

Asymmetric absorption at custom wavelengths via a layered magnetoelectric window

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The controllable nonreciprocal propagation of light is an intensively investigated field of optics, with studies motivated both by fundamental questions and possible telecommunication applications. So far, polarization-independent, switchable one-way transparency has been demonstrated only at certain resonances of multiferroic crystals at cryogenic temperatures and in high magnetic fields, limiting the practical implementation of this effect. Here, as an alternative approach, we present the one-way transparency of an artificial layered structure consisting of split-ring metamaterial and magnetic substrate layers interacting in the dynamic regime. As evident from our quasioptical experiments in the GHz frequency range, this unique combination breaks time and space inversion symmetries in external magnetic field. The ease of tuning the dynamic response and the possibility of controllable one-way transparency make this approach promising for real-world applications.

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I. INTRODUCTION

Creating windows that are transparent in one direction and opaque in the opposite direction remains a difficult task in the fields of optics and material design. Ideally, the effect of such a window should not depend on the polarization state of the incident radiation. As a counterexample, consider the Faraday isolator [1], i.e., a transparent window that rotates the plane of polarization of light in a suitable magnetic field by 45° and is sandwiched between a pair of polarizers also rotated by 45° with respect to each other. This structure is only transparent to a particular linear polarization state of light in one direction, while it blocks the propagation of the perpendicularly polarized light beam coming from both directions. Therefore, even with a linear sequence of Faraday isolators operating with different polarization states, it is not possible to achieve polarization-independent one-way transparency. In the language of Maxwell's electromagnetic equations, the problem can be reformulated as an asymmetry of the refractive indices of the eigenmodes of the window material in the forward and backward propagation directions. In a completely anisotropic case, this can even lead to four different refractive indices belonging to the combinations of orthogonal polarization states

and opposite propagation directions along a given optical axis [2].

Substantial progress in the control and manipulation of light has been achieved in recent years using metamaterials [3–6] and metasurfaces [7,8]. In these systems an asymmetric transmission for specific polarization may be achieved via an appropriate design [9–14]. However, most artificial materials are built from time-reversal symmetric components, i.e., nonmagnetic dielectrics and metals. Thus, both the building blocks and the structures are reciprocal, and they are symmetric from the point of view of strict definitions [15–18]. Although an asymmetry in transmission may be achieved in a specific experiment and for a specific polarization, after reversing the time axis or after interchanging the source and detector, the new experiment would result the same transmission coefficient. As obvious from symmetry considerations [19,20], polarization-independently nonreciprocal optical systems have to violate all possible symmetries connecting the opposite directions of the wave propagation axis, such as the spatial-inversion and time-reversal symmetries.

Corresponding to the breaking of the time-reversal symmetry, it can be shown by rigorous calculations [15,21] that one necessary condition for nonreciprocity is the violation of one of the following relations for material susceptibilities:

$$\hat{\epsilon} = \hat{\epsilon}^T, \quad \hat{\mu} = \hat{\mu}^T, \quad \hat{\chi}_{me} = -\hat{\chi}_{em}^T. \quad (1)$$

Here $(\dots)^T$ denotes the transposed matrix, and material equations are written in the form

$$\begin{aligned} \mathbf{B} &= \mu_0 \hat{\mu} \mathbf{H} + \sqrt{\epsilon_0 \mu_0} \hat{\chi}_{me} \mathbf{E}, \\ \mathbf{D} &= \sqrt{\epsilon_0 \mu_0} \hat{\chi}_{em} \mathbf{H} + \epsilon_0 \hat{\epsilon} \mathbf{E}. \end{aligned} \quad (2)$$

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$\mathbf{B}$ is the magnetic flux density, $\mathbf{D}$ is the electric displacement field, $\mathbf{E}$ and $\mathbf{H}$ are the electric and magnetic fields, $\hat{\epsilon}$, $\hat{\mu}$, and $\hat{\chi}_{me,em}$ are the tensors of electric permittivity, magnetic permeability, and magnetoelectric susceptibilities, respectively.

One possible way to make a material asymmetric would be including magnetic ordering into the system, thus, leading to asymmetric contributions to the dynamic magnetic susceptibility, which in the case of magnetic resonances violates Eq. (1):

$$\hat{\chi}_m \equiv \hat{\mu} - \hat{\mathbf{1}} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & \Delta\mu_{\perp} & i\alpha \\ 0 & -i\alpha & \Delta\mu_{\perp} \end{pmatrix}. \quad (3)$$

For clarity, the frequency dependence of the matrix elements is not written explicitly. $\Delta\mu_{\perp}$ corresponds to the strength of the magnetic resonance and α to the magnetic optical activity, $\hat{\mathbf{1}}$ is the three-dimensional identity matrix. This material realizes transmission that violates time-reversal symmetry in the Faraday geometry with light propagating parallel to the magnetic field, $\mathbf{k} \parallel \mathbf{B} \parallel x$ axis. However, the space inversion symmetry is still preserved in the Faraday rotation experiment. Therefore, to build a Faraday isolator [1] with an asymmetric transmission, a pair of 45°-rotated polarizers must be added on the two surfaces of the Faraday rotating layer to violate the space inversion as well. We note that the magnetic resonance experiment with susceptibility from Eq. (3) in the Voigt geometry, i.e., with $\mathbf{k} \perp \mathbf{B} \parallel x$, leads to time-reversal symmetric, reciprocal, light propagation as the polarization rotation due to Voigt effect is quadratic in the magnetic field [1].

