Supporting Information

Creating Two-Dimensional Quasicrystal, Supercell and Moiré Lattices with Laser Interference Lithography: Implications for Photonic Bandgap Materials

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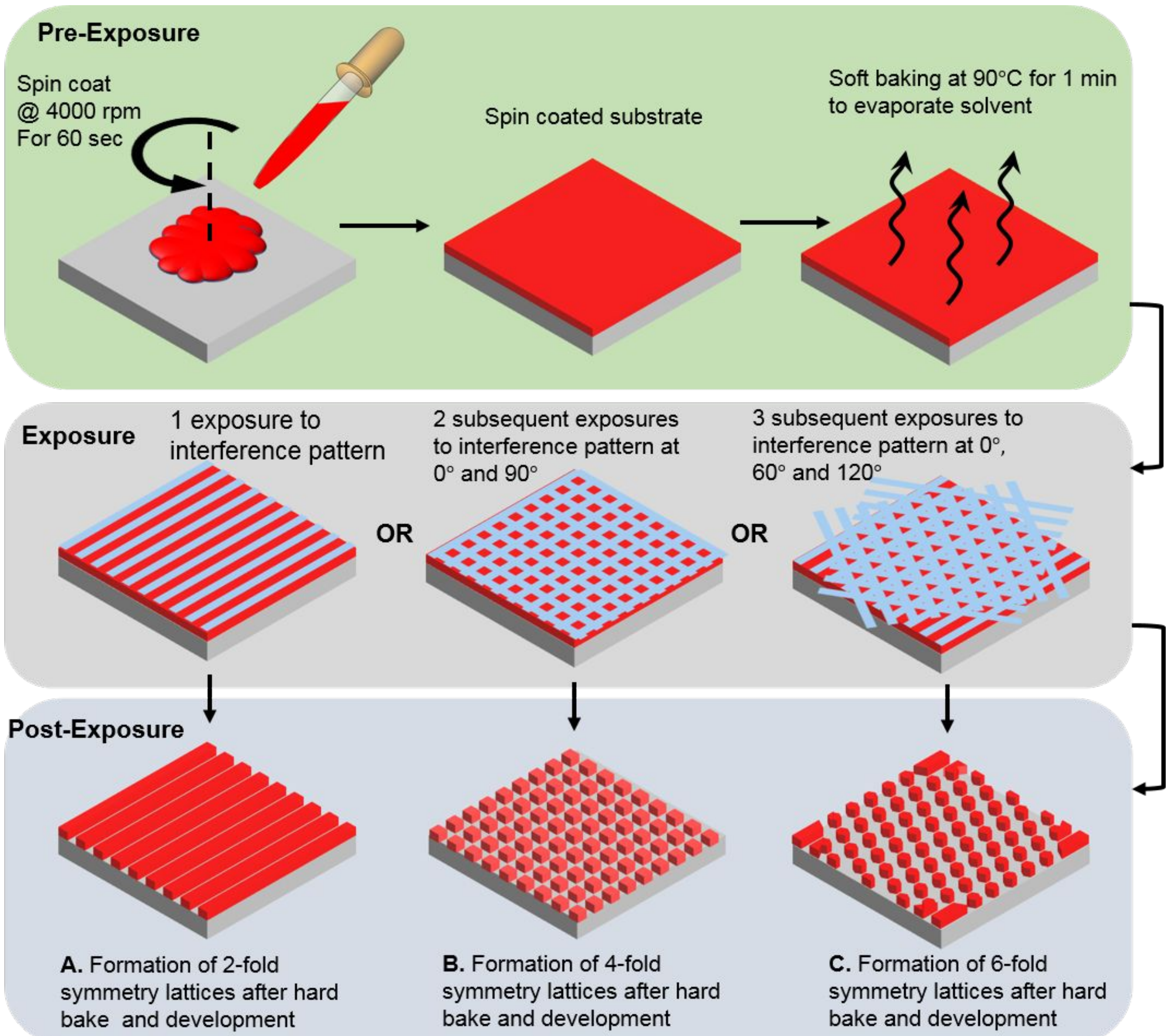

Figure S.1 Process flow of Interference Lithography. Pre-Exposure: applying, spin-coating and soft-baking of photoresist. Exposure: Multiple exposures of photoresist to create interference patterns. Post-exposure: Hard baking of the photoresist and development in developer to dissolve the exposed areas (A) Single exposure of interference pattern produces single pitch gratings, (B) two subsequent exposures at 0°and 90° produce 4-fold symmetry square lattice, and (C) three subsequent exposures at 0°, 60° and 120° produce 6-fold symmetry hexagonal lattice.

