Twisted Polaritonic Crystals in Thin van der Waals Slabs

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1. Introduction

In the last decade, polaritons in 2D and van der Waals (vdW) materials\textsuperscript{[1,2]} have emerged as a promising means to manipulate light at the nanoscale.\textsuperscript{[3]} In particular, some natural uniaxial and biaxial polar crystals (such as, for example, h-BN,\textsuperscript{[4]} MoO\textsubscript{3},\textsuperscript{[5–7]} V\textsubscript{2}O\textsubscript{5},\textsuperscript{[8]} or calcite\textsuperscript{[9]}) support phonon polaritons – lattice vibrations coupled to electromagnetic fields – (PhPs) with directional out-of-plane or in-plane propagation. In-plane anisotropic PhPs are particularly attractive because they manifest intriguing optical phenomena, such as light canalization,\textsuperscript{[10,11]} topological transitions\textsuperscript{[12]} or negative refraction\textsuperscript{[13]}, among others, which are directly accessible in real space by near-field nanoimaging techniques.

PCs based on anisotropic vdW materials (such as h-BN) have shown interesting possibilities for enhancing light–matter interactions\textsuperscript{[14–16]} and topological photonics\textsuperscript{[17]}. On the other hand, PCs in in-plane anisotropic materials, where polariton manipulation can exhibit more degrees of freedom, have not been addressed until now.

Here we introduce the concept of PCs created in biaxial crystal slabs (supporting in-plane anisotropic polaritons), with the lattice vectors twisted with respect to the optical axes of the crystal. We demonstrate that the polaritonic Bragg resonances arising in these PCs can be efficiently tuned, offering a new way to control light at deep subwavelength scales. We develop a simple theoretical approach to treat the diffraction of light by these twisted PCs and demonstrate its validity for realistic structures (e.g., hole arrays) by comparing analytical results with full-wave electromagnetic simulations.

2. Concept and General Approach

We first briefly outline the theoretical technique we have developed in this work. It has some important approximations, which allow us to significantly simplify the mathematical treatment of diffraction by an arbitrary lattice made in a thin biaxial crystal slab. Specifically, let us consider a double-periodic structure such as, for example, repetition of holes with periods \( L_1 \) and \( L_2 \) along two perpendicular directions, made in a thin biaxial slab (Figure 1a). The slab is sandwiched between two semi-infinite dielectric media with dielectric permittivities \( \varepsilon_1 \) and \( \varepsilon_2 \). For sufficiently thin slabs (with the thickness much smaller than the polariton wavelength), we can avoid considering the fields inside the slab, thus replacing it in the theoretical formalism by a 2D in-plane anisotropic conducting sheet placed in the \( xy \)-plane (see Section S1 in the Supporting Information) with an effective 2D conductivity tensor \( \sigma = \frac{\varepsilon e^2}{2 i \omega} \) (for convenience, we will...
use hereafter its normalized value, \( \alpha = \frac{\varepsilon}{\varepsilon_0} \). In this formalism, it is sufficient to properly match the fields in the effective conducting sheet by means of the boundary conditions, considering the continuity (discontinuity) of the in-plane electric (magnetic) fields. The introduction of the effective 2D conductivity tensor is valid for the description of the lowest electromagnetic waveguide mode in the slab (the one with the longest wavelength). Then, in general, we assume that the normalized effective conductivity tensor of the slab is a periodic function of the coordinates, \( \alpha(r) = \alpha(r + n_1L_1 + n_2L_2) \), being \( n_1, n_2 \) integers, so that it can be expanded in the Fourier series

\[
\alpha(r) = \sum_N \alpha_N e^{iG_N r} = \sum_N \left( \frac{a^x_N}{a^y_N} \alpha^z_N \right) e^{iG_N r}
\]

(1)

where the capital letter \( N \) is a compact multi-index signifying both indices \( n_1 \) and \( n_2 \) \((N \equiv \{n_1, n_2\})\) and \( G_N \) is a general reciprocal vector \( G_N = n_1G_1 + n_2G_2 \). For compactness reasons, we will use Dirac notations, in which the in-plane components of the s- and p-polarization basis vectors for each multi-index \( N \) read as

\[
|sN\rangle = \frac{1}{k_{IN}} \begin{bmatrix} -k_{pN} \\ k_{sN} \end{bmatrix} e^{i(\omega t + \beta_{sN} z)}
\]

