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Published in: iScience

Published: 21/07/2023

Document Version:

Final Published version, also known as Publisher's PDF, Publisher's Final version or Version of Record

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Publication record in CityU Scholars: Go to record

Published version (DOI): 10.1016/j.isci.2023.107032

Publication details:

Pan, A., Lin, K., Chen, S., & Tso, C. Y. (2023). Investigations on asymmetric transmittivity of optical devices and different diode-like behaviors. *iScience*, *26*(7), [107032]. https://doi.org/10.1016/j.isci.2023.107032

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Pan et al., iScience 26, 107032 July 21, 2023 © 2023 The Author(s). https://doi.org/10.1016/ j.isci.2023.107032

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Investigations on asymmetric transmittivity of optical devices and different diode-like behaviors

Aiqiang Pan,¹ Kaixin Lin,¹ Siru Chen,¹ and Chi Yan Tso^{1,2,*}

SUMMARY

This study theoretically proved that although reciprocal optical devices can show asymmetric transmittivity (AT) under controlled incident modes (i.e., conditional AT), they cannot guarantee AT with arbitrary incident light modes, whereas only nonreciprocal optical devices can possibly guarantee AT. Besides, the thermodynamics of both reciprocal and nonreciprocal optical devices were discussed to show that the second law of thermodynamics is valid anyway. Furthermore, the diode-like behaviors of optical and electronic devices were compared. Electrons are identical to electronic devices, so electronic devices could have asymmetric conductance regardless of electrons. In contrast, electromagnetic waves are different from optical devices as transmittivity of different modes can be different, so reciprocal optical devices showing conditional AT cannot guarantee AT when incident modes are arbitrary. The mathematical proof and characteristic comparisons between electronic and optical diodes, which are firstly presented here, should help clarifying the necessary nonreciprocity required for being optical diodes.

INTRODUCTION

Many optical devices were demonstrated to have asymmetric transmittivity (AT) of light in the two opposite directions with experiments or numerical calculations.¹ AT is also referred to as nonsymmetric, unequal, or unidirectional transmittivity. Specifically, optical devices having AT allow the transmission of light in one direction but block the transmission of light in the opposite direction.² This AT property is useful in some applications such as laser systems and optical communication systems.³

Optical devices having AT were called optical diodes (or optical isolators) by some researchers. In general, the existing literature reported AT with optical devices that either break time-inverse symmetry (i.e., break the Lorentz reciprocity)^{2,4} or break space-inverse symmetry (structurally asymmetric).^{3,5} The optical devices breaking the Lorentz reciprocity are made with nonreciprocal materials. The nonreciprocity of the materials is achieved by being time-dependent,^{6,7} nonlinear,⁸⁻¹⁰ or anisotropic with asymmetric permittivity and permeability tensors. Magneto-optical materials are one of the most common materials that are anisotropic with asymmetric permittivity.^{2,11–13} On the other hand, some optical devices breaking space-inverse symmetry are made with reciprocal materials. The AT achieved by reciprocal optical devices is diffractive AT in general.⁵ Such kinds of reciprocal optical devices take various forms, one main group is diffraction gratings, which can be classified into gratings with tapered structures¹⁴⁻¹⁹ or corrugated surfaces,²⁰ gratings with a subwavelength slit,²¹⁻²³ dual or double gratings,²⁴⁻²⁶ photonic crystal gratings with corrugated surfaces²⁷⁻³⁰ or with other asymmetric structures,^{31,32} and some other special gratings.^{33,34} Other forms include multilayer crystals containing defects, ^{35–37} metasurfaces, ^{38,39} and photonic crystals.^{40–42} Despite the various forms, these reciprocal optical devices are commonly spatial asymmetric. More importantly, the AT of reciprocal optical devices is usually numerically simulated with controlled incident light (for example, polarized light),³ or experimentally measured under controlled incident light where polarizers are frequently used to polarize the incident light.^{1,37}

Although reciprocal optical devices can have AT of light under incident light with certain modes in the two opposite directions, they cannot guarantee AT of light in real applications where incident light modes can be arbitrary, for example, parasitic reflections can excite an arbitrary ensemble of modes.⁴³ In other words, reciprocal optical devices cannot consistently have the diode-like behavior, and their AT of light is conditional

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https://doi.org/10.1016/j.isci. 2023.107032









on the incident light in optical circuits. Despite this, reciprocal optical devices with AT are still being developed by different researchers and are referred to as optical diodes. This raises the question of whether reciprocal optical devices with conditional AT should be named optical diodes. One thing that can be referred to concerning the terminology of "diode" and the common diode-like behavior is the electronic diode. In comparison, for electronic diodes that are fabricated to have asymmetric electrical conductivity (AT of electrons), their diode-like property is consistent (below the breakdown voltage), and not conditional on the flowing electrons in electric circuits. Apparently, there never comes a similar question in the field of electronics as in the field of optics.

