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Optics Letters

Quasiperiodic light

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Quasiperiodicity is a form of spatial order that has been observed in quasicrystalline matter but not light. We construct a quasicrystalline surface out of a light emitting diode. Using a nanoscale waveguide as a microscope (NSOM), we directly image the light field at the surface of the diode. Here we show, using reciprocal space representations of the images, that the light field is quasiperiodic. We explain the structure of the light field with wave superposition. Periodic ordering is limited to at most six-fold symmetry. The light field exhibits 12-fold quasisymmetry, showing order while disproving periodicity. This demonstrates that a new class, consisting of projections from hyperspace, exists in the taxonomy of light ordering. © 2022 Optica Publishing Group

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Introduction. Using macroscopic ray optics, it is trivial to create radiative fields with arbitrary structure. Shadows can have any shape. On the nanoscale, light cannot be as readily controlled because wave superposition modulates the field. In nanoscience, creating fields with new structures is a matter of intense interest. Photonic crystals, which use superposition to control waves, can in principle be prepared with any shape [1], but unlike a shadow the corresponding radiative field does not directly adopt that shape [2].

Quasiperiodicity [3] is incompatible with symmetry and is not random in real space [4]. Figure 1 contrasts symmetric spatial correlations with quasiperiodic correlations. However, viewing quasiperiodic structure in reciprocal space [5] reveals order. In simple cases, the reciprocal space representation of quasiperiodic structure is rotationally symmetric [6]. Quasiperiodic structures have space group symmetry, but it is the symmetry of a projection of a nonphysical object with more spatial dimensions than the physical structure [3]. The reciprocal space representation demonstrates that quasiperiodicity is a unique category of correlation. Quasiperiodic matter templated by light [7] and light propagation in quasiperiodic dielectric structures [1,8] have been reported previously. Here we show quasiperiodicity by directly imaging light with (a) rotational symmetry in reciprocal space, (b) long-range order developed through wave interference, and (c) the absence of all other symmetries.

Quasiperiodic matter is well known [6,9–11], but it is more difficult to demonstrate quasiperiodicity with light. To create a quasiperiodic light field, we design a photonic surface in the shape of a complementary quasicrystal with 12-fold symmetry (Fig. 2) [12–15]. By filling the holes of the complementary quasicrystal with quantum dots, we also create a luminescenent direct quasicrystal [16]. The complementary and direct quasicrystals are used to demonstrate distinct light fields with the same class of order.

Results. We fabricate a 5 mm by 5 mm quasiperiodic conventional LED from n-GaN/$In_xGa_{1-x}N$/p-GaN as illustrated in Fig. 2(a) [16]. The charge carriers flowing through the diode meet at a stack of $In_xGa_{1-x}N$ quantum wells, where they recombine into blue light. Further detail is given in the Supplementary material. The quantum well layer is penetrated by holes [14,15]. Figure 2(c) shows that the holes create a complementary quasicrystalline lattice. Supplementary Fig. S7 shows that the quantum dots occupy the holes.

For the LED, the emission pattern viewed from a distance greater than 10 mm is exceptionally uniform owing to wave superposition [17]. Measurement out of the plane of the surface distinguishes ray-optics phenomena from superposition-driven structure. The evanescent light at a distance less than 1 µm is not uniform. In order to discover the correlations in the radiative field, we insert a nanoscale waveguide as a scanning probe (NSOM) [18]. The probe is necessary because the radiative fields associated with a structure evolve as a consequence of superposition and therefore cannot be completely determined from far-field measurement [19]. The probe collects electroluminescence at different positions to image in three dimensions. The image resolution (150 nm, see Supplementary material) is aperture limited. This approach achieves direct detection of quasiperiodic structure in the radiative near field. The results include evolution to uniformity as a function of height above the sample's surface. Figure 3 shows the pattern of spots that forms in the radiative field at the surface without [Fig. 3(a)] and