(2)

where \( k_{sN} = (k_{sN}, k_{IN}) \) are the in-plane momenta, and \( k_{IN} \) represents their norm. Next, the in-plane components of the electric fields in the superstrate (medium 1, \( z > 0 \)) and in the substrate (medium 2, \( z < 0 \)) can be represented as a Fourier–Floquet series constituting the superposition of plane waves of both s- and p-polarizations

\[
E_{sN}(z) = (L_s |s0\rangle + L_p |p0\rangle) e^{-i\beta_{sN} z} + \sum_N \left( R_{sN} |sN\rangle + R_{pN} |pN\rangle \right) e^{-i\beta_{sN} z} z \geq 0 \tag{3}
\]

\[
E_{pN}(z) = \sum_N \left( T_{sN} |sN\rangle + T_{pN} |pN\rangle \right) e^{i\beta_{sN} z} z < 0 \tag{4}
\]

where \( I_{sN} \) represents the amplitudes of the incident field and \( R_{sN}(T_{sN}) \) represents the amplitudes of the reflected (transmitted) plane waves. The out-of-plane momentum is given by \( k_{zN} = \sqrt{\varepsilon_2 k_0^2 - k_{IN}^2} \) where \( j = \{1, 2\} \) marks the medium, and \( k_{\text{inc}} = \sqrt{\varepsilon_1 k_0^2 - k_{\text{inc}}^2} \).

From Equations (3) and (4) we can find similar expressions for the magnetic fields, with the help of Maxwell’s equations (see Section S1 in the Supporting Information). Using the following boundary conditions

\[
E_{sN}(z = 0) = E_{pN}(z = 0) = 0 \tag{5}
\]

\[
e_{\times} \left( \mathbf{H}_{sN}(z = 0) - \mathbf{H}_{pN}(z = 0) \right) = 2\alpha E_{sN}(z = 0) \tag{6}
\]

and after some straightforward algebra (see Section S1 in the Supporting Information), we arrive at a linear system of algebraic equations for the amplitudes of the scattered plane waves, which can be compactly written as

\[
\sum_{\alpha} D^{\alpha}_{s\beta} \left( Y^s_{\alpha N} T_{\beta N} + Y^p_{\alpha N} R_{\beta N} \right) \delta_{\alpha\beta} \delta_{KN} = 2 I_{pN} Y^{p\alpha}_{\beta N}, \tag{7}
\]

\[
D^{\alpha}_{s \beta} = \left( Y^s_{\alpha N} + Y^p_{\alpha N} \right) \delta_{\alpha\beta} \delta_{KN} + 2 M^{\alpha}_{sN} \tag{8}
\]

where for each polarization we defined \( Y^s_{\alpha N} = k_{\text{inc}}/k_{sN} \) and \( Y^p_{\alpha N} = \varepsilon_{\text{inc}}/k_{pN} \), and \( Y^{p\alpha}_{\beta N} \) takes the values \( Y^{p\alpha}_{\alpha N} = k_{\text{inc}}/k_{sN} \) and \( Y^{p\alpha}_{\beta N} = \varepsilon_{\text{inc}}/k_{pN} \), for s- and p-polarization, respectively. The matrix elements \( M_{sN}^{\alpha\beta} \) are composed of the products between the conductivity tensor and the in-plane momentum

\[
M^{\alpha\beta}_{sN} = \frac{1}{k_{IN} k_{sN}} \left[ \left( -1 \right)^{1+\beta} \varepsilon^x_{\alpha N} k_{IN}^x k_{\beta N}^x + \varepsilon^y_{\alpha N} k_{IN}^y k_{\beta N}^y \right]
\]

(9)

where \( \gamma \) holds for \( x \) or \( y \) when \( \beta \) takes \( s \) or \( p \) values, respectively, while conversely, \( \gamma \) holds for \( x \) or \( y \) when \( \beta \) takes \( p \) or \( s \) values, respectively. Note that \( \gamma \) is a function of \( \beta \), while \( \gamma' \) depends on \( \beta' \). By truncating the infinite system of Equation (7) to a maximum
order, $N_{\text{max}}$ (large enough to achieve convergence, see Section S3 in the Supporting Information), one can directly invert the matrix $D$ by standard numerical procedures and obtain all the unknown amplitudes of the diffracted waves. The reflection and transmission coefficients can then be calculated, as well as the field distribution above the structure. Furthermore, as we demonstrate below, the system of equations in Equation (7) can also be solved analytically in the tight-binding approximation. Regardless of the solution method, it is important to note that, as follows from Equation (9), the nondiagonal elements of the matrix $D$, which describe the interaction between the diffracted plane waves, are proportional to the Fourier harmonics of the normalized conductivity. Therefore, the Fourier decomposition of the periodic structure plays a crucial role in the diffraction of an incident plane wave and the resonant excitation of polaritonic modes.

