

Holographic assembly of quasicrystalline photonic heterostructures

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Quasicrystals have a higher degree of rotational and point-reflection symmetry than conventional crystals. As a result, quasicrystalline heterostructures fabricated from dielectric materials with micrometer-scale features exhibit interesting and useful optical properties including large photonic bandgaps in two-dimensional systems. We demonstrate the holographic assembly of two-dimensional and three-dimensional dielectric quasicrystalline heterostructures, including structures with specifically engineered defects. The highly uniform quasiperiodic arrays of optical traps used in this process also provide model aperiodic potential energy landscapes for fundamental studies of transport and phase transitions in soft condensed matter systems.

Quasicrystals have long-ranged orientational order even though they lack the translational periodicity of crystals. Not limited by conventional spatial point groups, they can adopt rotational symmetries that are forbidden to crystals. The resulting large number of effective reciprocal lattice vectors endows quasicrystals' effective Brillouin zones with an unusually high degree of rotational and point inversion symmetry [1]. These symmetries, in turn, facilitate the development of photonic band gaps (PBG) [2] for light propagating through quasicrystalline dielectric heterostructures [3–5], even when the dielectric contrast among the constituent materials is low. Photonic band gaps have been realized in one- [6] and two-dimensional [7] lithographically defined quasiperiodic structures. Here we demonstrate rapid assembly of arbitrary materials into two-dimensional and three-dimensional quasicrystalline heterostructures with

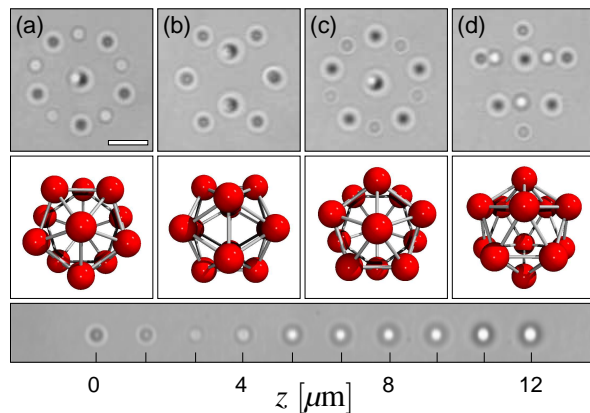


FIG. 2: Four views of a rolling colloidal icosahedron. (a) 5-fold axis. (b) 2-fold axis. (c) 5-fold axis. (d) Midplane. Scale bar indicates $5 \mu\text{m}$. The complete assembly and rolling process is shown in the associated movie.

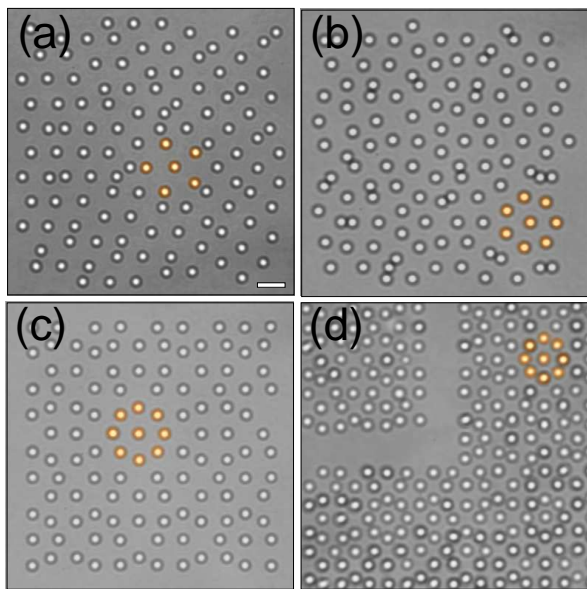


FIG. 1: Two-dimensional colloidal quasicrystals organized with holographic optical traps. (a) 5-fold. (b) 7-fold. (c) 8-fold. (d) An octagonal quasicrystal with an embedded structured defect. The scale bar in (a) indicates $5 \mu\text{m}$.

features suitable for obtaining interesting and potentially useful diffractive effects in infrared and visible light.

