Supplementary Information:

The Origin and Limit of Asymmetric Transmission in Chiral Resonators

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I. DERIVATION OF AT FROM QUASINORMAL MODES

We have recently shown [S1] that building upon Ref. [S2], the scattering matrix for a system with a set of complex eigenfrequencies $\tilde{\omega}_j (j=1,\ldots,m)$ can be written as:

$$S(\omega) = C + i \sum_{j=1}^{m} a_j \frac{\mathbf{b}_j \mathbf{b}_j^{\mathsf{T}}}{\omega - \tilde{\omega}_j},$$
 (S1)

where C is the direct-transport matrix (assumed to be a scattering matrix itself, thus unitary and symmetric) and $\tilde{\omega}_j$ are the complex eigenfrequencies of the quasinormal modes of the system (we assume the convention $\text{Im}(\tilde{\omega}_j) > 0$). The general expression for the coefficients a_j is given in Ref. [S1]. For a single isolated mode (with eigenfrequency $\tilde{\omega}_0$), this expression reduces to $a_0 = 2\text{Im}(\tilde{\omega}_0)/(\mathbf{b}^{\mathsf{T}}C^{\dagger}\mathbf{b})$. Then, at the resonance frequency ($\omega = \text{Re}(\tilde{\omega}_0)$), Eq. (S1) becomes:

$$S = C - 2 \frac{\mathbf{b} \mathbf{b}^{\mathsf{T}}}{\mathbf{b}^{\mathsf{T}} C^{\dagger} \mathbf{b}}.$$
 (S2)

Both the direct-coupling matrix C and the resulting scattering matrix S are unitary. From the relation $\mathbf{b}^{\mathsf{T}} S^{\mathsf{T}} S \mathbf{b}^* = \mathbf{b}^{\mathsf{T}} \mathbf{b}^*$, by replacing Eq. (S2), we obtain that

$$|\mathbf{b}^{\mathsf{T}} C^{\dagger} \mathbf{b}|^2 = |\mathbf{b}^{\dagger} \mathbf{b}|^2. \tag{S3}$$

The scattering eigenvector \mathbf{b} consists of a set of electric field components along $\hat{\mathbf{s_1}}$ and $\hat{\mathbf{s_2}}$ (b_{1U} and b_{2U} , respectively) which represents the eigenmode polarization above the structure and a set of field components (b_{1L} and b_{2L}) which represents the eigenmode polarization below the structure, in the far-field. Thus, \mathbf{b} can be written as: $\mathbf{b} = \begin{bmatrix} b_{1U} & b_{2U} & b_{1L} & b_{2L} \end{bmatrix}^\mathsf{T}$. As it can be seen by direct inspection, Eq. (S2) is independent of the choice of the normalization of \mathbf{b} .

As mentioned in the main text, for reciprocal materials the asymmetric transmission (AT) can be defined as the difference in orthogonal transmittances for two mutually perpendicular polarized incidences in a given direction. For generality, let us define the two orthogonal linearly polarized input vectors as:

$$\hat{\mathbf{s}}_{1in} = \begin{bmatrix} \cos \psi \\ \sin \psi \\ 0 \\ 0 \end{bmatrix}, \hat{\mathbf{s}}_{2in} = \begin{bmatrix} -\sin \psi \\ \cos \psi \\ 0 \\ 0 \end{bmatrix}, \tag{S4}$$

where the angle ψ gives the polarization angle of input light with respect to the geometrical reference x axis of the structure. Let t_{21} be the transmission component along $\hat{\mathbf{s}}_2$ when the input is $\hat{\mathbf{s}}_{1in}$ and t_{12} be the transmission component along $\hat{\mathbf{s}}_1$ when the input is $\hat{\mathbf{s}}_{2in}$. The cross-polarized transmission coefficients, t_{21} and t_{12} , can then be calculated as:

$$t_{21} = \begin{bmatrix} 0 & 0 & -\sin\psi & \cos\psi \end{bmatrix} S \,\hat{\mathbf{s}}_{1in}$$

$$t_{12} = \begin{bmatrix} 0 & 0 & \cos\psi & \sin\psi \end{bmatrix} S \,\hat{\mathbf{s}}_{2in}.$$
 (S5)

The expression for AT can thus be written as:

$$AT = |T_{21} - T_{12}| = ||t_{12}|^2 - |t_{21}|^2|.$$
 (S6)

The symmetry properties of the example structure can be used to derive a relation between the polarizations of the eigenmode above and below the structure. The structure possesses certain symmetry properties such that it returns to the original configuration after a possible series of operations as illustrated in the left panel of Fig. S1. For simplicity, in the figure we show only the configuration of holes inside a unit cell. The right panel of the figure contains the corresponding transformation matrices: T_1 (an inversion along z), T_2 (an inversion along x), and T_3 (a clockwise rotation of 90°).