Figure 1a shows an example of a square hole array (HA), with its Fourier coefficients depicted in Figure 1b. The lattice vector $\mathbf{L}$ forms an angle $\phi$ (twist angle) with respect to the [100] in-plane crystallographic axis. The HA can be described by the space-dependent normalized conductivity, $\alpha(r) = \alpha(1 - f(r))$, where the step function $f(r)$ takes a value of 1 (0) inside (outside) the holes with radius $a$, respectively. As is typical for such periodic functions, the Fourier transform (FT) coefficient $\alpha_{\mathbf{k}}$ takes the highest value, while the amplitudes of the other Fourier harmonics decrease with $N$. Without loss of generality and for illustrative purposes, we will focus in this work on the excitation of polaritons in a periodically structured $\alpha$-MoO$_3$ slab.

The vdW $\alpha$-MoO$_3$ crystal has several optical phonons at mid-IR frequencies, which open frequency bands (Reststrahlen bands) in the transverse direction (perpendicular to the slab) [20, 21]. The lattice parameters in a 100 nm thick $\alpha$-MoO$_3$ slab (with period $L = 900$ nm and hole radius, $a = 200$ nm) have been chosen to ensure the occurrence of PhP resonances within the second hyperbolic Reststrahlen band, RB2, of $\alpha$-MoO$_3$ [19], i.e., in the frequency range 840 – 960 cm$^{-1}$. At frequency $\omega = 909.9$ cm$^{-1}$ all field amplitudes show a prominent and narrow resonant peak (with a quality factor $Q$ for $(100)_p$ of $\approx 150$), with the largest value reached by the first-order field harmonic, indicating that the latter has the largest contribution to the resonance. Moreover, the peak position coincides with the condition of the first-order Bragg resonance, $k_{\mathbf{B}} = \mathbf{G}$, as is evident from the comparison with the hyperbolic isofrequency curve (IFC) – a slice of the 3D dispersion surface at a constant frequency – of the M0 mode in the $\alpha$-MoO$_3$ slab (Figure 2b) and its dispersion (Figure 2d). Finally, the electric field distribution above the lattice (Figure 2a), reconstructed with the help of Equation (3), visualizes a standing wave whose oscillation length (distance between maxima with the same polarity – blue or red in the figure) coincides with the lattice period and is much larger than the slab thickness, which guarantees the validity of our approximation. These observations clearly demonstrate that the spectra in Figure 2c manifest the excitation of a first-order narrow Bragg PhP resonance in a lattice realized in the $\alpha$-MoO$_3$ slab.

Surprisingly, the numerical solution of the linear system of Equation (7) (continuous curves in Figure 2c) shows an excellent agreement with our analytical approximation (square symbols in Figure 2c), in which only zero-order and first-order field harmonics are retained (see Section S2 in the Supporting Information). This agreement demonstrates the validity of the perturbative approximation (analogous to ref. [18, 22]), in which it is sufficient to retain only a few field Fourier harmonics.

Let us now study the dependence of the emerging Bragg PhP resonances on the twist angle, $\phi$ (angle between the grating vector, $\mathbf{G}$, and the [100] direction of the $\alpha$-MoO$_3$ crystal slab) in the case of the simplest 1D harmonic lattice (see the schematic in Figure 3a). As before, we assume illumination of the structure by a normally incident wave with the electric field polarized along the $x$-axis, the latter aligned with the [100] crystal direction in $\alpha$-MoO$_3$. Figure 3a shows the numerically and analytically calculated $\delta R$ surrounding the slab, we assume that both dielectric media have a dielectric permittivity equal to 1. On the one hand, this choice simplifies the interpretation and analysis of the resonances obtained and, on the other hand, it mimics the typical experimental scheme in which the $\alpha$-MoO$_3$ slab is placed on a highly transparent substrate (e.g., BaF$_2$) [18]. Note that as soon as the lattice cannot provide any momentum in the $x$-direction (and the incident wave is polarized along the $x$-direction), the $k$-vectors of all diffracted waves, as well as their electric fields, belong to the $xz$-plane. Since for normal incidence the incident plane is not well defined, we assign the $p$-polarization to the plane waves that have their electric fields within the $xz$-plane, while the “s-polarized” waves are completely absent.