The nonreciprocal dispersion of the excitation spectrum in one-dimensional photonic crystals, comprising a periodic arrangement of nonspontaneously magnetized and anisotropic dielectric layers, was proposed decades ago [19]. In that work, the two fundamental prerequisites for optical nonreciprocity were identified: the absence of symmetry operations that reverse the propagation vector, and the interplay between magnetic and electric effects. Despite this early theoretical insight, the experimental demonstration of nonreciprocal wave propagation in artificial structures only followed a few years later.

Several methods have been suggested for creating materials and systems with nonreciprocal propagation. As magnetic ordering breaks the time-inversion symmetry, magneto-optical interactions have been utilized extensively to investigate and produce microstructures and photonic crystals with nonreciprocal propagation [19,22–27]. Polarized light scattering on a two-dimensional array of magnetized, non-centrosymmetric chevron-shaped submicron islands has been shown to violate reciprocity [28]. Numerical simulations on metamaterials [29] and metasurfaces [30–32] in magnetic field can also indicate nonreciprocal wave propagation. Further mechanisms have been investigated [6] that utilize active elements [33–35], nonlinearity [36], time modulation [37], or mechanical coupling [38].

Nonreciprocal propagation due to magnetoelectric coupling is known from experiments on multiferroics, i.e., materials with simultaneous electric and magnetic ordering that are mutually connected [39,40]. Asymmetric absorption

in multiferroics is often referred to as directional dichroism [2,41–44], and can be reduced to magnetoelectric susceptibility violating Eq. (1). In such a case, the refractive indices for the propagating solutions of the Maxwell equations are asymmetric with respect to the reversal of the light propagation [20,45]. In several cases [2,43,44,46], even one-way transparency is achieved with practically no absorption in one and strong absorption in the opposite propagation direction. However, most of these experiments [2,43,46] were performed in the Faraday geometry, where the polarization state of the incoming light was not preserved. With few exceptions [46,47], nonreciprocity in multiferroics occurs in the low-temperature magnetically ordered phase. In all reported cases, the effect is confined to relatively narrow, magneto-optical resonances at material-specific frequencies. Thus, from the perspective of potential applications, the typically cryogenic temperature range and limited frequency tunability of these systems are the main technological challenges.

As compared to the above examples on multiferroic crystals, we explore here a fundamentally different route to the asymmetric propagation of light based on a metamaterial with magnetoelectric interaction [29,48–51]. In the following we are going to show that appropriate coupling of an array of split-ring resonators with a paramagnet violates the reciprocity condition of Eq. (1). The spatial inversion symmetry is broken by the sandwich structure of the multilayer sample consisting of split-ring and magnetic layers, while the time-reversal asymmetry is produced by the nonspontaneously magnetized layer, leading to different transmission for forward and backward propagation, as illustrated in Fig. 1. The nonreciprocal effect of this sandwich structure, although once speculatively proposed based on a simulation result on an array of spontaneously magnetized ferrite slabs [29], has not been investigated experimentally in any of the former studies on split-ring metamaterials [29,48–51]. On the other hand, nonreciprocal electromagnetic wave propagation was proposed based on a theoretical model [52] and numerical simulations on various artificial structures [53,54], and demonstrated on a minimal model consisting of a ferrite sphere and a metal stripe [55]. While our experiments were performed at cryogenic temperature ($T = 5.5$ K), by the choice of a magnetically ordered substrate layer the method might have the potential to realize one-way transparency at room temperature as well. In this sense, integrating a layer of frequency-tailored electric resonators with a suitable magnetic substrate offers a promising alternative to multiferroic crystals in nonreciprocal devices.

A potential candidate material is ferrimagnetic yttrium iron garnet (YIG), whose ferromagnetic resonance frequency can be tuned by an external magnetic field. However, due to the exceptionally low damping in thin YIG films [56], the resulting absorption line is extremely narrow, which limits the operational frequency range of a one-way transparent device based on YIG at room temperature. Similar to YIG, room-temperature ferromagnetic insulators, such as barium hexaferrite, typically exhibit narrow magnetic resonance lines in the microwave regime. In contrast, in thin films of titanium- and manganese-substituted barium hexaferrite, $\text{Ba}(\text{MnTi})_x\text{Fe}_{12-x}\text{O}_{19}$, the resonance linewidth can be significantly broadened [57], making this material a promising