In this paper, AT of optical devices was theoretically analyzed based on the scattering matrix theory. Although the Lorentz reciprocity theorem is strong evidence that reciprocal optical devices breaking space-inverse symmetry cannot guarantee AT regardless of the incident modes, the theorem itself only extends to the symmetry of scattering matrixes describing the transmission property of optical devices, i.e., symmetric modal transmission. Integrated mathematical derivations as solid proof instead of evidence of this point are lacking. Therefore, in this study, it is mathematically proven that although reciprocal optical devices breaking space-inverse symmetry can show AT under controlled incident modes (i.e., conditional AT), reciprocal optical devices whose transmission properties are described by symmetric scattering matrices cannot guarantee AT regardless of incident light modes, whereas only nonreciprocal optical devices whose transmission properties are described by asymmetric scattering matrices can possibly guarantee AT with arbitrary incident modes. To the best of our knowledge, it is the first time that this argument is proven mathematically. The clarification of requirements for optical diodes can help avoid further futile efforts on developing optical diodes with reciprocal materials. Besides, the thermodynamics related with AT of optical devices are discussed. This discussion can help prevent the improper use of reciprocal optical devices with conditional AT to function as optical diodes. Furthermore, by comparing the AT of light of optical devices with the AT of electrons of electronic devices, the awareness of their intrinsic differences was raised for the first time to suggest the accurate use of the terminology "diode". Flowing electrons in electric circuits make no difference to electronic devices, so electronic devices being fabricated to have asymmetric electrical conductivity (AT of electrons) would have the property consistently. In contrast, electromagnetic waves in optical circuits are characterized by different modes, which may have different transmittivities even in a same waveguide. In other words, they make a difference to optical devices, so reciprocal optical devices showing conditional AT would fail to exhibit AT consistently when incident modes change. It is argued that reciprocal optical devices, even having conditional AT, are intrinsically different from electronic diodes in terms of the transferring physical quantities, and hence, should not be called optical diodes.

RESULTS

AT of optical devices

As mentioned previously, one main group of the reciprocal optical devices with AT is diffraction gratings. The common feature of these diffraction gratings is that they are structurally asymmetric in the two opposite directions. As shown in Figure 1A, a diffraction grating with a tapered structure is taken as an example to represent the various spatially asymmetric and reciprocal diffraction gratings (SARDG). Assuming a SARDG is passive (the grating adds no energy to the light), the outgoing light L_{out} with different modes (a'_i, b'_i) can be related to the incoming light L_{in} (a_i, b_i) as $L_{out} = SL_{in}$, where **S** is the scattering matrix.

The corresponding expanded form of $L_{out} = SL_{in}$ is given in Equation 1, where the on-diagonal $S_{a'i,ai}$ and $S_{b'i,bi}$ are reflection coefficients from a_i and b_i to a'_i and b'_i , respectively, the off-diagonal coefficients are transmission coefficients, for example, $S_{a'i,aj}$ ($i \neq j$) and $S_{a'i,bj}$ are transmission coefficients from a_j and b_j to a'_i , respectively.

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Figure 1. Schematic of SARSG and two different cases of light scattering

(A) SARDG with incoming and outgoing light on the upper side $(a_i \text{ and } a'_i)$ and lower side $(b_i \text{ and } b'_i)$, respectively. (B) Case 1. The incoming light on the upper side is a single mode plane wave only, as denoted by a_1 . The reflected light back into the same mode is a'_1 . The transmitted light to the lower side of various modes, i.e., different diffraction orders, are denoted by b'_1 , b'_2 , b'_3 , \cdots , where b'_1 denotes the zero-order diffraction light and b'_2 , b'_3 , \cdots denotes nonzero-order diffraction light.

(C) Case 2. The incoming light on the lower side is a single mode plane wave only, as denoted by b_1 . The reflected light back into the same mode is b'_1 . The transmitted light to the upper side of various modes, i.e., different diffraction orders, are denoted by a'_1 , a'_2 , a'_3 , \cdots , where a'_1 denotes the zero-order diffraction light and a'_2 , a'_3 , \cdots denotes nonzero-order diffraction light. Although it should have other reflection modes on the upper side (a'_2 , a'_3 , \cdots) in Case 1 and on the lower side (b'_2 , b'_3 , \cdots) in Case 2, they are deliberately neglected as the analysis focuses on the transmission to the other side only.

Consider Case 1 and Case 2 as shown in Figures 1B and 1C, respectively. According to the scattering matrix in Equation 1, the transmitted power into the other side in Case 1 and Case 2 is calculated with Equations 2 and 3, respectively.

 $P_{Case1} = \begin{bmatrix} b_1' \\ b_2' \\ b_3' \\ \vdots \end{bmatrix}^{T} \begin{bmatrix} b_1' \\ b_2' \\ b_3' \\ \vdots \end{bmatrix} = |b_1'|^2 + |b_2'|^2 + |b_3'|^2 + \dots = (|S_{b'1,a1}|^2 + |S_{b'2,a1}|^2 + |S_{b'3,a1}|^2 + \dots)|a_1|^2,$

(Equation 2)

$$P_{Case2} = \begin{bmatrix} a_1' \\ a_2' \\ a_3' \\ \vdots \end{bmatrix}^+ \begin{bmatrix} a_1' \\ a_2' \\ a_3' \\ \vdots \end{bmatrix} = |a_1'|^2 + |a_2'|^2 + |a_3'|^2 + \dots = (|S_{a'1,b1}|^2 + |S_{a'2,b1}|^2 + |S_{a'3,b1}|^2 + \dots)|b_1|^2,$$

(Equation 3)

In Equations 2 and 3, the superscript ' + ' denotes the conjugate transpose of matrix. To compare the transmitted power in Case 1 and Case 2, the transmission coefficients in Equations 2 and 3 are marked with a red and blue solid line box in the corresponding scattering matrix, respectively, as shown in Figure 2. As the SARDG is reciprocal, the scattering matrix must be symmetric according to the Lorentz reciprocity theorem. So only $S_{b'1,a1}$ in Equation 2 equals $S_{a'1,b1}$ in Equation 3, while $S_{b'2,a1}$ does not necessarily equal $S_{a'3,b1}$, and so on. This explains the different transmitted power in Case 1 and Case 2, which is attributed to the different transmittivities of nonzero-order diffraction modes





Figure 2. Scattering matrix of a reciprocal optical device

As the SARDG is reciprocal, the transmission coefficients in the red solid line box equal those in the red dash line box, respectively, but do not equal those in the blue solid line box. $S_{b'1,a1} = S_{a'1,b1}$ means that the transmittivities of the zero-order diffraction mode in the two opposite directions are equal. $S_{b'j,a1}$ does not necessarily equal $S_{a'j,b1}$ ($j \neq 1$), meaning that the transmittivities of nonzero-order diffraction modes in the two opposite directions are not necessarily equal.

in the two opposite directions. Clearly, when the modal amplitude of a_1 is equal to b_1 , i.e., $|a_1|^2 = |b_1|^2$, transmittivities in Case 1 and Case 2 are different, i.e., AT. Actually, as the scattering matrix is symmetric for reciprocal optical devices according to the Lorentz reciprocity theorem, the transmission coefficients must satisfy $S_{b'j,ai} = S_{a'i,bj}$. This equality is the symmetric model transmittivity. In contrast, $S_{b'j,ai}$ does not necessarily equal $S_{a'j,bi}$. The equality and inequality are further explained in Figure 3.

