Local self-uniformity in photonic networks

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The interaction of a material with light is intimately related to its wavelength-scale structure. Simple connections between structure and optical response empower us with essential intuition to engineer complex optical functionalities. Here we develop local self-uniformity (LSU) as a measure of a random network’s internal structural similarity, ranking networks on a continuous scale from crystalline, through glassy intermediate states, to chaotic configurations. We demonstrate that complete photonic bandgap structures possess substantial LSU and validate LSU’s importance in gap formation through design of amorphous gyroid structures. Amorphous gyroid samples are fabricated via three-dimensional ceramic printing and the bandgaps experimentally verified. We explore also the wing-scale structuring in the butterfly *Pseudolycaena morsyas* and show that it possesses substantial amorphous gyroid character, demonstrating the subtle order achieved by evolutionary optimization and the possibility of an amorphous gyroid’s self-assembly.
A complete photonic bandgap (PBG) is frequency window within which a material, by virtue of its structure, supports no propagating electromagnetic modes. Typically, structures which possess complete PBGs are periodic arrays of dielectric material; such arrays are called photonic crystals (PhCs). PhCs have the potential to play a key role in the development of next-generation photonic integrated circuits\textsuperscript{1–3}. However, although the complexity of PhC-based technologies continues to grow, questions regarding the fundamental mechanisms of PBG formation remain unresolved\textsuperscript{4–6}.

The formation of PBGs is conventionally interpreted as a result of coherent scattering by a PhC’s lattice planes\textsuperscript{7,8}. In this picture, a plane wave may be scattered onto its counter-propagating equivalent when the wavevector of the initial state lies on the edge of the PhC’s Brillouin zone (BZ). When this condition is met, a pair of orthogonal standing wave modes, each possessing a distinct electromagnetic field profile, is formed\textsuperscript{9,10}. Energetic interaction between the electric field and the underlying dielectric distribution then breaks the degeneracy of the standing wave states. For the specific propagation direction under consideration, the resulting forbidden spectral range defines a photonic stop gap.

To engineer a complete PBG, photonic stop gaps must open along all propagation directions. Further, these stop gaps must be spectrally aligned. Both these considerations can be addressed by designing PhCs to possess maximally spherical BZs\textsuperscript{11}. The search for the first complete PBG thus focussed on face-centred cubic crystals—the most isotropic of the three-dimensional (3D) Bravais lattices—and discovered a large complete gap in a diamond-like network of dielectric material\textsuperscript{12}. In spite of the many PhC designs that have since been discovered, those based on the diamond network remain the champion, possessing the largest complete PBGs\textsuperscript{13,14}.

There is, however, much evidence to suggest that PBG formation is governed by more than just coherent scattering processes. PhCs derived from the body-centred cubic single-network gyroid (SNG) structure (triamond) and low-symmetry diamond embeddings all possess near-champion PBGs in spite of their less spherical BZs\textsuperscript{15}. Although based on a face-centred cubic lattice, the inverse opal network exhibits a complete PBG only one quarter the size of the champion diamond gap\textsuperscript{1}. Most tellingly, a glassy 3D network—dubbed photonic amorphous diamond (PAD)—exhibits a sizeable complete PBG\textsuperscript{4}. This gap exists despite PAD’s diffuse primary diffraction maximum which spreads the structure’s coherent scattering power of a range of wavevectors\textsuperscript{5}.

Similar evidence is found in two-dimensional structures that possess PBGs for light with a transverse electric (TE) polarization. Amongst these structures, the champion PhC design is a honeycomb network of dielectric material\textsuperscript{5,15}. Many glassy networks that can be broadly styled as ‘hyperuniform disordered honeycombs’ have been found to possess sizeable TE PBGs\textsuperscript{6,16–20}. As with PAD, these gaps exist despite the diminished coherent scattering power of each structure.

Here, we address the mechanisms governing PBG formation by re-formulating the ideal structural properties of a PBG-forming network. To achieve this, we introduce the concept of local self-uniformity (LSU). LSU measures the geometrical and topological similarities of the local environments in a connected network of uniform valency. We note that existing sizeable PBG networks possess significant LSU. We demonstrate the connection between LSU and PBG forming ability by designing novel amorphous gyroid (amorphous triamond) connected networks. Specifically, amorphous gyroids can possess sizeable PBGs and an amorphous gyroid’s LSU is strongly correlated with its PBG width. This correlation is explained by recognizing the contribution of spatially localized resonant scattering processes to PBG formation in connected networks. Locally self-uniform ceramic 3D-printed amorphous gyroids are characterized through microwave transmission experiments. We explore also the possibility that amorphous gyroid exists within the wing scales of butterflies. In particular, we reveal that the microstructure in the scales of *Pseudolycaena marsyas* possesses substantial amorphous gyroid character and demonstrate that the butterfly’s reflectance spectrum can be effectively reproduced by amorphous gyroid microstructures.

**Results**

**Local self-uniformity.** The exact structure of glasses has long been debated\textsuperscript{21}. Recent research has demonstrated the complex interplay of ordered and disordered phases in the vitreous state\textsuperscript{22–24}. The disparate variety of bulk metallic glasses in particular has challenged researchers to develop predictive theories of an alloy’s glass-forming ability\textsuperscript{25}. The existence of sizeable PBGs in glassy networks exposes an analogous deficiency in current understanding of a structure’s PBG forming ability. Unlike silicate and metallic glasses, the structures of glassy PBG materials are designed. Nevertheless, the structural characteristics that render these glasses amenable to PBG formation remain mysterious. With the aim of clarifying these PBG forming characteristics, here we develop LSU a general measure of structural order in connected networks.

