Nonreciprocal infrared absorption via resonant magneto-optical coupling to InAs

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Nonreciprocal elements are a vital building block of electrical and optical systems. In the infrared regime, there is a particular interest in structures that break reciprocity because their thermal absorptive (and emissive) properties should not obey the Kirchhoff thermal radiation law. In this work, we break time-reversal symmetry and reciprocity in n-type–doped magneto-optic InAs with a static magnetic field where light coupling is mediated by a guided-mode resonator structure, whose resonant frequency coincides with the epsilon-near-zero resonance of the doped indium arsenide. Using this structure, we observe the nonreciprocal absorptive behavior as a function of magnetic field and scattering angle in the infrared. Accounting for resonant and nonresonant optical scattering, we reliably model experimental results that break reciprocal absorption relations in the infrared. The ability to design these nonreciprocal absorbers opens an avenue to explore devices with unequal absorptivity and emissivity in specific channels.

INTRODUCTION

There has been great interest and numerous theoretical proposals for nonreciprocal absorbers on the grounds that they do not obey the Kirchhoff thermal radiation law (1–4). Interest in this phenomenon stems from fundamental considerations and applications ranging from infrared (IR) spectroscopy, sensing, and thermal energy conversion efficiency (5, 6). For IR sensing applications, the amount of light absorbed in each channel is something that could be controlled in a nonreciprocal device, where the absorptivity can be tuned from strong in one channel and weak in another and vice versa (7, 8). As we review in the coming section, nonreciprocal absorption achieved through an external magnetic field necessitates that the emission is channeled through the opposite channel. Nonreciprocal absorption is a required functionality to enable thermal energy harvesting exceeding the conversion efficiency constrained by the Shockley-Queisser limit (9) and reaching higher theoretical efficiency limits, such as the Landsberg limit (10–12).

The Kirchhoff thermal radiation law can be expressed as an equality of the absorptivity and emissivity at a given wavelength (λ), polarization, and angle of incidence (θ0) (13–15)

\[ \alpha(\theta_0, \lambda) = e(\theta_0, \lambda) \]  

This equality between the angular and spectral distributions of the emissivity and absorptivity is a direct consequence of reciprocity, which, in a scattering context, takes the form

\[ r(\theta_0, \lambda) = r(-\theta_0, \lambda) \]  

where \( r(\theta_0, \lambda) \) is the incident radiation from \( \theta_0 \) that is not absorbed by the absorber/emitter and reflected through the \( -\theta_0 \) channel and vice versa for \( r(-\theta_0, \lambda) \). The above relationships, however, assume that the emitter/absorber obeys Lorentz reciprocity and does not transmit any of the incident radiation (16). In a nonreciprocal system, the equality in Eq. 2 is broken, and the Kirchhoff law is violated (17). The nonreciprocal behavior of the reflection directly relates to the nonreciprocal thermal radiation. This relation is visualized in Fig. 1 (A and B).

Achievement of nonreciprocity for IR radiation is a subject of widespread investigation. Reciprocity can be broken with linear time–invariant, nonlinear, and linear time–varying platforms (18–21). The use of magneto-optical materials, e.g., ferrites, in basic optical elements such as isolators and circulators has been a fundamental building block in integrated photonics for many decades. The use of bare magneto-optical materials to observe free-space propagation from coupled plasmon-phonon modes has been demonstrated; however, the wavelength regimes are limited to the far IR, and the dependence on magnetic field was not measured (22). Prior experimental work on free-space magneto-optical elements has investigated coupling to surface plasmon resonances at a fixed incidence angle in the Otto configuration in the far-IR regime (23) and polarization rotation in the Faraday geometry (24, 25). Pairing magneto-optical semiconductors with photonic crystals and examining the magnetic field and angular dependence in the IR have not yet been experimentally explored. Furthermore, to date, there has not been a measurement of the interaction of thermal radiation with materials that break reciprocal relations in the far and mid-IR despite an abundance of theoretical designs reporting free-space propagation with magneto-optical elements in this wavelength regime (5, 26, 27). Here, we experimentally demonstrate strong nonreciprocal behavior, breaking the reflectivity relationship in Eq. 2 over a wide range of incident angles in an IR thermal photonic absorber that incorporates magneto-optic InAs with a static external magnetic field (28). To apply the external magnetic field, we use a Halbach array consisting of three permanent magnets with their fields oriented toward a pole piece (Fig. 1C) (29). The pole piece is made of a soft ferromagnetic alloy, which focuses the field so that it is uniform over the sample. We control the strength of the field through the sample by tuning the length of the gap, \( l \), between the two pole pieces (Fig. 1C and D). The maximum field used in this paper is limited by the sample size, 5 mm.