Mathematical proof

AT realized by reciprocal optical devices as shown previously are under the conditions that the incident light on the upper and lower sides is the same single mode plane wave. Under a different condition, the AT will no longer exist. This is proven as follows. For convenience, Equation 1 is rewritten as:

$$\begin{bmatrix} A'\\B' \end{bmatrix} = \begin{bmatrix} S_{A'A} & S_{A'B}\\S_{B'A} & S_{B'B} \end{bmatrix} \begin{bmatrix} A\\B \end{bmatrix},$$
 (Equation 4)

where A and B stand for the column vector of $[a_1 a_2 a_3...]^T$ and $[b_1 b_2 b_3...]^T$, respectively (the superscript 'T' denotes the matrix transpose); A' and B' stand for the column vector of $[a'_1 a'_2 a'_3 ...]^T$ and $[b'_1 b'_2 b'_3 ...]^T$, respectively; $S_{A'A}$ stands for the coefficients in the upper left quadrant of the scattering matrix in Equation 1, and $S_{A'B}$ for the upper right quadrant, $S_{B'A}$ for the lower left quadrant, and $S_{B'B}$ for the lower right quadrant. Considering Case 3 where there is a light containing some modes incident on the upper side only, i.e., $[A \ 0]^T$, and Case 4 where the incident light is on the lower side only, i.e., $[0 \ B]^T$, the corresponding relationships between outgoing and incoming light for Cases 3 and 4 according to Equation 4 are given in Equations 5 and 6, respectively.

$$\begin{bmatrix} A'\\B' \end{bmatrix} = \begin{bmatrix} S_{A'A} & S_{A'B}\\S_{B'A} & S_{B'B} \end{bmatrix} \begin{bmatrix} A\\0 \end{bmatrix},$$
 (Equation 5)

$$\begin{bmatrix} A'\\B' \end{bmatrix} = \begin{bmatrix} S_{A'A} & S_{A'B}\\S_{B'A} & S_{B'B} \end{bmatrix} \begin{bmatrix} 0\\B \end{bmatrix},$$
 (Equation 6)

Then, the transmitted power to the lower side in Case 3 and to the upper side in Case 4 is given in Equations 7 and 8, respectively.

$$B'^{+}B' = (S_{B'A}A)^{+}S_{B'A}A = A^{+}S_{B'A}^{+}S_{B'A}A,$$
 (Equation 7)

$$A'^{+}A' = (S_{A'B}B)^{+}S_{A'B}B = B^{+}S_{A'B}^{+}S_{A'B}B,$$
 (Equation 8)

And the transmittivities for Cases 3 and 4 are given in Equations 9 and 10, respectively.

$$\tau_{Case3} = \frac{A^+ S^+_{B'A} S_{B'A} A}{A^+ A},$$
 (Equation 9)







Figure 3. Schematic of equality and inequality in transmission coefficients

(A) $S_{b'j,ai}$ is the transmission coefficient of the incoming light a_i to the outgoing light b'_j . $S_{a'i,bj}$ is the transmission coefficient of the incoming light b_j to the outgoing light a'_i . The equality $S_{b'j,ai} = S_{a'i,bj}$ means that transmitted light can always trace the same way back to its original incident light with the same transmittivity in reciprocal optical devices. (B) $S_{a'j,bi}$ is the transmission coefficient of the incoming light b_i to the outgoing light a'_j . The inequality between $S_{b'j,ai}$ and $S_{a'j,bi}$ ($i \neq j$) means even when two incident light in the two opposite directions have the same mode, their transmittivities to the same other modes are not necessarily the same in reciprocal optical devices.

$$\tau_{Case4} = \frac{B^+ S_{AB}^+ S_{A'B} S_{A'B} B}{B^+ B},$$
 (Equation 10)

As the scattering matrix is symmetric for the SARDG, $S_{A'B}$ equals $S_{B'A}^T$. τ_{Case4} in Equation 10 can be rewritten as:

$$\tau_{Case4} = \frac{B^+ S_{B'A} S_{B'A}^+ B}{B^+ B},$$
 (Equation 11)

Using *M* to denote $S^+_{B'A}S_{B'A}$ in Equation 9, *N* to denote $S_{B'A}S^+_{B'A}$ in Equation 11, since *M* and *N* are Hermitian matrices, *M* and *N* have the same real eigenvalues λ_i . The incident light $[A 0]^T$ in Case 3 and the incident light $[0 B]^T$ in Case 4 can be represented as $A = \sum_{i=1}^{n} x_i u_i$ and $B = \sum_{i=1}^{n} y_i v_i$, respectively, where u_i and v_i are the orthogonal eigenvectors group of *M* and *N*, respectively, in corresponding to the eigenvalues λ_i , and x_i and y_i are the corresponding coefficients. Subsequently, τ_{Case3} in Equation 9 and τ_{Case4} in Equation 11 can be rewritten as Equations 12 and 13, respectively.