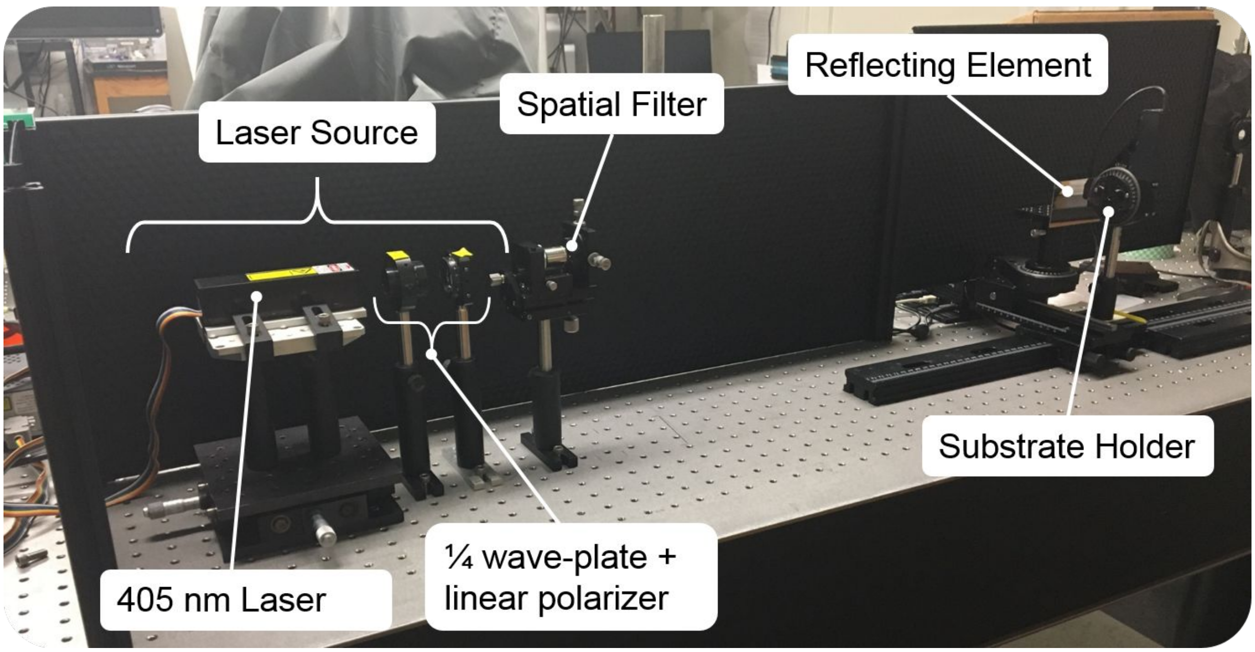

Figure S.2 Laser Interference Lithography set-up. The laser source consists of a 405 nm diode laser source. The light passes through a ¼ wave plate and a linear polarizer. The spatial filter expands the beam into a gaussian profile beam. The beam the goes to the reflecting element and substrate holder arrangement. The substrate is mounted on a rotatable substrate holder to control orientation of the substrate during exposures. The mirror is also mounted on a rotatable holder to control the angle of the mirror with respect to the incoming beam and therefore control the pitch value of the interference pattern.

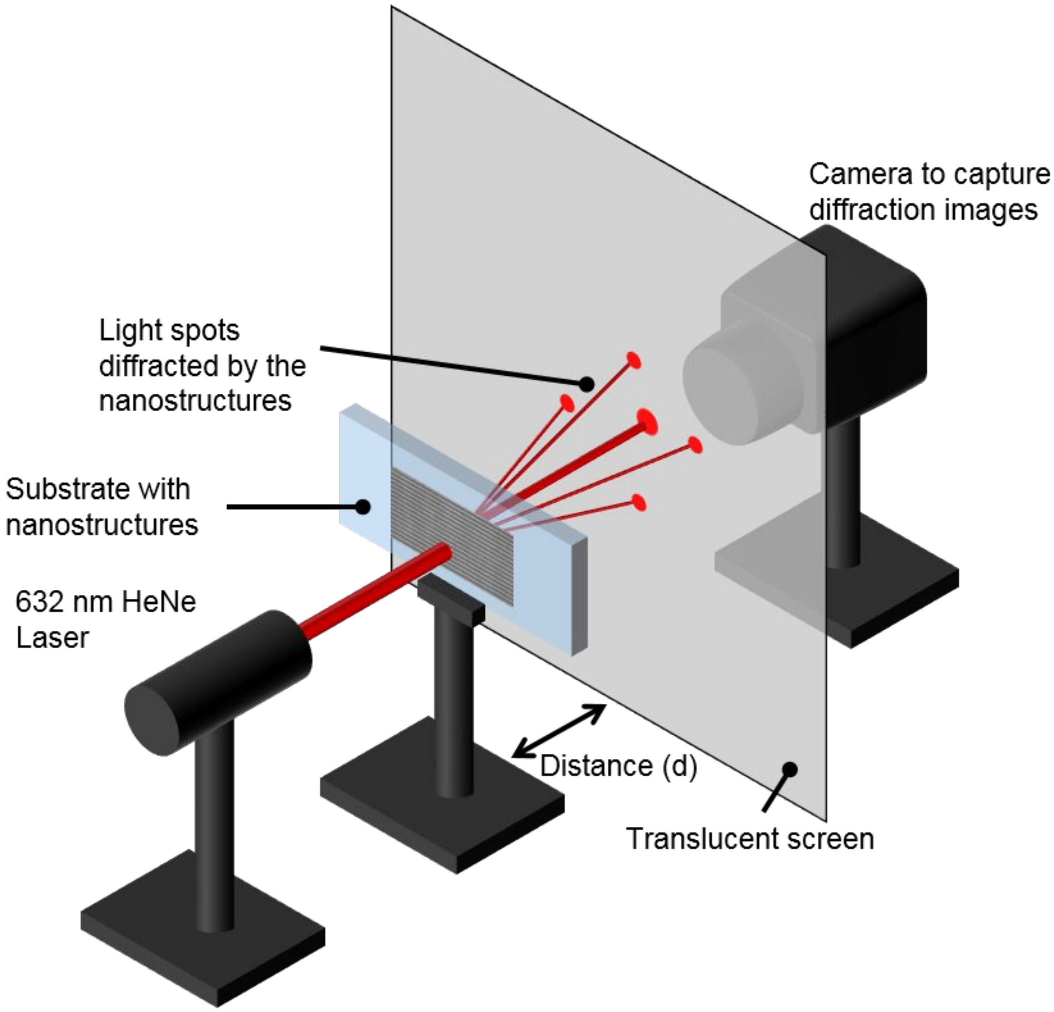

Figure S.3 Experimental setup to record diffraction characteristics of fabricated nanostructures. The setup consists of a 632 nm HeNe laser source. The beam from laser is transmitted perpendicularly through the nanostructures. The diffracted light is projected on a translucent screen made of a thick paper sheet. The images of the diffraction spots are recorded by a camera from the opposite side of the screen.

Intensity Distribution of Single Exposure

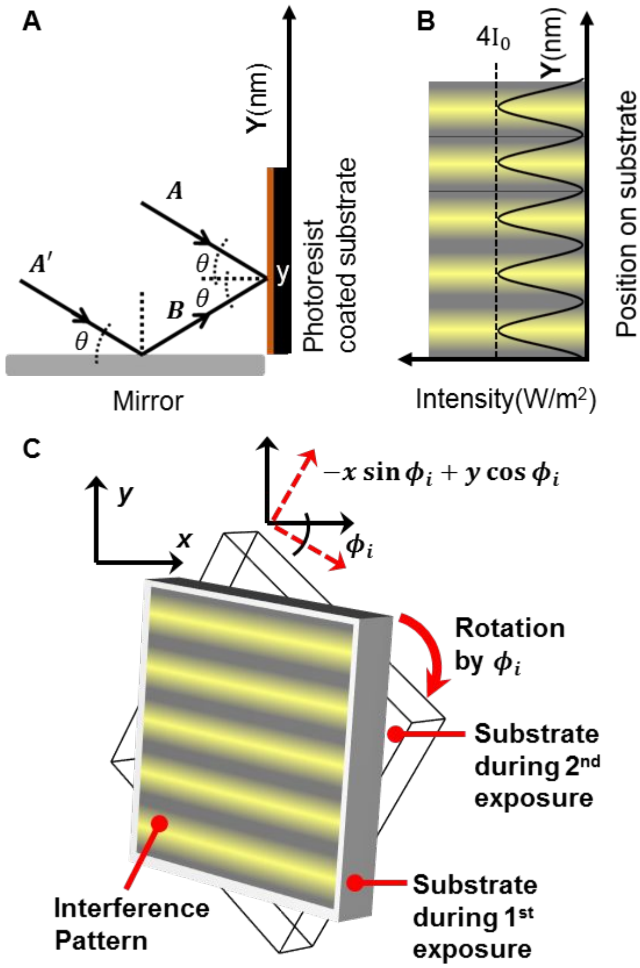

Figure S.4 Diagram to illustrate theory of LIL A. Sideview of incoming beam A and A' being incident on the photoresist and mirror respectively. Beam A' is reflected from the mirror as beam B onto the photoresist surface. B. Intensity distribution of the two interfering beams on the sample assuming that the beams are homogenous and have equal intensities. C. The solid colored substrate shows the orientation of the substrate during first exposure. During second exposure, the substrate is rotated by angle ϕ_i as shown by the outline of the substrate. The rotation changes the coordinate of the intensity distribution on the substrate by $-x$ sin $\phi_i + y\cos\phi_i$.