Our approach is based on the holographic optical trapping technique [8–10] in which computer-generated holograms are projected through a high-numerical-aperture microscope objective lens to create large three-dimensional arrays of optical traps. In our implementation, light at 532 nm from a frequency-doubled diode-pumped solid state laser (Coherent Verdi) is imprinted with phase-only holograms using a liquid crystal spatial light modulator (SLM) (Hamamatsu X8267 PPM). The modified laser beam is relayed to the input pupil of a $100\times$ NA 1.4 SPlan Apo oil immersion objective mounted in an inverted optical microscope (Nikon TE-2000U), which focuses it into traps. The same objective lens is used to form images of trapped objects, using the microscope's conventional imaging train [10].

We used this system to organize colloidal silica microspheres $1.53 \mu\text{m}$ in diameter (Duke Scientific Lot 5238) dispersed in an aqueous solution of $180 : 12 : 1$ (wt/wt) acrylamide, N,N' -methylenebisacrylamide and diethoxyacetophenone (all Aldrich electrophoresis grade). This solution rapidly photopolymerizes into a

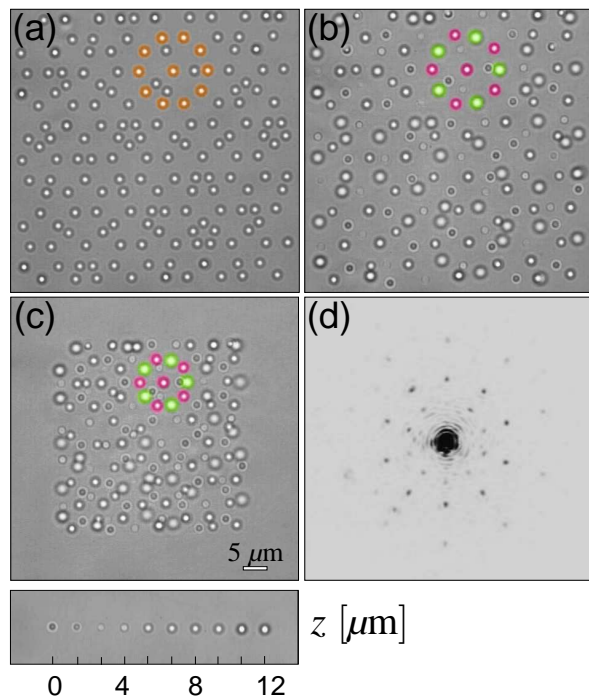


FIG. 3: Holographic assembly of a three-dimensional colloidal quasicrystal. (a) Colloidal particles trapped in a two-dimensional projection of a three-dimensional icosahedral quasicrystalline lattice. (b) Particles displaced into the fully three-dimensional configuration. The shaded region identifies one embedded icosahedron. (c) Reducing the lattice constant creates a compact three-dimensional quasicrystal. (d) Optical diffraction pattern showing ten-fold symmetric peaks. The three-dimensional assembly process is shown in the associated movie.

transparent polyacrylamide hydrogel under ultraviolet illumination, and is stable otherwise. Fluid dispersions were imbibed into $30\ \mu\text{m}$ thick slit pores formed by bonding the edges of #1 coverslips to the faces of glass microscope slides. The sealed samples were then mounted on the microscope's stage for processing and analysis.

Silica spheres are roughly twice as dense as water and sediment rapidly into a monolayer above the coverslip. A dilute layer of spheres is readily organized by holographic optical tweezers into arbitrary two-dimensional configurations, including the quasicrystalline examples in Fig. 1. Figures 1(a), (b) and (c) show planar pentagonal, heptagonal and octagonal quasicrystalline domains [11], respectively, each consisting of more than 100 particles. Highlighted spheres emphasize each domain's symmetry. These structures all have been shown to act as two-dimensional PBG materials in microfabricated arrays of posts and holes [3, 12–14]. As a soft fabrication technique, holographic assembly requires substantially less processing than conventional methods such as electron-beam lithography, and can be applied to a wider range of materials. Unlike complementary optical fabrication techniques such as multiple-beam holographic photopoly-

merization [14–17], assembly with holographic optical traps lends itself to creating nonuniform architectures with specifically engineered features, such as the channel embedded in the octagonal domain in Fig. 1(d). Similar structures of comparable dimensions created lithographically have been shown to act as narrow-band waveguides and frequency-selective filters for visible light [12, 13, 18, 19].