$$\tau_{Case3} = \frac{\sum_{i=1}^{n} \lambda_i |x_i|^2}{\sum_{i=1}^{n} |x_i|^2},$$
 (Equation 12)
$$\tau_{Case4} = \frac{\sum_{i=1}^{n} \lambda_i |y_i|^2}{\sum_{i=1}^{n} |y_i|^2},$$
 (Equation 13)

Clearly, $\lambda_{min} \leq \tau_{Case3} \leq \lambda_{max}$ and $\lambda_{min} \leq \tau_{Case4} \leq \lambda_{max}$, where λ_{min} and λ_{max} are the minimal and maximal values of the eigenvalues λ_i , respectively. This means τ_{Case3} and τ_{Case4} have the same upper and lower limit values. Since the coefficients y_i can be arbitrary, for the light having any given modes incident on the upper side only, i.e., $[A \ 0]^T$, there must exist a light with a combination of modes incident on the lower side only, i.e., $[O \ B]^T$, that their transmittivities to the opposite side being equal. Consequently, it can be proved that the AT of the SARDG is not guaranteed. In contrast, when the optical device is nonreciprocal, the corresponding scattering matrix is asymmetric, and hence $S_{A'B}$ does not equal $S_{B'A}^T$. Then, the value range of τ_{Case3} can be larger than λ_{max} of τ_{Case4} . In this way, τ_{Case3} can have a value where τ_{Case4} can never be ($\lambda_{min,Case4} - \lambda_{min,Case3}$ and $\lambda_{max,Case3} - \lambda_{max,Case4}$). Consequently, it is proven that nonreciprocity is necessary for optical devices to guarantee AT.



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Figure 4. A SARDG fabricated with a 3D printer

(A) Cross-sectional geometry of the SARDG. The grating is composed of isosceles triangles placed equidistantly, the height, bottom span, and space of the grating are 50 μ m, 12 μ m, and 12 μ m, respectively.

(B) The 3D model of the SARDG. The gratings having an area of 5.6 mm \times 6 mm were surrounded by a rectangular frame as a support.

(C) The real object of the 3D-printed SARDG.

(D) The SEM image (top view) of the 3D-printed SARDG after sputtering with 1 μm silver on its surface. The white stripes represent the two equal sides of the isosceles triangles of the gratings.

(E) The microscope image (side view) of the 3D-printed SARDG.

Experimental and numerical demonstrations

To further support this argument, a SARDG was fabricated using a 3D printer (BMF NanoArch S130) and sputtering machine, as shown in Figure 4. The grating was first printed by the 3D printer with the UV-curing resin and then sputtered with silver. The SARDG is composed of isosceles triangles placed equidistantly, the height, bottom span, and space of the grating are 50 μ m, 12 μ m, and 12 μ m, respectively. The geometry of the SARDG was based on the optimized finite-difference time-domain (FDTD) simulation results that the SARDG has a high contrast ratio of AT under controlled incident modes, as indicated in Figure 5A. In the FDTD simulation, the modes of the incident linearly polarized plane waves in the forward and backward directions were the same. Clearly, the transmittance in the forward direction is about 2 times larger than the transmittance in the backward direction. However, the transmittances of the fabricated SARDG in the forward and backward directions measured by the FTIR (Fourier Transform Infrared Spectroscopy, SHIMADZU IRaffinity-1S) were almost the same, as shown in Figure 5B. In other words, the AT shown in the FDTD simulation by the SARDG is conditional. Under a different condition of incident waves in the forward and backward directions, for example, the condition in the experimental measurement where incident waves in the forward and backward directions are nonpolarized and contain a combination of arbitrary modes, the SARDG shows nearly no AT.







Figure 5. Transmittance of the SARDG in the forward and backward directions (A) Numerical results by the FDTD simulation. (B) Experimental results measured by the FTIR.

DISCUSSION

Discussions on thermodynamics

As the transfer of light is closely related with radiative heat transfer, AT of light of optical devices raises confusion about whether optical devices with AT will break the second law of thermodynamics,⁴⁴ which in the Clausius statement is that heat can never transfer from a colder to a warmer body without some other change.⁴⁵ For example, when it is assumed that two bodies with different temperatures only exchange heat radiatively through an optical device with AT in an isolated system, whether the heat will transfer from Body 1 with lower temperature (T_1) to Body 2 with high temperature (T_2) spontaneously if the transmittivity is larger from Body 1 to Body 2 than the transmittivity in the reverse direction, as shown in Figure 6. The discussions can be separated for cases of reciprocal optical devices with conditional AT and cases of nonreciprocal optical diodes. As the system is isolated, it can be further assumed that one body's radiations reach the other body through the optical devices only.

For cases of reciprocal optical devices with conditional AT, the reciprocal optical devices with conditional AT are assumed to have no radiation absorption ideally. Even when the two bodies managed to only emit thermal radiations whose modes have AT (the transmittivity from Body 1 to Body 2 is larger than that in the reverse direction) through the reciprocal optical devices, heat can still never flow from colder Body 1 to warmer Body 2 spontaneously. This is because the reflected thermal radiations of Body 2 by the reciprocal optical device with conditional AT would be continuously reflected by Body 2. As parasitic reflections contain an arbitrary combination of modes, the reflected modes which do not have AT in the reciprocal optical device with conditional AT. Therefore, even under the ideal cases where the two bodies managed to only release thermal radiations







Figure 6. Isolated system consists of one optical device with AT and two bodies

whose modes have AT through the reciprocal optical devices, the second law of thermodynamics holds and reciprocal optical devices with conditional AT cannot realize the function of optical diodes.

The argument about parasitic reflections explaining why the second law of thermodynamics holds for cases of reciprocal optical devices with conditional AT fails to explain the cases of nonreciprocal optical diodes, as the optical diodes having consistent AT anyway and hence the second law of thermodynamics would be violated seemingly. However, the fact is that the optical diodes themselves would have absorptions in real cases. The radiation from Body 2 would be absorbed by the optical diodes. Nonetheless, optical diodes can help protect the body emitting the light from back reflections, which is useful in some applications such as laser systems.