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Shayegan et al., Sci. Adv. 8, eabm4308 (2022) 6 May 2022
RESULTS

Experiment

Our device consists of a low-loss, amorphous silicon (α-Si) guided mode resonance (GMR) waveguide structure fabricated on the surface of a 500-μm-thick, degenerately n-type–doped InAs wafer (Fig. 1, D and E) serving as our magneto-optical material resonantly excited in the Voigt geometry. The patterned α-Si structure on the InAs gives a clear angle-dependent guided mode dispersion relation with distinct resonances for both the zero and nonzero magnetic field cases. While most of the modal field of a guided mode is confined within the low-loss α-Si resonator, some of the modal field penetrates the magneto-optic InAs layer below, which produces the nonreciprocal effect in the presence of a magnetic field (Fig. 1F).

Optical characterization of InAs and resonant IR device design

The doping concentration of the InAs is chosen such that the GMR wavelength coincides with the epsilon-near-zero (ENZ) wavelength (17.3 μm) in the IR. The degenerately doped InAs exhibits a Drude-like optical response with nonzero off-diagonal permittivity values in an applied magnetic field:

\[
\varepsilon(\omega) = \begin{bmatrix} \varepsilon_{xx} & \varepsilon_{xy} & 0 \\ \varepsilon_{yx} & \varepsilon_{yy} & 0 \\ 0 & 0 & \varepsilon_{zz} \end{bmatrix}
\]

In Eqs. 4 to 6, the plasma and cyclotron frequencies are \(\omega_p = \sqrt{\frac{ne^2}{(m_e\varepsilon_0)}}\) and \(\omega_c = eB/m_e\), respectively. Four-point probe measurements of the InAs wafer in conjunction with ellipsometry fits of the values Ψ and Δ, the complex ratio amplitude and phase responses, give a relaxation rate \(\Gamma = 4.5\) THz, a high-frequency limit dielectric constant \(\varepsilon_\infty = 12.3\), a carrier concentration \(n = 1.5 \times 10^{18} \text{ cm}^{-3}\), and an effective electron mass \(m = 0.033 m_e\), where \(m_e = 9.109 \times 10^{-31} \text{ kg}\) (30). The Drude model fits to the experimental values is shown in Fig. 2A. Furthermore, our designs targeted the ENZ wavelength regime (17.3 μm) since the off-diagonal terms of the dielectric function \((\varepsilon_{xy}, \varepsilon_{yx})\) are largest relative to the on-diagonal terms when a magnetic field is applied to the InAs at this wavelength (Fig. 2B).

We focused on controlling the nonreciprocal response around 17.3 μm since it corresponds to the portion of the electromagnetic spectrum exhibiting thermal radiation at or below room temperature.
We implemented this control with a GMR structure, which is commonly used to enhance absorption/reflection by critical coupling in IR applications (31–34). We designed the GMR structure to critically couple to p-polarized free-space radiation (i.e., electric field oscillating in the x-y plane) near the ENZ wavelength over a range of angles, with the guided resonance in the thermal emitter. Because the resonator has been designed around the ENZ wavelength, we see a strong detuning of a resonant peak with a magnetic field. If no patterned α-Si was used, then the plasma edge of the InAs would have no distinct resonance that could be detuned with a magnetic field. This effect is discussed in more detail in the following section.

