

Supplementary Information for Chiral Diffraction from Aperiodic Monotile Structure

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S1 Structure of hat tiling and monotile quasilattice

S1.1 Weak C_3 symmetry in the hat tiling and monotile quasilattice

Many quasicrystalline structures are characterized by their unique rotational symmetries. The rotational symmetry of quasicrystalline order can sometimes be described by the symmetry of the higher-dimensional space. For example, the structure based on the Penrose tiling exhibits C_5 symmetry in real space, but possesses C_{10} symmetry in higher-dimensional space, and even in real space it shows “weak” C_{10} symmetry. Here, “weak” means that, when the aperiodic tiling is extended infinitely, the set of points that break the rotational symmetry has a scale smaller than the spatial dimension of the tiling.

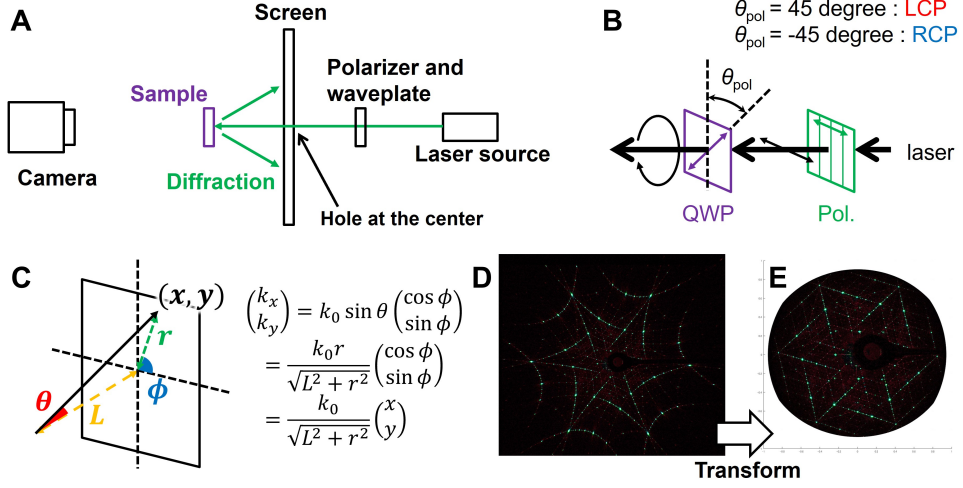


Fig. S1 Details of experiments and analysis. (A) Experimental setup for diffraction measurements. (B) Definition of the circular polarization in the experimental setup. (C) Coordinate transformation from the screen coordinates to wavenumber coordinates. Typical diffraction images (D) before and (E) after transformation.

Such a feature can also be found in the hat tiling and in the monotile quasilattice studied in this work. Figures S2(A) and (B) show superpositions of the hat tiling of the H_2 metatyle with its copies rotated by 120° and 240° , respectively. The regions where the two tilings do not completely overlap are highlighted in color. Figure S2(C) shows a similar superposition for the H_3 metatyle. An important observation is that while the total area increases with the square of the spatial scale from Fig. S2(A) to (C), the size of the non-overlapping (light-blue) defective regions increases only linearly with the scale. This implies that, as the hat tiling is expanded infinitely through the inflation rule, the effect of the defect regions becomes negligible. Therefore, the hat tiling has weak C_3 symmetry.

A similar property is found in the monotile quasilattice. In the case of the monotile quasilattice, perfect C_3 symmetry appears at the center of the H metatyle, whereas weak C_3 symmetry, as illustrated in Fig. S2(D), can be found at specific points other than the center. Such weak C_3 symmetry appears at multiple points. Therefore, since the rotational symmetry of the monotile quasilattice emerges not only at the central point but also at several other positions, the positions of the diffraction peaks do not depend on the illumination position and always exhibit a C_6 -symmetric diffraction pattern (see section S2). Ref. [1] has shown that a quasicrystalline structure with C_3 symmetry in real space can exhibit C_6 symmetry in higher-dimensional space. In a similar manner, a key tiling derived from the hat tiling can be constructed by projection from six-dimensional space [2].