Discussions of different diode-like behaviors

As mathematically proven and discussed previously, although reciprocal optical devices can have AT under conditions of controlled incident modes, they cannot have AT consistently like optical diodes which have nonreciprocity or modal asymmetry. Hence, it is suggested that reciprocal devices with conditional AT cannot be called optical diodes. Note that the widely used electronic diodes nowadays also have the diode-like property, i.e., asymmetric electric conductivity (AT of electrons), as shown in Figure 7. Flowing electrons can easily pass in one direction but hardly pass in the opposite direction. This diode-like behavior of unidirectional conductance is called rectification.

Electronic diodes will always exhibit the asymmetric electric conductivity property consistently in any electric circuit (excluding the cases above breakdown voltage). There is no electronic diode that has asymmetric electric conductivity in one electric circuit but does not have asymmetric electric conductivity in another electric circuit. In contrast, the so-called optical diodes (reciprocal optical devices with conditional AT) can exhibit AT of light in some cases of controlled incident light, but cannot exhibit AT of light consistently.

In view of this, we believe that the inappropriate use of the terminology "optical diode" for reciprocal optical devices having conditional AT could be related with insufficient awareness of the difference in the physical quantity transferring in optical and electronic devices. From the perspective of mathematics, the electrical conductivity of electronic diodes is a scalar (some materials have electrical conductivity in tensor form, but it is not relevant to our discussions here), though they have a nonlinear behavior (nonlinear current-voltage characteristics of electronic diodes), whereas the optical transmittivity of optical devices should be a matrix, for example, $L_{out} = SL_{in}$ as presented in section of AT of optical devices. The difference between the scalar of the electrical conductivity of electronic diodes and the matrix of the optical transmittivity of optical devices reveals the difference in the physical quantity transferring in optical and electronic devices, but traveling light in optical circuits is different for optical devices, since traveling light, considered as electromagnetic waves, has different modes. Actually, electromagnetic waves are characterized by different modes, which may have different transmittivities even in the same waveguide of optical devices.

Based on the aforementioned observations, it can be easily understood that when a reciprocal optical device is demonstrated to have AT under incident light which has certain modes, it cannot guarantee AT regardless of the modes of incident light. The modes of incident light of an optical device can be manipulated, but the electrons in an electrical circuit cannot since the electrons make no difference to electric devices in electrical circuits. Therefore, even when a reciprocal optical device is demonstrated to have AT, it cannot consistently have AT regardless of incident modes. Such kind of circumstance does not happen to electronic devices. The practice of naming optical devices as optical diodes when demonstrating that they can exhibit AT of light, without checking the reciprocity or scattering matrix of the optical



Figure 7. Schematic of electronic diodes

(A) Forward direction. Electrons can easily pass through the electronic diode in the forward direction, forming an electric current.

(B) Reverse direction. Electrons can hardly pass through the electronic diode in the backward direction, blocking the transmission of electrons effectively.

devices, is natural given the idea of electronic diodes, but it is actually not accurate. Reciprocal optical devices with conditional AT cannot function as optical diodes. Instead, only nonreciprocal optical devices which have modal asymmetry may function as optical diodes. Hence, it is suggested that reciprocal devices with conditional AT cannot be called optical diodes.

Conclusions

In conclusion, this study mathematically proved that AT of reciprocal optical devices, even breaking spaceinverse symmetry, is only realized by limiting incident light modes, reciprocal optical devices cannot guarantee AT regardless of incident light modes. Nonreciprocity (breaking time-inverse symmetry or the Lorentz reciprocity) is necessary for optical devices to be optical diodes. The thermodynamics related with the AT of optical devices was also discussed to explain the validity of the second law of thermodynamics. Additionally, it explains why calling reciprocal optical devices as optical diodes, even with conditional AT, is inaccurate through the comparison with electronic diodes and thermal diodes. The key point is that traveling light has different modes, which may have different optical transmittivities in optical devices, but flowing electrons do not have different transmittivities in electric devices. When a reciprocal optical device is shown to have AT under incident light with certain modes, it will not necessarily have AT under incident light with other modes. But electric diodes will have this AT property consistently. Therefore, reciprocal optical devices with conditional AT cannot function as optical diodes. Only nonreciprocal optical devices that have asymmetric scattering matrix, i.e., the asymmetrical modal transmission can be called optical diodes. The idea of electronic diodes and the unawareness of the difference in the physical quantities, i.e., traveling light and flowing electrons, might cause the incautious use of the terminology "optical diode" for reciprocal optical devices with conditional AT. This study can help to avoid the futile efforts of developing optical diodes with reciprocal materials and avoid the mistaken use of reciprocal optical devices to have AT function in real applications where incident light can be arbitrary. Also, this study can help settle the dispute over the inaccurate use of "optical diode" for reciprocal optical devices with conditional AT.

Limitations of the study

In this study, due to the unavailability of the nonreciprocal materials and limited fabrication equipment, real optical diodes are not fabricated for demonstration of AT, which can be found in some existing research.^{4,11-13}

STAR*METHODS

Detailed methods are provided in the online version of this paper and include the following:

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SUPPLEMENTAL INFORMATION

Supplemental information can be found online at https://doi.org/10.1016/j.isci.2023.107032.

ACKNOWLEDGMENTS

The funding for this research is provided by the Hong Kong Research Grant Council (RGC) via General Research Fund (GRF) account 16200518.

AUTHOR CONTRIBUTIONS

Conceptualization, C.Y.T.; Methodology, C.Y.T. and A.P.; Investigation, C.Y.T., K.L., and A.P.; Writing— Original Draft, A.P.; Writing—Review & Editing, C.Y.T., A.P., K.L., and S.C.; Supervision, C.Y.T.; Project Administration, C.Y.T.; Funding Acquisition, C.Y.T.