A typical PhC consists of a connected distribution of dielectric material surrounded by air. As an example, Fig. 1a,b show the champion PhC diamond; it is a connected network of dielectric cylinders (green) arranged as in a diamond crystal. A PBG’s size is usually measured as a dimensionless width given by \( \Delta \omega / \omega_0 \) —the frequency width divided by the central frequency. Figure 1c displays an amorphous version of the diamond network. The complete PBG in diamond has been shown to have a width of 30% for dielectric material of refractive index 3.6 (ref. 13). We focus our discussion of PBG properties hereafter in this high-refractive index regime.

To describe a general connected network, we decompose it into a set of vertices and edges. A vertex is a point at which two or more distinct lobes of material intersect. An edge is a vector between two vertices that specifies the central axis of a lobe of material. As an illustration, Fig. 1a shows the fundamental unit of a diamond network in its Wigner–Seitz (WS) cell. A vertex, which sits at the centre of the cell, has four edges connecting it to its nearest neighbour vertices. The four nearest neighbours sit at the corners of a tetrahedron and the four edges define a tetrahedral unit. Similarly, Fig. 1d shows the fundamental unit of a SNG structure in its WS cell. The central vertex has three nearest neighbour vertices to which it is connected by three edges. These edges are of equal length and are separated by inter-edge angles of 120°; we call this configuration a trihedral unit.

The WS cell is the basic building block from which an extended periodic network can be assembled. Stacking the WS cells of diamond and SNG produces extended diamond (Fig. 1b) and gyroid (Fig. 1e) networks, respectively. An extended periodic network of this type is a highly ordered case of a continuous random network (CRN). A general CRN is a collection of vertices, connected by edges, which are arbitrarily positioned in space\textsuperscript{26}. CRNs do not typically possess translational periodicities as the diamond and gyroid crystals do. Instead, Fig. 1c illustrates a classic CRN—an amorphous diamond network, studied extensively as a model of amorphous silicon\textsuperscript{27,28}. Every vertex in amorphous diamond is tetravalent and has four nearest neighbours that describe the points of a deformed tetrahedron. The resulting network is a connected assembly of deformed...
tetrahedral units. Similarly, an amorphous gyroid is a CRN comprising deformed trihedral units as depicted in Fig. 1f. Amorphous gyroid (f) comprises deformed trihedral units connected in a CRN.

Figure 1 | Strongly isotropic networks and their amorphous derivatives in three dimensions. The WS cell of a diamond PhC (a) contains a 1-tree of dielectric material (green); the edges of the tree define a tetrahedral unit. Stacking of the WS cells generates the champion diamond PhC (b). Amorphous diamond (c) consists of deformed tetrahedral units connected in a CRN. Similarly, the WS cell of a single-network gyroid PhC contains a 2-tree (d), comprising dielectric material (blue), in which each vertex is a trihedron. When stacked, the WS cells generate a single-network gyroid PhC with a near-champion PBG (e). Amorphous gyroid (f) comprises deformed trihedral units connected in a CRN.

To build a robust measure of network structural order, we require a formalism that can compare the trees of arbitrarily disordered networks. We thus build upon the measurement of tree spatial similarity statistics in the following ways. We stipulate that the CRN from which the trees are drawn comprises only vertices with a fixed number of edges. We then generalize the measurement of tree spatial similarities to \( n \)-trees of any size. In three dimensions, these networks are amorphous. Amorphous gyroid (f) comprises deformed trihedral units connected in a CRN.
which determines the natural edge overlaps; this is detailed in the Methods section. We also allow the similarities of arbitrarily disordered trees to be measured. Here we acknowledge that the root edges of two trees will not necessarily overlap perfectly. Instead, for each permutation we find a transformation that yields an approximate overlap. We then quantify the quality of this overlap according to a metric of spatial similarity (see Methods section).

We now define a CRN’s LSU distribution $\Phi_{nl}$ to depth $n$ and locality $l$ as the set of $n$-tree spatial similarities $\Phi_{nl}$, for all pairs of trees whose root vertices are within $l$ edges of one another. The LSU distribution is a set of spatial similarities that describes the extent to which the CRN’s geometry is uniform on the length-scale $l$. For example, $\Phi_{22}$ describes the spatial similarities for all pairs of trees of depth 2 separated by two edges or less.

We present a set of example LSU distributions in Fig. 3. Figure 3a shows $\Phi_{22}$ for an amorphous diamond network. The trees in amorphous diamond are non-identical and present a broad distribution of spatial similarity statistics. However, the spatial similarities remain relatively large and show that the local geometries in the CRN are similar. Fig. 3b compares three LSU distributions, $\Phi_{12}$, $\Phi_{22}$ and $\Phi_{32}$, for amorphous diamond. Amorphous networks have strong positional correlations at local length-scales that fade with distance; as a result, trees become

![Figure 2](image_url)