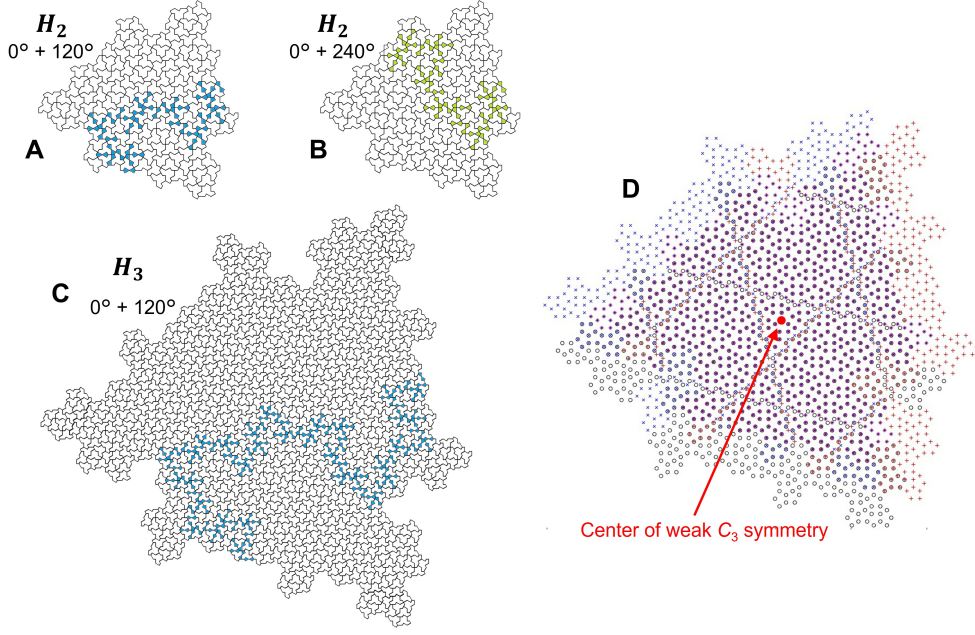


Fig. S2 Weak C_3 symmetry in the hat tiling and monotile quasilattice. (A) The H_2 hat tiling overlaid with its 120° -rotated copy. Colored regions indicate the areas where the two do not fully overlap. (B) The H_2 hat tiling overlaid with its 240° -rotated copy. (C) The H_3 hat tiling overlaid with its 120° -rotated copy. (D) The monotile quasilattices rotated by 120° (red hollow circles) and 240° (blue crosses) around a point (red point) off the center of the H_3 metatile.

S1.2 Calculation of θ_{chiral}

Figure S3 illustrates the generation of H_n metatiles using T , H , P , and F metatiles [3]. As shown in Fig. S3(A), the four initial metatiles (outlined in bold black) have shapes aligned with the underlying honeycomb framework. From Fig. S3(B), the combination of the metatiles is C_3 symmetric, which is the reason why the monotile quasilattice has C_3 symmetry. When the inflation rule is applied to generate the H_1 metatile, we notice that the H_1 metatile is rotated clockwise relative to the base honeycomb framework. This rotation angle gradually converges to θ_{chiral} as the inflation rule is repeatedly applied. The chiral nature of the hat tiling and the monotile quasilattice results from this rotation.

To calculate θ_{chiral} , we focus on the H_{n-1} metatile outlined in red, which constitutes H_n metatiles (Fig. S3(B)-(D)). The $\vec{A}_n$, the vector from the center of the H_n metatile to the center of the red H_{n-1} metatile, can be expressed by a linear combination of two basis vectors $\vec{a}_1$ and $\vec{a}_2$ aligned with the underlying honeycomb lattice as shown in Fig. S3. Interestingly, the coefficients A_{n1} and A_{n2} follow the Fibonacci