DECLARATION OF INTERESTS

The authors declare no competing interests.

INCLUSION AND DIVERSITY

We support inclusive, diverse, and equitable conduct of research.

Received: November 10, 2022 Revised: February 10, 2023 Accepted: May 30, 2023 Published: June 7, 2023

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STAR*METHODS

KEY RESOURCES TABLE

REAGENT or RESOURCE	SOURCE	IDENTIFIER
Chemicals, peptides, and recombinant proteins		
HTL resin	BOSTON MICRO FABRICATION	https://bmf3d.com/micro-3d-printing-materials/
Silver	中诺新材 (http://www.znxc.com/)	CAS: 7440-22-4
Software and algorithms		
Ansys Lumerical FDTD	Ansys	https://www.ansys.com/products/photonics/fdtd
Other		
3D printer (BMF nanoArch S130)	BOSTON MICRO FABRICATION	https://www.bmftec.cn/3d-printers/s130
Fourier-transform infrared spectroscopy (IRAffinity-1S, SHIMADZU)	SHIMADZU	https://www.shimadzu.com/an/products/molecular- spectroscopy/ftir/ftir-spectroscopy/iraffinity-1s/index.html

RESOURCE AVAILABILITY

Lead contact

Further information and requests for resources and reagents should be directed to and will be fulfilled by the lead contact, Chi Yan Tso (chiytso@cityu.edu.hk).

Materials availability

This study did not generate new unique reagents.

Data and code availability

- All data reported in this paper will be shared by the lead contact upon request.
- This paper does not report original code.
- Any additional information required to reanalyze the data reported in this paper is available from the lead contact upon request.

METHOD DETAILS

Basic knowledge for theoretical analysis

Light can be modeled by Maxwell equations when considered as electromagnetic waves. Maxwell equations for electromagnetic waves in a medium with no source (no free charge or conduction current) are:

$$\nabla \times E = -\frac{\partial B}{\partial t},$$
 (Equation 14)

$$\nabla \times H = \frac{\partial D}{\partial t},$$
 (Equation 15)

$$\nabla \cdot \boldsymbol{D} = \boldsymbol{0}, \tag{Equation 16}$$

$$\nabla \cdot \boldsymbol{B} = \boldsymbol{0}, \tag{Equation 17}$$

In Equations 14, 15, 16, and 17, E and H are the electric and magnetic fields respectively, D and B are the electric and magnetic flux density respectively, and t is the time. For time-harmonic electromagnetic fields (field's time dependence $e^{j\omega t}$, where j is an imaginary unit, ω is frequency), Maxwell curl equations can be written as:

$$\nabla \times E = -j\omega\mu H$$
, (Equation 18)

$$\nabla \times H = j\omega \varepsilon E, \qquad (Equation 19)$$





In Equations 18 and 19, ϵ and μ are the medium permittivity and permeability respectively. They are in bold to indicate that they are tensors, but they could also be scalars (ϵ and μ), depending on the medium properties which will be explained later on in detail. One assumption has been made for Equations 18 and 19 that the medium permittivity and permeability, whether they are tensors or scalars, are assumed to be time-invariant/time-independent when calculating the partial derivative of **B** and **D** with respect to time.

Next, the concept of electromagnetic waveguides and modes are introduced. In general, an optical waveguide is defined as a physical structure that guides electromagnetic waves in the directions parallel to its axis and confines the waves within or near its surfaces.⁴⁶ More generally, it can be defined as a channel that is uniform along the direction of its axis.⁴⁷ Specifically, a free space can also be considered as a waveguide because it is uniform along all directions. The propagating electromagnetic waves have various field patterns, which can be described by electromagnetic modes. Mathematically, electromagnetic modes are the eigen solutions of sourceless Maxwell equations. Electromagnetic modes are normal modes for electromagnetic fields with no source, having a $e^{-j\gamma z}$ z-dependence, where z is the propagation direction and γ is the propagation constant of the modes.⁴⁷ Modes in waveguides can be classified into transverse modes and hybrid modes. Transverse modes have either their electric or magnetic field, or both fields entirely transverse to their propagation directions, while hybrid modes have both electric and magnetic fields in their propagation direction. Specifically, modes in free space are plane waves with both their electric and magnetic fields orthogonal to the propagation direction. Free space plane waves vary as $e^{-j(\alpha x + \beta y + \gamma z)}$, where α and β are purely real numbers. Different waveguide modes may have various phase velocity, group velocity, cross-sectional intensity distribution, and polarization.⁴⁶ In optics, a mode represents an electromagnetic wave with a certain frequency, polarization, and direction of propagation.

Scattering matrix theory

Scattering matrices represent relationships between input and output electromagnetic waves of passive optical devices/circuits, providing the global transmission property of passive optical circuits. Consider an optical circuit as shown in Figure S1, it is enclosed inside the fictious surface Ω . The optical circuit has incoming and outgoing electromagnetic waves through waveguides, which can be physical optical structures or free space channels. In general, waveguides of optical circuits can be considered as "ports" having pieces of cross-section planes. The theory of scattering matrix is based on the following assumptions: all the electromagnetic waves in the ports contain no evanescent mode.

