

Thue-Morse nanostructures for tunable light extraction in the visible region



M. Rippa^a, R. Castagna^a, A. Marino^a, V. Tkachenko^a, G. Palermo^b, A. Pane^c, C. Umeton^b,
N. Tabiryan^d, L. Petti^{a,*}

^a Institute of Applied Sciences and Intelligent Systems “E. Caianiello” of CNR, Via Campi Flegrei 34, Pozzuoli 80072, Italy

^b Department of Physics – University of Calabria, Centre of Excellence for the Study of Innovative Functional Materials, Arcavacata di Rende 87036, Italy

^c CNR - NANOTEC - UOS di CS – Licryl c/o Department of Physics – University of Calabria, Arcavacata di Rende 87036, Italy

^d Beam Engineering for Advanced Measurements, Co.809 South Orlando Ave., Suite I, Winter Park, FL 32789, USA

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ABSTRACT

Controlling light propagation at the nanoscale is a fascinating opportunity offered by modern photonics, more than a challenge to face off. This study is aimed at investigating a particular kind of nanocomposite and reconfigurable optical metamaterials that can be exploited for the realization of a new class of switchable photonic devices, representing a breakthrough with respect to the state of the art. Existing photonic devices exhibit, in general, a drawback in the absence of tunability; this work aims to the design and characterization of metamaterials exploiting reconfigurable media, like LCs, which enable realization of a tunable, high quality, photonic quasi-crystal based switchable mode selector. It turned out that, starting from an unpolarized white light source, through a light extraction mechanism based on the diffraction of light, the high quality structure, combined with a uniformly aligned Photo-responsive Liquid Crystal (PLC), is able to give rise to an extremely narrow (FWHM ≈ 5 nm) and linearly polarized single mode peak of the extracted light intensity. Moreover, we have shown that the spectral properties (switching) of the samples can be finely controlled by using both an external applied voltage and a suitable pump light source with a maximum increase of 45% of the extracted light. Finally, both Scanning Electron Microscopy (SEM) and Far Field Diffraction (FFD) analysis have shown the high quality morphology of the realized structure.

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1. Introduction

Engineering efficient nanostructures for light manipulation offers a great potential to develop devices with high performance, which go from optical components to sophisticated sensors [1–4]. The light interaction at nanoscale gives rise to a variety of interesting physical effects that allow to control and tune both intensity and phase of an optical signal as well as its propagation properties. Periodic arrangements of nano-elements, known in literature as Photonic Crystals (PhCs), have been extensively used to control the spectral properties of guided light in different kind of devices, by improving its confinement or, on the contrary, by preventing its propagation [5,6]. In particular, the use of PhCs for filtering and selecting light extraction, has contributed to develop and boost the performance of different types of devices, for example by enhancing the optical absorption in solar cells [7–9], by improving the efficiency of both semiconductor and organic light-emitting diode (LED and OLED) [10,11], and by enabling the design of compact low cost spectrometer [12], the engineering of colorimetric sensors [13,14], the realization of tunable color filters and polarimetry [15], up to showing

great potential in the development of high-resolution display technologies [16]. However, the interests and efforts of many research groups that are focused on these issues aim to the development of new nanosystems that enable more efficient and compact devices. In this framework, aperiodic nanostructures may represent a good solution due to their unique optical properties. It has been shown that nanostructures based on aperiodic geometries can allow an exceptional degree of control of light characteristics, so to provide the realization of devices with performance strongly enhanced respect to the periodic counterpart [17–23]. While periodic systems are of fundamental importance in various phenomena that govern wave transport and interference, deviations from periodicity may result in high complexity and give rise to a number of surprising effects. Photonic Quasi - Crystals (PQCs) exhibit unique optical properties because: (i) by using well-defined algorithms, they can be designed to give rise to controlled interference patterns; (ii) they possess unique and rich symmetries in the Fourier space, which are not allowed in a periodic lattice. In fact, the structural complexity of PQCs is measured by their spatial Fourier spectra, which can be discrete (singular), singular-continuous or absolutely continuous for pseudorandom structures of increasing complexity. Among the various types of aperiodic patterns, the optical properties of the Thue-Morse (ThMo) geometry have been widely investigated, and their peculiarities as self-similar

* Corresponding author.