**Figure 2 | Comparison of 1-trees illustrated with trihedra, tetrahedra and octahedra.** Two identical trihedral trees (a) are labelled by their edges. The edges of tree b are then permuted. Tree b can then be rotated around the edge $3_b$ axis and made to perfectly overlap tree a. Similarly, two identical tetrahedral trees are labelled (b), and then the edges of tree b permuted. Reflection of tree B in the plane of edges $2_b$ and $4_b$, followed by rotation around the new edge $3_b$ axis, brings the two trees into alignment. Two identical octahedral trees (c) can also be compared. We apply a permutation to tree B’s edges. When overlapped, the two trees are now mismatched; their yellow and purple edges are not aligned and cannot be made so by any congruent transformation without creating a new mismatch.
Overall, evidence suggests that the extent of a network’s LSU influences its PBG forming ability. Hence, we expect that a hypothetical amorphous SNG, analogous to PAD4 and disordered honeycomb6, should possess both a high degree of LSU and a sizeable complete PBG. To the best of our knowledge, amorphous gyroids have not been observed in any context. We thus undertake the design of amorphous gyroids as a means of testing the relationship between LSU and PBG forming ability. To achieve this, we apply the Wooten-Winer-Weaire (WWW) algorithm to anneal amorphous gyroids from random seed networks27,28. In order to yield faithfully gyroidal local geometries, networks are annealed using a modified potential energy function that is distinct to the regular Keating energy27 (see also Supplementary Methods).

We generated a set of amorphous gyroid models of varying total size as measured by their number of component vertices N. In particular, we generated an ensemble of 57, 216-vertex models across a spectrum of disorder. PBGs were probed by numerical solution of the Maxwell equations via a plane wave expansion method29. A refractive index contrast of 3.6:1 was used for all calculations. Bandgap widths were measured as a percentage of their central frequency.

We demonstrate that amorphous gyroid networks can possess sizeable PBGs. The average PBG measured for a set of high-quality 1,000-vertex networks was 16%. The largest single gap observed was for a well-annealed 216-vertex network and had a size of 21%. These gap widths compare favourably with the 18% gap in PAD at the same refractive index contrast4.

We investigated our ensemble of 216-vertex networks closely, calculating the LSU \( \Phi_{22} \), distribution for each network. In Fig. 4a we plot the mean value \( \Phi_{22} \) of each distribution against PBG width for all 57 networks. We see that gap width is strongly correlated with network LSU. We expect PBGs in amorphous gyroid to open within the gap region of a SNG PhC of equivalent index contrast. We thus defined the SNG gap as a critical frequency region, and counted the number of photonic bands that each of our 216-vertex networks support within this window. The number of bands within the critical frequency region can be interpreted as an integrated density of states (DOS). We plot the integrated DOS against \( \Phi_{22} \) in Fig. 4b. We see that a network’s inability to support electromagnetic modes is strongly correlated with its LSU (see also Supplementary Figs 2 and 3 and Supplementary Notes 1 and 2). Together these results demonstrate the power of LSU as a predictor of PBG forming ability.

We now discuss two characteristic types of amorphous gyroid in detail; we call these type-1 and type-2 networks. We characterize the networks through histograms of their geometrical properties. In particular, we measure edge lengths (\( d \)), inter-edge angles (\( \theta \)), dihedral angles (\( \phi \)) and skew angles (\( \chi \)). Note that the skew angle measures the coplanarity of a trihedral unit, with \( \chi = \pi/2 \) representing a flat trihedron (see Supplementary Methods).

Figure 4c,d show typical frequency distributions of \( d \), \( \theta \), \( \phi \) and \( \chi \) for type-1 networks. They are characterized by strongly peaked edge-length and inter-edge angle distributions, but have non-uniform dihedral and skew angles. Type-1 networks can thus be considered high-quality amorphous networks of trihedral 1-tree units. Figure 4f,g show the same distributions for a typical type-2 network. Its \( d \), \( \theta \), \( \phi \) and \( \chi \) distributions are all peaked around the ideal values for gyroidal vertices. Compared with type-1 networks, the local geometries of type-2 structures are much closer to an ideal strongly isotropic configuration. They possess gyroidal structural order on the length-scale of a 2-tree unit—the fundamental building block of SNG (Fig. 1d). Type-1 networks have \( \Phi_{22} \) values around 0.72. Type-2 networks have significantly higher LSUs with \( \Phi_{22} \)

**Figure 3 | Example LSU distributions.** (a) The distribution of spatial similarity statistics for tree comparisons of depth 2 and locality 2 (\( \Phi_{22} \)) for an amorphous diamond network. (b) LSU distributions \( \Phi_{nl} \) of amorphous diamond for different tree depths \( n \). (c) \( \Phi_{22} \) for a 216-vertex sample comprising a single gyroid crystallite suspended in amorphous gyroid, demonstrating LSU’s ability to differentiate between phases. (d) \( \Phi_{22} \) distributions for two characteristic types of amorphous gyroid network—type-1 (1-tree local order) and type-2 (2-tree local order).
Figure 4 | LSU and PBG forming ability. The LSU of amorphous gyroid networks, as measured by the mean of their $\Phi_{22}$ distributions, is strongly correlated with PBG width (a). Similarly, the integrated DOS $N(\omega)$ decreases smoothly with increasing $\Phi_{22}$ (b). Fit lines (cubic polynomials) are for visualization purposes only. Approximate LSU regions for type-1 and type-2 networks are indicated (a, orange). We present also the edge length frequency ($f$) distributions (c,f), inter-edge ($\ell$), dihedral ($\phi$) and skew ($\chi$) angle frequency distributions (d,g) and structure factor slices (e,h) for typical type-1 and type-2 networks, respectively. Structure factor intensities $I$ are plotted on logarithmic colour scales and data in both panels is normalized to the maximum intensity $I_0$ of $h$.

around 0.89. Typical $\Phi_{22}$ distributions for these networks are shown in Fig. 3d. Figure 4.e,h show planar slices through the structure factors of type-1 and type-2 networks, respectively. The structure factor is a quantity that characterizes a structure’s coherent scattering power as a function of wavevector. We see that the peak scattering power of both network types is distributed in a circular ring on account of the average isotropy of the structures; wavevectors that lie on this ring are strongly scattered. We averaged the full structure factor across all propagation directions and measured the peak scattering power of type-2 networks to be approximately 1.6 times greater than type-1. In spite of this small increase, type-2 networks take the PBG width from practically zero to a maximum of 21%. We attribute these radically different PBG widths to the formation of locally self-uniform trees within type-2 networks.

