



Soft self-assembly of Weyl materials for light and sound

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Soft materials can self-assemble into highly structured phases that replicate at the mesoscopic scale the symmetry of atomic crystals. As such, they offer an unparalleled platform to design mesostructured materials for light and sound. Here, we present a bottom-up approach based on self-assembly to engineer 3D photonic and phononic crystals with topologically protected Weyl points. In addition to angular and frequency selectivity of their bulk optical response, Weyl materials are endowed with topological surface states, which allow for the existence of one-way channels, even in the presence of time-reversal invariance. Using a combination of group-theoretical methods and numerical simulations, we identify the general symmetry constraints that a self-assembled structure has to satisfy to host Weyl points and describe how to achieve such constraints using a symmetry-driven pipeline for self-assembled material design and discovery. We illustrate our general approach using block copolymer self-assembly as a model system.

topological matter | metamaterials | polymers | colloids | semimetal

The propagation of waves in spatially periodic media is described by band theory, which determines the frequencies that can propagate in a given direction and at a given wavelength. While band theory was first developed to understand the behavior of electrons and phonons in solids, it applies to all kinds of waves. For example, photonic crystals are spatially periodic structures encompassed by such a description (1), which most notably can host a photonic band gap resulting in peculiar optical properties, such as the structural coloration of several butterflies' wings (2, 3).

A band structure is typically a complicated set of bands, which to a casual observer, seem to cross each other every possible way. This is, in fact, not the case: degeneracies in a band structure mainly appear at highly symmetric points and stem from the existence of additional symmetries (beyond translation invariance). However, so-called accidental degeneracies also exist, which are not enforced by the presence of a particular symmetry (4–6). Accidental degeneracies are generically rare and unstable, as they require a fine tuning of the system parameters to exist. However, such degeneracies may be protected by the existence of a particular singular configuration of the Bloch eigenstates in the neighborhood of the degenerate point (similar to a vortex or a hedgehog), which confers them with a topological character and hence, robustness against certain perturbations.

The simplest of such topological degeneracies are so-called Weyl points, where a 3D band structure locally exhibits a linear band crossing in all directions (7–9). Crucially, such a Weyl point is characterized by a topological charge, which describes the singularity in the Bloch eigenstates near the crossing point. Weyl points are robust against perturbations, which means that they can be moved in momentum space but not made to disappear unless they annihilate with a Weyl point of opposite charge, similar to hedgehog–antihedgehog pairs in real space in liquid crystals (10). Note that the Weyl points that we consider here generically occur at finite frequency and do not require a particular symmetry. In contrast, mechanical symmetry-protected Weyl points (similar to Dirac points in graphene) and Weyl lines occur at zero frequency (11–15). There, a chiral symmetry is essential to define the topological quantities and in turn, reveals a duality

between zero-frequency free mechanical motions and so-called self-stress modes (16–21).

Excitations following the Weyl equation (7–9) have been experimentally observed in electronic condensed matter in the so-called Weyl semimetal tantalum arsenide (22–26) as well as in photonic (27–32), phononic, and acoustic (33–36) crystals and in homogeneous magnetized plasma (37). Beyond their fundamental importance, such discoveries may pave the way for multiple applications enabled by the peculiar properties of Weyl points, such as their angular and frequency selective response and the existence of topologically protected arc surface states (called Fermi arcs in the electronic context) that appear at the boundary of finite samples, even when time-reversal invariance is not broken (38–43). This is in sharp contrast with gapped topological materials, where the existence of one-way channels requires breaking time-reversal invariance in some way, such as with external drives (44–47), magnetic or rotation fields (48–51), or active materials (52, 53).

All photonic Weyl materials designed up to now are based on top-down approaches (28–32, 39, 54–59). In this article, we show how soft matter self-assembly (60) provides a viable bottom-up strategy to realize Weyl materials for sound and light. Block copolymers are used as a paradigmatic example of soft materials that self-assemble into a variety of highly structured phases arising from the competition between elastic energy and surface tension (61). However, our strategy is applicable to a wider

Significance

Soft materials self-assemble into complex structures that can replicate on a larger scale the symmetric arrangements of atomic crystals. Harnessing soft self-assembly for material synthesis requires controlling the symmetries of such structures, a job tailored for group-theoretical methods traditionally used to describe crystal symmetries and their breaking. We illustrate this approach by providing a blueprint for self-assembled materials with an exotic feature of wave propagation known as Weyl points. The presence of such topological objects means that, at certain wavelengths, light or sound can only propagate inside the material in a handful of directions. Most crucially, sound and light waves can propagate on the material surface without backscattering from imperfections.

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Data deposition: The code used to compute the photonic band structures, the charges of the Weyl points, the irreducible representations of the numerical eigenvectors, the space group of the structures, and the spectra of the effective Hamiltonians is available on Zenodo at <https://doi.org/10.5281/zenodo.1182581>.

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1C) of topological origin. Let us consider a monochromatic beam of light or sound shone on the system. Depending on the wave frequency and its wave vector, it may be either reflected or transmitted in the bulk material as ruled by the band structure. When the beam hits the interface, it may also excite the arc surface states. At all frequencies close to the Weyl points, there is an arc-shaped curve in momentum space connecting the locations of the Weyl cones, representing the set of wave vectors at which topologically protected surface states are present.