Lloyd's mirror interferometer consists of a mirror perpendicularly mounted to the sample coated with photoresists as shown in Figure S.1A. The mirror reflects a portion of the beam (represented by A') onto the substrate. On the substrate, the direct beam (A) and the reflected beam (B) interfere creating Bragg interference pattern, which is recorded by the photoresist. The following two equations represent the two time-independent linearly polarized interfering plane waves:

$$\boldsymbol{E_A}(\boldsymbol{r}) = \boldsymbol{E_A}\cos\left(\boldsymbol{k_A}\cdot\boldsymbol{r} + \phi_A\right) \tag{S1.1}$$

$$\boldsymbol{E_B}(\boldsymbol{r}) = \boldsymbol{E_B}\cos\left(\boldsymbol{k_B}\cdot\boldsymbol{r} + \phi_B\right) \tag{S1.2}$$

where $\boldsymbol{E_A}(\boldsymbol{r})$ and $\boldsymbol{E_B}(\boldsymbol{r})$ are the electric fields of interfering waves at the intersection point y of beam A and B. $\boldsymbol{E_A}$ and $\boldsymbol{E_A}$ are the amplitudes of the two waves. $\boldsymbol{k_A}$ and $\boldsymbol{k_B}$ are wave vectors whose directions represent the direction of propagation of the waves and their magnitudes determines wavenumber of the waves. The magnitude of k is given by $k = \frac{2\pi}{\lambda}$. $\boldsymbol{r}$ is the position vector. ϕ_A and ϕ_B are the initial phase angles of the two waves. At point y, where the two propagating waves A and B meet, we can apply the principle of superposition to find the total electric field. Principle of superposition states that at any point in space the total electric field is given by the vector sum of the electric fields of the individual plane waves [18]. Therefore, the total electric field $\boldsymbol{E_T}(\boldsymbol{r})$ at y is given by:

$$\boldsymbol{E_T}(\boldsymbol{r}) = \boldsymbol{E_A}(\boldsymbol{r}) + \boldsymbol{E_B}(\boldsymbol{r}) \tag{S1.3}$$

Or, equivalently,

$$\boldsymbol{E_T}(\boldsymbol{r}) = \boldsymbol{E_A}\cos\left(\boldsymbol{k_A}\cdot\boldsymbol{r} + \phi_A\right) + \boldsymbol{E_B}\cos\left(\boldsymbol{k_B}\cdot\boldsymbol{r} + \phi_B\right) \tag{S1.4}$$

The irradiance or intensity, I, of an electric field is given by the following equation: $I = \frac{1}{2}\sqrt{\frac{\varepsilon}{\mu}}|\boldsymbol{E_T}|^2$, where ε is the dielectric constant (also called electric permittivity) and μ is the magnetic permeability of the material. Since it is common in optics to compare intensities of electromagnetic waves propagating in the same material, we can forgo the common factors in equation 1.5:

$$I = |\boldsymbol{E_T}|^2 \tag{S1.5}$$

By expanding equation 1.5 using equation 1.4, we have

$$I = \left[\boldsymbol{E_A}\cos\left(\boldsymbol{k_A}\cdot\boldsymbol{r} + \phi_A\right) + \boldsymbol{E_B}\cos\left(\boldsymbol{k_B}\cdot\boldsymbol{r} + \phi_B\right)\right]^2 \tag{S1.6}$$

Or, equivalently,

$$I = |\boldsymbol{E_A}|^2 + |\boldsymbol{E_B}|^2 + 2\boldsymbol{E_A}\boldsymbol{E_B}\cos\left((\boldsymbol{k_A} - \boldsymbol{k_B})\cdot\boldsymbol{r} + (\phi_A - \phi_B)\right) \tag{S1.7}$$

$$= I_A + I_B + I_{AB} \tag{S1.8}$$

where, I_A and I_B are the irradiances of the individual waves and I_{AB} is known as the interference term. From the definition of irradiance, I_{AB} can be expressed as $I_{AB} = 2\sqrt{I_A I_B}\cos(\delta)$, where $\delta =$ (

$\boldsymbol{k_A} - \boldsymbol{k_B}) \cdot \boldsymbol{r} + (\phi_A - \phi_B)$. If we assume that the interfering beams have equal amplitude, $I_A = I_B = I_0$, Equation 1.8 simplifies to:

$$I = I_A + I_B + 2\sqrt{I_A I_B}\cos(\delta) = 4I_0\cos^2\left(\frac{\delta}{2}\right) \qquad \text{(S1.9)}$$

The value of δ can be found from Fig. S.5A. Beam A' undergoes reflection at the mirror surface and therefore beam B attains a phase shift of π with respect to beam A. The position vector $\boldsymbol{r}$ is the path difference between beam A and B and is given by $(2y\sin\theta)$. Using these two expressions, equation 1.9 further simplifies to:

$$I = 4I_0\cos^2\left(\frac{\delta}{2}\right) = 4I_0\cos^2\left(\frac{1}{2}\left((\boldsymbol{k_A} - \boldsymbol{k_B}) \cdot \boldsymbol{r} + (\phi_A - \phi_B)\right)\right)$$

$$= 4I_0\cos^2\left(\frac{1}{2}\left(\left(\frac{2\pi}{\lambda}\right) \times (2y\sin\theta) + \pi\right)\right)$$

$$= 4I_0\sin^2\left(\frac{2\pi y}{\lambda}\sin\theta\right) \qquad \text{(S1.10)}$$

Equation 1.10 expresses the spatial intensity distribution of interfering beam A and beam B. A two-dimensional plot of the intensity distribution is shown in Fig. S.5B.