We now demonstrate the natural connection between LSU and PBG formation in connected networks. In addition to the coherent scattering (Bragg) mechanism, there exists a Mie scattering mechanism of PBG formation. The Mie mechanism is known to be the dominant formation process for transverse magnetic (TM) polarization gaps in dielectric cylinder arrays. Specifically, sizeable PBGs exist in periodic, quasicrystalline and random cylinder arrangements and are observed to be of the same origin in each case.

The gap originates from resonant scattering by the Mie modes of a single cylinder. For all TM-gap cylinder arrays, it is clear from the electric field profiles at the edges of the fundamental PBG that Mie scattering mediates light propagation. Just below the gap, modes are characterized by localization of field nodes in the vicinity of the cylinder surfaces. Just above the gap, field nodes consistently bisect the dielectric cylinders. These two node profiles derive from the interaction of a plane wave with an isolated cylinder, and are associated with the first and second Mie resonances, respectively. Just above the first Mie resonance, incident and scattered fields are in antiphase at the cylinder surface; this creates a localized standing wavefront which inhibits propagation and leads to PBG formation.

Existing evidence suggests that PBG formation in two-dimensional and 3D dielectric networks is governed by a similar mechanism of resonant scattering. Specifically, careful examination of the gap-edge eigenmodes of all honeycomb-derived networks presents a consistent picture of the nature of these scattering resonances. In both crystalline and hyperuniform disordered honeycombs, modes just below the fundamental PBG are characterized magnetic field nodes localized within the dielectric network (conversely, field anti-nodes focus within the air cells). Just above the PBG, magnetic field nodes pass between air cells, cutting the inter-vertex dielectric walls almost normally. These gap-edge node characteristics are consistent across honeycomb-derived trivalent networks and, by analogy to the TM case, evidence the significance of spatially localized resonant scattering processes.
We therefore view CRNs as a connected ensemble of distinct scattering units (trees) which, in isolation, exhibit a number of resonant electromagnetic modes. For frequencies just above a resonance, scattered and incident fields are in antiphase and interfere destructively, localizing a field node in the vicinity of the scatterer and suppressing propagation through it.

A potential champion PBG structure comprises geometrically identical scattering units. All scattering centres thus possess degenerate electromagnetic resonances, and the spectral ranges for which each scatterer inhibits transmission are maximally aligned. Fixed valence networks comprising non-identical scattering units exhibit smaller PBGs than their crystalline precursors. Structural deformation of the scattering centres breaks the degeneracy of their scattering resonances; the resulting precursors. Structural deformation of the scattering centres aligns. Fixed valence networks comprising non-identical degenerate electromagnetic resonances, and the spectral ranges identical scattering units. All scattering centres thus possess scatterer and suppressing propagation through it.

The set of networks comprising geometrically identical scattering units exhibits a clear hierarchy of PBG size (see Supplementary Table 1). Specifically, networks built from vertices with a low coordination number possess the largest PBGs. Accordingly, the diamond and SNG architectures are champion and near-champion, respectively; these networks are strongly isotropic due to the simplicity of their vertices and comprise scattering units which are perfectly superimposable under permutation. This combinatorial symmetry has a strong influence on the PBG width. We argue that symmetry under permutation minimizes the number of distinct scattering resonances that a scattering centre supports. As a result, the frequency gaps between scattering resonances are maximized, together with the width of the spectral region above resonance for which transmission is suppressed.

Fundamentally, both Bragg and resonant scattering mechanisms contribute to PBG formation. The largest PBGs are obtained by optimization of a structure’s dielectric fill fraction to overlap the spectral range associated with the two mechanisms. We note, however, that strong diffraction rings in the structure factor of amorphous materials do not directly lead to PBGs, but reflect the presence of local order that, depending on its LSU, may favour gap formation. This observation clarifies the relationship between LSU and work on PBG formation in hyperuniform structures. Architectures derived from disordered hyperuniform point patterns possess significant local structural correlations and local geometrical order; these characteristics have proven essential in establishing sizeable PBGs. However, hyperuniform point patterns must be tessellated in an ad-hoc way to produce viable PBG-forming networks. This tessellation protocol naturally creates nearly-optimal network topologies, but these networks are successful only because they possess locally self-uniform structural order. In contrast to hyperuniformity, LSU measures both geometrical and topological order simultaneously and is thus an effective measure of PBG forming ability. Hyperuniformity and LSU remain compatible; we note the emergence of a hyperuniform-like exclusion domain around \( k = 0 \) in the structure factor of networks with significant LSU (Fig. 4h). The association of LSU with PBG formation parallels the proof that amorphous materials with well-defined atomic connectivity can possess an electronic bandgap.