The total transformation matrix for such a series of operations can be written as:

$$T = T_3 T_2 T_1 = \begin{bmatrix} 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \\ 0 & 1 & 0 & 0 \\ 1 & 0 & 0 & 0 \end{bmatrix}.$$
 (S7)

From this symmetry it follows that the eigenvectors of the system must satisfy the relation $T\mathbf{b} = \pm \mathbf{b}$ [S3]. For this relation to hold, the eigenmode electric field values above and below the structure must be related according

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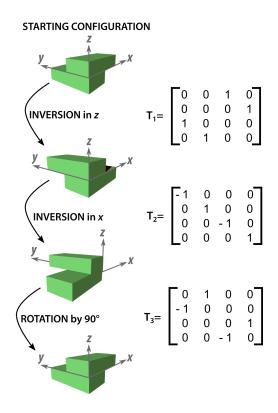


FIG. S1. Series of symmetry operations for the proposed structure: A cartoon illustrating one possible series of symmetry operations which would return the proposed structure to it's original configurations. For simplicity, the cartoon shows the arrangement of holes inside the unit cell. Corresponding transformation matrices are shown alongside.

to Eq. (4) in the main text, which we repeat here for convenience:

$$b_{1L} = \pm b_{2U}, \quad b_{2L} = \pm b_{1U}.$$
 (S8)

As a consequence of this symmetry relations, we can represent the scattering eigenvector \mathbf{b} only in terms of the polarization parameters above the structure:

$$\mathbf{b} = \begin{bmatrix} \cos \theta \ e^{-i\frac{\phi}{2}} \\ \sin \theta \ e^{i\frac{\phi}{2}} \\ \pm \sin \theta \ e^{i\frac{\phi}{2}} \\ \pm \cos \theta \ e^{-i\frac{\phi}{2}} \end{bmatrix}, \tag{S9}$$

where $\tan(\theta)$ is the ratio of amplitudes of the field components b_{1U} and b_{2U} and ϕ is the relative phase between them.

In order to compute the AT, the cross-polarized transmittances T_{21} and T_{12} can be obtained from Eq. (S2), (S3), (S5), and (S9):

$$T_{21} = |t_{21}|^2 = |(\pm \cos^2 \theta \cos^2 \psi e^{-i\phi} \mp \sin^2 \theta \sin^2 \psi e^{i\phi})|^2$$

$$T_{12} = |t_{21}|^2 = |(\pm \sin^2 \theta \cos^2 \psi e^{i\phi} \mp \cos^2 \theta \sin^2 \psi e^{-i\phi})|^2.$$
(S10)

AT can then be calculated from Eq. (S6):

$$AT = |T_{12} - T_{21}| = \cos(2\theta) \cos(2\psi).$$
 (S11)

This equation gives us an insight on the choice of input polarization direction for maximizing the AT. The maximum AT corresponds to the choice of the input polarization along the geometrical reference x axis of the structure ($\psi = 0$). In this case, we have a final expression for AT as:

$$AT = \cos(2\theta). \tag{S12}$$

The polarization of the far-field of eigenmodes can also be expressed in terms of normalized Stokes parameters, which, using the notation of Eq. (S9), are defined as

$$S_0 = \cos^2 \theta + \sin^2 \theta = 1,$$

$$S_1 = \cos^2 \theta - \sin^2 \theta = \cos(2\theta),$$

$$S_2 = 2\cos \theta \sin \theta \cos \phi = \sin(2\theta)\cos \phi,$$

$$S_3 = 2\cos \theta \sin \theta \sin \phi = \sin(2\theta)\sin \phi.$$
(S13)