The optimal dimensions for the GMR structure are found on the basis of an analysis of the guided mode in a uniform α-Si slab atop n-InAs. In this approximation of the periodic α-Si, the uniform α-Si has a thickness $t = d_1/2 + d_2$, where $d_1$ is the periodic element depth and $d_2$ is the supporting layer thickness of the α-Si. The dispersion relation, $\omega(k_x)$, of the fundamental guided mode is then found by solving the equation (26, 35, 36)

$$\tan(k_x t) = \frac{k_{Si} \varepsilon_{Si} [k_{air} + k_{InAs,xx}]}{k_{Si}^2 - \varepsilon_{Si} k_{air} k_{InAs,xx}}$$

where $k_{Si} = \sqrt{Re(\varepsilon_{Si}) (\omega/c)^2 - k_x^2}$, $k_{air} = \sqrt{k_x^2 - (\omega/c)^2}$, $k_{InAs} = \sqrt{k_x^2 - (\omega/c)^2 Re(\varepsilon_{InAs})}$, $\varepsilon_{Si}/2 = 1/Re(\varepsilon_{InAs})$, and $k_x$ is the wave vector along the direction of propagation. In this derivation, we focus on the reciprocal case to get a sense of matching the slab waveguide to the Drude-like resonance of the InAs and neglect the off-diagonal terms of the InAs, which are zero when there is no applied magnetic field. In (26), the derivation for the nonzero magnetic field case shows that $\omega(k_x) \neq \omega(-k_x)$ and the symmetry of the dispersion relation is broken.

A plot of the symmetric angular dispersion for varying slab thicknesses of the α-Si waveguide is provided in fig. S1. In the calculations, we used the fitted dielectric constant values of the n-InAs from ellipsometry.

We chose a periodicity (Λ) of 7 μm such that the guided mode can be folded in the light cone and couple with the free-space radiation. We fine-tuned the other dimensions and used a GMR structure depth $d_1 = 0.72$ μm and supporting layer thickness $d_2 = 2.1$ μm.

Since the periodicity of the GMR structure itself is much smaller than the wavelength of interest, we expect to see only specular reflection in the considered wavelength range (37). We note that the linewidth of the resonance created by the GMR structure is sensitive to both the patterned area widths (Λ/2) and the scattering rate of the n-InAs underneath (38).

**Magnetic field–dependent absorption tuning in magneto-optic GMR structure**

The Voigt geometry used in these experiments means that the non-reciprocal behavior of electromagnetic radiation inside of the InAs will be confined to p-polarized scattering, where the electric field oscillates perpendicular to the applied magnetic field (Fig. 3A). Conversely, we expect reciprocal behavior for s-polarized scattering, as the applied magnetic field is aligned with the oscillations in the electric field, and the cross product of the electric and magnetic fields is zero. The reciprocal behavior for s-polarized light is shown in Fig. 3B, where the absorptivity maxima for all magnetic field values are marked with a dashed gray line at approximately 17 μm. In Fig. 3 (A and B), we emphasize the spectral shift in the peak position of the absorptivity and offset the data for clarity. The overall absorptivity for s-polarized light does not change with varying magnetic field. Furthermore, we do not observe any detectable polarization conversion from p to s or s to p polarization, confirming the proper alignment of our GMR structure relative to the optical beam path of the light (Fig. 3, C and D). The InAs layer is optically thick, and therefore, no light is transmitted through the sample, allowing us to only measure the absorptivity and reflectivity. We observed a substantial redshift of the absorptivity peak due to the GMR, especially in the positive field case (0 to 1.2 THz). The strong magnetic dependence of the absorptivity spectrum clearly demonstrates the reciprocity breaking effect. This nonreciprocal effect is observed in a wide angular and wavelength range.

To deconvolve the contribution coming from the n-type InAs and the entire GMR structure (n-type InAs with α-Si on top), we simulated angular absorption spectra as a function of magnetic field for both cases and compared the simulations to our measurements (Fig. 4). The simulations were performed using a finite-difference, frequency-domain electromagnetic simulation tool and incorporated the measured InAs optical properties and α-Si grating parameters. The simulated and measured plasma edge splitting for the unpatterned InAs wafer are in good qualitative agreement, with a broad resonance visible for the positive-field surface magnetoplasmon and a shorter wavelength onset (16.5 μm) for the negative-field surface magnetoplasmon (Fig. 4, A and C). The addition of the periodic α-Si does not increase the wavelength shift induced by the plasma edge splitting into two separate surface magnetoplasmons observed in the unpatterned InAs case. However, clear maxima are seen from the coupled GMR structure that can be evaluated for multiple angles and compared to the data, which is further discussed in the following section.