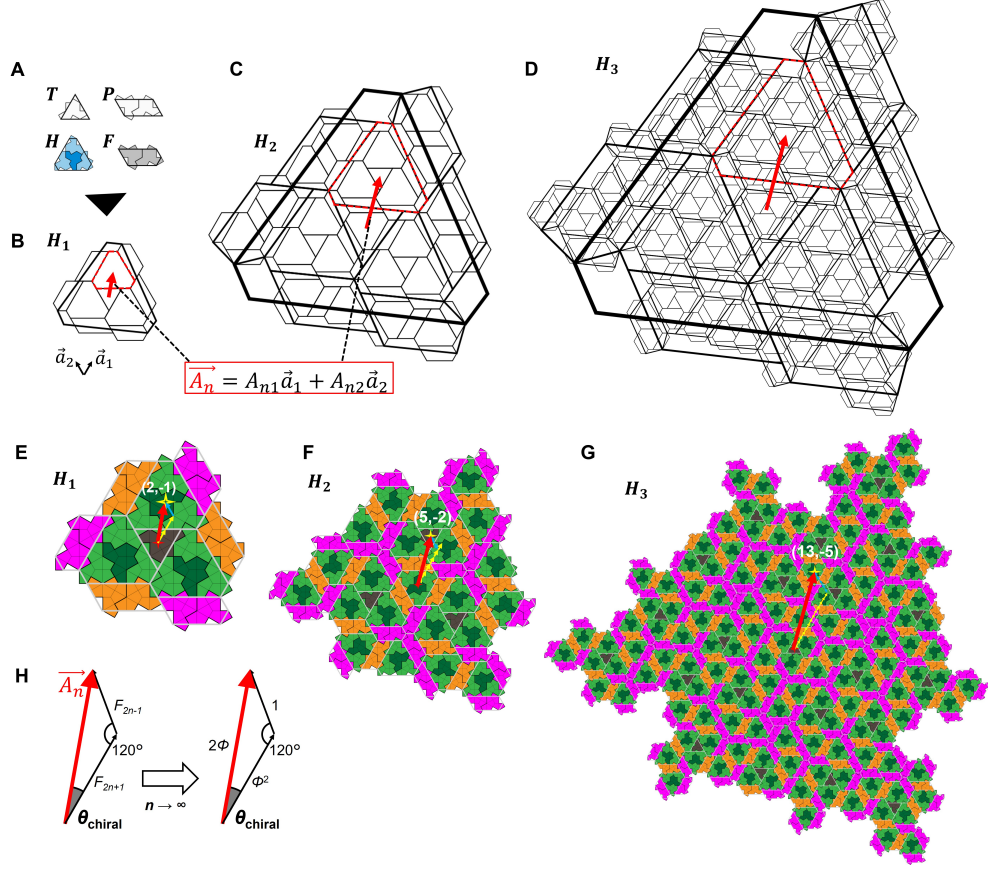


Fig. S3 Generation of hat tiling and its twisting nature. (A)-(D) Frames of metatiles for H_n metatiles. The pictures of the hat tiling are generated by a browser-based visualization tool provided by Smith's team (<https://cs.uwaterloo.ca/%7Ecsk/hat/>). (E)-(G) Hat tilings of H metatiles. We can confirm the appearance of the Fibonacci sequence in the vector $\vec{A}$. (H) The triangle to calculate θ_{chiral} .

sequence, $F_{n+2} = F_{n+1} + F_n$ (1, 1, 2, 3, 5, 8, 13, 21, 34, 55, 89, ...), as shown in Table 1, which can be confirmed by Fig. S3(E)-(G). In the limit of $n \rightarrow \infty$, the angle θ_{chiral} can be calculated by the triangle shown in Fig. S3(H). Using the law of cosines and $\phi^2 = \phi + 1$,

$$\cos \theta_{\text{chiral}} = \frac{\phi^4 + (2\phi)^2 - 1}{2\phi^2 \cdot 2\phi} = \frac{3\phi - 1}{4}. \quad (1)$$

S1.3 SEM image of fabricated sample

Figure S4 is an SEM image of the fabricated monotile quasilattice of H_3 . The corresponding hat tiling is also drawn over the SEM image.

Table 1 Fibonacci sequence appearing in $\vec{A}_n$.
The coefficients, A_{n1} and A_{n2} for H_n metatiles. We can see the Fibonacci sequence in these coefficients.

	H_1	H_2	H_3	H_4	H_5	H_6	H_n
A_{n1}	2	5	13	34	89	233	F_{2n+1}
A_{n2}	1	2	5	13	34	89	F_{2n-1}

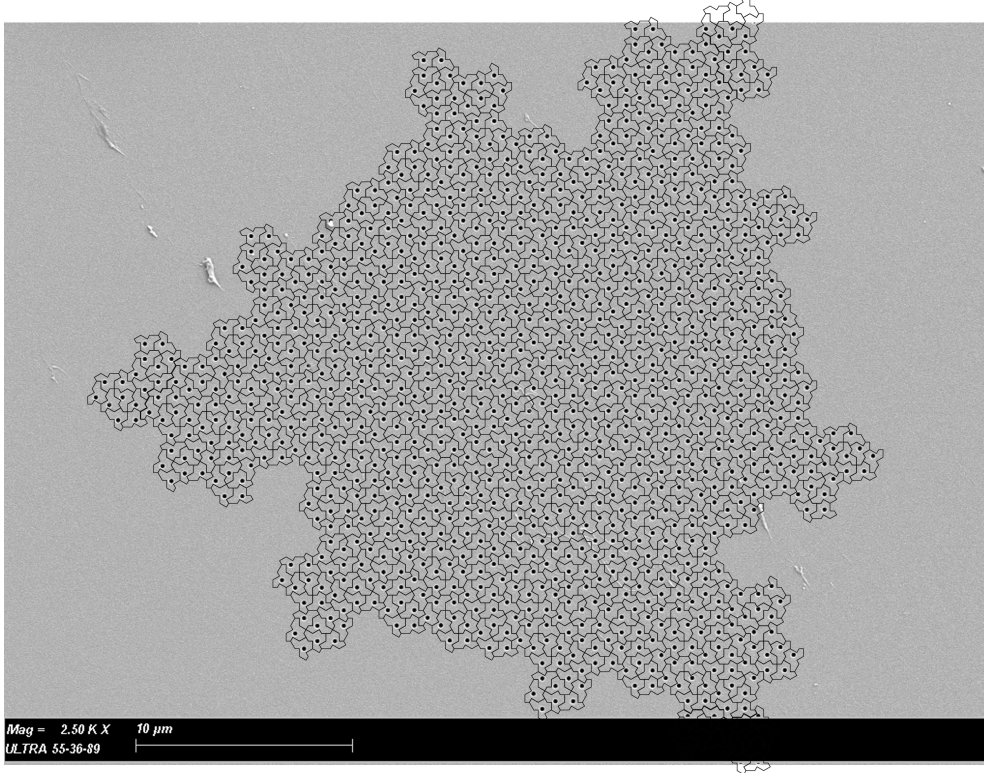


Fig. S4 SEM image. An SEM image of the fabricated monotile quasilattice of H_3 metatiles. A hat tiling is overlaid on the SEM image.

