17-, 23-fold symmetries in this order; Fig. 2(f) shows a 2D Thue-Morse quasicrystal. The similarity between the positions and cone angle of the spots in the calculated Fourier transform FT and the observed diffraction pattern DP demonstrates the good quality of the quasicrystals written with our holographic technique. These structures are two-dimensional phase gratings in which the modulation profile of the average refractive index is not easily accessible with the scanning electron microscopy. Nevertheless, the observed optical diffraction patterns doubtless substantiate the presence of the index profile. The diffraction spots in first and higher orders show the expected *N*-fold symmetry and have *N* points for even symmetry and 2*N* points for odd symmetry (Figs. 1 and 2). The magnitudes of the basic reciprocal vectors in the diffraction patterns are related to sensible lengths of the crystal structures, such as a tile side [16,33]. In this way we estimated the size *d* of the self-similarity cell (the "unit cell") to be in the range between 4.8 and 8.6 µm, increasing with the order of the symmetry, as reported in Figs. 1 and 2 (IP insets). It is worth noticing that, even if the typical length of the obtained structures was in the range 5-10 µm, the typical separation between equal dielectric regions in the fine structure of the self-similarity cell was in the range of ~1-2 µm. In fact, we were able to realize 1D Bragg grating and 2D periodic square lattice with a pitch of ~1µm. The scale length of the realized structures depends on the lateral magnification of the relay lenses, which also affects the extension of the writing area, as long as the spatial resolution is far from the diffraction limit. The ultimate limit in the achievable resolution depends on the SLM pixel size and on the wavelength of the writing light. Using UV light and state-of-the-art SLM

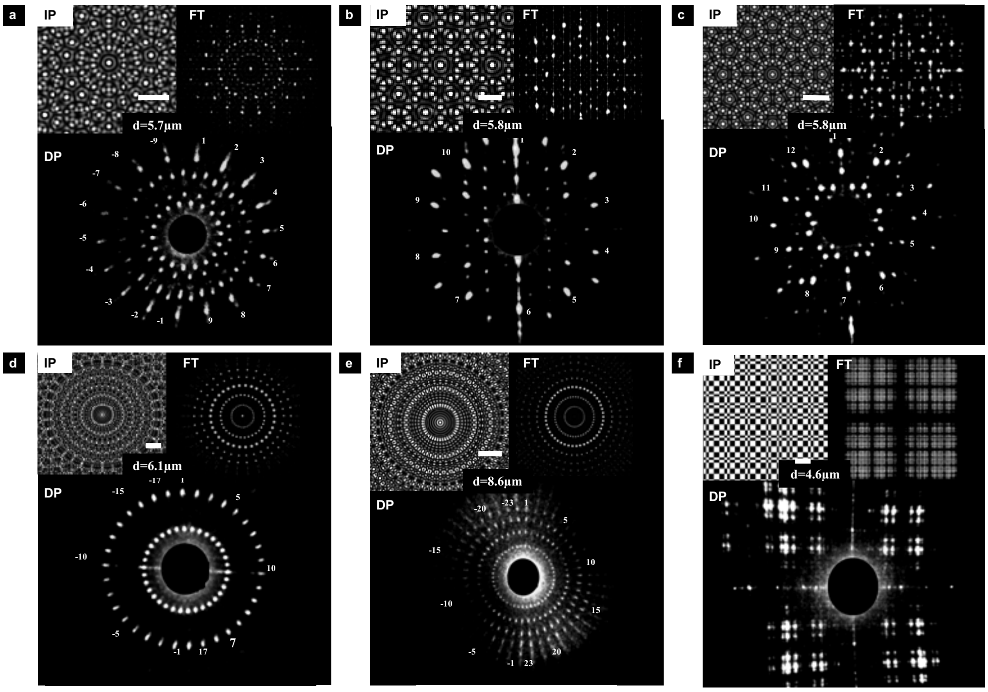

Fig. 2. (a) Penrose-tiled quasicrystal structure with 9-fold rotational symmetry, (b) 10-fold symmetry, (c) 12-fold symmetry, (d) 17-fold symmetry, (e) 23-fold symmetry. (f) Two-dimensional Thue-Morse quasicrystal structure. (a)-(f) Top left inset: calculated irradiance profile (IP), top right inset: 2D Fourier transform (FT) of the irradiance profile, bottom inset: observed diffraction pattern (DP); *d* estimates the self-similarity cell size of the structures.

pixel size [34], the limit in the spatial resolution could be improved by a factor of 3-4. Since now however, the scalability of the optical properties permits to use our quasiperiodic structures to study their interesting yet unknown optical behavior.

3. Conclusion and discussion

In conclusion, we used Computer-Generated Holograms to drive a liquid crystal Spatial Light Modulator that permits to write arbitrary structures with simple and reliable optical setup. The writing light intensity pattern can be modified at will by real time change of the CGH sent to the programmable SLM. Accurate control of arbitrary pattern designs can be achieved without mechanical motion and optical realignment. Furthermore, highly reproducible patterns can be obtained over an area of hundreds of square millimeters with micrometric resolution and without drawbacks due to the long term stability of the holographic process. We were able to obtain with a single beam technique quasiperiodic Penrose-tiled structures with unprecedented rotational symmetry up to 17- and 23-fold and two-dimensional Thue-Morse structures too. To realize a 23-fold quasicrystal with the usual multi-beam holographic lithography, 23 interfering laser beams would have been necessary. Furthermore, now it is open the possibility of a full characterization of quite interesting aperiodic structures, like two-dimensional Thue-Morse, which have been studied until now only theoretically [24]. In fact, this kind of pattern could not be realized by multiple beam holography even in principle. The quality of our structures was demonstrated by the good agreement between the observed and calculated optical diffraction pattern. Our structures were written into polymeric liquid crystal films, so to permit switching by external fields [16]. The PBG is not expected for the small index contrast (~0.2) we can achieve, even though small index contrast have produced absolute PBGs in 12-fold symmetric photonic quasicrystals [4]. We are working in order to obtain 2D quasicrystals with improved features not only in terms of spatial resolution along the line mentioned above but also in terms of better optical contrast by choosing a different material, so to match the demands for practical applications. In fact, our holographic technique could be applied, in principle, to any photosensitive material (e.g. with a larger index contrast), or to produce patterned masks and templates for use in lithography of hard materials [9-16,27]. We expect that the SLM based CGH technique may have a large impact on the production of complex photonic structures.

Acknowledgments

This work has been partially supported by INTAS under the project no. 05-1000008-7902 and by the NATO grant CBP.NUKR.CLG.981968.

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