Comparing the measured positive field for the GMR structure (Fig. 4B, red curve) and the simulation (Fig. 4D, red curve), we note that both maxima occur at a resonance centered at 17.5 μm. However, a second shoulder resonance is present in the simulation at 16.7 μm, which is not easily visible in the experiment. The shoulder resonance is much weaker in the experiment likely because of the larger material loss in both the α-Si and InAs. Going from zero to negative applied magnetic field at a $\theta_i = 50^\circ$, we did not see a large
shift in the resonant peak position for either the simulation or the measurement but we do observe the resonance narrowing for the negative field case. The simulated and experimental traces for more angles of incidence and an intermediary 0.8-T field are included in figs. S2 to S4.

To further highlight the effect of the GMR structure on nonreciprocal absorption, we look at the intensity effect on the absorption resonance at more oblique angles. Taking $\theta_i = 70^\circ$, we can subtract the absorption of the bare InAs around the plasma shoulder from the GMR structure absorption and fit a Lorentzian to the difference (Fig. 5A). When the magnetic field is then turned on, the resonant intensity peak can be markedly tuned from absorption that greatly exceeds the plasma shoulder (positive field) to well below (negative field).

**Angle dependence of nonreciprocal absorption behavior**

To more comprehensively understand and characterize the nonreciprocal response of the hybrid GMR structure, we compare the experimental and simulated absorptivity maxima for p-polarized light over a range of incidence angles for $B = 0, 0.8,$ and $1.2$ T. To map our dispersion across positive and negative angles, we confirm that Onsager-Casimir relations hold (fig. S2). When $B = 0$ T, reciprocity is preserved, and we do not expect to see any reciprocity-breaking behavior. Consequently, the angular dispersion relation of the GMR at the plasma edge of the InAs remains symmetric. Figure 6A shows the symmetric case measured in our experiment and the corresponding absorptivity maxima from simulations (Fig. 6D). We also observe a narrowing of the resonant linewidth for more oblique angles of incidence in both the simulated and experimental data when no magnetic field is applied, as shown by the vertical bars marking the full width at 5% maximum of each absorptivity spectrum. When the in-plane magnetic field is applied, reciprocity is broken, and the measured dispersion of the absorptivity maxima becomes asymmetric (Fig. 6, B and C). The degree of reciprocity breaking grows at narrower angles, which is expected from the absorptivity maxima found from the simulations (Fig. 6, E and F).

We also observe a clear detuning of the absorptivity maxima with an increase in the magnetic field. For both the experimental (Fig. 6B)
and simulated (Fig. 6E) absorptivity maxima at the intermediate 0.8-T magnetic field, the lineshape width for positive angles of incidence grows considerably. This makes the detuning of the absorptivity maxima at narrow incidence angles difficult to resolve.

Increasing the magnetic field to 1.2 T separates the maxima at narrow angles such that the 5% bars do not overlap.

Unfortunately, our measurement system only allows for incidence angles down to 35°, limiting the ability to compare experiments and simulations at these angles.
DISCUSSION
In this study, we use a degenerately doped magneto-optical semiconductor paired with a GMR coupled to free-space incident radiation to demonstrate nonreciprocal absorption in the IR regime when a moderate magnetic field (B up to 1.2 T) is applied. The degree of reciprocity breaking is largest at narrower incidence angles \( \theta \) (45° to 55°), making the design potentially useful for cascading multiple emitters/absorbers to achieve directional flow of energy (39). The methods used to model, design, and fabricate the structure presented in this manuscript can be used for future implementations of nonreciprocal absorbers. As a future direction for engineering higher efficiency thermal radiators, direct measurement of the emissivity could be used to demonstrate applications of systems that violate the Kirchhoff thermal radiation law. Higher quality factors of the structure could be obtained using a thinner magneto-optic layer with a back reflector or a lower doping concentration of InAs; however, this would result in a longer working wavelength. A higher mobility magneto-optic material, such as InSb, could also be used to achieve a narrower linewidth (40). The easily identifiable absorptivity peaks at the plasma edge are a consequence of using a periodic \( \alpha \)-Si structure supporting a GMR, while the nonreciprocal behavior comes from the plasma-edge splitting of the magneto-optic \( \alpha \)-type InAs. The temperature dependence of the optical properties of the \( n \)-InAs is included in the Supplementary Materials as a reference for designing at elevated temperatures to measure emissivity (fig. S5). Beyond demonstrating nonreciprocal absorption in the IR, hybrid magneto-optic and photonic crystal structures such as those reported here are of potential interest for thermal radiation control and free-space information processing owing to the polarization-dependent response and low required magnetic fields for resonance tunability.