Assuming the propagation axis of each waveguide mode points toward the optical circuit, the local modal expansion of the modes in each waveguide can be written as:

$$\boldsymbol{E}_{T,i}(\boldsymbol{x},\boldsymbol{y},\boldsymbol{z}) = (a_i \boldsymbol{e}^{-j\gamma_i \boldsymbol{z}} + a'_i \boldsymbol{e}^{j\gamma_i \boldsymbol{z}}) \boldsymbol{e}_{T,i}(\boldsymbol{x},\boldsymbol{y}), \qquad (\text{Equation 20})$$

$$H_{T,i}(x,y,z) = (a_i e^{-j\gamma_i z} + a'_i e^{j\gamma_i z}) h_{T,i}(x,y),$$
 (Equation 21)

In Equations 20 and 21, x, y, z are local coordinates for each waveguide mode, *i* is the mode index, a_i and a'_i are the complex amplitude of the incoming (z) and outgoing (-z) light of mode *i* respectively. In each waveguide, any two modes, as indicated by mode index *i* and *j*, satisfy the following orthogonal condition that

$$\iint_{\Omega} \mathbf{e}_i \times \mathbf{h}_j^* d\mathbf{\Omega} = 2p_0 \delta_{ij}, \qquad (\text{Equation 22})$$

In Equation 22, * is the complex conjugate, p_0 is the unit power, and δ_{ij} is the Kronecker delta. Based on Equation 22, the electromagnetic waves in each waveguide of the optical circuit can be represented by a set of modes, each of which can be as expressed with Equations 20 and 21. Furthermore, by assigning the same modes in different waveguides with different mode indices, Equation 22 is valid on the whole $\boldsymbol{\Omega}$. Then, the whole set of local model expansions of the modes in the waveguides of the optical circuit, describing all the incoming and outgoing electromagnetic waves, can be expressed as:

$$\boldsymbol{E}_{T} = \sum_{i} (\boldsymbol{a}_{i} + \boldsymbol{a}_{i}') \boldsymbol{e}_{T,i}, \qquad (\text{Equation 23})$$





$$H_{T} = \sum_{i} (a_{i} - a_{i}') h_{T,i}, \qquad (Equation 24)$$

Based on Equations 23 and 24, the power carried by the incoming and outgoing electromagnetic waves of an optical device can be described by the amplitudes (a_i, a'_i) of the mode *i* in the waveguides of the optical device as:

$$P_{in} = \sum_{i} a_{i}^{+} a_{i}, \qquad (Equation 25)$$

$$P_{\text{out}} = \sum_{i} a'_{i} a'_{i}, \qquad (\text{Equation 26})$$

In Equations 25 and 26, + denotes the complex conjugate transpose. Using L_{in} to represent the column vector containing a_i , and L_{out} to represent the column vector containing a'_i , the relationship between L_{in} and L_{out} can be written as:

$$L_{out} = SL_{in},$$
 (Equation 27)

where **S** is the scattering matrix of the optical circuit. The scattering matrix is a fundamental tool to analyze the transmission property of optical devices. Then, the power that enters and leaves the optical device can be calculated as:

$$P_{in} = L_{in}^+ L_{in}, \qquad (Equation 28)$$

$$P_{out} = L_{out}^+ L_{out}, \qquad (Equation 29)$$

Lorentz reciprocity theorem

The derivation of Lorentz reciprocity starts from Maxwell curl equations in Equations 18 and 19. Consider two different states of a passive optical circuit, the electromagnetic fields in correspondence to the two different states are denoted by subscripts 1 and 2, respectively. According to Equations 18 and 19, the relation between electric fields and magnetic fields are:

$$\nabla \times E_1 = -j\omega\mu H_1,$$
 (Equation 30)

$$\nabla \times H_1 = j\omega \varepsilon E_1,$$
 (Equation 31)

$$\nabla \times E_2 = -j\omega\mu H_2, \qquad (Equation 32)$$

$$\nabla \times H_2 = j\omega \varepsilon E_2,$$
 (Equation 33)

Consider the quantity $\nabla \cdot (E_1 \times H_2 - E_2 \times H_1)$, expanding which gives:

$$\nabla \cdot (E_1 \times H_2 - E_2 \times H_1) = H_2 \cdot (\nabla \times E_1) - E_1 \cdot (\nabla \times H_2) - H_1 \cdot (\nabla \times E_2) + E_2 \cdot (\nabla \times H_1), \quad \text{(Equation 34)}$$

Substituting Equations 30, 31, 32, and 33 into Equation 34, it can be derived that:

$$\nabla \cdot (\mathbf{E}_1 \times \mathbf{H}_2 - \mathbf{E}_2 \times \mathbf{H}_1) = j\omega(-\mathbf{H}_2 \mu \mathbf{H}_1 - \mathbf{E}_1 \boldsymbol{\varepsilon} \mathbf{E}_2 + \mathbf{H}_1 \mu \mathbf{H}_2 + \mathbf{E}_2 \boldsymbol{\varepsilon} \mathbf{E}_1), \quad (\text{Equation 35})$$

When the medium permittivity and permeability are scalars (ϵ and μ) or symmetric tensors, i.e., $\epsilon = \epsilon^T$ and $\mu = \mu^T$, the right-hand side of Equation 35 equals zero. Then,

$$\nabla \cdot (\mathbf{E}_1 \times \mathbf{H}_2 - \mathbf{E}_2 \times \mathbf{H}_1) = \mathbf{0}, \tag{36}$$

It should be noted that Equation 36 is the Lorentz reciprocity theorem. It can be seen from the derivation of the Lorentz reciprocity theorem that it applies to optical devices which are made of materials whose permittivity and permeability are time-invariant and described by either scalars or symmetric tensors.