The arc surface states arise from the topology of bulk states: pictorially, they can be seen as the projections of a bulk “Dirac string” connecting the Weyl points.* As the Dirac string is not gauge invariant, however, topology only determines the connectivity of the surface states (i.e., which Weyl points are connected). Beyond their fundamental importance, one of the main interests of arc surface states is the fact that their topological origin confers them a certain robustness to perturbations. Such arc states were experimentally observed in 3D materials both for light (29–32, 58) and for sound (33, 35). Although time-reversal invariance is preserved, unidirectional wave propagation immune to backscattering can be observed at the interface (30, 31, 33). This robustness is, however, not as strong as in a system with broken time-reversal invariance, as a component of the tangent momentum has to be (at least partially) conserved (30, 31, 33).

While the bulk Weyl points are most useful when they are spectrally isolated from other bands (27, 28), the arc surface states do not require such a frequency isolation (31). At microwave frequencies, arc surface states were observed by plugging an antenna into the sample (31). At lower wavelengths, they could be observed as follows: a defect, such as a small hole drilled at the surface of a Weyl material, allows a coupling between the surface states and freely propagating light on the outside. Let us consider several of such holes drilled at different places on the surface of the material as represented in Fig. 1D. In the absence of surface states at the light’s frequencies, a light beam shone on one of such holes propagates in the bulk and quickly disappears from the interface: all of the defects but the source are dark. In contrast, when surface states are present, a sizeable part of the beam intensity propagates at the interface in directions controlled by the positions of the arc states in the surface Brillouin zone. As a consequence, a handful of the holes are illuminated. It is worth noting that the surface of a Weyl metamaterial can only support arc surface modes if the conservation laws prohibit hybridization of such modes with the electromagnetic continuum outside. For the interface between Weyl materials and the vacuum, this requires the Weyl points to be below the light cone in the reciprocal space. When this is not the case, one has to consider an actual interface with a band gap material. In this case, the hole can simply be extended into the band gap material.

Self-Assembling Inversion-Asymmetric Gyroids

In this article, we assume time-reversal invariance and concentrate on inversion symmetry breaking to avoid the need for external drives, magnetic fields, or active materials. However, generically breaking inversion symmetry leads to uninteresting band structures. We adopt the following strategy: (i) start with highly symmetric structures possessing particular degeneracies and (ii) split such degeneracies into Weyl points by applying carefully chosen symmetry-breaking perturbations. Implementing both steps through a bottom-up strategy is very challenging. Our goal is to overcome this difficulty using soft self-assembly.

The first example of a photonic crystal displaying Weyl points was engineered by milling and stacking dielectric layers into a

highly symmetric structure called a double gyroid, in which additional holes were deliberately drilled at strategic points to reduce the symmetry of the system (28). A gyroid is an infinitely connected triply periodic surface of zero mean curvature discovered by Alan Schoen (67). The surface of the gyroid divides space into two regions corresponding to the interpenetrating labyrinth structures shown in Fig. 2.

A remarkable fact from soft matter science is that double gyroids naturally self-assemble in situations where two or several linked components have repulsive interactions with each other. In such circumstances, the minimization of surface energy constrained by the presence of links between the immiscible components can lead to a variety of minimal surfaces, among which is the gyroid surface. Gyroids generically appear in various soft materials, such as liquid crystals (70–72), amphiphilic surfactants (73, 74), dispersions of anisotropic and patchy colloids (75–79), and block copolymers (61, 80, 81) to name but a few.

AB diblock copolymers are the archetypal example of such a self-assembling soft material. They are composed of two immiscible polymer blocks denoted by A and B glued together by covalent bonds. For a well-chosen set of system parameters (typically the average degree of polymerization, the relative fractions of A and B, and the Flory–Huggins parameter characterizing the interaction energy between the blocks A and B), the constrained minimization of the interface energy leads to a double-gyroid structure, where two minority networks of opposite chirality are interwoven inside a matrix majority network (82, 83). The interface between one of the minority networks and the matrix is a gyroid surface, a triply periodic constant curvature surface (67, 84, 85), which is well-approximated by the isosurface $g(x, y, z) \equiv \sin(2\pi x) \cos(2\pi y) + \sin(2\pi y) \cos(2\pi z) + \sin(2\pi z) \cos(2\pi x) = t$ (with $0 \leq t < \sqrt{2}$) (86), where x , y , and z are measured in units of the unit cell size a . The second minority network is obtained from the first through space inversion. Hence, one of the gyroidal minority networks is described by $g(x, y, z) \geq t$, while its chiral partner, obtained by space inversion, is described by $g(-x, -y, -z) \geq t$. Both are composed, say, of the A blocks, while the majority matrix is composed of the B blocks.

Crucially, the resulting structure has inversion symmetry that is almost impossible to get rid of without local modifications. This is certainly possible in engineered structures like the milled structures in ref. 28, where one has direct control on the shape of the unit cell, but it is not compatible with a bottom-up material synthesis scheme. To take advantage of a self-assembly scheme, we instead choose to use ABC triblock terpolymers, which self-assemble in a double gyroid where two chemically distinct gyroid-shaped minority networks of opposite chirality are interwoven inside a matrix majority network (82, 83). For instance, one of the gyroidal labyrinths may be composed of A blocks, but its image by space inversion is then composed of C blocks, and the matrix is still composed of B blocks. The resulting structure is called an asymmetric double gyroid or an alternating double gyroid. After the polymer self-assembly, standard techniques allow us to selectively etch one of the gyroidal minority networks and to replace it with a high-permittivity material (87, 88) [for example, by metal (68, 69) or dielectric (89, 90) deposition]. Crucially, the chemical difference between both gyroidal networks allows us to induce an optical asymmetry between them either by depositing materials of different dielectric constants or through the use of a mild etching agent to tune their respective radii. The last step is to get rid of the majority network matrix. The whole process is summarized pictorially in Fig. 2. After this process is complete, we are left with a structure where the dielectric constant is ϵ_A for $g(x, y, z) \geq t_A$, ϵ_B for $g(-x, -y, -z) \geq t_B$, and $\epsilon_{\text{air}} = 1$ outside of such regions. Similarly, a phononic crystal can be obtained by inducing an asymmetry in the elastic properties of the two gyroidal networks. In the following, we focus on photonic crystals for concreteness, but full details on acoustic and phononic crystals are provided in *SI Appendix*.