I. Intensity Distribution of i Subsequent Exposures

If the sample is placed on the xy plane and rotated by angle ϕ_i, the y coordinate transforms to $-x\sin\phi_i + y\cos\phi_i$

$$I_i = 4I_0\sin^2\left[\frac{2\pi\sin\theta}{\lambda}(-x\sin\phi_i + y\cos\phi_i)\right]$$

Where, i is the number of exposures, $4I_0$ determines the intensity and can be replaced with a constant β_i which represents the sensitivity of the photoresist during each exposure. For Multi-exposure, the intensity is given by the sum of intensity at each exposure:

$$I_{total} = \sum_i I_i = \sum_i \beta_i \sin^2\left[\frac{2\pi\sin\theta}{\lambda}(-x\sin\phi_i + y\cos\phi_i)\right] \qquad \text{(S1.11)}$$

where, I_{total} is the sum of the intensity distribution on the substrate from all the exposures. Figure S.6 in the text illustrates intensity distribution for single (i = 1), two (i = 2) and three exposures (i = 3) at specific rotation angles.

Matlab code for simulating total intensity distribution after *i* subsequent exposures:

```
clear all;% clears any stored values of variables
x=[-10000:50:10000];%the x-length of the lattice (nm)
y=[-10000:50:10000];%the y-length of the lattice (nm)
I_total=zeros (length(y));%creates an empty array of length y
[xx,yy]=meshgrid(x,y);%creates a 2D mesh of x by y elements

%%%system parameters%%%%%
lambda = 405;%wavelength of the laser beam
theta = pi/180*[7 7 7];% input the incident angle of the incoming beam
            %on the mirror assuming that Lloyd's mirror config %for each exposure. We did 3 exposures here each
at 7 deg incident angle
rotation_angle = [0 60 120];% angle by which the substrate is rotated at
              %each exposure with respect to the original
              %position
n = length(rotation_angle);%finds out number of exposures

%%Loop to sum up the intensities%%%%
for i = 1:n
  t_i = 0.1/(i)^0;%a variable which can be used to account for decreased
  %sensitivity after each exposure. here we considered constant. changing
  %the power of i from 0 to any value will reduce t_i during each loop.
  current_angle = rotation_angle(i);%assigns a
  phi_i = pi/180*current_angle;
  term1 =-xx*sin(phi_i)+yy*cos(phi_i);
  term2 = 2*pi*sin(theta(i))/lambda;
  I_i = t_i*sin(term2*term1).^2;
  I_total = I_i+I_total;

end

%%Outputting the intensity distribution%%%%
figure
alw = 2;    % AxesLineWidth
fsz = 11;      % Fontsize
set(gca, 'FontSize',fsz,'LineWidth',alw);%sets the graph properties
mesh(xx/1000,yy/1000,I_total)% outputting the graph in um, therefore
%divided everything by 1000
ylabel("Y(\mum)");
xlabel("X (\mum)");
zlabel("Intensity (arbitary unit)");
colormap hot;%sets the color properties of the plot. could be changed to
%gray for grayscale. there are other color schemes as well
view(0, 90);%gives a surface view of the 3D plot
```

Simulated intensity distributions can be determined using Equtions 2 and 3. Figure S.6 provides example results for intensity distributions after n number exposures at different angular orientations ϕ, where n was varied from 1 to 4. The angle θ was set at 1°, which produced interference patterns with a pitch value of 11.6 μm. A single exposure contains a sinusoidal intensity profile of constant periodicity in the x-direction (Figure S.2A) and produces a simple grating with 2-fold symmetry. A second exposure with the substrate rotated by $\phi = 90°$ with respect to the first creates a total intensity distribution with 4-fold symmetry and a square lattice (Figure S.2B). For the example shown in Figure S.2C, second and third exposures were performed at 60° and 120° with respect to the first exposure, which resulted in a total intensity distribution with 6-fold symmetry akin to a hexagonal lattice.

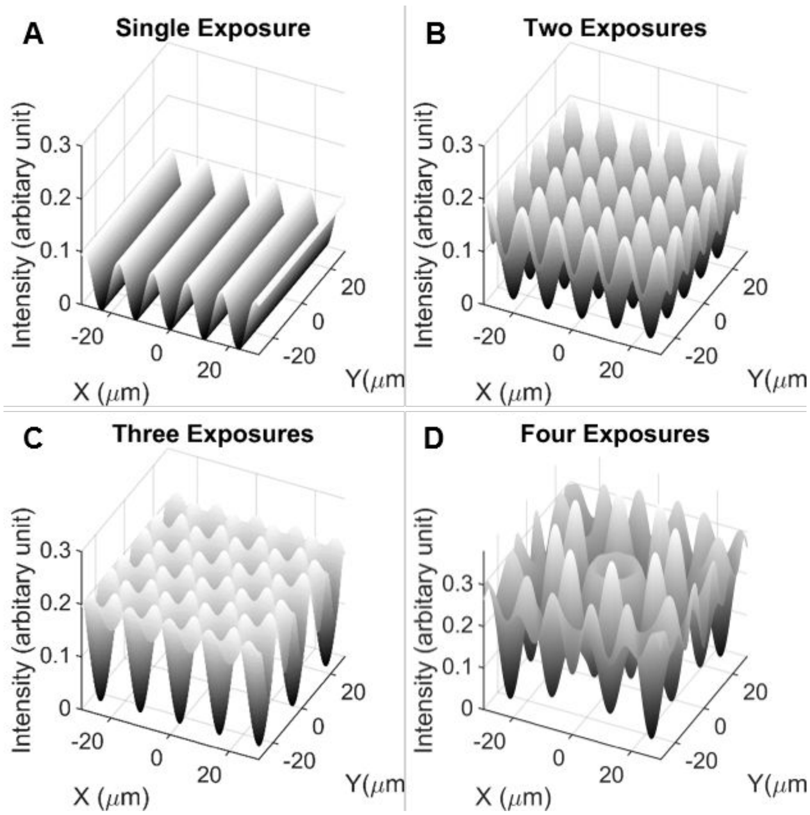

Figure S.5 Intensity distribution of representative n-number of exposures using Interference Lithography. (A) Single exposure intensity distribution of a traditional LIL technique for fabrication of 2-fold symmetry gratings, (B) two subsequent exposures at 0° and 90° produced 4-fold symmetry square lattice intensity profiled, (C) three subsequent exposures at 0°, 60° and 120° produced 6-fold symmetry hexagonal lattice intensity profile, and (D) four subsequent exposures at 0°, 45°, 90° and 135° producing 8-fold symmetry quasicrystal lattice intensity profile.