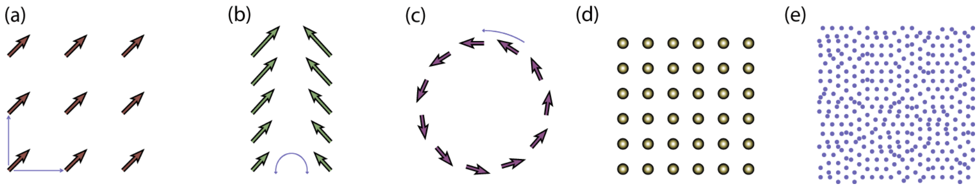

Fig. 1. Correlation in two-dimensional real space: (a) translation symmetry; (b) reflection symmetry; (c) rotation symmetry; (d) translation, reflection, and rotation; (e) quasiperiodicity.

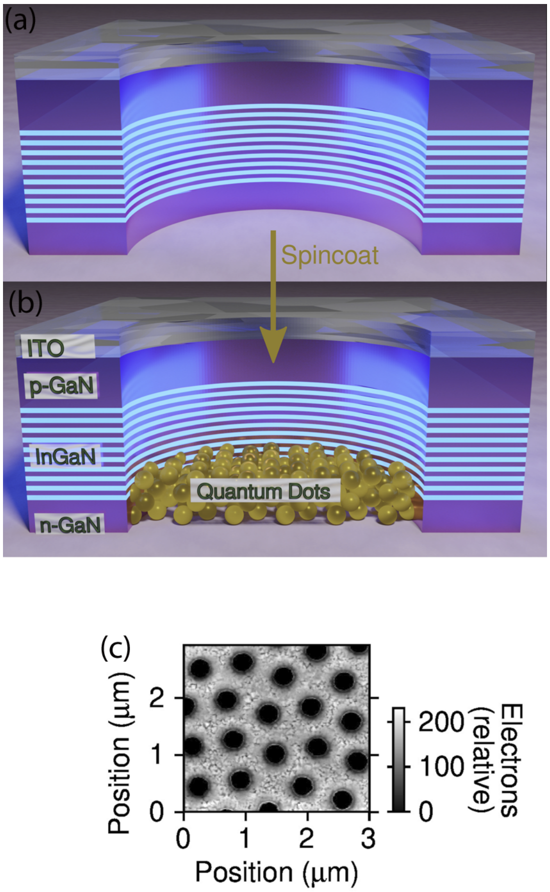

Fig. 2. (a) Cutaway illustration of the light emitting diode (LED) with a hole and (b) with quantum dots added. (c) Everhart–Thornley scanning electron image showing a top view of the LED with holes arranged to form a complementary quasicrystal.

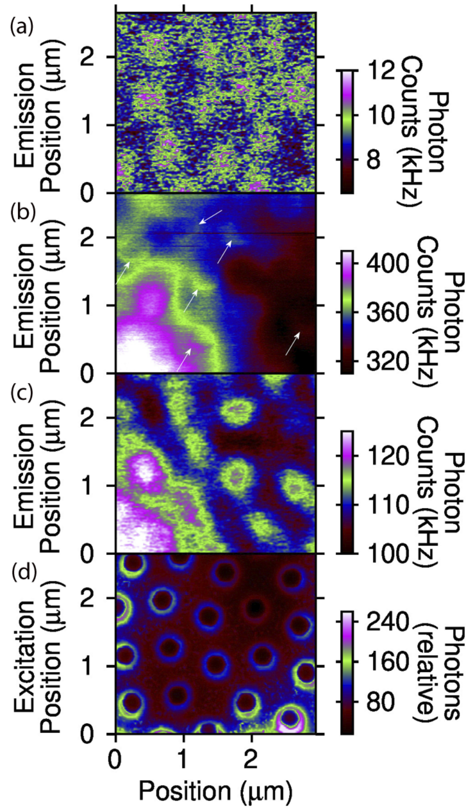

Fig. 3. (a)–(c) Scanning near field electroluminescence images of (a) complementary photonic quasicrystal electroluminescence, (b),(c) complementary quasicrystal and direct quasicrystal [(b) blue well electroluminescence; (c) orange quantum dot photoluminescence]. (d) Cathodoluminescence image. Arrows point toward the convex side of crescent-shaped features.

with [Fig. 3(b)] the addition of quantum dots. Crescent-shaped features (white arrows) were only observed in the sample with quantum dots.