Figure 2c shows the spectra of the normalized reflection coefficient (introduced as $\delta R = |R_{\mathbf{p}}(\omega) - R_{\mathbf{p}}^0(\omega)|$, where $R_{\mathbf{p}}^0$ represents the $p$-polarization reflection coefficient of a bare slab) and the amplitudes of the first, $(\pm 1, 0)$, and second order, $(\pm 2, 0)$, field harmonics. Due to the symmetry provided by the normally incident wave, the amplitudes of the $(n, 0)$ and $(-n, 0)$ field harmonics are identical. The lattice parameters in a 100 nm thick $\alpha$-MoO$_3$ slab (with period $L = 900$ nm and hole radius, $a = 200$ nm) have been chosen to ensure the occurrence of PhP resonances within the second hyperbolic Reststrahlen band, RB2, of $\alpha$-MoO$_3$ [19], i.e., in the frequency range 840 – 960 cm$^{-1}$. At frequency $\omega = 909.9$ cm$^{-1}$ all field amplitudes show a prominent and narrow resonant peak (with a quality factor $Q$ for $(100)_p$ of $\approx 150$), with the largest value reached by the first-order field harmonic, indicating that the latter has the largest contribution to the resonance. Moreover, the peak position coincides with the condition of the first-order Bragg resonance, $k_{\mathbf{B}} = \mathbf{G}$, as is evident from the comparison with the hyperbolic isofrequency curve (IFC) – a slice of the 3D dispersion surface at a constant frequency – of the M0 mode in the $\alpha$-MoO$_3$ slab (Figure 2b) and its dispersion (Figure 2d). Finally, the electric field distribution above the lattice (Figure 2a), reconstructed with the help of Equation (3), visualizes a standing wave whose oscillation length (distance between maxima with the same polarity – blue or red in the figure) coincides with the lattice period and is much larger than the slab thickness, which guarantees the validity of our approximation. These observations clearly demonstrate that the spectra in Figure 2c manifest the excitation of a first-order narrow Bragg PhP resonance in a lattice realized in the $\alpha$-MoO$_3$ slab.

3. Single Lattice

We will focus on the polaritonic effects originating from the first-order scattering processes of the polaritonic lowest-order mode, i.e., the processes provided by the first-order Fourier harmonics of the 2D lattice. For this reason, we will investigate an artificial lattice composed exclusively of first-order Fourier coefficients of $\alpha$ (a “harmonic” lattice), i.e., with the orders $(0, 0)$, $(\pm 1, 0)$ and $(0, \pm 1)$. On the other hand, we will take the amplitudes of these Fourier harmonics from the realistic expansion of the HA (marked by the dashed white circles in Figure 1b). Furthermore, let us first assume that the Fourier harmonics of the lattice along the $y$-direction have zero amplitude, $\alpha_{0,1} = 0$, so that the lattice is 1D. Its schematic is shown in Figure 2a. The 1D lattice is aligned along the $[100]$ crystal direction (coincident with the $x$-axis), and the polarization of the normally incident light is parallel to the $x$-axis, thus being aligned with the grating vector, $\mathbf{G}$. Although our theoretical approach is valid for arbitrary isotropic dielectric materials, the following discussion will be restricted to the case of $\alpha$-MoO$_3$. In this case, the Fourier decomposition of the periodic structure plays a crucial role in the diffraction of an incident plane wave and the resonant excitation of polaritonic modes.
Figure 2. PhP Bragg resonance in a 1D harmonic lattice in an $\alpha$-MoO$_3$ slab. a) Schematic of the 1D lattice with its vector aligned to the [100] $\alpha$-MoO$_3$ crystallographic axis. The lattice period is $L = 900$ nm, the permittivities of the surrounding dielectric media are $\varepsilon_1 = \varepsilon_2 = 1$ and the thickness of the MoO$_3$ slab is $d = 100$ nm. The lattice Fourier harmonic amplitude $a_{\pm 10}$ is taken for a HA with hole radius $a = 200$ nm. The normally incident light is polarized along the $x$-axis. The vertical electric field distribution shown is calculated at $\omega = 909.9 \text{ cm}^{-1}$. b) Isofrequency curve (IFC) of the M0 PhP mode at $\omega = 909.9 \text{ cm}^{-1}$. $G = \pm G_1$ represents the reciprocal lattice vector. The reciprocal lattice points corresponding to the lattice Fourier coefficients with nonzero values are highlighted in red. c) Spectra of the $(\pm 1,0)$ and $(\pm 2,0)$ diffracted waves amplitudes ($T_{p10}$ and $T_{p20}$, respectively) and the relative reflection coefficient, $\delta R$. d) Dispersion relation for the M0 PhP mode.}