Using the definition of Stokes parameters, Eq. (S12) can be recast in the form reported in Eq. (5) of the main text:

$$AT = |S_1|. (S14)$$

II. PRINCIPLE OF RECIPROCITY AND THE LIMIT OF AT

According to Ref. [S2], the principle of reciprocity in coupled-mode theory states that the direct process matrix and the resonant process are related by the expression

$$\tilde{C}\mathbf{d}^* = -\mathbf{d},\tag{S15}$$

where \mathbf{d} is the vector containing coupling coefficients which relate the resonance to the input and output waves and \tilde{C} is the direct coupling matrix of the system. For a single mode, the coupling vector is proportional to the far-field amplitudes of the eigenmode with a complex proportionality coefficient having unit magnitude. i.e., $\mathbf{d} = e^{i\zeta} \mathbf{b}$. By expressing the direct matrix as $\tilde{C} = e^{i\chi} C$, where C is the direct matrix defined in Eq. (2) of the main text, we can write the principle of reciprocity as:

$$e^{i\chi}C\mathbf{b}^* = -e^{2i\zeta}\mathbf{b}.\tag{S16}$$

The C-matrix phase factor can be incorporated without loss of generality in a total phase factor $\xi = 2\zeta - \chi$. In this way, we arrive at Eq. (6) of the main text:

$$C\mathbf{b}^* = -e^{i\xi}\mathbf{b}.\tag{S17}$$

Using Eq. (S9) and the definition of C (Eq. 2 in the main text) in Eq. (S17), we can write:

$$r\cos\theta \,e^{\mathrm{i}\frac{\phi}{2}} \pm \mathrm{i}t\sin\theta \,e^{-\mathrm{i}\frac{\phi}{2}} = -e^{\mathrm{i}\xi}\cos\theta \,e^{-\mathrm{i}\frac{\phi}{2}} \tag{S18}$$

$$r\sin\theta e^{-i\frac{\phi}{2}} \pm it\cos\theta e^{i\frac{\phi}{2}} = -e^{i\xi}\sin\theta e^{i\frac{\phi}{2}}$$
 (S19)

The ratio of Eq. (S18) and Eq. (S19), after some algebraic manipulations, gives Eq. (7) in the main text:

$$\frac{t}{r} = \left| \frac{2\cos\theta\sin\theta\sin\phi}{\cos^2\theta - \sin^2\theta} \right| = \left| \frac{S_3}{S_1} \right| \tag{S20}$$

From this equation and the normalization of the Stokes parameters $(S_1^2 + S_2^2 + S_3^2 = 1)$ the intrinsic limit on AT discussed in the main text, AT $\leq r$, is derived.

III. AT AND ITS LIMIT FOR GENERAL STRUCTURES

For general structures without any symmetry properties, we are no longer allowed to use the symmetry condition given by Eq. (S8); however, we can express the polarization components of the eigenmode field below the structure in terms of the components above the structure using principle of reciprocity in Eq. (S17). From Eq. (S17), it can be shown that:

$$b_{1L} = \frac{b_{1U} r + b_{1U}^* e^{-i\xi}}{i t}$$

$$b_{2L} = \frac{b_{2U} r + b_{2U}^* e^{-i\xi}}{i t}.$$
(S21)

Using the same notation of Eq. (S9) for the polarization of the field above the structure, we can write **b** as:

$$\mathbf{b} = \begin{bmatrix} \cos \theta \ e^{-i\frac{\phi}{2}} \\ \sin \theta \ e^{i\frac{\phi}{2}} \\ \frac{1}{it}(r + e^{i(\phi - \xi)})\cos \theta \ e^{-i\frac{\phi}{2}} \\ \frac{1}{it}(r + e^{-i(\phi + \xi)})\sin \theta \ e^{i\frac{\phi}{2}} \end{bmatrix}. \tag{S22}$$

AT can be obtained from Eq. (S6), together with Eqs. (S2) and (S5), as

$$AT = \left| \frac{4r t^2 \cos^2 \theta \sin^2 \theta \sin \phi \sin \xi}{(1 + r (\cos^2 \theta \cos(\phi - \xi) + \sin^2 \theta \cos(\phi + \xi)))^2} \right|.$$
(S23)

In deriving this equation we have assumed $\psi = 0$ without loss of generality in the definition of the input vectors in Eq. (S4).

We can write Eq. (S23) in terms of the Stokes parameter for the polarization above the structure, obtaining Eq. (9) in the main text, which we repeat here:

$$AT = \left| \frac{r t^2 (1 - S_1^2) \sin \phi \sin \xi}{(1 + r (\cos \phi \cos \xi + S_1 \sin \phi \sin \xi))^2} \right|.$$
 (S24)

As stated in the text, the maxima of AT corresponds to the points $|S_1| = r, |S_3| = t, \psi = \pm \pi/2$, as it can be verified by computing the first derivatives of Eq. (S24). In a similar fashion, it can be verified that all these points are equivalent global maxima.