E-mail address: l.petti@isasi.cnr.it (L. Petti).

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hierarchy of pseudoband-gap regions, omnidirectional reflectivity and high localized optical state make such geometries an attractive candidate for the realization of devices with high efficient light extraction manipulation [24–29]. Electromagnetic properties of PhCs and PQC's can be controlled by properly designing their geometry: almost each single parameter of these structures, related to periodicity or aperiodicity, shape, material, etc., can be managed to achieve a desired propagation or extraction property. In this way, by finely controlling lattice parameters of a given photonic structure, it is possible to optimize its extraction characteristics at a precise wavelength, or minimize the extraction of undesired modes. In general, however, once a structure is realized, its extraction properties cannot be varied. To cross this inconvenient, it is possible to combine capabilities offered by both PhCs and PQC's with the reconfigurable properties of smart materials, such as Liquid Crystals (LCs); in this way, a completely new class of “reconfigurable metamaterials” can be realized. This synergy opens up the possibility to realize new generation devices such as tunable metamaterials or active filters; moreover, LCs enable the possibility to control the surface plasmon polaritonic (SPP) properties by applying an external perturbation (electric or magnetic field, temperature variations, etc.).

Recently, we have already reported on the realization and characterization of a switchable photonic device, working in the visible range, based on nanostructured metal Thue Morse PQC's, layered with a PLC [30]. Both experimental characterization and numerical simulations have shown that excitation spectra can be controlled by applying an external voltage or by means of a laser light. In this work, we have characterized some engineered 2D compact ThMo nanostructures with different minimum interparticle distance (*mid*), along with their performance, in terms of light extraction, compared with those of a periodic pattern. To fabricate the nanostructures we used electron beam lithography (EBL) technique. Using the aperiodic ThMo geometry, we demonstrated, the possibility to realize devices with a vertical light extraction of a filtered color, working on all visible spectral region. Moreover, by comparing their performance with those of a PhC, in the case of ThMo patterns, we found extracted signals having double energy and narrower full width at half maximum (FWHM) with respect to those of the periodic counterpart. Successively, we have tested one of the characterized ThMo nanopattern inside a real device based on a PLC cell. By using the peculiar optical properties of liquid crystals in term of director reorientation and phase transition, we show the possibility to tune the extracted light through the use of both an external applied voltage (electrical tuning) and an incident laser light (optical tuning), achieving an increase up to almost 50% of the peak intensity. Obtained results demonstrate how the greater complexity required in the design and manufacture of aperiodic nanostructures can be well repaid by outstanding optical performance.

2. Materials and methods

2.1. Fabrication of nanostructures and morphological characterization

We used Electron Beam Lithography (EBL) technique (Raith 150 EBL system) to fabricate three $400 \times 400 \mu\text{m}$ gold nanostructures based on ThMo arrays with circular Nano Particles (NPs) and minimum interparticle distances (*mid*) of 50, 100 and 150 nm. The diameter of NPs was $d = 300 \text{ nm}$. Moreover, in order to make a comparison, we fabricated a $400 \times 400 \mu\text{m}$ gold periodic photonic crystal (PhC) based on circular NPs with size optimized by simulation and comparable to those used for ThMo patterns with $d = 300 \text{ nm}$ and *mid* of 100 nm.