S2 Real-space position dependence of the diffraction pattern

Here, we show that the diffraction pattern of the monotile quasilattice does not depend on the illumination position in real space. This insensitivity is strongly related to the presence of rotational symmetry at multiple positions (see section S1.1). This position independence is consistently confirmed by both numerical calculations and

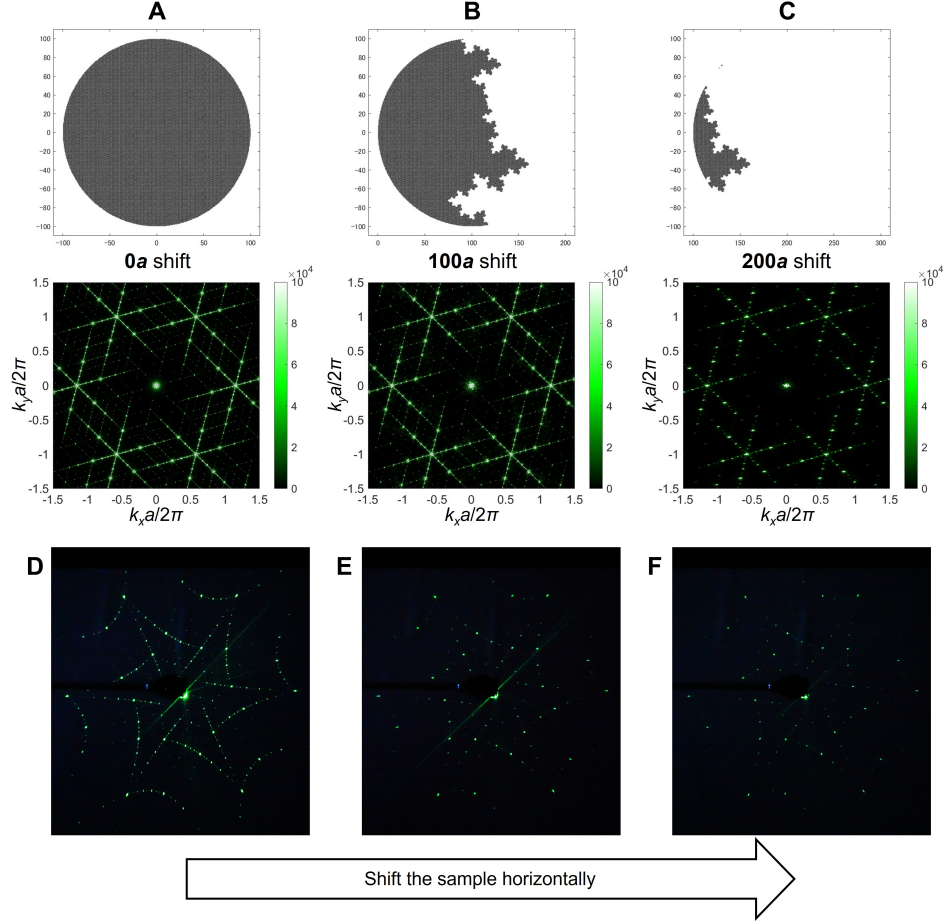


Fig. S5 Position independence of the diffraction pattern. Calculated diffraction patterns when the center position is shifted by (A) $0a$, (B) $100a$, and (C) $200a$ along the x direction. Only the points within a circular region of radius $100a$ are used in the calculation. (D)-(F) Experimentally obtained diffraction images while the beam spot was shifted laterally. (D) at the center, (E) shifted by 0.5 mm, and (F) shifted by 1 mm from the center.

experimental observations, providing strong evidence that the monotile quasilattice exhibits quasicrystalline order.

The diffraction pattern was obtained by Fourier transform of the quasilattice confined within a circular region with a radius of $100a$. To investigate the position dependence, the center of this circular region was shifted in the x direction from the center of the H metatiles as shown in Fig. S5(A)-(C). Figures S5(A), (B), and (C) correspond to the calculated diffraction patterns with the shift of $0a$, $100a$, and $200a$, respectively. It is observed that as the shift becomes larger, the diffraction intensity gradually decreases, whereas the positions of the diffraction peaks do not change.

Figures S5 (D)-(F) show the experimentally obtained diffraction images from the H_6 metatiles. From S5(D) to (F), the illumination position was shifted horizontally by 0.5 mm increments from the center of the metatiles. The spot size width of an incident laser is about 1 mm. Consistent with the numerical results, the diffraction intensity decreases with increasing shift, whereas the positions of the diffraction peaks and the overall symmetry remain unchanged. These numerical and experimental results indicate that the rotational symmetry of the monotile quasilattice is preserved not only around specific central points but also over a global region, suggesting its quasicrystalline order.

S3 Comparison of diffraction peaks between experiments and calculations

Figure S6 compares the diffraction peaks obtained experimentally and numerically. Figures S6(C) and (D) show the experimental and calculated diffraction intensities along the lines indicated in Figs. S6(A) and (B), respectively. Figures S6(C) and (D) are displayed on a logarithmic scale, and the positions of the corresponding peaks are indicated by red dashed lines. As indicated by the dashed lines, the experimental and calculated k -space peak positions, as well as the relative magnitudes of the peak intensities, show excellent agreement.