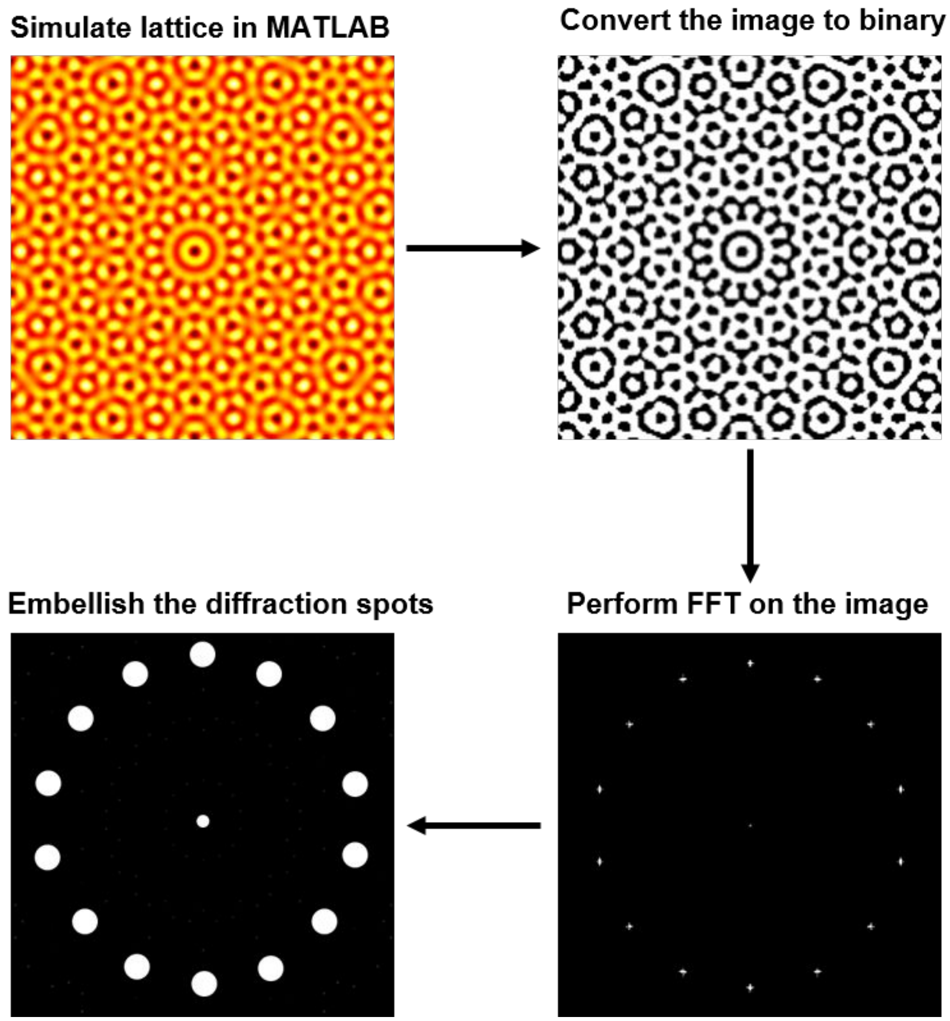

Figure S.6 Simulating diffraction characteristics of the simulated lattice structures. The images from MATLAB were converted into binary images. FFT was performed on the binary images to find the diffraction spots. The diffraction spots were embellished to make them more conspicuous.

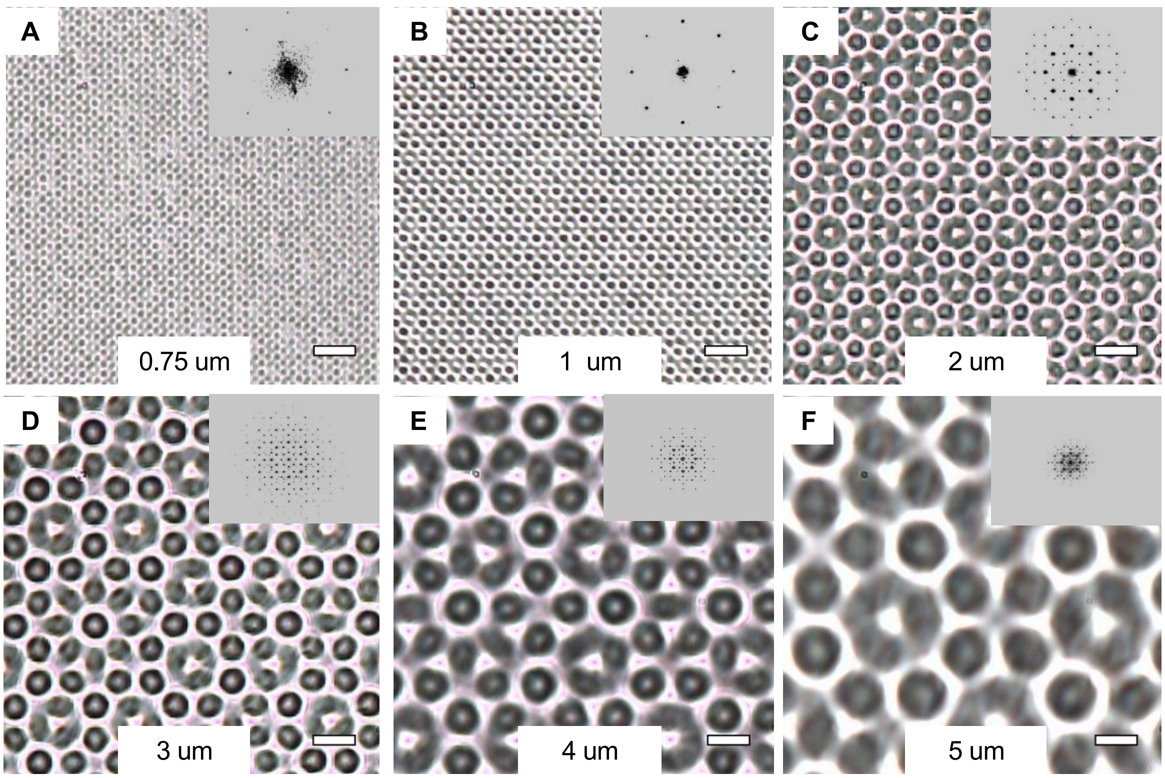

Figure S.7 Optical microscope and optical diffraction (insets) images of 8-fold lattice with different pitch values ranging from 750 nm up to 5 microns.