As an example, in Fig. S2, we plot the maximum AT (maximized over ξ) given by Eq. (S24) as a function of

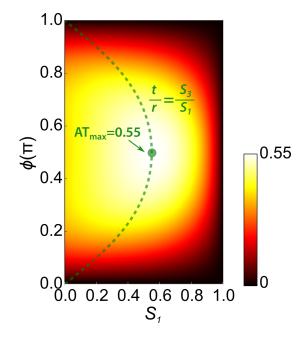


FIG. S2. Numerical maximization of AT: The 2-D Plot shows numerically calculated maximum values of AT given by Eq. (S24) over the total phase factor $\xi \in (0, 2\pi)$ as a function of eigenmode polarization parameters S_1 and ϕ . Bright green spot marks the maximum value of AT_{max} in the 2-D space. Green dashed line corresponds to the points in the polarization space which follow the relation $\frac{t}{r} = \frac{S_3}{S_3}$.

 S_1 and ϕ , for a direct reflection coefficient r=0.55. The global maximum of AT is marked by the green dot and corresponds to the value AT=r=0.55, as expected. The dashed green line indicates the curve in polarization space where the ratio between the Stokes parameters is equal to the ratio of the direct-process reflection and transmission coefficients, i.e., it corresponds to Eq. (S20). It can be seen that most of this line is along near-the-limit regions of the maximum AT. As discussed beforehand, all the structures with the same symmetry as the one proposed in this work follow the same relation. As a consequence, structures having symmetries of this kind have significant prospects for offering AT near the fundamental limit.

IV. IMPLICATIONS OF ANGULAR DEPENDENCE

In all the simulations and calculations presented in the main text, we consider light which is incident normally onto the sample. However, a real experimental setup involves the use of finite-size incident beam with a finite angular spread. It is thus important to calculate the variation of AT as a function of angle of incidence. This is particularly relevant to the example of a planar periodic sample as we used in our example, as its modal bandstructure will depend on angle.

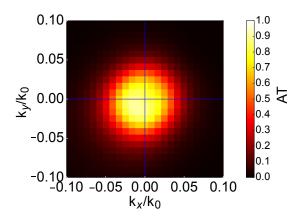


FIG. S3. AT at oblique incidence: Variation of AT as a function of in-plane wave-vector at a frequency of $\omega_0/2\pi=363.78$ THz from FEM simulations. In plane wave vector is shown in units of $k_0=\omega_0/c$.

We consider a beam of light which is polarized perpendicularly to the in-plane axis $\hat{\mathbf{y}}$ and propagates along either the $\hat{\mathbf{z}}$ or $-\hat{\mathbf{z}}$ directions. As a consequence of the finite size of the beam, the incident electric field can be described as the superposition of plane waves with different in-plane wavevectors $\mathbf{k}_{\parallel} = (k_x, k_y)$:

$$\mathbf{E}(\mathbf{r}) = \int dk_x dk_y \, \frac{\mathcal{E}(k_x, k_y)}{\sqrt{k_x^2 + k_z^2}} \begin{bmatrix} -k_z \\ 0 \\ k_x \end{bmatrix} e^{i(k_x x + k_y y + k_z z)},$$
(S2)

where $\mathcal{E}(k_x, k_y)$ corresponds to the amplitudes of the inplane Fourier components and $k_z = \pm (k_0^2 - k_x^2 - k_y^2)^{1/2}$.

In Fig. S3, for a given frequency ($\omega_0/2\pi = 363.78 \text{ THz}$), we calculate the AT of the system introduced in Fig. 2 of the main text as a function of the in-plane wavevector $\mathbf{k}_{\parallel} = (k_x, k_y)$. The calculations have been performed with the finite element method. As evident from the figure, AT varies with the variation of the incident angle with respect to normal incidence $(k_x = k_y = 0)$. This is likely result of the fact that for finite angles the mode frequency is expected to shift away from the resonance frequency ω_0 at normal incidence, as well as a potential evolution of the mode's polarization along the photonic crystal band. From the data in the figure, we can estimate a maximal half opening angle for the incident field of the order of NA $\simeq 0.06$, which corresponds, assuming a Gaussian envelope for the field, to a minimal beam waist of the order of $w_0 \simeq 5\lambda_0$. This value also constrains the minimal size of the sample, and it seems fully compatible with realistic experimental and fabrication conditions.

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