*More precisely, the topology of the band structure is fully characterized not only by the charges of the Weyl points but by weak first Chern numbers (or weak Fu–Kane–Mele invariants) defined on 2D planes or surfaces of the bulk Brillouin zone (63–65). In contrast, the exact shape of the topological surface states is indeed not determined by the topology and depends on the boundary conditions (31). In particular cases, it can, however, be predicted from the bulk through the entire data of the Berry connection (66).

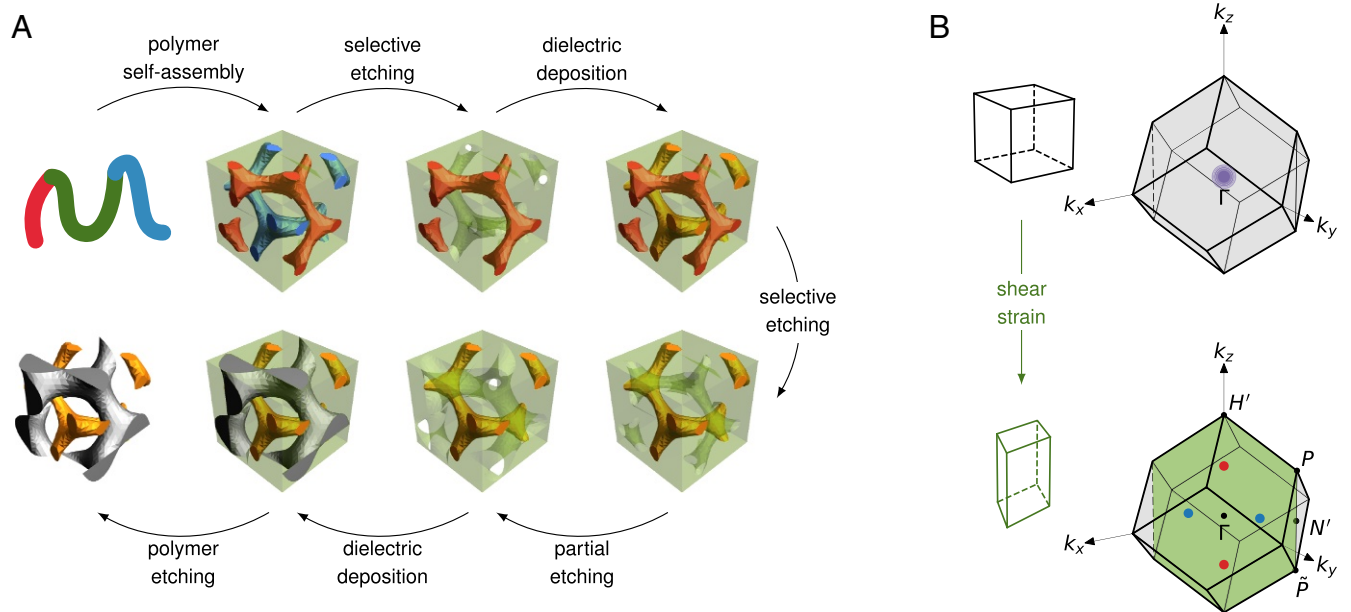


Fig. 2. Self-assembly process and effect of strain. (A) The self-assembly of triblock terpolymers leads to “colored” double gyroids, where the two minority networks (red and blue) are chemically distinct (68, 69). Starting from the self-assembled structure, a series of selective etching, partial dissolution, and backfilling steps leads to an asymmetric double gyroid made of high dielectric constant materials, which constitute a 3D photonic metacrystal. Crucially, the photonic band structure of such a system has a threefold degeneracy at the center of the Brillouin zone (the Γ point), which is represented in purple in *B, Upper Right*. This threefold degeneracy can be split into a set of Weyl points by an appropriate strain (in this case, pure shear) represented in red and blue (for Weyl points of charge 1 and -1 , respectively) in *B, Lower Right*.

Effective Description of the Band Structure

To obtain Weyl points, the symmetry of the double gyroid must be reduced further. Full-wave numerical simulations reveal that the photonic band structure of a dielectric double gyroid has a threefold quadratic degeneracy at the Γ point (the center of the first Brillouin zone) (91). From the point of view of symmetries, the threefold degeneracy is allowed by the existence of 3D irreducible representations of the subgroup of symmetries leaving the Γ point invariant, namely the irreducible representation T_{1g} of the full octahedral group $m\bar{3}m$ (or O_h in Schoenflies notation) (*SI Appendix*). This threefold degeneracy can be split into pairs of Weyl points by symmetry-breaking perturbations (27, 39) as represented in Figs. 2*B* and 3. The systematic description of a band structure near a high-symmetry point of the Brillouin zone as well as the effect of symmetry-breaking perturbations can be obtained from group theory. This approach, known as the method of invariants, originated within condensed matter physics (92–96), but it also applies to photonic systems (97, 98) and more generally, to all kinds of waves in periodic media.