S4 Additional data of diffraction calculations

S4.1 Additional data of calculated diffraction patterns

Figure S7(B), (C), and (D) show the diffraction patterns of the H_6 monotile quasilattice, its complementary quasilattice, and combined lattice (honeycomb lattice), respectively. As shown in Fig. S7(A), the monotile quasilattice can be considered a honeycomb lattice with many defects. We call the quasilattice generated by the defect points the complementary quasilattice hereafter. The combined point set of the monotile quasilattice and its complementary quasilattice is the same as the honeycomb lattice. This means that the diffraction amplitude (Fourier coefficients) of the honeycomb lattice is given by the sum of the diffraction amplitudes of the monotile quasilattice and its complementary quasilattice. In the diffraction pattern of the honeycomb lattice (Fig. S7(D)), Bragg peaks appear only at wave vectors exhibiting C_6 symmetry. Consequently, the diffraction pattern of the complementary quasilattice (Fig. S7(C)) has a shape similar to that of the monotile quasilattice (Fig. S7(B)). In the asterisk-shaped pattern, the diffraction amplitudes of the monotile quasilattice and the complementary quasilattice have opposite signs, leading to their cancellation. As a result, these features disappear in the diffraction pattern of the honeycomb lattice. Fig. S7(E) presents the diffraction pattern over an extended wavevector space, showing periodic repetitions of the diffraction patterns [4]. This periodicity reflects the fact that the monotile quasilattice possesses C_3 symmetry, which is allowed in conventional crystals, distinguishing it from conventional quasicrystals.

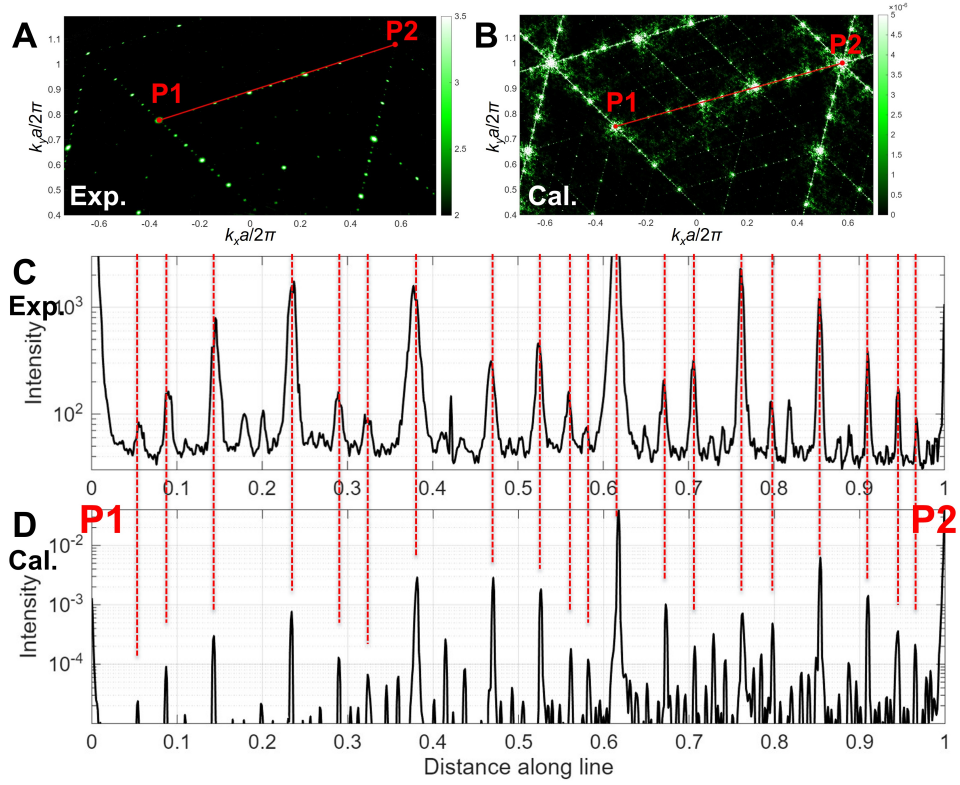


Fig. S6 Comparison of diffraction peaks between experiments and calculations. (A), (B) Lines from P1 to P2 used to extract the diffraction peaks from the experimental and calculated diffraction patterns, respectively. Panel (A) is shown on a logarithmic scale. The diffraction intensities along these lines are presented in panels (C) and (D), respectively. The red dashed lines indicate peaks that coincide between the experiment and the calculation.

S4.2 Meandering of diffraction peaks

Figure S8 presents magnified views of calculated diffraction patterns to see meandering of weak diffraction peaks. Figures S8(A), (B), and (C) correspond to the calculated diffraction patterns for (A) H_6 , (B) H_5 , and (C) H_4 metatiles, respectively. Relatively strong peaks indicated by light-green arrows are aligned along the white lines tilted by θ_{chiral} . In contrast, it can be observed that the relatively weak peaks between the strong ones deviate slightly from the white line, exhibiting a meandering alignment. This feature can be observed for other generations of H_n metatiles.