Microwave experiments with amorphous gyroid. To verify our theoretical calculations, we fabricated millimetre-scale amorphous gyroid samples and experimentally characterized their PBGs. Samples were produced at the Fraunhofer Institute for Ceramic Technologies and Systems using a 3D ceramic printing technique. The samples were made from alumina (\( \text{Al}_2\text{O}_3 \)), whose permittivity was experimentally determined to be \( \varepsilon_r = (9.5 \pm 0.3) \) at frequencies in the microwave band (18–26.5 GHz). Two types of sample were made: cuboidal samples of SNG (Fig. 5c) and amorphous gyroid (Fig. 5a,d), and a cylindrical sample of amorphous gyroid (Fig. 5b). The internal network, comprising cylinders with diameter \( D = 2.03 \text{ mm} \), was well formed (Fig. 5c,d). The gyroid primitive cell parameter was

![Figure 5](image-url)
Measurable properties of the gyroid network were determined by performing finite-difference time-domain (FDTD) electromagnetic simulations. Specifically, we placed a number of dipole sources inside our amorphous gyroid cylinder and record the power flux some distance from the cylinder using a circular array of detectors (see Supplementary Methods). This result is presented as a second polar false-colour map in which the radial coordinate represents frequency and the angular coordinate records the cylinder’s rotation angle. Here, the rotational isotropy of the PBG is clear; the blue- and green- colored transmission gap for the SNG sample agrees very well with the simulated PBG, highlighted by vertical dashed lines in the figure.

We now turn our attention to the Cambridge Blue butterfly (Callophrys gryneus) and model its reflectance spectrum with an amorphous gyroid structure. Many butterfly species are known to derive colouration from gyroidal networks of chitin within their scales, most famously Parides sesostris (the emerald-patched cattleheart) and numerous species of the genus Callophrys (the green hairstreaks). Microscopy has demonstrated that the scales contain numerous crystallites of a well-ordered network of chitin in air. Small angle X-ray scattering (SAXS) has assigned the symmetry group of SNG to the structures.

In this regard, self-assembly is a particularly attractive fabrication method. The three strongly isotropic networks have all been observed to self-assemble. Interestingly, amorphous honeycomb and diamond have also been observed in the natural world, it is thus clear that a self-assembly pathway capable of producing complex short-range order exists. Here, we explore evidence that an amorphous gyroid could be similarly self-assembled. First, we demonstrate that topological defects exist in the gyroidal microstructures of green hairstreak butterflies. We then present a disordered network structure in the scales of the Cambridge Blue butterfly and model its reflectance spectrum with an amorphous gyroid structure.

To model the SAXS results, we approximate the scale as an ensemble of separate crystallites which contribute to the diffraction spectrum independently. The positional correlations between adjacent crystal domains are destroyed by the matching defects at the grain boundaries; the SAXS spectrum can thus be considered an incoherent superposition of the diffraction patterns of distinct crystallites. To fit the experimental results, we generate a number of distinct crystallites across a range of chitin fill fractions and sum their diffraction patterns incoherently, optimizing their weights in the summation to minimize the Reitveld weighted profile $R$ factor, $R_{wp}$ of the fit. We calculate two types of fits, described in detail in the Methods section. In the first case, we model the scales as perfect gyroids, employing the level-set approximation of the gyroidal minimal surface (dotted orange curve in Fig. 6a). In the second case, we model the tomographic data using partially disordered gyroid (PDG) networks (dashed blue line in Fig. 6a). PDG is generated by introducing a small number of topological defects into a perfectly ordered gyroid net. Specifically, one defect is introduced for every 100 network vertices together with a small amount of vertex positional disorder.

Note that, at publication, no SAXS data for C. rubi was available. Instead we compare our models to SAXS results for Callophrys gryneus (solid green line in Fig. 6a); we expect the microstructure in C. gryneus to comprise gyroid grains connected by topological matching defects, just as in C. rubi. $R_{wp}$ values for our level-set and PDG fits are 980 and 645, respectively, suggesting that PDG is a superior model of the green hairstreak scale structure. The pure level-set model produces overly prominent peaks, particularly between the (110) and (211), and (400) and (420) reflections. The inclusion of topological and positional disorder dampens the network correlations and reduces this contrast across the whole spectrum. The quality of the disordered PDG fit is most evident for high $(hkl)$ values; here, the overall profile of the pattern, in particular the double peak of the (321) and (400) planes, is well-captured.

Taken together, the tomographic data and the results of our scale modelling (Fig. 6a) suggest that topological imperfections form, to a limited degree, at the grain boundaries in the gyroidal microstructures of the green hairstreaks. The existence of these topological defects renders the hairstreaks (sub-family Theclinae) a promising family of butterflies within which to search for an amorphous gyroid.
Figure 6 | LSU in butterfly scales. Comparison with SAXS data of level-set and partially disordered gyroid models of wing scales in the butterfly C. gryneus (a). Specimen of Ps. marsyas (b) and FIB section of one of its blue scales (c), imaged with 52° tilt in the FIB. Detail (d), taken from (c), compared with a projection of an amorphous gyroid model (e). Transmission electron micrograph of a Ps. marsyas scale cross-section (f). Comparison of experimental reflectance data from a large wing area to an amorphous gyroid scale model (g). Scale bars, 1 cm (b); 1 μm (c); 500 nm (d,e); and 1.5 μm (f). The TEM section (f) is reproduced with permission from Ver¬tesy et al.52. TEM, transmission electron microscopy.

We now model scales of Ps. marsyas as amorphous gyroid CRNs and investigate the compatibility of these models with experimental reflectance measurements. We employ a number of amorphous gyroid CRNs with Φ2 as around 0.88. We calculate their far field reflectance spectra by FDTD solution of the Maxwell equations.