For example, in the absence of charges and currents, Maxwell equations can be written in a convenient way as

$$i\partial_t \begin{pmatrix} \mathcal{E} \\ \mathcal{H} \end{pmatrix} = \left[\begin{pmatrix} \epsilon & 0 \\ 0 & \mu \end{pmatrix}^{-1} \begin{pmatrix} 0 & \text{rot} \\ -\text{rot} & 0 \end{pmatrix} \right] \begin{pmatrix} \mathcal{E} \\ \mathcal{H} \end{pmatrix}, \quad [3]$$

where \mathcal{E} and \mathcal{H} are the electric and magnetic fields, respectively, while ϵ and μ are the spatially varying permittivity and permeability of the medium, respectively. In this form, the operator in square brackets, called the Maxwell operator, plays the role of a Hamiltonian[†] (1, 99, 100). This full-wave Maxwell equation is usually impossible to solve analytically: one has to

resort to numerical simulations. However, with minimal input from a numerical full-wave solution complemented with the full knowledge of symmetries in the problem, one can determine an effective Hamiltonian, which is sufficient for perturbative design purposes. Similar considerations apply to other kinds of waves propagating in periodic media (for example, elastic waves) (*SI Appendix*).

By reducing the full description of the system (contained in the Maxwell operator) to the subspace spanned by a few relevant degrees of freedom, one obtains an effective Hamiltonian describing a few bands in the vicinity of a (usually high-symmetry) point k_0 of the Brillouin zone. For example, the eigenstates involved in a degeneracy at k_0 can then be used as a basis to describe the effective Hamiltonian operator as a matrix $H(q)$, where $q = k - k_0$. Both the Maxwell operator and the effective Hamiltonian operator are invariant with respect to the symmetries g of the group of the wave vector k_0 , defined as the subgroup of symmetries that leave k_0 invariant. The general idea of the theory of invariants is that symmetries can be used to construct the effective Hamiltonian matrix from scratch by combining a set of basis matrices X (which form a basis of, say, the space of 3×3 Hermitian matrices) and irreducible functions $\mathcal{K}(q)$ of the wave vector components (like $q_x^2 + q_y^2 + q_z^2$). The basis matrices represent operators in the basis of eigenstates at k_0 . As such, they change when a symmetry operation is applied. This is also the case in irreducible functions as the symmetry operation is applied to the momentum vector. As the effective Hamiltonian operator is invariant when a symmetry is applied, it is possible to determine all terms allowed by symmetry by selecting all combinations of the form $\mathcal{K}(q)X$, which are left invariant by the action of the symmetries.

More precisely, if the eigenstates at k_0 form an irreducible representation Γ , then the matrix representation $H(q)$ of the effective Hamiltonian operator describing the corresponding bands will be covariant with respect to the symmetries. Namely, $D(g)H(g^{-1}q)D(g)^{-1} = H(q)$, where D is a representation of Γ acting on the effective Hamiltonian by its adjoint action as a representation of $\Gamma \times \Gamma^*$. The effective Hamiltonian can then be

[†]For normal materials where permittivity and permeability are strictly positive, the Maxwell operator is Hermitian with respect to a relevant scalar product (1, 99). An additional constraint stemming from the source-free equations has to be taken into account, which commutes with the Maxwell operator.

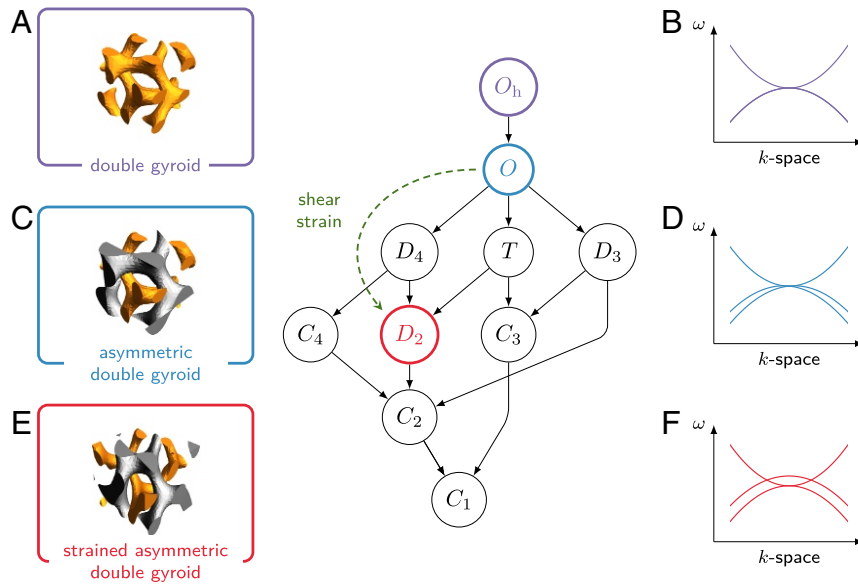


Fig. 3. Reducing the symmetry. In A–C, we show the various structures of interest: (A) the (symmetric) double gyroid, (B) the asymmetric double gyroid, and (C) the strained asymmetric double gyroid (with shear strain). For the double gyroid, the group of the wave vector Γ is the octahedral group O_h ($m\bar{3}m$ in Hermann–Mauguin notation), while it is the octahedral rotation group O (432) for the asymmetric double gyroid. When strain is applied to the asymmetric double gyroid, its symmetry is reduced, which corresponds at the Γ point to a subgroup of the octahedral rotation group O . Such subgroups are organized in a Hasse diagram. (The octahedral rotation group is indeed a subgroup of the full octahedral group O_h , which has more subgroups that are not relevant here and were not represented. As strain preserves inversion symmetry, any strained symmetric double gyroid still has an inversion center. The point group at Γ is then the product of the inversion group S_2 with a subgroup of O . Such situations can be achieved by starting from the symmetric double gyroid. In addition, there are other mixed subgroups of O_h , which cannot be directly realized through our method.) In D–F, the method of invariants predicts the qualitative features of the band structure of the modified double gyroids (B and C).