S4.3 Self-similar structure in diffraction patterns

We found self-similar structures in the diffraction patterns of the monotile quasitile. Figure S9 shows the magnified views of the asterisk shapes in the calculated diffraction patterns. Both axes are scaled with ϕ^{-1} , ϕ^0 , and ϕ^1 in Figs. S9(A), (B), and (C), respectively. As multiplying the golden mean ϕ , the positions of the strong

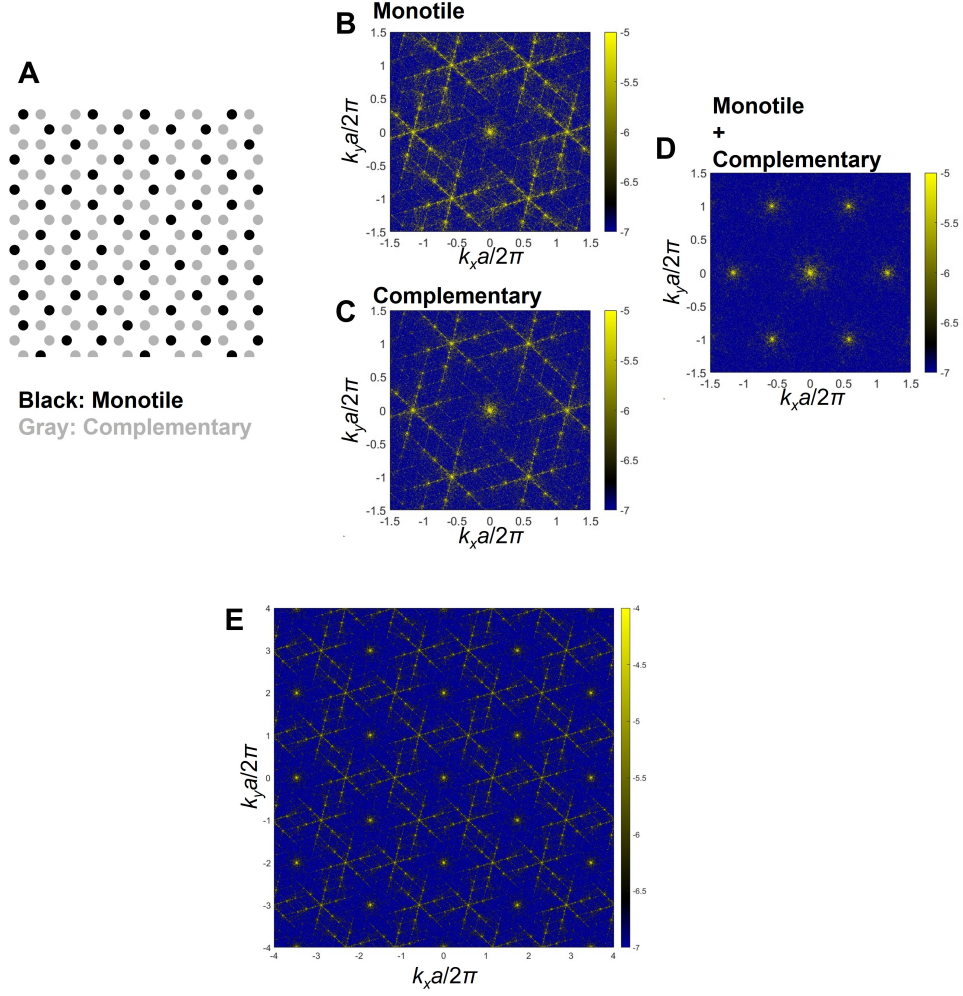


Fig. S7 Calculated diffraction patterns in logarithmic scale. (A) The monotile quasilattice and its complementary quasilattice. These two quasilattices form a honeycomb lattice. Calculated diffraction patterns from the H_6 metatile for the cases of (B) the monotile quasilattice, (C) its complementary quasilattice, and (D) honeycomb lattice. (E) Diffraction in wide wavenumber range.

diffraction peaks are unchanged. This kind of self-similar structure can be found in the diffraction patterns from a one-dimensional Fibonacci lattice and two-dimensional Penrose structure, which are characterized by the golden mean. The key difference between these two lattices and the monotile quasilattice is that the self-similar structure is observed not around the center of reciprocal space, but around the center of the asterisk pattern.