as a function of frequency (solid curves and square symbols, respectively) for different values of $\phi$. For $\phi \neq 0$, polarization conversion takes place so that cross-polarized ($s$-) components of the diffracted plane waves appear. The amplitude of the $s$-polarized reflected wave of zero-diffraction order is plotted in Figure 3a by dashed lines, indicating a polarization conversion of the order of 5%. More importantly, the resonant peak associated with the first-order PhP Bragg resonance redshifts strongly with increasing $\phi$. The resonance redshift can be understood by the Bragg resonance condition, i.e., matching of the $k_{\pm 10}$ in-plane wavevectors with the IFC of the M0 mode (see Figure 3b). In fact, due to the rotation of the grating (vector $G$) with increasing $\phi$, the hyperbolic IFC meets the points $(1,0)$ and $(-1,0)$ in the reciprocal space at a lower frequency (the asymptote of the hyperbola becomes more “aligned” along the vector $G$), the latter matching the position of the resonant peaks in Figure 3a. Obviously, for the extreme case of $\phi = 90^\circ$, the hyperbolic IFC does not meet the reciprocal vector of the lattice for any frequency and, therefore, the PhP Bragg resonance does not appear (the coefficient $\delta R$ is close to zero). Since in general the resonances in Figure 3a only depend on the relative angle between the crystallographic axes and the reciprocal lattice, an alternative to Figure 3b could be constructed by fixing the angle of the grating while the crystallographic axes are twisted (see Section S5 in the Supporting Information).

The dispersion of the polaritons supported by our lattice (Bloch modes) presents the band structure. The latter is visualized in Figure 3c,d by the near-field maxima as a function of in-plane momentum, $k_x$, and frequency, $\omega$. The largest bandgap is observed at the symmetry point X ($k_x = G/2$), due to a strong interaction between the “bare” counterpropagating polaritons [represented by the field harmonics ($-1,0$) and $(1,0)$], while in the center of the Brillouin zone ($k_x = 0$) the splitting between the dispersion branches is much smaller. The insets to Figure 3c,d show in detail the dispersion branches within the light cone region. As is typical in polaritonic crystals, only one of the split modes can effectively couple to the normally incident plane wave\textsuperscript{[15]}, and thus only one flat maximum of $\delta R$ is observed. More importantly, the band structure manifests the strong frequency shift with the change of the twist angle, consistent with the shift of the Bragg resonance in Figure 3a. Taken together, our results show, for the first time, the tunability of the PhP Bragg resonance by simply twisting the photonic lattice with respect to the vdW crystal slab. In practice, such tuning can be realized for lattices made directly on a biaxial vdW crystal slab or on a periodically structured substrate with a rotatable crystal slab placed on top: a “dielectric engineering” concept.

