Realization of optical periodic quasicrystals using holographic lithography

Xia Wang, a) Jun Xu, Jeffrey Chi Wai Lee, Yee Kwong Pang, Wing Yim Tam, b) C. T. Chan, and Ping Sheng

Department of Physics and Institute of Nano Science and Technology, Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, Hong Kong, China

(Received 16 June 2005; accepted 5 December 2005; published online 30 January 2006)

We have fabricated three-dimensional periodic quasicrystals exhibiting quasiperiodicity of the Penrose structure in the x-y plane and periodic along the z axis using ten-beam visible light holographic lithography. The quasicrystals show photonic band gaps in the visible range. The band gaps are found to follow a simple relation as a function of periodicity and polymeric volume fraction, in accord with the Bragg’s diffraction relation. © 2006 American Institute of Physics.

Holographic lithography, a method combining holography and photoinduced polymerization techniques, has attracted much interest recently in fabricating mesoscale structures such as photonic materials like photonic crystals. Holographic lithography has certain advantages over the conventional techniques, such as the self-assembly of colloidal microspheres or microfabrication in the fabricating of photonic crystals, even though it is more difficult to incorporate defects in the crystals as compared to, for example, the microfabrication technique. In this method, holographic pattern formed by the interference of multiple coherent beams is projected onto a photosresist which, after exposure, can be developed as a “record” of the interference pattern. The holographic pattern displays maxima at locations satisfying the Bragg diffraction condition, thus creating periodic mesoscale structures when exposed onto photosresist. For example, a fcc lattice can be obtained from a bcc reciprocal lattice constructed from a four-beam interference with an “umbrella” configuration. The holographic lithography method can be extended to a periodic quasicrystals as demonstrated recently in making two-dimensional (2D) quasicrystals with Penrose symmetry by using a five-beam configuration and a dual beam multiple exposure technique. Quasicrystals are attractive in photonic applications because they have higher rotational symmetries than conventional photonic crystals, implying more isotropic photonic band gaps. They also exhibit interesting wave propagation properties. Holographic lithography offers some advantage in the fabrication of mesoscale quasicrystals. The 2D Penrose quasicrystals fabricated by the five-beam configuration, and the dual beam multiple exposure technique, are steps in this direction.

In this contribution, we report the use of a ten-beam visible light holographic lithography method to fabricate a three-dimensional (3D) polymeric resin structure that has a quasiperiodic (i.e., Penrose) pattern in 2D and is periodic in the third (z) direction. Such structure is denoted a periodic quasicrystal, and can be regarded as a variant of the icosahedral quasicrystal. The periodic quasicrystals fabricated exhibit colorful photonic band-gap characteristics in the visible range.

To fabricate the periodic quasicrystal, we use a ten-beam configuration for our holographic lithography setup as shown in Fig. 1(a). The ten beams can be represented by

\[ \mathbf{k}_n = k \left( \cos \frac{2n\pi}{5} \sin \varphi, \sin \frac{2n\pi}{5} \sin \varphi, \cos \varphi \right), \]

and

\[ \mathbf{k}_n' = k \left( \cos \frac{2n\pi}{5} \sin \varphi, \sin \frac{2n\pi}{5} \sin \varphi, -\cos \varphi \right), \]

for \( n = 0 \rightarrow 4 \). In Eqs. (1) and (2) \( k = 2\pi/\lambda \) (\( \lambda \) is the wavelength of the laser light inside the photosresist) and \( \varphi \) is the angle between each beam with the vertical (z) axis. We have recently fabricated 2D Penrose quasicrystals using a five-beam holographic lithography method with wave vectors given by Eq. (1). Here, \( \mathbf{k}_n' \) are simply the reflection images of the \( \mathbf{k}_n \).