The diode transfers energy to the quantum dots. The transfer mechanism is both radiative energy transfer and Förster transfer [14,16]. The quantum dots have orange photoluminescence. Using optical filters, the electroluminescence [Fig. 3(b)] and photoluminescence [Fig. 3(c)] were measured in the same location. The complementary (holey) and direct quasicrystals produce different radiative fields. In particular, the crescent-shaped features are not present in the photoluminescence from the quantum dots and the locations of the brightest areas are different.

Direct measurement shows the radiative field of each structure is quasiperiodic, as might be expected. We contrast direct measurement of light with diffractive measurement or measurement of matter ordering. Diffractive measurement is unable to distinguish clusters as small as 11 elements across [8] from patterning of the full diode. In reciprocal space images, spots indicate order. To determine the correlations in the radiative field at the surface of the quasicrystal, we Fourier transformed luminescence images. The reciprocal space representation of the radiative field image in Fig. 4(a) is 12-fold rotationally symmetric. This proves a quasiperiodic light field at the photonic surface. Two-dimensional periodic systems have reciprocal space representations with at most six-fold (but never five-fold) symmetry [20].

The expected quasicrystal structure is demonstrated by a visible-frequency Laue [8,21] diffractogram with sharp spots [Fig. 4(b)]. To show the 12-fold symmetry of the quantum well excitation, we measured the cathodoluminescence of a scanning

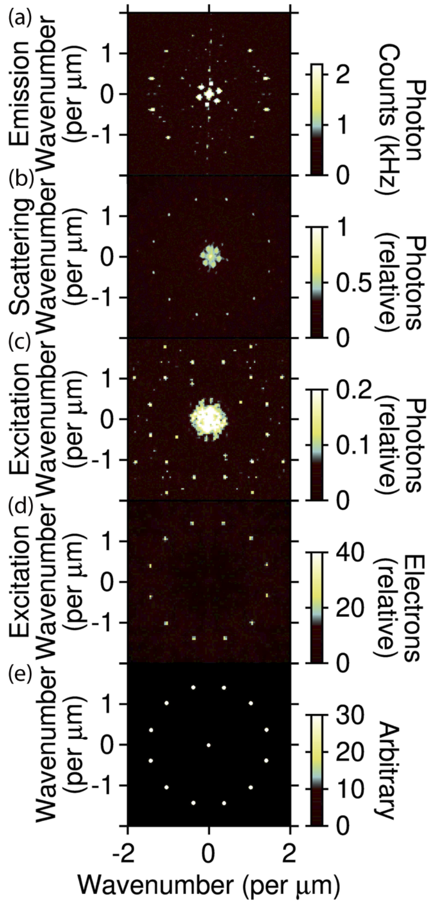

Fig. 4. Reciprocal space images are evidence of spatial order and nonlocal rotational symmetry, which are the distinguishing properties of quasiperiodicity: (a) scanning near field photoluminescence; (b) Laue; (c) cathodoluminescence; (d) Everhart–Thornley scanning electron; (e) theoretical reciprocal space image.

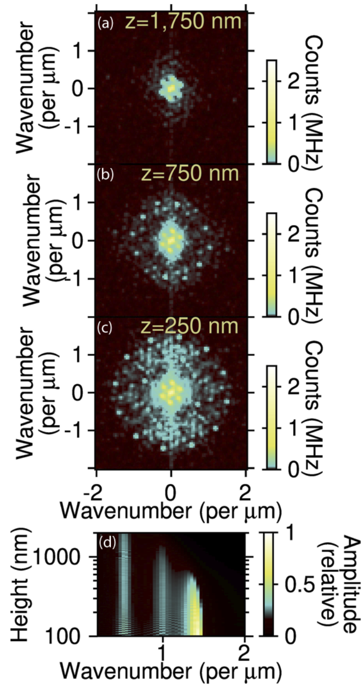

Fig. 5. (a)–(c) Reciprocal space near-field electroluminescence images recorded at different distances z from the surface of the device. (d) A side view computed using Eq. (1). Since the 12-fold symmetry is lost 1750 nm above the surface, we conclude the quasiperiodicity is evanescent, or short ranged.