4. Two Crossed Lattices

A much richer family of rotationally tunable Bragg resonances appears when the periodicity in the biaxial crystal slab is formed simultaneously in two crystal directions. An example of such 2D lattice (consisting of the two simplest 1D harmonic lattices with mutually perpendicular Bragg vectors and twisted at an
Figure 3. PhP Bragg resonance in a twisted 1D harmonic lattice in an α-MoO3 slab. a) Numerically and analytically calculated spectra of \( \delta R \) (shown by continuous curves and square symbols, respectively) for different values of the twist angle \( \phi \). Dashed lines represent \( R_{00} \) coefficient for the same angles. The schematic of a 1D grating with its basis twisted an angle \( \phi \) with respect to the crystallographic axes is shown as an inset. A normally incident light with a linearly polarized electric field along the x-axis is considered. The parameters of the structure are: L = 900 nm, \( \epsilon_1 = \epsilon_2 = 1 \), and \( d = 100 \) nm. The amplitude \( \alpha_{-10} \) is taken for a HA with \( a = 200 \) nm. b) IFCs for fixed frequencies and different twist angles \( \phi \) between the grating basis and the crystallographic axes. Points in reciprocal space with corresponding nonzero Fourier coefficients of the conductivity tensor are highlighted in red. The points on the circle are \((1,0)\) and \((-1,0)\) for different angles \( \phi \). c,d) The dispersion branches visualized by the near-field color plot (the sum of the amplitudes of the Fourier harmonics of the field, \( \sum |T_{n0}| \)) as a function of \( \omega \) and \( k \) for twist angles \( \phi = 0^\circ \) and \( \phi = 45^\circ \). The electric field is calculated within the first Brillouin zone with \( k_2 \) aligned with the reciprocal vector \( G \). The dashed blue lines are the dispersion of the bare MoO3 slab. The insets show \( \delta R \) as a function of \( \omega \) and \( k_2 \) = \( k_0 \) sin \( \theta \) within the light cone. The parameters of the lattice and MoO3 slab are the same as in (a).

angle \( \phi = 60^\circ \) with respect to the [100] crystallographic axis in \( \alpha\)-MoO3) is depicted as a schematic in Figure 4a. Illumination of the periodic structure by a normally incident plane wave polarized along the [100] crystallographic axis in \( \alpha\)-MoO3 leads to the appearance of multiple resonant peaks in the \( \delta R \) spectra, as shown in Figure 4b (red curve and square symbols for the numerical and analytical solutions, respectively). We assume that the two most prominent peaks appearing at 860 and 895.6 cm\(^{-1}\) can be associated with the first-order Bragg resonances (\( \pm 1,0 \)) and (\( 0,\pm 1 \)), respectively, as can be concluded from the matching of the reciprocal lattice vectors \( G_{10} \) and \( G_{01} \) with the hyperbolic IFC in the two orthogonal directions (Figure 4c,d). It is noteworthy that the transmission coefficient \( T_{310} \) (Figure 4b, black curve) has a single maximum at 860 cm\(^{-1}\), while the coefficient \( T_{210} \) (Figure 4b, green curve) has a single maximum at 895.6 cm\(^{-1}\). Since these transmission coefficients characterize the amplitudes of the first-order diffracted waves (\( \pm 1,0 \)) and (\( 0,\pm 1 \)), their resonance character indicates the formation of intense polaritonic standing waves (Bloch waves) along the x and y directions. The latter can be clearly seen in the calculated field distribution shown in Figure 4a, confirming our assumption regarding the nature of the resonances responsible for the \( \delta R \) peaks. Note that while the amplitudes of the (\( \pm 1,0 \)) and (\( 0,\pm 1 \)) diffraction orders are dominating throughout the whole Fourier–Floquet expansion given by Equations (3) and (4) (particularly, at resonance frequencies, as expected), the amplitudes of the higher diffraction orders decay rapidly with \( N \) (see, for example, the amplitude of the second order, shown by the magenta curve in Figure 4a). In fact, this behavior of the diffraction amplitudes is expected from the Fourier composition of our diffraction grating. The latter generates mainly the first-order diffracted waves via the first-order scattering processes (proportional to \( \alpha_{-10} \)), while the higher-order diffracted waves are generated by the higher-order scattering processes (proportional to higher powers of \( \alpha_{-10} \)) and thus giving rise to much smaller diffraction amplitudes, particularly for rather “weak” gratings\(^{18,22}\). The noncollinearity of the incident wave polarization with the lattice vectors (analogous to the case of the twisted 1D grating) allows polarization transformation, as illustrated by the spectra of the amplitude of the s-polarization component of the zero-order reflected wave, \( R_{s00} \) (magenta curve in Figure 4). Although the efficiency of this transformation is small, it can be substantially improved by properly tuning the grating parameters, as well as its orientation with respect to the crystal axes and the incident field. Overall, our analysis of PhP Bragg resonances in the 2D lattice of the \( \alpha\)-MoO3 slab, twisted with respect to the crystal axes, suggests interesting possibilities for versatile
control of the PhPs by means of the twist angle or the lattice amplitude and periods. In particular, twisting the lattice with respect to the crystal axes may allow the excitation of PhPs in predetermined directions. That is, PhPs can be excited in different directions at the same frequency, or in different directions at different frequencies, both of which are attractive scenarios for potential near-field electromagnetic routing applications.