[FIG. 1. (a) Schematic setup for the ten-beam interference. The \( \mathbf{k}_n' \) beams are obtained by reflections of the \( \mathbf{k}_n \) beams. (b) SEM image of periodic quasicrystal. The inset is a normal view of the sample. The scale bar is 2 \( \mu \)m and the circle shows the tenfold symmetry of Penrose. (c) Simulation of the periodic quasicrystal using ten-beam interference. The image is an intensity contour of the 3D structure. The inset is a normal view. (d) Optical image (reflection) of the periodic quasicrystal. The inset is a diffraction pattern obtained by focusing a He–Ne laser beam between regions (1) and (2) as indicated. The scale bar is 500 \( \mu \)m. Reflection and transmission spectra and SEM images are obtained along the dashed lines.

---

a) On leave from the Department of Mathematics and Physics, QingDao University of Science and Technology, China.
b) Author to whom correspondence should be addressed; electronic mail: phtam@ust.hk

DOI: 10.1063/1.2168487
from the x-y plane. The interference pattern of the ten beams in Eqs. (1) and (2) is given by the following intensity pattern:

$$I(r) = \sum_{n,m} (E_n e^{-ik_n r} + E_n^* e^{ik_n^* r}) \cdot (E_m e^{ik_m r} + E_m^* e^{-ik_m^* r})$$  \(3\)

for \(n, m=0\ldots4\) with polarizations \(E_n\) in the planes of incident of each beam. This pattern is characterized by quasiperiodicity in the x-y plane and periodicity along the z axis with period given by \(\lambda/2 \cos \phi\). Patterns given by Eqs. (1)–(3) represent a class of periodic quasicrystals for different values of \(\phi\).

The five direct beams, \(k_n\) in Eq. (1), were obtained by splitting the 488 nm line of an argon ion laser using a reflection grating. The splitted beams (beam size 4 mm, power 1.55 mW, and polarization on the plane formed by each beam with the axis of symmetry), were placed symmetrically at 72° from neighbouring beams around an axis. In order to obtain an incident angle \(\phi (~60°)\) greater than the critical angle of the photoresist (on glass substrate), an inverted pentagonal prism, similar to that used in Miklyaev et al., was employed such that the five laser beams each entered at normal to one of the five tilted surfaces of the prism and collimated with other beams at a point inside the photoresist. The configuration in Fig. 1(a) was realized by simply reflecting the five direct beams \(k_n\) by a mirror placed in contact with the photoresist to obtain the five reflected \(k'_n\). Index matching fluids were placed between interfaces (prism to substrate and photoresist to mirror) to reduce multiple reflections from the interfaces.

We used photoresist “SU8” (from Shell) as the raw polymer resin. The SU8, dissolved in \(\gamma\)-butyrolactone (from Aldrich) (1:1) with 2 wt % of photoinitiator Irgacure 261 (from Ciba Co.) sensitive to the 488 nm line, was spin-coated on glass plates to form ~15 \(\mu\)m thick samples. The samples were heated to ~90 °C for about an hour to remove any solvent left. After exposure, a post-thermal treatment at ~90 °C for about 20 min was needed to complete the polymerization. Polymerization occurred only at regions where dosage of exposure from the interfering pattern exceeded a critical value, while underexposed regions were washed away first by developing the sample in propylene-glycol-methyl-ether-acetate (developer) for more than 10 h and then cleaned with acetone for 5–10 min, creating a copy of the 3D interference pattern. The cleaning with acetone turned out to be a crucial step to obtain lattice spacing in the optical range. During the acetone cleaning process the sample was found to shrink substantially, about two times more than the 20% shrinkage without the acetone, creating periodicitities and features much smaller than the wavelength, 0.301 \(\mu\)m (0.488 \(\mu\)m/\(n_{SU8}=1.62\)), of the laser light inside the photoresist.