constructed by combining basis matrices $X^{(\gamma,\mu)}$ of the irreducible representations Γ_γ appearing in decomposition of this product and irreducible functions of the wave vector components $\mathcal{K}^{\gamma,\mu}(q)$ as

$$H(q) = \sum_{\gamma} a_{\gamma} \sum_{\mu} X^{(\gamma,\mu)} \mathcal{K}^{\gamma,\mu*}(q), \quad [4]$$

where a_{γ} indicates arbitrary constants chosen such that $H(q)$ is Hermitian and where μ labels the basis elements in the same irreducible representation.

Given (i) the input of the space group $Ia\bar{3}d$ of the double gyroid and (ii) the fact, known from full-wave computations, that a threefold band crossing transforming according to the 3D irreducible representation T_{1g} exists at the Γ point, the method of invariants yields the following effective Hamiltonian, describing the band structure in the vicinity of this crossing (SI Appendix):

$$H_0(k) = a_1^{(0)} \text{Id} + a_1^{(2)} k^2 \text{Id} + a_{12}^{(2)} (\bar{K} \Lambda + K \Lambda^\dagger) + a_{25}^{(2)} (k_x k_y L_x L_y + \text{c.p.}) + O(k^3), \quad [5]$$

where $k^2 = k_x^2 + k_y^2 + k_z^2$ and c.p. stands for “circular permutation” (of the indices). Matrices L_i are 3×3 Hermitian angular momentum matrices satisfying $[L_i, L_j] = i \epsilon_{ijk} L_k$ and $L_x^2 + L_y^2 + L_z^2 = 2\text{Id}$, and Id is the identity matrix. We also defined $K = (k_x^2 + \bar{\omega} k_y^2 + \omega k_z^2)$ and $\Lambda = L_x^2 + \bar{\omega} L_y^2 + \omega L_z^2$ with $\omega = e^{i2\pi/3}$.

In this expansion, the indices of the coefficients $a_I^{(p)}$ refer to the irreducible representation Γ_I from which the invariant term was constructed and the exponent in parentheses to the order of the irreducible polynomial composed of the wave vector components (at the Γ point, $k_0 = 0$, so that $q = k$). Finally, in addition to space symmetries, time-reversal invariance is imposed by considering only time-reversal even combinations.

The main interest of the method of invariants is that it allows us to determine what new terms can be added to the preced-

ing effective Hamiltonian when the symmetry is reduced. This enables us to qualitatively predict the effect of perturbations on the band structure as illustrated in Fig. 3. As we have seen, the very first step toward inducing Weyl points is the removal of inversion symmetry from the structure (Fig. 3C). Hence, the point group at Γ becomes the chiral octahedral group 432 (or O in Schoenflies notation). However, this modification does not allow a constant term in the effective Hamiltonian: only a new linear term of the form $k_x L_x + k_y L_y + k_z L_z$ appears due to the reduction of the irreducible representation T_{1g} and T_{1u} of O_h to T_1 in O . Hence, the quadratic band crossing at Γ cannot be lifted by such a term (Fig. 3D), and a further reduction in symmetry is required.

Strain and Symmetries

The simplest yet global way to reduce the symmetry of a structure is to apply a mechanical strain. The key point is that this strategy is compatible with self-assembly, unlike local modification or patterning of the individual building blocks. The asymmetry between the enantiomeric (i.e., nonidentical mirror images of each other) gyroidal components described in the previous section reduces the space group $Ia\bar{3}d$ [International Union of Crystallography (IUC) no. 230] of the symmetric double gyroid to $I4_132$ (IUC no. 214). When strain is applied, this space group is further reduced.

We choose to apply the shear strain

$$\epsilon = \begin{pmatrix} \cos \theta & \sin \theta & 0 \\ \sin \theta & \cos \theta & 0 \\ 0 & 0 & 1 \end{pmatrix} \quad [6]$$

written in the Cartesian coordinates of the standard conventional cell (not the primitive cell). This transformation (illustrated in Fig. 3E) reduces the space group of the asymmetric double gyroid to $F222$ (IUC no. 22) when θ is nonzero [the