A 140 nm layer of styrene methyl acrylate based polymer (ZEP) electron-sensitive resist was spin coated on a 15 nm conductive ITO coated glass substrate, baked at 170° for 5 min and exposed to the 16 pA electron beam current. The patterns were generated in ZEP layer after a sequence of three steps: development in a *n*-Amyl acetate solvent for 90 s, then rinsed for 60 s in 1:3 MIBK:IPA solution (Methyl isobutyl ketone:Isopropyl alcohol), and finally rinsed in IPA for further 30 s. The gold nanostructure arrays were created by evaporating 2 nm Cr and,

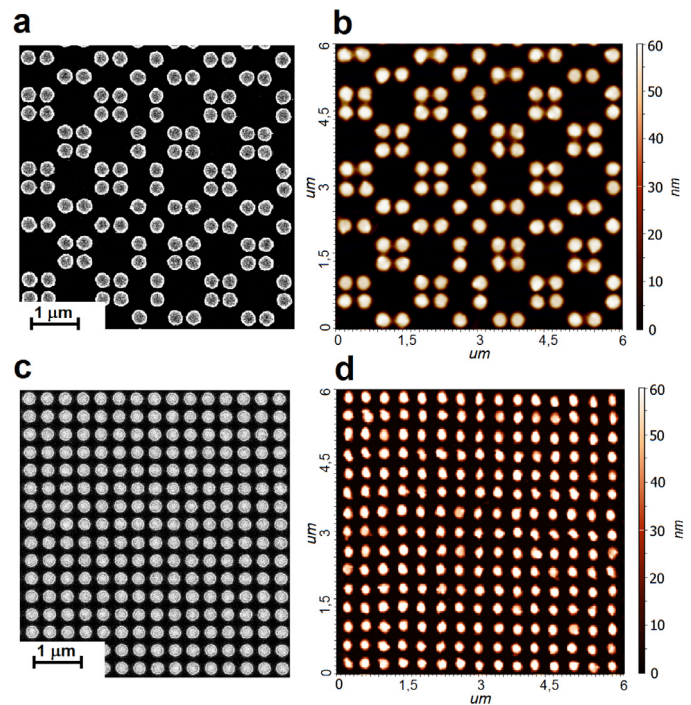


Fig. 1. Morphological characterization: (a) SEM and (b) AFM images of the ThMo nanopatterns with *mid* = 100 nm, (c) SEM and (d) AFM images of the PhC with *mid* = 100 nm.

subsequently, 60 nm of gold film (SISTEC CL-400C e-beam evaporator) to the ZEP surface and then with an additional lift-off step carried out by immersing the samples in *N*-Methylpyrrolidinone (NMP) heated at 80°C for 5 min, and then sprayed with a squizzle of NMP to remove the gold film [19,29]. Morphological characterization was performed both by SEM (Raith 150) and Atomic Force Microscopy (AFM - NT-MDT NTE-GRA Spectra) analysis. The microscope images in Fig. 1 refer to ThMo (a and b) and PhC (c and d) nanostructures both with *mid* = 100 nm. They show how both geometries are highly uniform and regular on the whole patterned area.

2.2. Optical setup for light extraction

An optical setup was used to evaluate the spectrum of the vertically extracted light at room temperature (Fig. 2). In the setup the white light of an unpolarized halogen source (HL 2000 Ocean Optics) is coupled from the edge of the glass substrate by a fiber. The light propagates in the glass due to the total internal reflection and reaches the nanostructure where a part of it is extracted by diffraction. The extracted light is collected by a fiber (N.A = 0.22) positioned 3 cm from the surface of the sample and analyzed by a spectrophotometer in the Vis-NIR region (USB 4000 Ocean Optics).

2.3. 3D-FDTD simulations for size optimization

Given the difficulties to achieve through numerical calculation an accurate prediction of the optical behavior of the considered aperiodic structures, we have firstly optimized the characteristic sizes of the periodic structure by numerical simulations, and, subsequently, we have fabricated ThMo patterns of size comparable to it. In order to achieve an optimized out-coupling efficiency in the visible region for the periodic crystals through an appropriate pattern design, we have calculated the vertically extracted light (with a resonant wavelength) by 3D numerical simulations employing a finite difference time-domain numerical method (FDTD). We have assumed that the glass substrate (BK7) and air are semi-infinite with constant refractive indices $n_{\text{glass}} = 1.51$ and

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