For linear materials, electric flux density D and magnetic flux density B are proportional to electric field Eand magnetic field H respectively. For isotropic materials, material permittivity and permeability are independent of fields directions. In other words, the permittivity and permeability of linear and isotropic materials are scalars that $D = \epsilon E$ and $B = \mu H$. So, Lorentz reciprocity applies to optical devices made by linear and isotropic materials. For nonlinear materials, permittivity is a function of field strength, for example, permittivity is $\epsilon(E)$ for nonlinear isotropic materials. As a result, in Equation 35, the terms $-E_1\epsilon E_2 + E_2\epsilon E_1$ should be $-E_1\epsilon(E)E_2 + E_2\epsilon(E)E_1$, which is not necessarily zero for arbitrary E_1





and E_2 . Hence, Lorentz reciprocity does not apply to optical devices made by nonlinear materials, no matter whether the materials are isotropic or anisotropic. For linear and anisotropic materials, permittivity and permeability are described by tensors. When $e = e^T$ and $\mu = \mu^T$, $E_1eE_2 = E_2eE_1$ and $H_1\mu H_2 = H_2\mu H_1$, implying that the right-hand side of Equation 35 also equals zero. This means that Lorentz reciprocity applies to linear and anisotropic materials with symmetric permittivity and permeability tensors.

In summary, Lorentz reciprocity applies to passive optical devices made of materials that are time-invariant, linear, and isotropic or anisotropic but with symmetric permittivity and permeability tensors. Conversely, Lorentz reciprocity does not hold in optical devices that are active, or made of materials that are time-dependent, or nonlinear, or anisotropic with asymmetric permittivity and permeability tensors. Examples of optical devices which Lorentz reciprocity does not apply to have been given in the literature,⁴³ for example, the magneto-optical materials which have asymmetric permittivity and permeability tensors.

Scattering matrix of reciprocal optical devices

It has been introduced that Lorentz reciprocity applies to passive optical devices constituting materials that are time-invariant, linear, and isotropic or anisotropic but having symmetric permittivity and permeability tensors. The optical devices satisfying Lorentz reciprocity are reciprocal. One important property of reciprocal optical devices is that their scattering matrix is symmetric. This property is proved as follows. Consider two different states of an optical circuit, one state has the electromagnetic fields denoted as E_1 and H_1 , and the incoming waves denoted as $L_{in,1}$ (a_1, a_2, a_3, \cdots) and the outgoing waves denoted as $L_{out,1}$ (a'_1, a'_2, a'_3, \cdots). The other state has the electromagnetic fields denoted as $L_{in,2}$ (b_1, b_2, b_3, \cdots) and the outgoing waves denoted as $L_{in,2}$ (b_1, b_2, b_3', \cdots). According to Equations 23 and 24, substituting the model expansions of the modes in correspondence to the two states into Equation 36 gives:

$$2\sum_{i}\sum_{j}\left(a'_{i}b_{j}-a_{i}b'_{j}\right)\iint_{\Omega}\mathbf{e}_{i}\times\mathbf{h}_{j}d\boldsymbol{\Omega}=0, \tag{Equation 37}$$

As the ports are assumed to be lossless, h_j equals h_j^* . Then, according to Equation 22, Equation 37 can be written as:

$$\sum_{i} (a'_{i}b_{i} - a_{i}b'_{i}) = 0, \qquad (Equation 38)$$

Substituting in the incoming and outgoing waves of the two different states into Equation 38, it can be derived that

$$L_{out,1}^{T}L_{in,2} - L_{in,1}^{T}L_{out,2} = (SL_{in,1})^{T}L_{in,2} - L_{in,1}^{T}SL_{in,2} = L_{in,1}^{T}(S^{T} - S)L_{in,2} = 0,$$
 (Equation 39)

 $L_{in,1}^{T}(S^{T} - S)L_{in,2} = 0$ holds for arbitrary L_{in} , so that the scattering matrix S must be symmetric. Therefore, the scattering matrix of a reciprocal optical device must be symmetric.

Materials

UV-curing resin (HTL resin), Silver.

Fabrication of SARDG

SARDG was fabricated using the 3D printer (BMF nanoArch S130) and sputtering machine. The grating was firstly printed by the 3D printer with the UV-curing resin and then sputtered with silver. The SARDG is composed of isosceles triangles placed equidistantly, the height, bottom span, and space of the grating are 50 μ m, 12 μ m, and 12 μ m, respectively.

FDTD simulation of SARDG

FDTD solver on the Ansys platform is used to simulate and optimize the structure of SARDG. The principle of the FDTD simulation is solving the Maxwell equations to acquire the transmittance. Maxwell equations are solved in the time domain for E-field E(x,t) and B-field intensities H(x,t) numerically with finite difference scheme, where x and t denote the position vector and time respectively. The Maxwell equations are expressed as $\frac{\partial D}{\partial t} = \nabla \times H$, $\frac{\partial H}{\partial t} = \frac{1}{\mu_0} \nabla \times E$, where $D(\omega) = \varepsilon_0 \varepsilon_r(\omega) E(\omega)$ (ε_0 is the free space dielectric permeability, $\varepsilon_r(\omega) = n^2$, where n is the refractive index), and μ_0 is the free space magnetic permeability. According to Discrete Time Fourier transform of time domain solutions, incident $P_i(\lambda)$, reflected $P_\rho(\lambda)$, and transmitted





power $P_{\tau}(\lambda)$ spectra can be evaluated by the EM energy flux $S(\omega)$ normal to the measurement planes. Subsequently, reflection $\rho(\lambda)$, transmission $\tau(\lambda)$ and adsorption/emission $\varepsilon(\lambda)$ spectra can be determined by $P_{\rho}(\lambda)/P_i(\lambda)$, $P_{\tau}(\lambda)/P_i(\lambda)$, and $1 - [P_{\rho}(\lambda)/P_i(\lambda)] - [P_{\tau}(\lambda)/P_i(\lambda)]$, respectively. In the simulation, we employed a 2-dimention simulation region. The refractive index of the surrounding is set to be 1. Plane wave sources are used to inject SARDG.

Transmittance measurement of SARDG

The transmittance of SARDG was characterized by using a Fourier-transform infrared spectroscopy (IRAffinity-1S, Shimadzu) with resolution of 0.5 cm^{-1} .