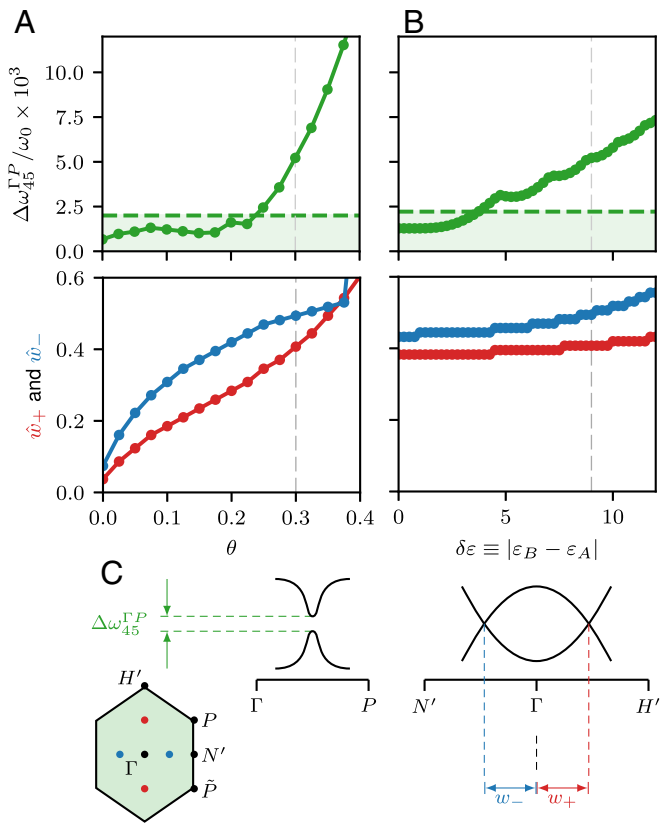


Fig. 5. Evolution of the main features of the photonic band structures. Evolution of characteristic features of the Weyl points, represented in (A) with the strain angle θ and (B) the dielectric asymmetry $\delta\varepsilon \equiv \varepsilon_B - \varepsilon_A$. Here, the band structures are computed with the set of parameters in Fig. 4B on a $(32 \times 3)^3$ grid. We plot both the minimum of the local gap between the fourth and the fifth bands, $\Delta f_{45}^{\Gamma P} = \min\{|f_5(k) - f_4(k)| \mid k \in \Gamma - P\}$, and the normalized positions of the two inequivalent Weyl points, $\hat{w}_+ = w_+ / \|\Gamma N'\|$ and $\hat{w}_- = w_- / \|\Gamma H'\|$. The gray dashed lines correspond to the value at which each parameter is kept constant in other figures. The abrupt jump in the position of one of the Weyl points near $\theta = 0.4$ is an artifact: another set of band crossings appears on the $\Gamma - N'$ and $\Gamma - H'$ lines near this value (*SI Appendix*). For the local gap, light green regions delimited by dashed lines correspond to the order of magnitude of symmetry-breaking numerical errors (*SI Appendix*). The data are not meaningless below this threshold, but the effects of the strain and structural asymmetry are not distinguishable from the spurious numerical reduction of symmetry. Similarly, \hat{w}_\pm should both vanish at $\theta = 0$ (which is clearly not the case). This provides an order of magnitude of the uncertainty on both observables.

Such tunable gap materials have been used to realize strain sensors (105). Here, we can envision a combination of such strain-sensing methods with an optical tracking of the strain-induced Weyl points to achieve a high-precision measure of mechanical properties.

Self-Assembled Weyl Materials for Light and Sound

While we focused on photonic systems, the same group-theoretical analysis applies to other kinds of waves. We consider three examples where Weyl points were already shown: (i) dispersive photonic media (37) (i.e., with a frequency-dependent dielectric tensor), (ii) phononic crystals (34), and (iii) acoustic crystals (33, 35, 36). To show the possibility of obtaining a self-assembled Weyl material, we only need to consider the band structure of an unperturbed double gyroid and look for an essential threefold degeneracy at the Γ point. The rest follows from our group-theoretical analysis. As we shall see, such a threefold degeneracy appears in both dispersive photonic and phononic

systems but does not seem to arise in the considered acoustic system, at least at reasonably low frequencies.

When light propagates in a structure made of a metal or in a dielectric at high frequency, the plasma oscillations of the electron density couple with the electromagnetic field, leading to a dispersive photonic crystal where the propagation of light is still described by Maxwell equations but with a frequency-dependent dielectric tensor (106, 107). We consider a double gyroid made of a Drude metal with the plasma frequency of gold, the band structure of which is represented in Fig. 6A. In dispersive photonic crystals, the scale invariance of Maxwell equations is not valid anymore, as the plasma frequency provides a length scale.