We estimated the scale thickness from FIB sections to be 1300 ± 200 nm. The lower half of the scale was taken to be absorbing in accordance with the pigment distribution observed by transmission electron microscopy (Fig. 6f). We suggest a plausible model for the complex refractive index of the Ps. marsyas pigment, derived from the extinction coefficient of the pigment in Papilio nireus (see the Methods section). Amorphous gyroid edge lengths for our Ps. marsyas sample were estimated from electron micrographs to be 117 ± 6 nm; this corresponds to an effective SNG primitive cell parameter of a = 166 ± 8 nm. The reflectance of a large wing area of Ps. marsyas was measured by a previous study52; scaling of the amorphous gyroid (110)-type SAXS peak to the wavelength of maximum reflectance suggests a = 169 nm. The theoretical reflectance presented (Fig. 6g) is an average over six amorphous gyroid models, all scaled by an a value of 166 nm.

Given the assumptions made in modelling the pigmentation, the general agreement between theoretical and experimental reflectance spectra suggests that an amorphous gyroid with a UV-absorbing pigment is a plausible model of the structure in Ps. marsyas scales. The divergence at UV wavelengths is attributable to uncertainty in the exact complex refractive index of the butterfly scale across a 300 to 700 nm range; future studies should measure this directly. The small divergence at red wavelengths is attributable to reflections from unmodelled melanized ground scales.

Two other neotropical hairstreaks, Arcas imperialis (Theclinae) and Evenus coronata (Theclinae)—close relatives of the Cambridge Blue—have previously been surveyed through microscopy56. Their structures appear to possess strong multilayer and gyroidal characteristics, respectively. It is therefore possible that amorphous gyroid has evolved in the Cambridge Blue by some small change in scale cell development conditions, leading to an evolutionary divergence. Indeed, such a divergence has been postulated in the case of the gyroid-containing Pa sesostris, whose-scale structuring has diverged from the perforated multilayers of many closely related Parides species57.

Our work indicates that the scale structuring in the Cambridge Blue is related to an amorphous gyroid. Electromagnetic modelling shows that amorphous gyroid models are consistent with observed reflectance data and the existence of topological matching defects between gyroid grains in green hairstreak scales suggests that the production of amorphous gyroid is developmentally possible.

Any further search for a natural amorphous gyroid should not be limited to butterflies. A thorough survey of avian feather barbs has revealed an abundance of colour-producing channel-type architectures. Several species—Diglossa cyanae58 (Thraupidae), Passerina cyanea58 (Cardinalidae) and Alcedo
atthis\textsuperscript{59} (Alcedinidae)—are themselves a brilliant blue and possess microstructures with striking similarity to amorphous gyroid. Experimentally, it may be possible to produce amorphous gyroid or a similar architecture via block co-polymer self-assembly\textsuperscript{59}. It is possible to generate mixed lamellar and gyroidal states that may resemble the Ps. marsyas structure\textsuperscript{61,62}. An alternative pathway may be to quench the phase transition between the gyroid and metastable perforated multilayer phases\textsuperscript{62}.

Discussion
We have introduced LSU as means of measuring the extent to which local environments in a CRN of fixed valency are spatially similar. When applied to strictly trivalent and tetravalent networks the states of maximum LSU are the strongly isotropic single gyroid and diamond networks, respectively. As a network’s LSU decreases from its maximum it becomes glassy and eventually chaotic. LSU can thus classify all CRNs of fixed valency by the extent of their internal order.

We designed LSU as a means of characterizing the optical properties of CRNs. In particular, we have shown that networks with maximal LSU possess champion PBGs in both 2D and 3D. Further, other known networks endowed with a complete PGB in 3D or a TE polarization PGB in 2D are characterized by high levels of LSU. PBGs result from scattering by the electromagnetic resonances of a network’s local scattering units. When these units are spatially similar, their resonances are maximally degenerate and complete PGB formation is favoured. LSU is thus a predictive measure of a network’s PGB forming ability. While here we focussed on trivalent and tetravalent networks, LSU can be generalized to include networks of arbitrary or mixed valency.

We have introduced designs of novel amorphous gyroid CRNs. Here we used ceramic 3D printing to fabricate amorphous gyroid samples in high-refractive index alumina and demonstrated their sizeable isotropic PBGs via microwave transmission experiments. The relevance of amorphous gyroid, and architectures which can be derived from it, is broad. In particular, its development, or that of a closely related CRN, appears to have occurred in the scales of the butterfly Ps. marsyas. That it is possible to self-assemble such a structure is a prerogative for its existence in natural systems. It may be possible to produce amorphous gyroid networks via block co-polymer experiments or equivalent self-assembly methods. This poses an interesting experimental challenge, the solution of which will facilitate the fabrication of advanced optical metamaterials for industrial applications\textsuperscript{43,60}.

Fundamentally, we have demonstrated that the tree comparison method is a powerful framework for controlling the LSU of the scattering centres in a CRN. Sculpting a network’s LSU distributions translates directly to advanced control over its optical properties. The optical properties controlled need not be limited to PGB forming ability; they could include structural colour, the scattering mean free path\textsuperscript{63} and random lasing\textsuperscript{64}. Moreover, similar design principles may be employed to control other wave phenomena in electronic, phononic, elastic and acoustic materials.

Methods

Definition of the LSU distributions. Consider a CRN $C$ with a set of defined vertex positions and inter-vertex connectivities, in which each vertex in $C$ has exactly $γ$ edges. We define an $n$-tree $T_n$ on vertex $a$ of $C$ as the set of vertices within $n$ edges of $a$. The root edges of $T_n$ are all the edges of $T_1$. Computationally, all information regarding $T_n$ can be obtained by performing a breadth-first graph search to depth $n$ from vertex $a$.