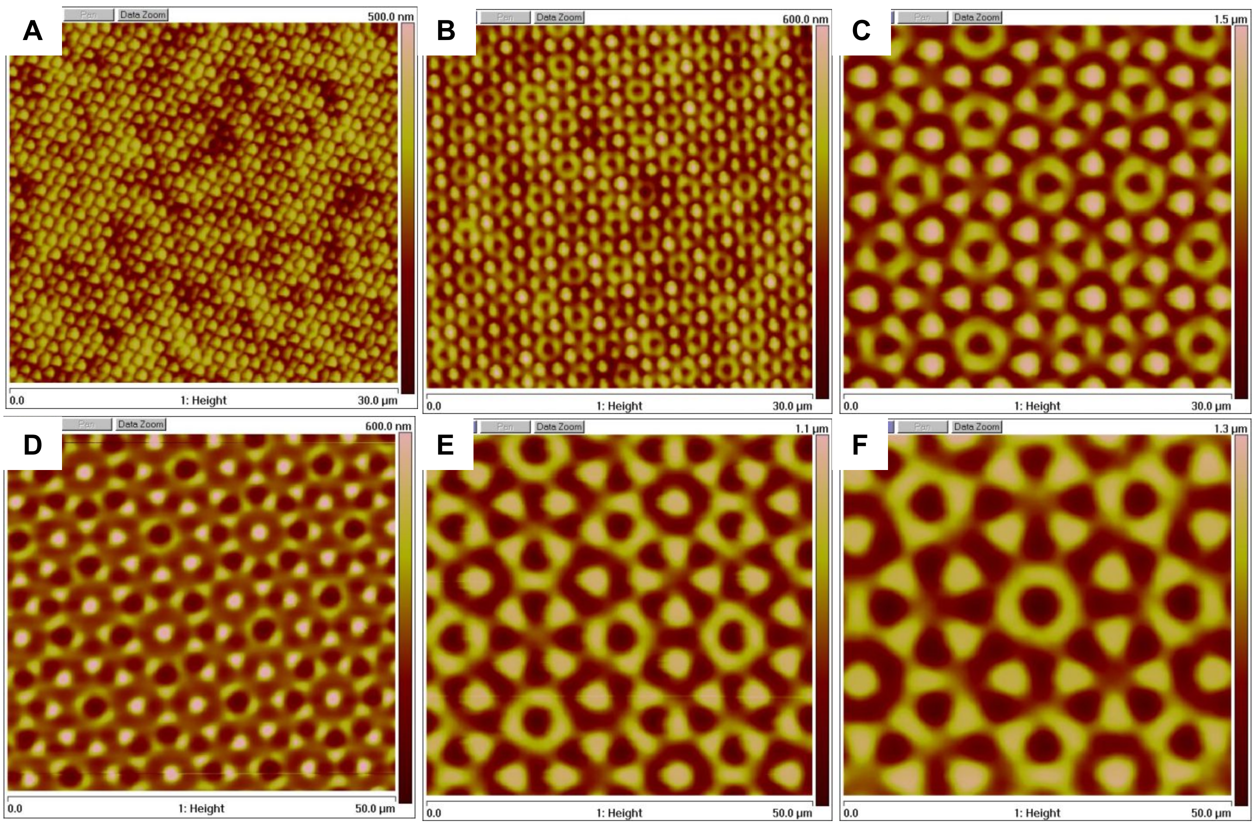

Figure S.8 Atomic force microscope (AFM) images of 8-fold lattices with different pitch values: A) 750 nm, B) 1 µm, C) 2 µm, D) 3 µm, E) 4 µm, F) 5 µm. Scale bars are noted in images.

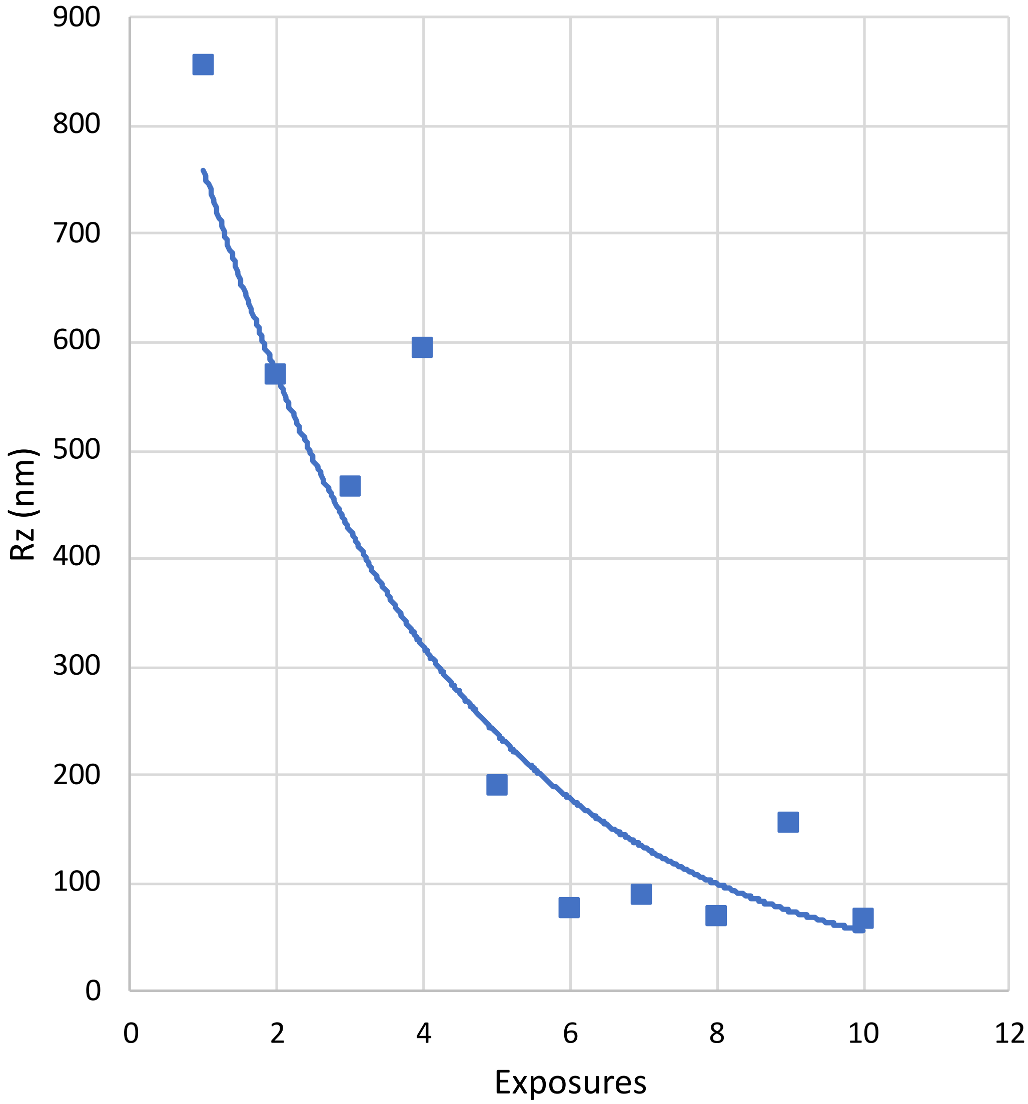

Figure S.9 Average maximum amplitude (Rz) as measured by atomic force microscopy for quasicrystal lattice structures fabricated by multiple rotational exposures from 1 up to 10 exposures (2 up to 20-fold quasilattices).