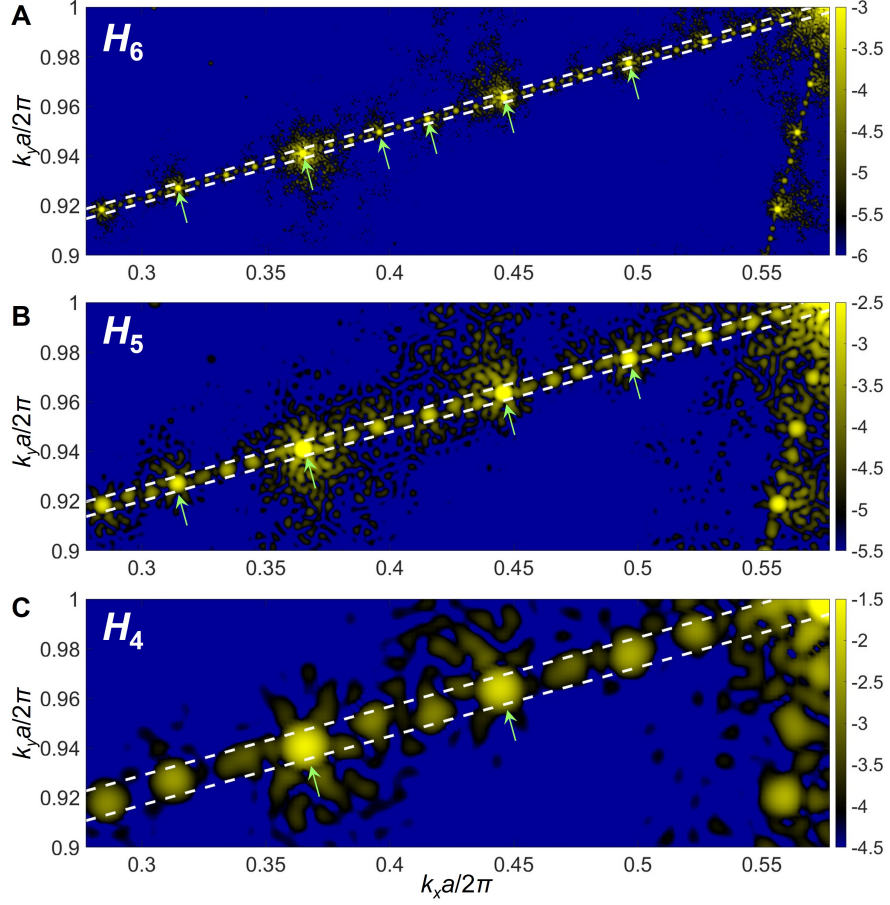


Fig. S8 Meandering of relatively weak peaks in diffraction patterns. Calculated diffraction patterns for the (A) H_6 , (B) H_5 , and (C) H_4 metatiles. Relatively strong peaks are indicated by light-green arrows.

S5 Additional data of polarization measurements

The diffraction pattern from the monotile quasilattice exhibits linear polarization dependence. Figures S10(A) and S10(B) show the calculated degree of linear polarization defined by

$$\frac{I_x - I_y}{I_x + I_y}, \quad (2)$$

and

$$\frac{I_{+45} - I_{-45}}{I_{+45} + I_{-45}}, \quad (3)$$

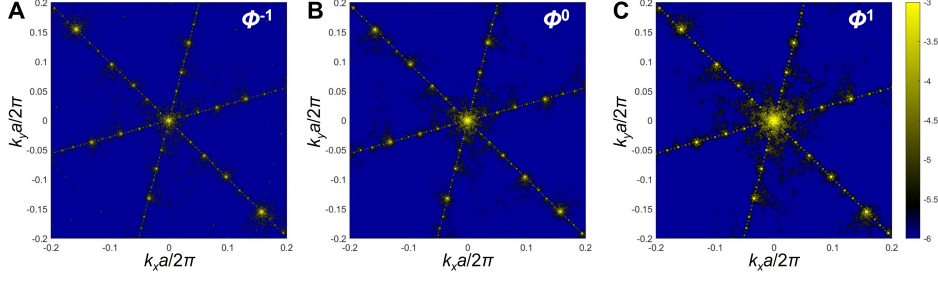


Fig. S9 Self-similar structures of the asterisk shape in diffraction patterns. Magnified view of the asterisk patterns in the calculated diffraction. Here, the origin of the graph is moved to the center of the asterisk. Both axes are scaled with (A) ϕ^{-1} , (B) ϕ^0 , and (C) ϕ^1 .

respectively. Diffraction in the vertical direction is slightly stronger for x -polarized light, while diffraction in the horizontal direction is slightly stronger for y -polarized light. For $\pm 45^\circ$ polarized light, this relationship rotates by 45° .

Measured results for circular-polarization incidence for the other a are shown in Fig. S11. Overall features of the helicity dependence are unchanged despite a . Namely, η maps for positive and negative η have different shapes in wavenumber space. For mirrored metatile $\bar{H}_6$, the shapes for positive and negative η are exchanged. By comparing the results for different a , one finds that the shape of each η map changes depending on a . This demonstrates that the intriguing circular-polarization dependence of the monotile quasilattice arises not only from the intrinsic properties of the monotile quasilattice itself, but also from the wave vector and other factors.

References

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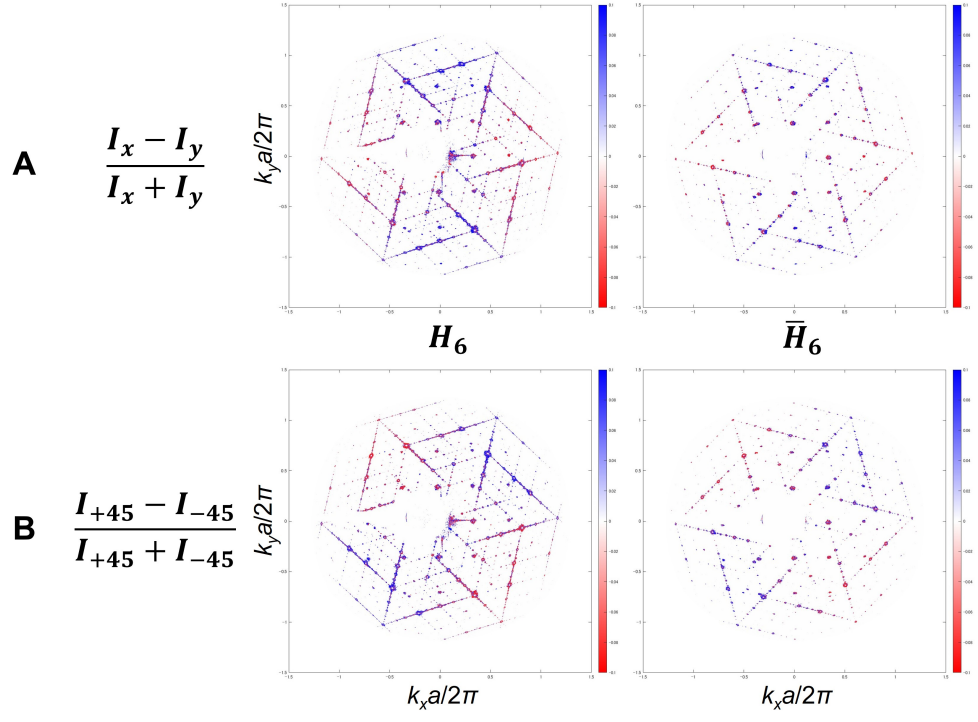