Note that our general analysis can also be applied to the other two Reststrahlen bands of the MoO₃ crystal, in particular to the elliptic frequency band (957 – 1007 cm⁻¹), where, although excitation of the Bragg resonances is allowed for any lattice orientation with respect to the crystal axes, the emerging resonant features depend strongly on the twist angle (see Section S4 in the Supporting Information).

5. Hole Array

Finally, we apply our theory to illustrate PhP Bragg resonances in an experimentally realizable structure consisting of a periodic HA in an α-MoO₃ slab, illustrated in Figure 1a. Such a HA can be realized, for example, by decorating the slab with periodically arranged circular holes milled by focused ion beam (FIB) high-resolution lithography. Figure 5a shows the spectra of the reflection, transmission and absorption coefficients (ρ, τ, and A, respectively) of normally incident light when a square HA with periodicity L = 900 nm and hole radius, a = 200 nm is considered (the unit cell together with the incident light polarization and axes orientation is outlined in the inset to Figure 5a). In all the spectra shown, other resonant features appear at different frequencies. Obviously, as all the spatial Fourier harmonics of the HA have a nonzero value, its optical response has a significantly more complex structure, compared to the harmonic lattices considered above. For the interpretation of such emergent resonances, in Figure 5b we plot the calculated IFCs of the M0 PhP mode in reciprocal space at the resonant frequencies. By tracking the crossings between the IFCs and the points in the reciprocal space, we assign the specific Bragg resonances (of certain orders) to the corresponding peaks/dips in the spectra in Figure 5a. Note that the resonance “visibility” (relative difference between the maximum and minimum values of the reflectivity/absorption within the resonance) decreases with the resonance diffraction order, in line with the decrease of the lattice Fourier harmonic amplitudes (Figure 1b) at higher orders, resulting in weaker resonant scattering processes. Remarkably, the strongest resonant feature in Figure 5a coincides with the Bragg condition for the resonance (±1,±2), corresponding to the nearly asymptotic regime of the hyperbolic IFC (a straight line), where the PhP momentum is already quite high. We speculate that, apart from the Bragg scattering processes, the high efficiency of the resonant PhP excitations in the diffraction orders (±1,±2) may be related to the dipole...
lar resonance of the holes (see the dipolar-like field pattern in the snapshot of the vertical electric field shown in the inset of Figure 5a), as well as to the flat bands arising in the polaritonic crystals.[14] However, detailed analysis of such an anomaly is beyond the scope of this manuscript and will be addressed in future work.

6. Conclusion

In summary, we have illustrated rotationally tunable anisotropic PhP Bragg resonances in twisted lattices formed in biaxial vdW crystal slabs. We have developed a simple theoretical approach suitable for both numerical and analytical analysis of "collective" polariton resonances in arbitrary lattices, which is valid for any biaxial crystal slab, provided its thickness is smaller than the wavelength of the polaritons. We have clarified the role of the spectral Fourier composition of the lattices by considering their specific Fourier harmonics. Our results open up a new research area of twisted polaritonic crystals, including those that support nontrivial topological polaritons. These crystals may be very attractive for some practical use, such as in tunable sensors or photodetectors.

7. Experimental Section

**Numerical Simulations:** Full-wave simulations based on the finite-element method (FEM) in the frequency domain were performed using COMSOL. simulated the 2D square periodic hole array in a MoO3 slab with a thickness d = 100 nm, period L = 900 nm, and hole radius a = 200 nm. The incident angle of an illuminating p-polarized plane wave, as well as the angle between one of the Bragg vectors of the hole array and the [010] crystal axis, were varied (see Section S4 in the Supporting Information). The dielectric permittivity of the superstrate was taken as air, ε₁ = 1, while ε₂ (substrate) was taken to be that of BaF₂.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

Bragg resonance, hyperbolic polaritons, polaritonic crystal, twisted lattices

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