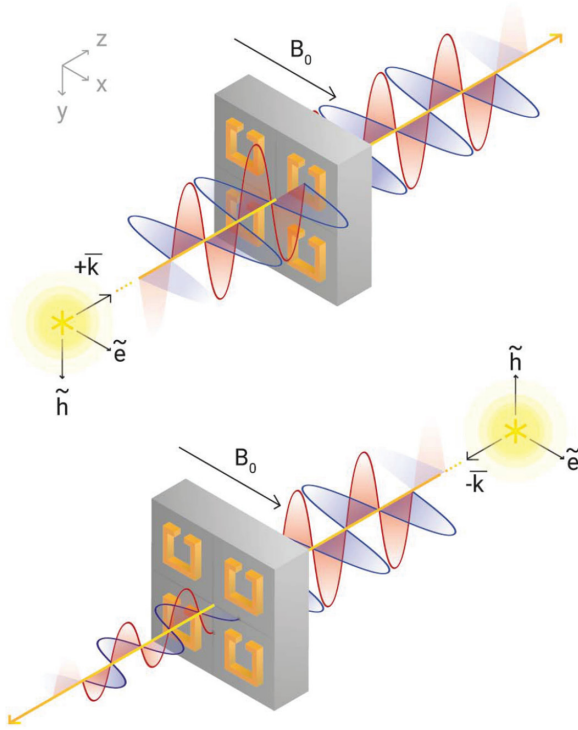


FIG. 1. Directional anisotropy of a bilayer structure built from a two-dimensional array of split-ring resonators and a paramagnetic film. Flipping the propagation direction of light results in a tremendous change in the transmission of the sample due to the interplay between the reciprocal magnetoelectric coupling of the split rings and the off-diagonal magnetic susceptibility of the paramagnetic film. Red and blue waves correspond to the oscillating magnetic and electric components of light, respectively. Split rings are in gold, while the magnetic layer is in gray.

candidate for the magnetic component of one-way transparent devices operating at ambient conditions.

An important consideration when evaluating nonreciprocal optical properties is the frequency range over which the effect is observed. In our system, nonreciprocity emerges from the coupling between an intrinsic magnetic resonance and a tunable electric resonance, allowing the operational frequency to be flexibly adjusted within the microwave regime. This tunability makes our approach particularly attractive for applications in telecommunication technologies. In contrast, in some multiferroic crystals, nonreciprocity originates from weak electronic orbital transitions, enabling the effect to be shifted toward significantly higher frequencies, including the visible spectral range. Such characteristics make these crystals promising candidates for implementation in integrated optical devices [58–61].

II. RESULTS

The one-way transparent window that we propose is a composite structure consisting of a paramagnetic gadolinium gallium garnet ($\text{Gd}_3\text{Ga}_5\text{O}_{12}$, GGG) layer coupled to a two-dimensional periodic array of split-ring resonators, as presented in Fig. 1. In contrast to most of the experimental studies of nonreciprocity in multiferroics [2,41–44] and

the calculations on ferrite slabs [29], our sample possesses no spontaneous magnetic or ferroelectric ordering, and the magnetoelectric interaction is manifested only in the dynamic regime. As we show in the following, the presence of both the paramagnetic and the split-ring layers and their electromagnetic coupling by the close vicinity are essential to the realization of the nonreciprocal propagation of light.

The interaction between the electric and magnetic resonances can be influenced by the broadening of their respective absorption peaks. To minimize damping-related effects [56] in our proof-of-concept study, all experiments were conducted at a cryogenic temperature of 5.5 K.

A. Long-wavelength electrodynamics of the hybrid heterostructure

The magnetic permeability of GGG in this geometry may be approximated by Eq. (3) with $\mathbf{B} \parallel x$ and $\mathbf{k} \parallel z$ (Fig. 1, Voigt geometry). In this arrangement the solutions of the Maxwell equations are linearly polarized waves with refractive indices $N_{x\pm} = \sqrt{1 + \Delta\mu_{\perp} - \alpha^2/(1 + \Delta\mu_{\perp})}$ and $N_{y\pm} = 1$ for light propagation along the $\pm z$ direction. Here x and y indicate the polarization state (oscillating electric component) of the beam. The propagation in positive and negative directions is completely symmetric.

Similarly, considering a split-ring metamaterial layer alone, the solution of the Maxwell equations results in symmetric wave propagation. In our experimental geometry for wavelengths exceeding the periodicity of the two-dimensional array of split rings, light-matter interaction can be described by four nonzero components of the susceptibility matrices [62,63]:

$$\begin{aligned} \chi_e^{xx} &= \Delta\epsilon, & \chi_m^{zz} &= \Delta\mu_r, \\ \chi_{em}^{xz} &= -\chi_{me}^{zx} = i\sqrt{\Delta\epsilon \cdot \Delta\mu_r}. \end{aligned} \quad (4)$$

Here $\hat{\chi}_e \equiv \hat{\epsilon} - \hat{1}$ is the electric susceptibility, $\Delta\epsilon$ and $\Delta\mu_r$ are the oscillator strengths of electric and magnetic modes, and the frequency dependence of the matrix components is omitted for simplicity. The electric and magnetic resonances are closely related, which intuitively can be seen when considering the split-ring resonator as a swing circuit. Oscillating currents, excited by an alternating electric field along the gap of the ring (x), produce an alternating magnetic field perpendicular to the plane of the ring (along z), and vice versa. Nevertheless, the magnetoelectric coupling fulfills Eq. (1), thus, it does not violate the reciprocity of light propagation either.

In the next step we combine the GGG with the split-ring metamaterial layers, as shown in Fig. 1, which results in a nonreciprocal magnetoelectric coupling between the layers due to the combination of the nonreciprocal χ_m^{zy} of the paramagnet and the reciprocal χ_{me}^{zx} of the split rings.