Fig. S10 Linear polarization dependence of diffraction patterns. Degree of polarization for (A) x and y polarizations and (B) $+45^\circ$ and -45° polarizations

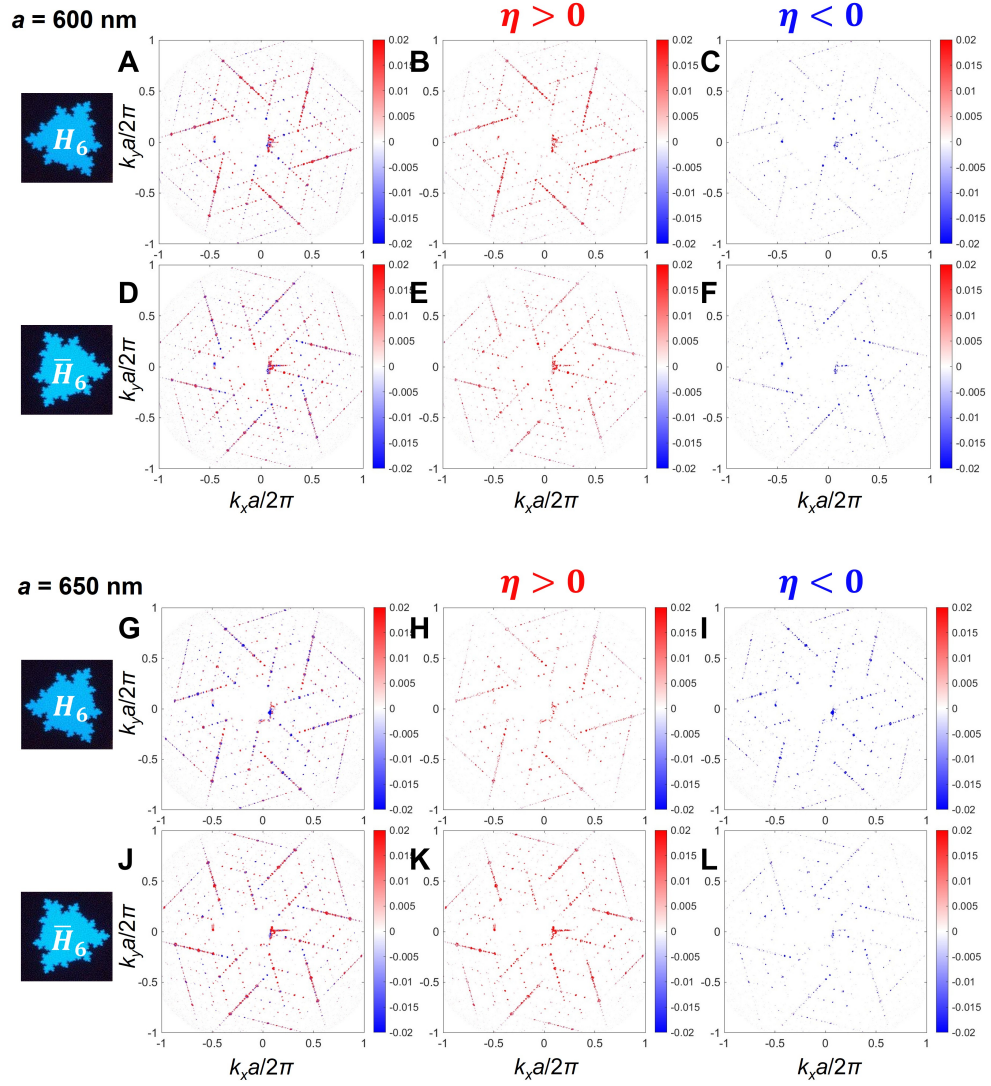


Fig. S11 Helicity dependence of measured diffractions for other a . (A)-(F) η maps in k space for $a = 600$ nm. (A)-(C) H_6 and (D)-(F) $\bar{H}_6$. (B) and (C) are positive and negative parts of η in (A), respectively. (E) and (F) are positive and negative parts of η in (D), respectively. (G)-(L) η maps in k space for $a = 650$ nm. (G)-(I) H_6 and (J)-(L) $\bar{H}_6$. (H) and (I) are positive and negative parts of η in (G), respectively. (K) and (L) are positive and negative parts of η in (J), respectively.