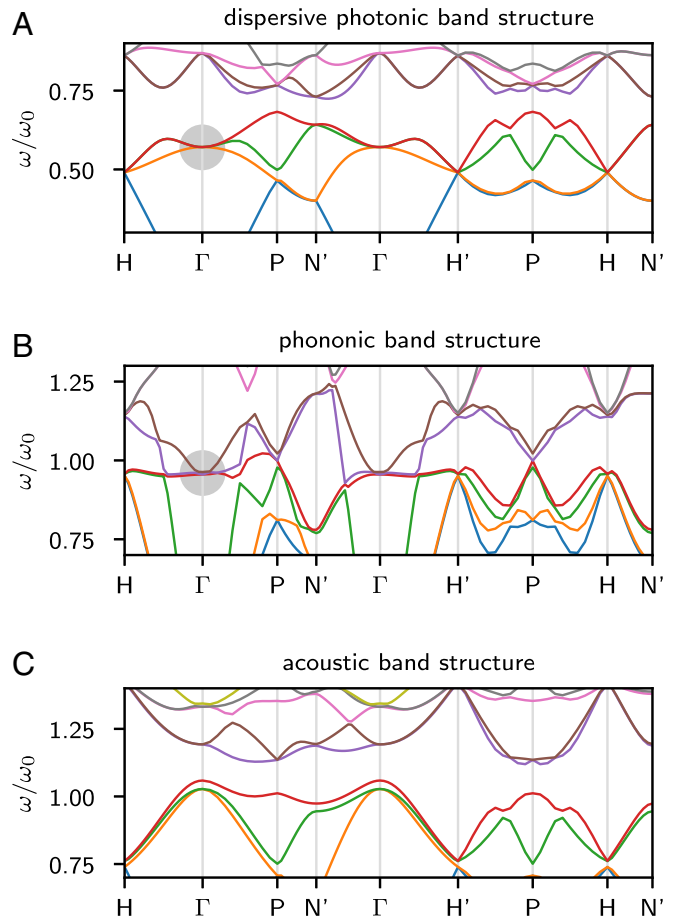


Fig. 6. Band structures of the unperturbed double gyroid for different waves. (A) Dispersive photonic band structure of a metallic double-gyroid structure made of a Drude metal with the plasma frequency of gold standing in vacuum. (B) Phononic band structure for an elastic double gyroid in steel embedded in an epoxy elastic matrix. (C) Acoustic band structure for sound in air confined outside of a double gyroid with hard wall boundary conditions. In the dispersive photonic and phononic band structures (A and B), a threefold degeneracy (highlighted by gray circles) is found. As such, we expect such systems to exhibit Weyl points when strained. In A, $\omega_0 = 2\pi c/a$, where c is the speed of light in vacuum. We use the plasma frequency of gold, $\omega_p/2\pi \approx 2.19 \times 10^{15}$ Hz (108), and $a \approx 500$ nm. The loss term Γ is initially set to 0, and the results show no significant deviations from the case computed with the tabulated value $\Gamma/2\pi = 5.79 \times 10^{12}$ Hz (108). In B, $\omega_0 = 2\pi c_t/a$, where c_t is the speed of transverse waves in epoxy. The values assumed for the longitudinal and transverse speeds of sound in steel and epoxy are obtained from the components of elastic tensor C_{ij} as $c_t^2 = C_{44}/\rho$ and $c_l^2 = C_{11}/\rho$ from the values in refs. 109–111, namely $\rho^{\text{epoxy}} = 1180$ kg m $^{-3}$, $C_{11}^{\text{epoxy}} = 7.61$ GPa, and $C_{44}^{\text{epoxy}} = 1.59$ GPa and $\rho^{\text{steel}} = 7780$ kg m $^{-3}$, $C_{11}^{\text{steel}} = 264$ GPa, and $C_{44}^{\text{steel}} = 81$ GPa. In C, $\omega_0 = 2\pi c_{\text{air}}/a$, where c_{air} is the speed of sound in air. All computations are performed with a $48 \times 48 \times 48$ grid. More details on the model and computation are in *SI Appendix*.

The case of a unit cell of size $a = 100$ nm was previously considered in ref. 68. At such scales, the threefold degeneracy may still be present but is overlapped by highly dense plasma bands, and it cannot be identified. Here, we consider a unit cell of size $a = 500$ nm. A threefold degeneracy appears near $\omega/\omega_0 \sim 0.5$. Inspection of the eigenvectors shows that the electric field transforms along the 3D irreducible representations T_{1u} (more details are in *SI Appendix*). Similarly, in a phononic crystal, elastic waves propagate in a spatially periodic structure. Here, we consider a double gyroid made of steel embedded in an epoxy matrix, which couples elastically the two enantiomeric gyroids (110). As observed in Fig. 6 *A* and *B*, a threefold degeneracy is found near $\omega/\omega_0 \sim 0.95$. Inspection of the numerical eigenvectors shows that they also transform according to the 3D irreducible representation T_{1u} (more details are in *SI Appendix*). According to our analysis, such threefold degeneracies will be split into Weyl points by inducing an asymmetry in the enantiomeric gyroid networks and applying an appropriate strain in both the dispersive photonic and phononic systems.

By contrast, we consider the case of an acoustic system, where sound propagates in air outside a double gyroid-shaped labyrinth. Here, no threefold degeneracy at Γ seems to appear in the band structure (at least below $\omega/\omega_0 \sim 1.75$) (Fig. 6 *C* and *SI Appendix*) for the values of the parameters we considered. As a consequence, we do not expect Weyl points to appear under strain at those frequencies. Finally, the band structure of electrons constrained to move on gyroid-shaped nanostructured semiconductors displays multiple degeneracies (113, 114), which could also give rise to Weyl points under strain. In this situation, however, one has to take into account the spin degrees of freedom of the electrons, which are also affected by the curvature, and therefore, we can draw no definitive conclusion from our analysis, which would have to be adapted to include double-group representations.

The self-assembly of asymmetric double-gyroid structures has already been shown experimentally in block copolymers (69). Directed self-assembly can induce mechanical strains in the direction of growth (115), which according to our symmetry analysis, would automatically lead to the appearance of Weyl points without the need of applying external perturbations. Moreover, gyroid-based systems appear to be unusually resistant to the appearance of cracks when strained (116–118), possibly as a result of their 3D cocontinuous structure, making them a particularly good fit for our strain-based design. The size a of the unit cell of the structures obtained by block polymer self-assembly crucially depends on the blocks' molar mass. With current experimental techniques, the accessible unit cell sizes range from a few nanometers to a hundred nanometers (83, 119). In photonic crystals, this constraint on the unit cell size means that we can expect Weyl points to appear at wavelengths of order $\lambda_{\text{opt}} \sim a/0.5 \simeq 200$ nm (or smaller), which are at UV wavelength. Depending on the materials used in the process, the light frequency may be high enough for the dielectric function not to be constant anymore, but as we have shown, Weyl points can also occur in dispersive photonic crystals. While the direct observation of a Weyl band structure at such frequencies is challenging, such self-assembled photonic crystals could be used in X-ray/UV optics (for example, to realize Veselago lenses as proposed in ref. 42).