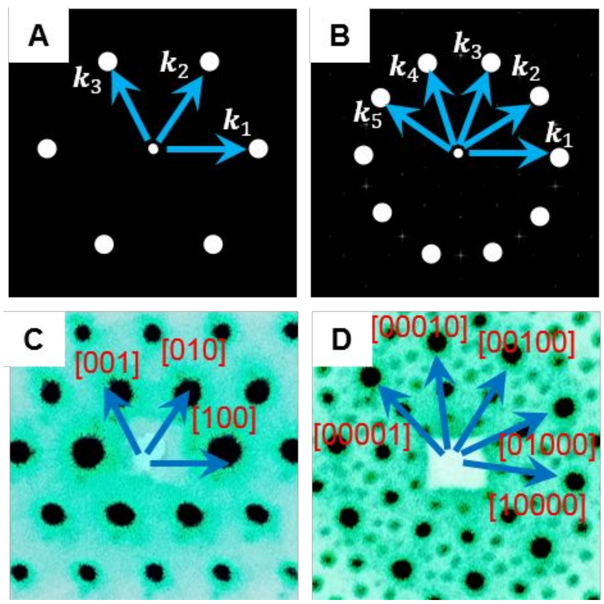

Figure S.10 (A) Fourier transform of a hexagonal lattice structure. The diffraction spots are labelled with basis vectors $\boldsymbol{k}_1$, $\boldsymbol{k}_2$ and $\boldsymbol{k}_3$; (B) Fourier transform of a 10-fold symmetry quasilattice structure. The diffraction spots are labelled with basis vectors $\boldsymbol{k}_1$, $\boldsymbol{k}_2$, $\boldsymbol{k}_3$, $\boldsymbol{k}_4$ and $\boldsymbol{k}_5$; (C) Diffraction image of a hexagonal lattice with indexed reciprocal basis vectors shown in A; (D) Diffraction image of a 10-fold symmetry quasilattice with indexed reciprocal basis vectors shown in B.

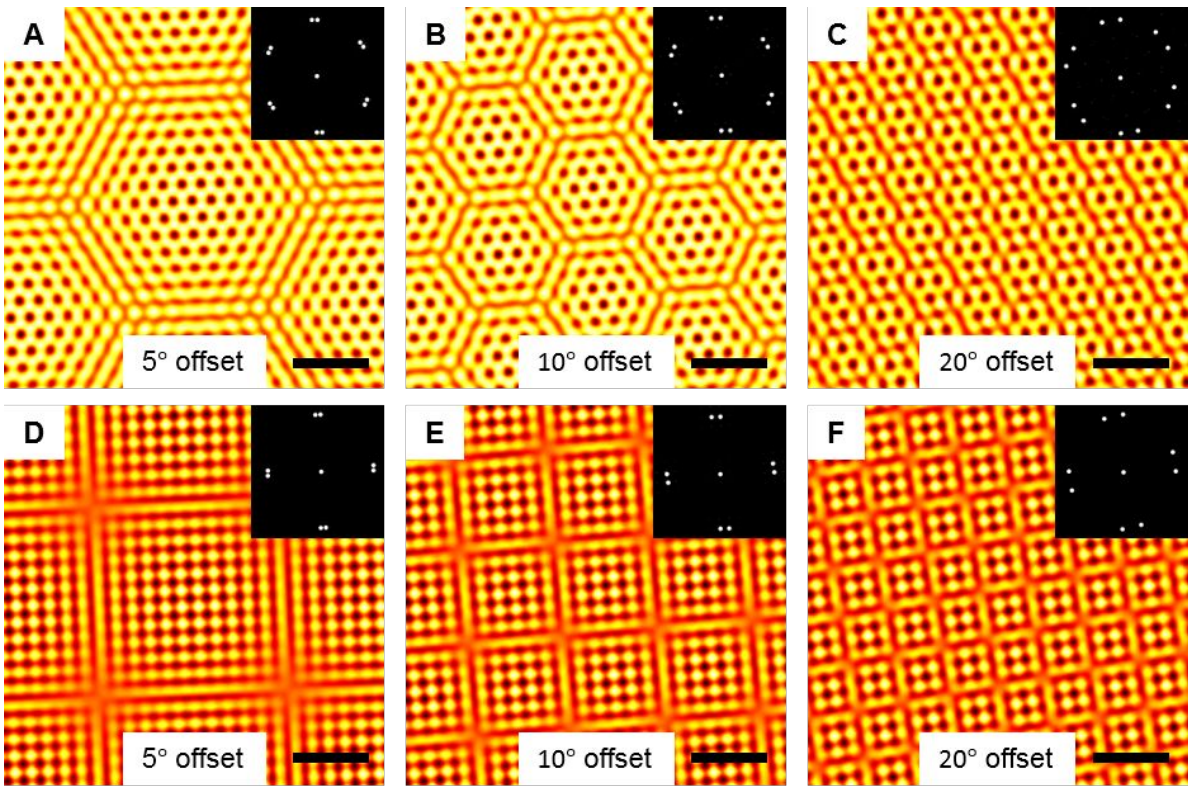

Figure S.11 Simulated Moiré patterns. Overlapped hexagonal lattices (A) with 5° offset, (B) with 10° offset, (C) with 20° offset. Overlapped square lattices (D) with5° offset, (E) with 10° offset, (F) with 20°offset. Scale bar: 10 μm.