The physical mechanism of the proposed effect can be understood within simple arguments as follows. A linearly polarized electromagnetic wave propagating along the z axis and with oscillating (ac) field components e_x and h_y would excite the split rings via the electric channel. Due to induction the currents in the rings will produce a secondary ac magnetic field along the z direction, which can be formulated by

the formal magnetoelectric coupling χ_{me}^{zx} as $h_z \sim \sqrt{\frac{\epsilon_0}{\mu_0}} \chi_{me}^{zx} e_x$. Combined with the h_y field of the wave an elliptically polarized magnetic field $h_{yz} = h_y + h_z$ is generated with rotational axis along x . This field will interact with the magnetic moments in GGG which are induced by the static external field $M_x \sim B_x$. The precession direction of the magnetic moments is determined by the sign of the external field and the rotation sense of the ac field h_{yz} is determined by the geometry of the experiment. Therefore, the two will rotate either in the same or in the opposite direction. Ideally, in the first case a strong magnetic absorption will be seen, and in the last case no absorption is expected. In a real experiment the asymmetry with respect to the propagation direction and with respect to the field direction should be observed.

More specifically, for wavelengths longer than the periodicity of the split-ring layer, the combined susceptibilities of the sandwich structure are given as

$$\begin{aligned} \hat{\chi}_m &= \begin{pmatrix} 0 & 0 & 0 \\ 0 & \Delta\mu_{\perp} & i\alpha \\ 0 & -i\alpha & \Delta\mu \end{pmatrix}, \\ \hat{\chi}_{em} &= \begin{pmatrix} 0 & 0 & i\sqrt{\Delta\epsilon \cdot \Delta\mu_r} \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} = -\hat{\chi}_{me}^T, \\ \hat{\chi}_e &= \begin{pmatrix} \Delta\epsilon_s + \Delta\epsilon & 0 & 0 \\ 0 & \Delta\epsilon_s & 0 \\ 0 & 0 & \Delta\epsilon_s \end{pmatrix}, \end{aligned} \quad (5)$$

where $\Delta\epsilon_s$ stands for the electric susceptibility of the substrate hosting the split rings and $\Delta\mu = \Delta\mu_{\perp} + \Delta\mu_r$ is the total magnetic contribution of GGG and split rings.

The solutions of the Maxwell equations (eigenmodes) for propagation along the $\pm z$ directions are linearly polarized transverse electromagnetic waves. While for the solutions with polarization e_y , h_x the counterpropagating beams have identical complex refractive indices ($N_{y\pm}$), for e_x , h_y we obtain different solutions ($N_{x\pm}$) for forward and backward propagation:

$$N_{y\pm} = \sqrt{(1 + \Delta\epsilon_s)(1 + \Delta\mu_{\perp})}, \quad (6)$$

$$N_{x\pm} = \sqrt{(1 + \Delta\tilde{\epsilon}_s)(1 + \Delta\tilde{\mu}_{\perp})} \pm \frac{\alpha\sqrt{\Delta\epsilon\Delta\mu_r}}{1 + \Delta\mu}. \quad (7)$$

Here $\Delta\tilde{\epsilon}_s = \Delta\epsilon_s + \frac{\Delta\epsilon(1 + \Delta\mu_{\perp})}{1 + \Delta\mu}$ and $\Delta\tilde{\mu}_{\perp} = \Delta\mu_{\perp} - \frac{\alpha^2}{1 + \Delta\mu}$ are renormalized electric and magnetic contributions, respectively. We note here that both magnetoelectric susceptibility χ_{zx}^{me} of the split rings and the magnetic susceptibility χ_{yx}^m of the GGG must simultaneously be present to provide the directional anisotropy $\Delta N = N_{x+} - N_{x-} = \frac{2\alpha\sqrt{\Delta\epsilon\Delta\mu_r}}{1 + \Delta\mu}$. In contrast to all former studies on multiferroic crystals [2,41–44,64], where the strong directional anisotropy is produced solely by the nonreciprocal magnetoelectric coupling, in the case of metamaterials [55] it is resulted from a higher-order process, namely, the interplay between the reciprocal magnetoelectric coupling due to induction of the split rings and the nonreciprocal off-diagonal magnetic susceptibility of the magnetic film.

The working principle of the nonreciprocal hybrid structure can also be explained purely from the symmetry arguments. The system realized by sandwiching the magnetic film between the two layers of the split-ring resonators as shown in Fig. 2(b) possesses $m_x 2'_y m'_z$ symmetry, which is the symmetry group of the crossed magnetization $\mathbf{M}_x$ and electric polarization $\mathbf{P}_y$ along the x and y axes, respectively. Here $m_{x/z}$ denotes a mirror plane symmetry perpendicular to x/z , while 2_y means two-fold rotational symmetry around y and $'$ indicates the time-reversal operation, respectively. The cross product of the magnetization and electric polarization, termed as toroidal moment ($\mathbf{T}_z = \mathbf{M}_x \times \mathbf{P}_y$), has the same transformation properties as the $\mathbf{k}$ wave vector of light, thus, transmissions along and opposite to $\mathbf{T}$ are not connected by symmetry [20]. This geometry was utilized in multiferroic crystals [41,42,44,64] to obtain an asymmetric transmission along z . Therefore, from the symmetry point of view the structure in Fig. 2(b) is equivalent to an asymmetric crystal with finite toroidal moment.

We note here that the optical experiments according to Figs. 1 and 2(b) are performed in the Voigt geometry with light propagating perpendicular to external magnetic field, where the incoming linear polarization state polarized along x or y is preserved during propagation. In this geometry neither split rings nor GGG are producing the asymmetric propagation separately.