To generate an optical response in the visible spectrum, it would be interesting to explore hierarchical self-assembly of gyroids using soft building blocks larger than standard polymeric monomers, such as superstructures formed by anisotropic colloids (75, 78), or liquid crystalline phases (72).

Symmetry-Driven Discovery of Self-Assembled Materials

Both the possible existence of a threefold degeneracy and its splitting into Weyl points are predicted by group theory. In this study, we did not need to make an initial guess of a structure leading to a threefold degeneracy at Γ , because we used the well-known example of a double gyroid. Note, however, that symmetry

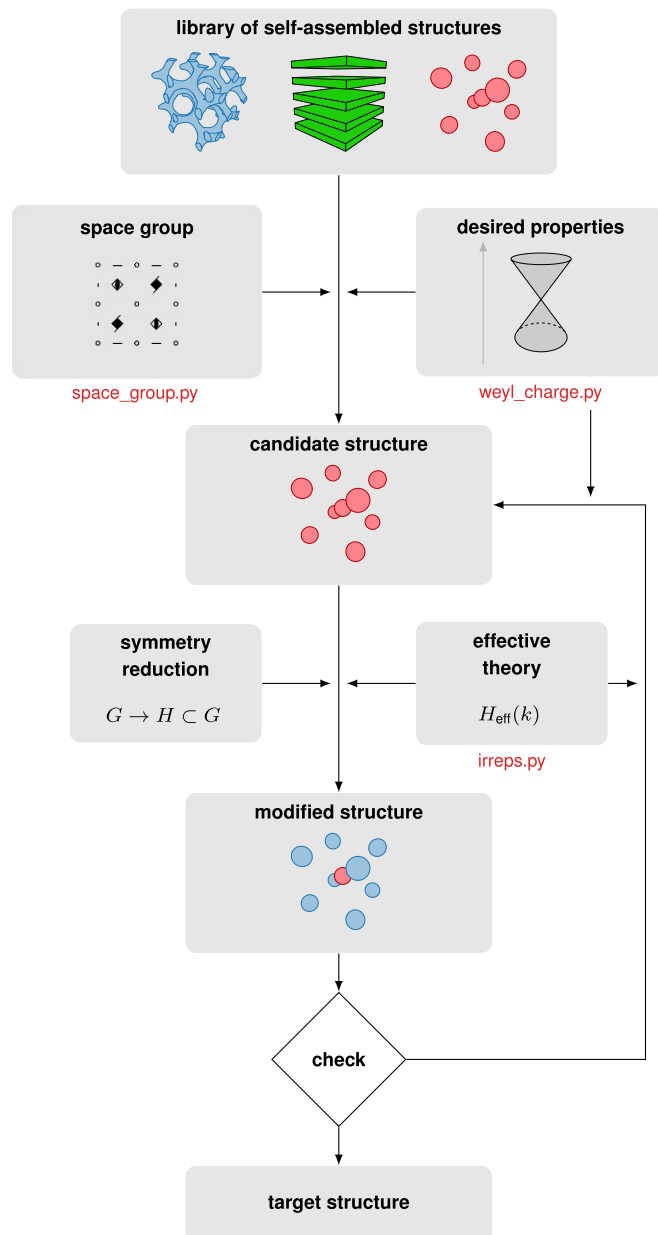


Fig. 7. Symmetry-driven mesostructured material discovery pipeline. To obtain mesostructured materials with a set of desired properties, we suggest the following automated discovery pipeline. We start from a library of self-assembled structures, which is scanned for candidates matching symmetry requirements for a set of target properties. This requires us to automatically determine the space group of each structure: a script `space_group.py` does this job for structures represented as a skeletal graph (*SI Appendix*). A best candidate for the initial structure is then selected, and its properties are numerically computed. For example, we compute the band structure, from which the topological charges of the Weyl points (if any) are determined by a script `weyl_charge.py`. An effective description is then extracted from the numerical data: here, we need to determine the irreducible representations of the numerical eigenvectors, a job performed by the script `irreps.py` (*SI Appendix*). The effective description then allows us to determine which modifications should lead to the desired properties (for example, through a symmetry reduction). Here, this step could also be automated using <https://github.com/greschd/kdotp-symmetry>. Finally, the properties of the modified structure are numerically determined and compared with the desired properties. In case of failure, a new initial structure is selected from the library, and the process is iterated.

considerations can also guide this first step, as they determine in which structures essential degeneracies can exist (120), such as frequently used both in solid-state physics (121–123) and for classical waves (124, 125). This approach combined with an iterative search through libraries of self-assembled structures could provide an extension of our results to different systems. We developed open source Python packages that perform some of the tasks required (details are in *SI Appendix*): (i) the script `space_group.py` numerically determines the space group of a structure represented as a skeletal graph in the presence and absence of mechanical deformations using the open source library `spglib` (101), (ii) the script `irreps.py` numerically determines the irreducible representations of the numerical eigenvectors, and (iii) the script `weyl_charge.py` computes numerically the charges of the Weyl nodes for an arbitrary band structure using a gauge-invariant method (126–132). Fig. 7 provides a schematic representation of

an automated self-assembled mesostructured material discovery pipeline, which would blend computationally intensive full-wave simulations and the group theoretical tools used in this study. In such a scheme as well as in our work, symmetries act as a powerful guide in the wealth of self-assembled structures (61, 83, 87, 133) by both identifying candidate systems and determining suitable perturbations to achieve a given response.

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