We consider now two $n$-trees of equal depth $T_{n}^a$ and $T_{n}^b$. We label the root edges of the trees $\{1_a, 2_a, \ldots, γ_a\}$ and $\{1_b, 2_b, \ldots, γ_b\}$, respectively. The spatial similarity statistic $φ_{ab}$ of these two trees is defined as

$$φ_{ab} = \frac{1}{T_n^a} \sum_{i=1}^{T_n^a} f(T_{n}^a, T_{n}^b; σ_i).$$

where $f$ is a similarity measure which grades the overlap of $T_{n}^a$ and $T_{n}^b$ when they are maximally aligned in a root edge permutation $σ$. We sum the similarities for all $γ_a$ overlap permutations of the trees’ root edges and then take the average.

The value of the spatial similarity statistic will depend both on the form that the measure $f$ takes and the method that is used to maximally align the two trees for each permutation. We describe our choice of $f$ and the alignment procedure we follow in the next section.

The LSU distributions of the CRN are particular sets of spatial similarity statistics. We define a new tree $T_{n}^l$ with depth $l$ on vertex $a$. The set of vertices in $T_{n}^l$ is the local neighbourhood of $a$ to depth $l$. The LSU distribution $Φ_{ab}$ can then be written as

$$Φ_{ab} = \{φ_{ab}\} : b \in T_{n}^a, a \in C.$$

Calculation of the overlap between two trees. Here we discuss in detail the exact form of our similarity measure $f$ and the process by which two trees are maximally overlapped. We note that the choice of $f$ is an important user-controllable degree of freedom. In general, $f$ must be maximal when two trees can be overlapped perfectly; this defines uniquely the maximal self-uniform configuration. However, the exact construction of which overlap is calculated can be optimized to suit the application and to possess meaningful properties as the network departs from maximal self-uniformity. Here, we adopt an intuitive framework in which tree edges are overlapped in pairs with their most natural partners.

Consider now an $n$-tree $T_{n}^a$ in the CRN $C$. We label its $|T_{n}^a|$ vertices with index $j$, subject to the constraint that indices $\{1, 2 \ldots γ\}$ represent the tree’s root edges. The branches of the tree are defined by its edge vectors. For $T_{n}^a$ we denote these as $r_a^j$, which defines the vector to vertex $j$ from its parent vertex as defined by a breadth-first graph search starting on $a$.

Consider now two $n$-trees $T_{n}^a$ and $T_{n}^b$. Before they can be compared, they must be maximally overlapped. This process consists of determining a congruent transformation which maximizes the overlap of their root edges in some chosen permutation $σ$. First, $T_{n}^a$ is translated such that vertices $a$ and $b$ overlap. Second, $T_{n}^a$ is rotated until $r_a^{(1)}$ and $r_b^{(1)}$ are parallel; this is always possible. Third, $T_{n}^b$ is rotated about the $r_b^1$ axis until $r_b^{(1)}$ and $r_a^{(1)}$ are maximally parallel, defined to be the configuration which minimizes their scalar product. For disordered trees, it is not usually possible to make these two vectors perfectly parallel. For trivalent networks, we deem these three steps sufficient to maximally align the two trees.

For tetravalent networks it is necessary to introduce a fourth alignment step. In this case, $T_{n}^a$ is reflected in the plane defined by $r_a^{(1)}$ and $r_a^{(2)}$ so as to bring $r_a^{(2)}$ into maximal alignment with $r_b^{(1)}$ and $r_b^{(2)}$. This step is performed only if the alignment between $T_{n}^a$ and $T_{n}^b$ is improved, as measured by an increase in the value of $F_{nl}$.

From this point, the two trees are considered to be maximally aligned. Once maximally aligned, we define our similarity measure $f$ for grading the overlap of two trees as

$$f(T_{n}^a, T_{n}^b; σ_i) = \frac{1}{|T_{n}^a|} \sum_{j=1}^{T_{n}^a} \frac{r_a^{(j)} \cdot r_b^{(j)}}{\left| \left| r_a^{(j)} \right| \right| \left| \left| r_b^{(j)} \right| \right|}.$$  

Overlap is calculated between edge pairs by taking their scalar product and normalizing it with the square of their mean norm; overlap of a single pairing is thus distributed on $[0, 1]$. The $|T_{n}^a| - 1$ edge pair comparisons are summed and averaged such that $f$ yields a maximum value of unity for perfectly overlapping trees.

Edge pairs are overlapped in the combination that is most natural. The whole overlap calculation is performed recursively in a depth-first sense, greatly simplifying the process of determining natural pairings. At a particular point in the algorithm’s execution, it has reached some vertex pair $j$ and $k(j)$ by comparison of the natural edge pair $r_j^i$ with $r_k(j)$. Overlap of all $(γ - 1)!$ possible sets of edge pair comparisons is calculated. The set that maximizes the sum of edge pair scalar products is chosen as the set of natural pairings; this result is accepted, and the algorithm proceeds to calculate overlap for the edges around each of the $(γ - 1)$ child vertices. This decision process is captured in the notation $r_k(j)$ to reflect that the choice of edge vector in tree $b$ is a function of the edge vector $r_j^i$ to which it is being compared.

Finally, the spatial similarity statistic $Φ_{ab}$ is determined by repeating this process for all root edge permutations according to equation (1).