Note that the spacing could also be modified also by changing the incident angle of the beams. Figure 1(b) shows cross-sectional scanning electron microscope (SEM) image of the fabricated 3D mesoscale periodic quasicrystal. It is clear that the crystal is periodic in the normal direction with -0.22 \(\mu\)m periodicity (corrected for the tilted angle 60° when taking the cross-sectional SEM image). The quasiperiodicity of Penrose in the x-y plane is better shown by a normal view SEM image in the inset. The tenfold symmetry (indicated by a white circle around a ring of ten holes) is clearly seen. The fabricated periodic quasicrystal resembles closely the simulated pattern, shown as a 3D intensity contour in Fig. 1(c), using the ten-beam interference given by Eqs. (1)–(3). Also shown is a normal view (inset) of the simulated periodic quasicrystal that is in excellent agreement with the experiment. The tenfold symmetry is further verified by diffraction pattern from a He-Ne laser focused to a beam size of around 300 \(\mu\)m normal to the sample as shown in the inset of Fig. 1(d) for the quasicrystal in Fig. 1(b). The tenfold symmetry is again obvious as seen from the rings of intense dots. Furthermore, the fabricated periodic quasicrystal shows colorful reflections in the visible range as shown in Fig. 1(d), viewed under a microscope. In addition, transmission image also shows complimentary colors, indicating that the color originates from the structure. The different color reflections are due to variations of periodicity and polymer volume fraction as a result of intensity inhomogeneities of the interfering laser beams and uneven shrinkage of the photoresist during the developing process. Normal reflection and transmission spectra in the visible range were obtained by using an optical microscope coupled to a spectrometer (Oriel Cornerstone 260) through an optical fiber. The microscope, with a pinhole installed in the optical path, could focus down to a size of 35 \(\mu\)m using a 20× objective. Reflectance and transmittance were normalized against backgrounds of silver mirror reflection and empty air transmission, respectively. Figure 2 shows four reflection and transmission spectra at locations indicated as circles in Fig. 1(d). Band gaps are clearly shown with gap wavelength \(\lambda_{gap}\) changing somewhat with the periodicity \(d_z\) in the z axis and polymeric volume fraction \(f\) as seen in the cross-sectional SEM images at the corresponding locations on the right-hand side of each figure. Since the refractive index of SU8 (~1.62) is not high, the dispersion should be linear except very close to the gap frequency. In that case, \(\lambda_{gap}\) should be related to the effective refractive index \(n_{eff}\) through the Bragg’s relation:

$${n_{eff}}^2 \sin^{2} \theta = n^2$$
In Fig. 3, we plot $n_{\text{eff}} = \lambda_{\text{gap}} / 2d_z$ as a function of the filling fraction $f$ from our experimental data. Also shown as a solid line in Fig. 3 is the value of $\langle n \rangle = \sqrt{n_{\text{sub}}^2 f + (1 - f)}$ (simple volume average of the dielectric constant), which should give the upper bound of the composite refractive index. Although there are scatters due to the rough estimation for the volume fraction obtained from the cross-sectional image (by counting the ratio of the bright and dark pixels), the experimental values of $n_{\text{eff}}$ have the same trend and are slightly smaller than $\langle n \rangle$, as expected. Note that this analysis is limited to the $z$ direction (normal reflectance and transmittance) and, due to the small size and the nonuniformity of the sample, it is difficult to get reasonable angular-resolved measurements to study the quasiperiodic effects due to the Penrose structure on the $x$-$y$ plane. Techniques reported recently by Wu et al. in Ref. 1 and Gauthier et al. in Ref. 5 could be used to fabricate larger and more uniform samples to fully explore the potential of such 3D quasicrystals.

To summarize, we have fabricated optical 3D periodic quasicrystals using a ten-beam visible light holographic lithography. The quasicrystals exhibit 2D quasiperiodicity of the Penrose structure and periodicity along the $z$ axis, in excellent agreement with simulated interference pattern. Furthermore, they are very colorful, exhibiting photonic band gaps in the visible range. The band gaps are found to follow a simple relation with periodicities and polymeric volume fractions, in accord with the Bragg’s diffraction relation.

Support from Hong Kong RGC grants HKUST6145/00P, N_HKUST033/00, CAO02/03.SC01, and HKUST603303 is gratefully acknowledged.


8See for example, L. Guidoni, C. Triche, P. Verkerk, and G. Gryenberg, Phys. Rev. Lett. 79, 3363 (1997) for using interference of five or six laser beams to create quasiperiodic optical lattice.