The above-introduced interaction between the split ring and GGG layers couples their resonances. Without the neighboring magnetic layer, the split ring is expected to show an electric excitation whose frequency is independent of the magnetic field, while the frequency of the paramagnetic resonance in GGG is proportional to the magnetic field. The split-ring frequency can be controlled by the geometrical parameters and the substrate permittivity, thus, the anticrossing of the two resonances can be observed in the GHz frequency range in conveniently achievable laboratory fields [48].

B. Experimental results

The directional anisotropy can be studied experimentally either by tuning the frequency of the electromagnetic wave at constant magnetic field or by sweeping the magnetic field and keeping the light frequency constant. The spectra obtained by the first approach are more straightforward to interpret using the Fresnel optical formulas for the transmittance of a plane-parallel sample [65] and assuming a Lorentzian form [66] of the excitations, while the second method offers significantly better signal-to-noise ratio due to the higher stability of the steady-frequency experimental setup. The effect of time reversal can be investigated either by reversing the propagation direction of light or by flipping the sign of magnetic field [67]. Due to the experimental convenience we applied the latter method and compared the transmission in positive and negative magnetic fields.

In our experiments the magnetic film is sandwiched between two layers of the two-dimensional periodic array of split-ring resonators, as shown in Fig. 2(b). On one hand, the doubling of the split-ring layers as compared to the schematic Fig. 1 increases the observed signal, on the other hand, it makes the sample more symmetric, emphasizing which

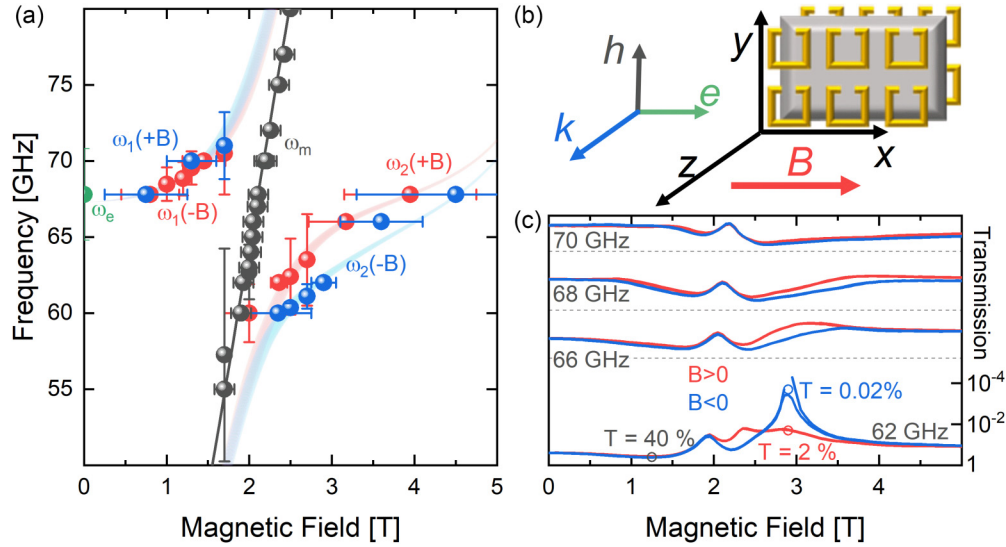


FIG. 2. Directional anisotropic resonances of the split-ring-paramagnet sandwich structure. (a) Magnetic field dependence of the observed (spheres) and calculated (lines) resonances. Red (blue) color corresponds to absorption maxima in positive (negative) magnetic fields. Linewidth of the light red (blue) lines is proportional to the calculated strength of the directional anisotropy for the magnetoelectric excitations. Black line shows the calculated magnetic resonance frequency. Due to the thickness of the magnetic layer, pure magnetic resonance (black spheres) was also observed, while green sphere corresponds to the zero-field electric resonance of the split rings. Error bars of the experimental symbols show the full width at half-maximum of the absorption peaks in field sweeps (horizontal) or in the spectra in constant field (vertical). (b) Geometry of the experiment showing the magnetic GGG layer (gray) sandwiched between two layers of 2D split-ring arrays (gold). (c) Observed magnetic field-dependent transmission in inverse logarithmic scale at different frequencies in positive (red) and negative (blue) fields. The field sweeps measured at 66, 68, and 70 GHz are shifted vertically in proportion to the frequency, where the corresponding perfect transmission baselines are indicated by dashed lines, while absolute transmission values corresponding to the 62-GHz experiment are shown in the right scale. The missing segment of the 62-GHz transmission curve recorded at negative magnetic fields corresponds to signal levels falling below the minimum detectable threshold of our experimental setup. Circle symbols indicate the respective transmission levels of the background ($T = 40\%$), and of the transmitting ($T = 2\%$) and blocking ($T = 0.02\%$) directions at the resonance with the highest contrast.

symmetry violation is necessary to achieve nonreciprocal light propagation. Figure 2(c) shows the magnetic field-dependent transmission at different frequencies for positive (red) and negative (blue) magnetic fields. Aside from the reciprocal peak around $B_x \approx 2$ T corresponding to the bulk GGG, clear difference in the transmission in case of $\pm B_x$ can be observed in the magnetic field region of $B_x \approx 2$ –4 T. To better visualize the nonreciprocal phenomenon and remove the effect of the reciprocal baseline, in Figs. 3–5 we turn to the discussion of the difference signal between positive and negative fields.