MATERIALS AND METHODS
Device fabrication
Our fabrication of the GMR structure started by deposition of \( \alpha \)-Si on the InAs wafer using plasma-enhanced chemical vapor deposition. The deposition was carried out at a temperature of 200°C and a pressure of 800 mtorr with a flow rate of 250 standard cm\(^3\)/min (5% SiH\(_4\)/Ar) for 90 s. We then spin-coated 500 nm of ZEP 520A resist (1 min at 2000 rpm) and baked the sample for 5 min at 180°C on a hot plate.

Electron beam lithography was then used to write the desired pattern into the resist using a beam current of 100 nA with a 300-\( \mu \)m aperture and a dose of 240 \( \mu \)C/cm\(^2\). After writing the pattern into the resist, the sample was dipped in ZED N-50 for 2 min and 30 s for development. Following exposure, the sample was baked at 140°C for 3 min.

The measured maxima for negative incidence angles do not shift below the zero magnetic field maxima for oblique angles. This contrasts with the maxima extracted from simulations, which redshift for oblique (−65° and −70°) angles of incidence.

Measurements
To probe the sample, we used a two-theta stage with a silicon carbide Globar source and focusing optics, exciting the sample at an incident angle \( \theta \), relative to the x-z plane and collect the specular reflected light at −\( \theta \). For the zero and low (up to but not including 0.8 T) magnetic field measurements, the original sample holder of the system (J.A. Woollam I.R. VASE Mark II) was used. For the higher magnetic field measurements, we used a separate adapter to accommodate the Halbach array. The lower magnetic field measurements are obtained using a specially designed aluminum holder to separate the two neodymium magnets while holding the sample in between the two magnets’ poles. The field strength was tuned by inserting aluminum spacers to increase the gap between the magnets. We used a Halbach array magnet configuration with tunable pole pieces to apply 0.8 < \( B \) < 1.2 T (29). An image of the magnet assembly can be found in fig. S7. We measured the magnetic field strength and uniformity across the sample surface with a transverse Hall sensor made from InAs (Lakeshore HGT 1010).

The sample was in the Voigt geometry, with an in-plane magnetic field, \( B \), breaking time-reversal symmetry. By changing the sign of the magnetic field, we produced the same effect as keeping the sign of magnetic field constant while switching the positions of the source and detector (41–43). The use of linear polarizers at both the source output and detector input allows for a deconvolution of the physical effects (e.g., ensure that there is no cross-polarization due to sample misalignment).

Device simulations
We used the COMSOL electromagnetic waves, frequency domain package to simulate the optical response of the GMR structure. For the material properties and geometries of the structure, we used the measured values of the \( \alpha \)-Si periodicity and depth from SEM images and \( n \)-InAs optical properties from the model fits of ellipsometry data and four-point probe measurements. For each trace at a fixed angle of incidence, we swept the wavelength from 16 to 19 \( \mu \)m in 12-nm intervals. The absorptivity was found by subtracting the reflectivity from 1, as there was no transmission for the structure. This was verified by taking transmission measurements of the \( n \)-InAs wafer.

SUPPLEMENTARY MATERIALS
Supplementary material for this article is available at https://science.org/doi/10.1126/sciadv.abe4308

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**Acknowledgments:** Y.K. used the Kavli Nanoscience Institute (KNI) at Caltech for fabrication facilities. K.J.S. would like to acknowledge H. Akbari, D. H. Drew, and A. Laucht for discussions around the design of the experiment and application of the magnetic field. **Funding:** This work was supported by DARPA NLM, grant no. HR00118200046 (to K.J.S., B.Z., Y.K., S.F., and H.A.A.). K.J.S. would like to thank the support by the NSF GRFP for a Graduate Research Fellowship. **Author contributions:** The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript. Measurements were done by K.J.S. Fabrication was done by Y.K. The theoretical and modeling work was led by B.Z. and supported by K.J.S. **Competing interests:** The authors declare that they have no competing interests. **Data and materials availability:** All data needed to evaluate the conclusions in the paper are present in the paper and/or the Supplementary Materials.

Submitted 16 September 2021

Accepted 21 March 2022

Published 6 May 2022

10.1126/sciadv.abm4308
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Sci. Adv., 8 (18), eabm4308.

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