Microwave transmission measurement. Gyroid and amorphous gyroid models were printed and finished in the workshops of the Fraunhofer Institute for Ceramic Technologies and Systems, Dresden. The gyroid primitive cell parameter was
designed to be to be 3.08 mm. As part of the printing process, the samples were sintered at high temperature, shrinking in side-length by ~20% and introducing some uncertainty into their true sizes. Actual scaling values were thus measured and fill fractions were determined using a water displacement method, yielding an effective gyroid primitive cell parameter of $a = 3.13 \pm 0.05 \text{ mm}$, and alumina volume-fill-fractions between 27 and 29%. Amorphous gyroid samples were fabricated preferentially. We also produced one SNG model. This model was designed for measurement of transmission along the [111] axis as this axis was chosen because of its symmetry (see Supplementary Note 4).

Characterization of the samples was performed with microwave radiation using an HP-8510C VNA. A single polarization mode was coupled through a pair of rectangular horn antennae and a pair of custom-made Teflon microwave lenses. For the measurements shown in Fig. 5c, the beam was aligned along the short edge of the cuboidal samples (Fig. 5a) and perpendicular to a flat sample surface, and the frequency varied from 15 to 35 GHz. Note that frequencies beyond the standard microwave K band (18–26.5 GHz) have very low efficiency coupling through the horns and waveguides. For the measurements shown in Fig. 5g, the cylinder sample was aligned with the incident beam perpendicular to the cylinder axis, around which we rotated the sample and recorded the transmission every 2°. This set of measurements was performed across a frequency range of 17–27 GHz.

The total dynamic range of the HP-8510C VNA is from 0 to ~65 dB (measured dark noise). A large area of microwave-absorbing material, into which a window was cut to hold the sample, was used to prevent reflection and scattering of radiation into the environment. The normalized transmission is then defined as the ratio between detected intensities with and without the sample in place. The addition of microwave-absorbing material lowers the overall coupling-efficiency through the pair of horn-antennae. As a result, the actual accessible dynamic range through this measurement is only ~35 dB, which limits the measured gap-depth. This limited dynamic range is apparent in the amorphous gyroid transmission of Fig. 5e: the transmission bottoms out and becomes noisy for frequencies between 20 and 26 GHz. For the same reason, transmission results for the amorphous gyroid network (Fig. 5g) appear noisy in comparison to the theoretical results (Fig. 5h). An increased dynamic range could be accessed by amplifying the source power.

**SAXS pattern modelling.** The total scattered intensity measured in a SAXS experiment can be written as a Fourier transform of the electron density function $\rho(r)$:

$$I(q) = \frac{4}{\pi} \int_0^1 \int_0^{2\pi} \int_0^{2\pi} \rho(r) \cdot r^2 \cdot \sin^2 \phi \cdot \sin \theta \cdot d\phi \cdot d\theta \cdot d\phi,$$

for $q$ the scattering vector in reciprocal space, $r$ the radius of the electron and $V$ the total sample volume. We Fourier transform the difference between $\rho(r)$ and its volume average $\bar{\rho}$ to remove the forward scattering peak and access any diffraction signatures that may be present at small scattering vectors. The $\rho(r)$ functions for our 3D network structures were generated by voxelization and were distributed on a 3D grid. Representing the air and chitin distributions of the butterfly scale networks, our 3D network structures were generated by voxelization and were distributed on a 3D grid.

To make progress in modelling the reflectance of Ps. marsyas scales, we propose a plausible complex refractive index for the pigmented lower layer. We derive the extinction coefficient $k(\omega)$ of the ultraviolet-absorbing pigment in Papilio nireus using the Beer–Lambert law. The real component of the refractive index was taken to be equal to that of the upper layer of the scale. Lumerical’s in-built refractive index fitting algorithm was used to fit this combination of refractive indices. The resulting complex refractive index used for the lower half of the scales is inset into Fig. 6g. The reflectance spectrum of Fig. 6g should be interpreted in light of the assumptions we have made in modelling the complex refractive index of the pigment. Given these assumptions, the agreement between the theoretical and experimental reflectance data shows that an amorphous gyroid network is a plausible model for the structure in Ps. marsyas scales. The divergence between theoretical and experimental reflectance at ultraviolet wavelengths is a result of uncertainty in the true refractive index of the Ps. marsyas pigment in this wavelength range. Further modelling of Ps. marsyas scales should be informed by a direct measurement of their complex refractive index.

**Data availability.** The data underlying the findings of this study are available without restriction. Details of the data and how to request access are available from the University of Surrey publications repository: http://dx.doi.org/10.15126/ surreydata.0813094.

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**Psuedolycaena marsyas scale imaging.** A dried male specimen of Ps. marsyas was acquired online from The Bugmaniac Insect Shop. The specimen was spread and mounted for sample photography. Single scales were removed, mounted and gold sputtered to produce a 4 nm coating. Scales were then imaged via electron microscopy, and sectioned using a FIB.
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**Author contributions**

S.R.S. initiated the project, designed the local self-uniformity measure and the algorithm for its measurement, performed simulations and wrote the paper. W.M. designed and performed the microwave experiments, analysed the data and wrote the paper. S.S. performed the microwave experiments and analysed data. M.F. initiated the programme, oversaw and directed the project, designed the local self-uniformity measure and wrote the paper.

**Additional information**

Supplementary Information accompanies this paper at http://www.nature.com/naturecommunications

**Competing financial interests**: The authors declare that British patent application no. 1601838.4 has been filed in the name of the University of Surrey. The contributing authors Steven Sellers and Marian Florescu are recorded as the co-inventors. Weining Man and Shervin Sahba declare no competing financial interests.

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