It is important to note the low overall transmission level of the sandwich structure used in our proof-of-concept experiment. Specifically, even far from the resonant absorption frequencies and magnetic fields, the transmission does not exceed 50%. At the resonant frequency, where nonreciprocity is most pronounced, such as at 62 GHz and 2.9 T [see Fig. 2(c)], the transmission in the forward (transmitting) direction is only 2%, while that in the backward (absorbing) direction falls below our detection limit of 10^{-4} . Since the magnetic layer served as the mechanical support of the sample, the overall transmission could likely be improved by using a thinner magnetic film in combination with a nonmagnetic, transparent substrate. Compared to conventional Faraday isolators, our system differs in a key aspect: Faraday isolators operate in a nonresonant frequency regime, enabling near-unity transmission in the forward direction.

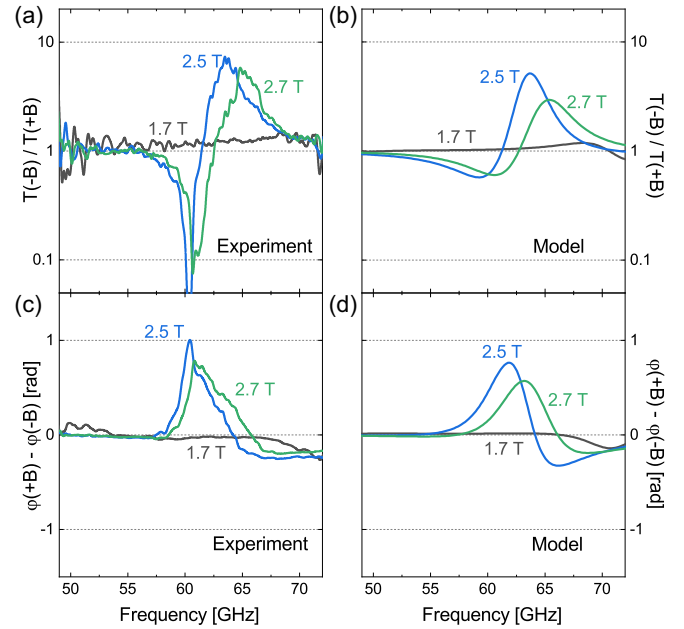


FIG. 3. Directional anisotropy spectra. Ratio of transmission in negative and positive magnetic fields in a logarithmic scale as observed (a) and calculated (b). Phase shift between light propagation for positive and negative fields as observed (c) and calculated (d). In all panels black, blue, and green lines correspond to external fields of $B_x = 1.7, 2.5$, and 2.7 T, respectively.

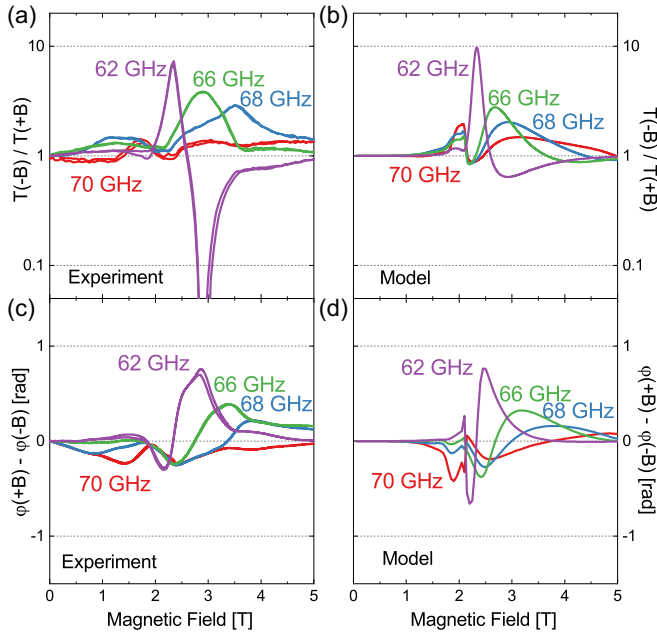


FIG. 4. Magnetic field dependence of the directional anisotropy at different frequencies. Ratio of transmission in negative and positive magnetic fields in a logarithmic scale as observed (a) and calculated (b). Phase shift between light propagation for positive and negative fields as observed (c) and calculated (d). In all panels red, blue, green, and purple lines correspond to frequencies of $\omega = 70$, 68, 66, and 62 GHz, respectively.