electron beam [Fig. 4(c)]. The electron scattering [Fig. 4(d)] also matches a theoretical quasicrystal in reciprocal space [Fig. 4(e)] [22,23]. The quantum dot photoluminescence and device scattering images [Figs. 4(a) and 4(b)] also exhibit a periodic reference pattern created by a hexagonal pattern on the diode substrate. The quasiperiodic reciprocal lattice constant is 1.478 μm^{-1} and the periodic reference pattern lattice constant is 0.26 μm^{-1}. The long-range order shown is characteristic of quasiperiodic order rather than amorphous disorder.

In order to model the quasiperiodic structure of the radiative field as a function of distance from the surface, we use Huygens wave theory [2]. As the detection waveguide's aperture is moved away from the surface, interference should transform the quasiperiodic structure of the light at the surface. This leads to a far field which is not quasiperiodic. Here S is a set of random 12-fold quasicrystalline lattice points in two dimensions [23]. The amplitude of the waves emitted from the lattice points is

$$A = \sum_{j \in S} \Re \left\{ e^{ik z_j} \right\}, \quad (1)$$

where z_j is the distance from the jth lattice point to the measurement aperture. Wave theory [Fig. 5(d)], shows that the in-plane reciprocal space representation of A exhibits a 2/3 subharmonic at 0.99 μm^{-1}. The subharmonic dominates over the fundamental at a height of 750 nm above the plane.

Intensity measurements as a function of distance from the surface demonstrate that the quasiperiodic field can be explained by wave superposition, but not by ray optics. In Fig. 5(c), we find that at $z = 250$ nm from the surface, the reciprocal space image is very similar to the image at the surface. This confirms the quasiperiodic intensity image is not caused by a mechanical interaction between the waveguide (NSOM tip) and the surface. In Fig. 5(b), at $z = 750$nm, interference has reduced the quasiperiodic reciprocal lattice constant by 2 /3, in agreement with the subharmonic computed using wave theory. Figure 5(a) shows that at $z = 1750$ nm, interference has made the quasiperiodicity of the light undetectable. The hexagonal reference pattern on the substrate, which is not nanostructured, remains detectable.

Conclusions. In summary, we demonstrate a taxonomically distinct ordering of light. Orderings in the quasiperiodic class are projections of objects in hyperspace [3]. Using reciprocal space representations of light captured by a scanning aperture, we show that a complementary quasicrystalline light emitting diode produced quasiperiodic electroluminescence. When the holes of the complementary quasicrystal are occupied by quantum dots, energy is transferred from the diode to the nanocrystals. The quantum dot photoluminescence produces a different real space structure from the electroluminescence, but the reciprocal space representations show the two fields have the same near-field correlations. The correlations in the evanescent light field agree with wave theory in three dimensions. Since the Huygens theory is not specific to light, the same approach can be used to generate quasiperiodicity in any physical field that supports superposition of classical waves. The resulting emergent phenomena are nonperiodic waves with long-range order. This work expands the study of highly ordered light from the standard 32 crystallographic point groups [20] to the much wider range of higher-dimensional point groups [3].

Experimental methods. Light emitting diodes were fabricated with molecular beam epitaxy. A quasiperiodic structure was generated with random Stampfli inflation [24,25]. The complementary quasicrystal shape was etched into the diodes using electron beam lithography. Trilite fluorescent CdS_xSe_{1-x}/ZnS nanocrystals purchased from Cytodiagnostics were dynamically spin coated at 1500 rpm without dilution [16]. Electroluminescence images were recorded using a customized Nanonics Multiview 4000 NSOM [18]. Scanning electron images were recorded using an FEI Nova NanoSEM FEG-SEM equipped with a Delmic SPARC cathodoluminescence hyperspectral imaging system. Further detail is in the Supplementary material.

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Disclosures. The authors declare no conflicts of interest.

Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

Supplemental document. See Supplement 1 for supporting content.

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