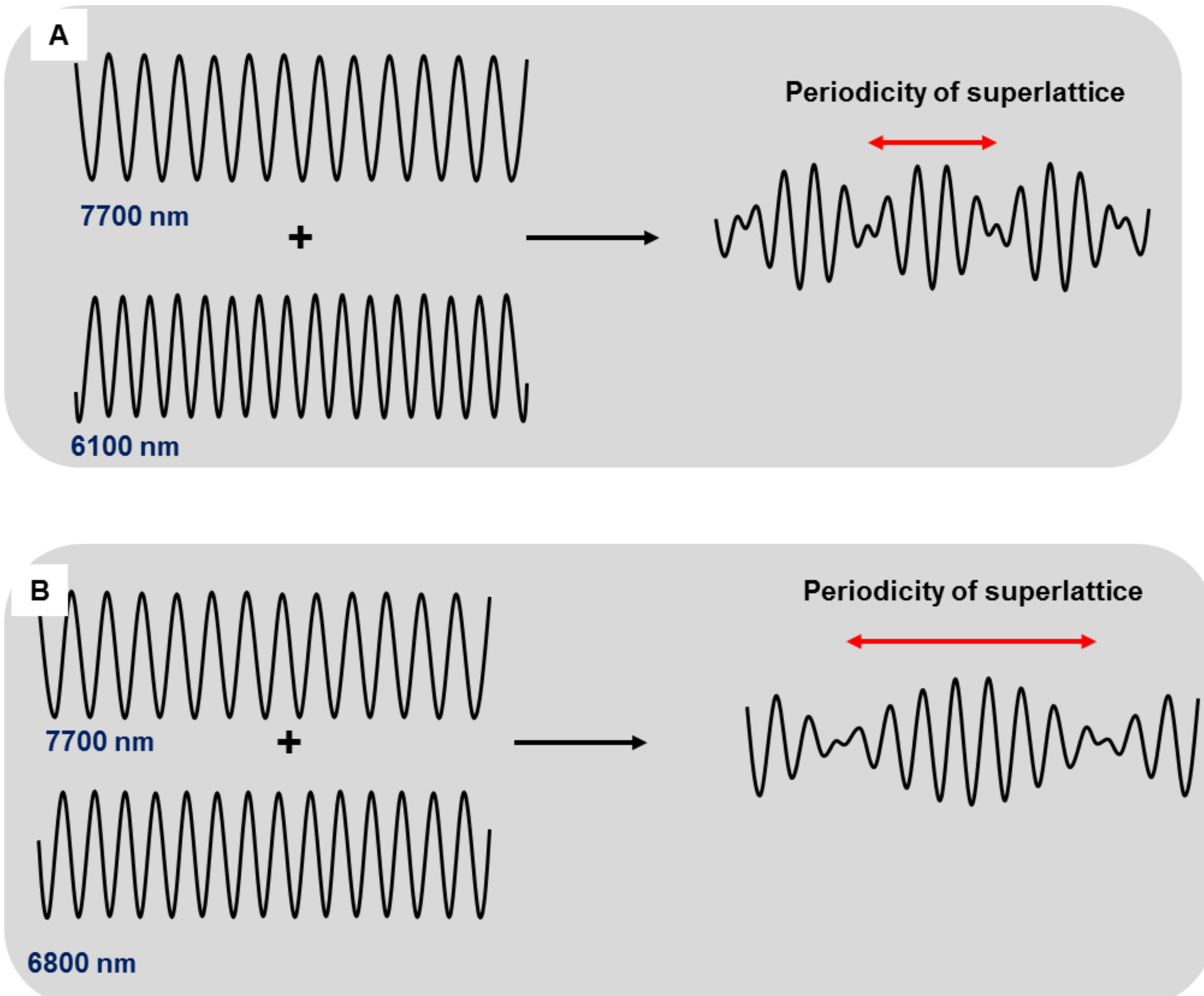

Figure S.12 Schematic illustrating the concept of superlattice formation due to mismatch of pitch values. (A) Superposition of two waves with large difference in their pitch values to produce superlattice structure with short superperiodicity. (B) Superposition of two waves with small difference in their pitch values to produce superlattice structure with long superperiodicity. The superperiodicities are indicated by the red double headed arrows.

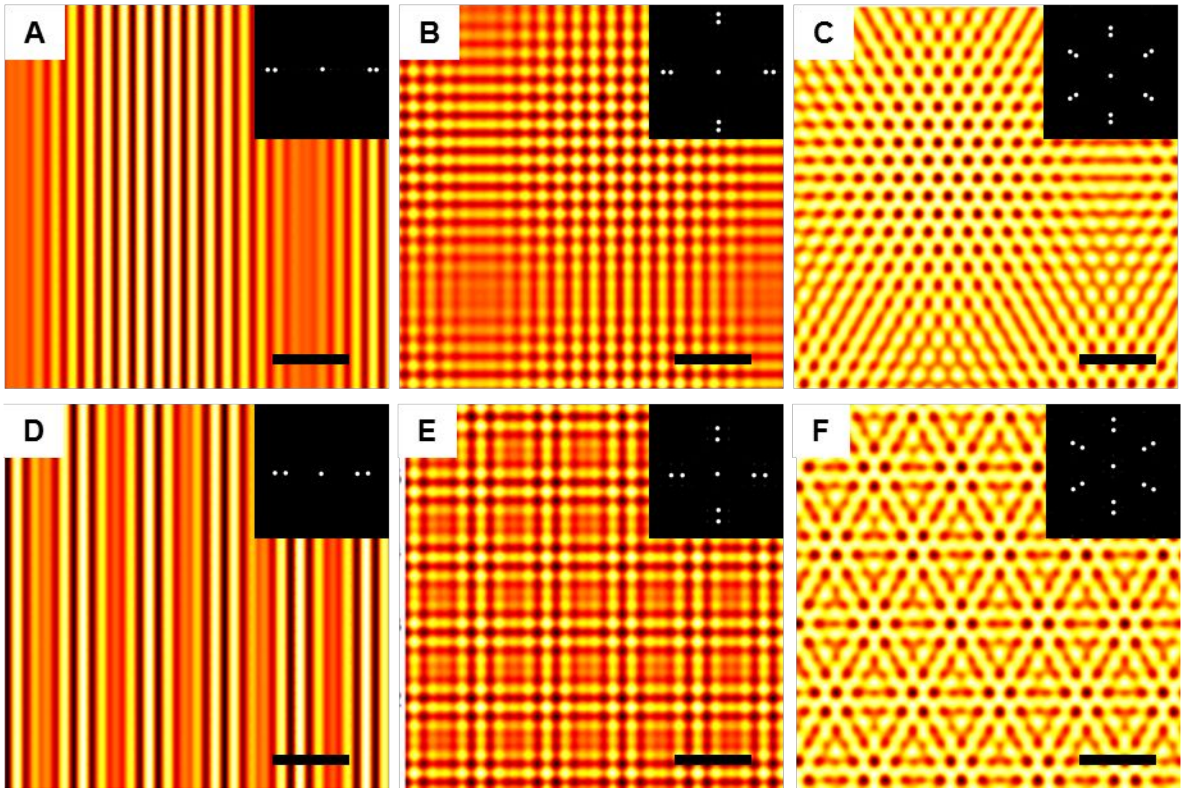

Figure S.13 Simulated superlattices. (A) 1D biperiodic patterns with small difference in pitch values. (B) Square lattice made from the biperiodic patterns in (A). (C) Hexagonal lattice made from the biperiodic patterns in (A). (D) 1D biperiodic patterns with large difference in pitch values. (E) Square lattice made from the biperiodic patterns in (D). (F) Hexagonal lattice made from the biperiodic patterns in (D). Scale bar: 10 μm.