The observed directional dichroism spectra of the three-layer sandwich structure are presented in Fig. 3. Figure 3(a) shows the difference in transmission for the opposite magnetic

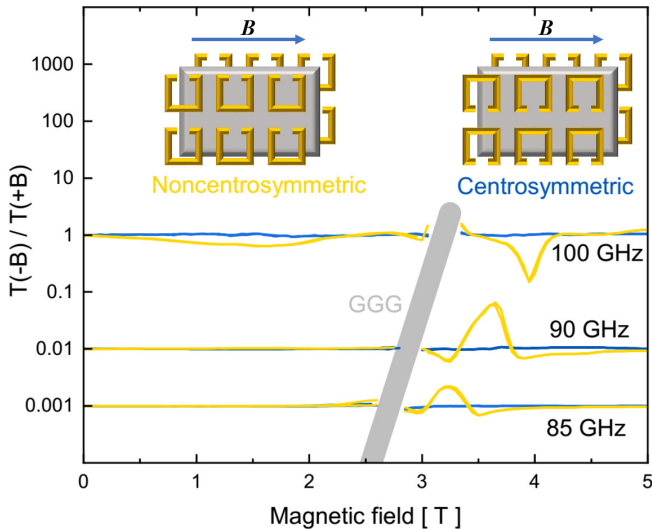


FIG. 5. Symmetry dependence of the directional anisotropy. Ratio of transmission in negative and positive magnetic fields in a logarithmic scale in the centrosymmetric (azure) and noncentrosymmetric (yellow) sample. Field sweeps at 85 and 90 GHz are vertically shifted with respect to the 100-GHz measurement. Gray area corresponds to the resonance of the GGG layer, where this sample is not transparent.

field directions reaching a two order of magnitude contrast in the vicinity of the anticrossing point of the magnetic and electric resonances. We note here that the observed difference is limited by the experimental setup, namely, there is practically zero transmission at the resonance peak for negative magnetic field. The directional anisotropy in transmission is due to the absorption difference and is related to the different imaginary parts of the two complex refractive indices $N_{\pm}$ corresponding to the forward and backward directions. The real part of N produces a difference in the phase shift of counterpropagating beams which reaches as high as $\Delta\varphi = 1$ rad, as shown in Fig. 3(c). Since the sample thickness (~ 0.25 mm) corresponds to a small fraction of the wavelength $\lambda \sim 5$ mm, the difference in both the real and imaginary parts of the refractive indices is extremely high, opening the path to the production of few-layer structures with one-way transparency and asymmetric phase shift.

Similarly to the directional anisotropy spectra in Fig. 3, the experimentally more stable field sweeps at constant frequencies also exhibit a transmission contrast [Fig. 4(a)] and a difference in phase shift $\Delta\varphi \sim 1$ rad [Fig. 4(c)] between positive and negative magnetic fields. In this experiment the one-way transparency is realized at $\omega = 62$ GHz and $B_x = 3$ T.

In order to verify that the observed nonreciprocity is a consequence of the noncentrosymmetric sandwich structure, and not inherently present in any of the constituent layers, we also performed control experiments with a centrosymmetric sample. Namely, by changing the relative orientation of the split rings in the two layers at the top and bottom of GGG, the inversion symmetry can easily be switched on (slits of the rings on top of GGG pointing down, on the bottom of GGG pointing up, Fig. 5) and off (slits of all rings pointing up, Fig. 5). Furthermore, to demonstrate the tunability of the characteristic frequencies of the structure, the control experiments were carried out on a sample with smaller split rings, thus, the resonances are shifted to higher frequencies. As presented in Fig. 5, the centrosymmetric sample does not show any difference between the transmission for positive and negative magnetic fields.

III. DISCUSSION

Both the directional dichroism field sweeps [Figs. 4(a) and 5] and the spectra [Fig. 3(a)] show a derivativelike line shape, which suggest a frequency shift of the resonance between positive and negative fields. To understand the form of the directional anisotropy curves, one has to consider the environment-dependent resonance frequency of the split rings. Modeling the ring by a swing circuit, the resonance frequency has an obvious dependence on the background permittivity and permeability via the capacitive and the inductive element, respectively. Taking both into account, one gets the $\omega_e \sim 1/N$ relation connecting the resonance frequency to the refractive index. In our experiment N has a definite magnetic field dependence due to the magnetic resonance of the GGG layer and due to the nonreciprocal contribution from the coupling of the split ring and GGG layers. This leads to field-dependent resonance frequency of the split rings. Thus, the hybridized magnetoelectric resonances, presented

also in Fig. 2(a), will have different frequencies depending on the sign of the magnetic field, which results in asymmetric transmission in the vicinity of those resonances. The observed peaks in Figs. 4(a) and 3(a) belong to the lower-frequency, quasimagnetic resonance of the structure [$\omega_2(\pm B)$ in Fig. 2(a)]. The resonance frequencies obtained by the model calculations (see Methods for the details) are compared to the experimental peak positions in Fig. 2(a), while the calculated directional anisotropy is presented in Figs. 3(b) and 3(d) and Figs. 4(b) and 4(d), showing a reasonable quantitative agreement with the experiments. However, the experimentally observed near-zero transmission for one propagation direction in the 60–62 GHz frequency range and at magnetic fields between 2.5 and 3 T is not accurately captured by our model. This discrepancy may stem from a resonance narrowing in the vicinity of the anticrossing point [68], which is not adequately accounted for in our phenomenological description.