Holographic trapping's ability to assemble free-form heterostructures extends also to three dimensions. The sequence of images of a rolling icosahedron in Fig. 2 shows how the colloidal spheres' appearance changes with distance from the focal plane. This sequence also recalls earlier reports [20, 21] that holographic traps can successfully organize spheres into vertical stacks along the optical axis, while maintaining one sphere in each trap.

The icosahedron itself is the fundamental building block of a class of three-dimensional quasicrystals, such as the example in Fig. 3. Building upon our earlier work on holographic assembly [22], we construct a three-dimensional colloidal quasicrystal by first gathering the appropriate number of sedimented spheres into a two-dimensional arrangement corresponding to the planar projection of the planned structure, Fig. 3(a). Next, we translate the spheres along the optical axis to their final three-dimensional coordinates in the quasicrystalline domain, as shown in Fig. 3(b). One icosahedral unit is highlighted in Figs. 3(a) and (b) to clarify this process. Finally, the separation between the traps is decreased in Fig. 3(c) to create an optically dense structure. This particular domain consists of 173 spheres in roughly 7 layers, with typical inter-particle separations of $3\ \mu\text{m}$.

The completed structure was gelled and its optical diffraction pattern recorded at a wavelength of $632\ \text{nm}$ by illuminating the sample with a collimated beam from a HeNe laser, collecting the diffracted light with the microscope's objective lens and projecting it onto a charge-coupled device (CCD) camera with a Bertrand lens. The well-defined diffraction spots clearly reflect the quasicrystal's five-fold rotational symmetry in the projected plane.

Quasicrystalline heterostructures are likely to be particularly well suited for photonic bandgap applications because their effective Brillouin zones are more nearly spherical than those for photonic crystals and thus optimize the overlap of local bandgaps [23]. This property has been exploited in lithographically defined two-dimensional devices with micrometer-scale features. Recently, the photonic band structure of a centimeter-scale three-dimensional icosahedral quasicrystal was measured in the microwave region and shown to feature prominent gaps at the effective Brillouin zone edge [23].

Although photonic band structure calculations generally are lacking for dielectric quasicrystals, the structure in Fig. 3 is similar to the polymeric quasicrystalline lattice in Ref. [23] and has a comparable mismatch in dielectric constant. It therefore should have comparable

optical properties at appropriately rescaled wavelengths in the infrared and visible bands. Moreover, we expect bandgaps to be more pronounced in the holographically defined colloidal quasicrystal because of its larger volume fraction. These benefits all could be further enhanced by drying the gel at the triple point of water to further increase both the volume fraction and the dielectric constant mismatch.

Sequential assembly starting from a planar starting configuration ensures that there is one particle at each vertex of the quasicrystal. A similar outcome could be achieved in a system with microfluidic sample control without this initial step, and indeed without imaging. Larger structures then can be fabricated by assembling smaller domains, fixing them through spatially-resolved polymerization [24] or by initiating inter-particle bonds, and then combining the sub-assemblies, in much the same spirit as shoot-and-step lithography commonly used for two-dimensional semiconductor devices.

The use of monodisperse silica spheres for this demonstration was dictated by their commercial availability, low optical absorption, and high density. Holographic assembly lends itself just as easily to other materials, and so can be tailored to particular applications. Deterministic organization of disparate components under holographic control could be used to embed gain media in photonic bandgap cavities, to install materials with nonlinear optical properties within waveguides to form switches, and to create domains with distinct chemical functionalization. Distinctly engineered domains can be combined into larger heterostructures through sequential assembly, with larger-scale structures lending themselves to conventional stamping technologies. In all cases, this adaptable soft fabrication process can be directed toward creating mechanically and environmentally stable materials that can be integrated readily into larger systems.

Beyond the immediate application of holographic trapping to fabricating quasicrystalline materials, the ability to create and continuously optimize such structures provides new opportunities for studying the dynamics [10] and statistical mechanics [25] of colloidal quasicrystals. The optically generated quasiperiodic potential energy landscapes developed for this study also should provide a flexible model system for experimental studies of transport [26] through aperiodically modulated environments.

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