IV. CONCLUSIONS

The multilayer structure of our study has significant benefits for technological applications compared to the other class of candidate materials for switchable one-way transparency, namely, multiferroic crystals. The electric resonance frequency can be easily tuned by the geometrical parameters of the split rings, while, in the case of crystals, the resonance is an intrinsic property of the material. Since the magnetic resonance frequency of the paramagnetic layer is proportional to the external field, the one-way transparency can also be realized in relatively low magnetic fields at the anticrossing of magnetic and electric excitations. With the suitable choice of the magnetic layer, for instance ferrimagnetic yttrium iron garnet (YIG) with magnetic ordering below $T_C = 560$ K [69] or titanium- and manganese-substituted barium hexaferrite, $\text{Ba}(\text{MnTi})_x\text{Fe}_{12-x}\text{O}_{19}$ [57], the effect can persist even at room temperature, which is difficult to realize in multiferroic crystals [46,64]. In contrast to the former examples of one-way transparency in the Faraday geometry [2,43,46], our Voigt-geometry design maintains the linear polarization state of light during the propagation, eliminating the intensity loss due to the analyzer behind the sample. Nonreciprocal transmission is realized for one linear polarization state and complete transparency for the orthogonal polarization state, thus, the anisotropy ratio $[T(+B) - T(-B)]/[T(+B) + T(-B)] \approx 1$ and $\approx \frac{1}{3}$ can be achieved for linearly polarized and unpolarized light beams, respectively. Furthermore, by stacking a pair of three-layer samples with 90° rotated orientation, polarization-independent one-way transparency might be achieved. Moreover, the tuning of the material's optical response by changing the split ring's size and the geometry is possible layer by layer, making the multilayer structure ideal for designing one-way transparent devices with customized optical spectrum.

V. METHODS

A. Measurement setup

Terahertz transmission experiments were carried out using quasi-optical terahertz spectroscopy [70,71]. This technique utilizes linearly polarized monochromatic radiation provided

by backward-wave oscillators. He-cooled bolometer was used as detector of the radiation. With the help of parallel wire-grid polarizer and analyzer, the complex transmission coefficient can be studied. The phase information was obtained by comparing the mirror positions necessary to reach an interference minimum between the two arms of our Mach-Zender interferometer for the sample and for the reference aperture. Static magnetic fields, up to ± 5 T, have been applied to the sample using a split-coil superconducting magnet. Instead of reversing the propagation direction, the sign of the magnetic field was inverted in order to study the effect of time reversal. The sample was in a He-cooled cryostat and all experiments were performed at $T = 5.5$ K temperature. Schematic diagram of the used measurement setup can be found in Figs. 1(a) of Refs. [56,72].

B. Phenomenological model

The long-wavelength electromagnetic response of a splitting-GGG heterostructure can be simulated using a model consisting of two coupled oscillators [48,73]. The electric resonance frequency of the split ring is $\omega_e(\pm B) = \omega_0/N(\pm B)$, while the paramagnetic resonance frequency of GGG is proportional to the field, $\omega_m(B) = \gamma B$, where γ is the gyromagnetic ratio. For a given magnetic field, the squares of the coupled resonance frequencies ω_1, ω_2 can be calculated as the eigenvalues of the dynamical matrix

$$\begin{pmatrix} \omega_e^2 & K \\ K & \omega_m^2 \end{pmatrix}, \quad (8)$$

where K corresponds to the coupling of the electric and magnetic oscillators:

$$\omega_{1,2}^2 = \frac{\omega_e^2 + \omega_m^2 \pm \sqrt{(\omega_e^2 - \omega_m^2)^2 + 4K^2}}{2}. \quad (9)$$

The mixing of the electric and magnetic character of the resonances can be determined from the eigenmodes of the matrix. Using the oscillator strengths of the electric and magnetic modes $\Delta\epsilon$ and $\Delta\mu_\perp$, respectively, one can write the formula

$$\begin{aligned} \Delta N_{1,2} \sim & \frac{8K \left(\omega_e^2 - \omega_m^2 \pm \sqrt{(\omega_e^2 - \omega_m^2)^2 + 4K^2} \right)}{4K^2 + \left(\omega_e^2 - \omega_m^2 \pm \sqrt{(\omega_e^2 - \omega_m^2)^2 + 4K^2} \right)^2} \\ & \times \sqrt{\Delta\epsilon \Delta\mu_\perp} \end{aligned} \quad (10)$$

for the oscillator strength of directional anisotropy for the two resonances. The overall static contribution of the two resonances is zero [74], resulting in an exclusively dynamic magnetoelectric response of the system.

In the model the lifetime of the coupled excitations has been considered by adding an $i\Gamma$ imaginary part to the resonance frequencies where Γ corresponds to the intrinsic linewidth [56]. Moreover, an imaginary part of the coupling has been applied to reproduce the resonance of lifetimes, namely, having narrower linewidth in the vicinity of the anticrossing point as the linewidth of either the pure electric or the magnetic excitation [68]. In our approach we considered the linewidths of the electric and magnetic resonances constant in

magnetic field, while the imaginary part of the coupling was set to be proportional to its real part. Since the nonspontaneous magnetization of the paramagnetic layer is proportional to the external field and the proposed coupling between the layers gets stronger with increasing ordered moment, we used $K \sim B$, equal for both split-ring–GGG interfaces. Moreover, the field-proportional coupling can also be motivated by comparing Eqs. (7) and (10), where in small fields on one hand $\Delta N \sim \alpha \sim B$, on the other hand $\Delta N \sim K$.

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A.M.K. performed the experiments. A.P. and A.A.M. designed and supervised the experiments. D.S. and A.M.K. analyzed the data. D.S., A.P., and A.S. developed the model. D.S. and A.P. prepared the manuscript with contributions from all authors.

The authors declare no conflict of interest.

DATA AVAILABILITY

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

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