

Broken Time Reversal and Parity Symmetries for Electromagnetic Excitations in Planar Chiral Nanostructures

Ewan M. Wright^{1,*} and Nikolay I. Zheludev^{2,†}

¹*Optical Sciences Center and Department of Physics, University of Arizona, Tucson, AZ 85721, USA*

²*School of Physics and Astronomy, University of Southampton, SO17 1BJ, UK*

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We elucidate the physical mechanisms by which electromagnetic excitations in planar chiral waveguides exhibit broken time reversal and parity symmetries as recently observed in metallic nanostructures (*Phys. Rev. Lett.* **91**, 247404 (2003)). Furthermore, we show that concomitant with these broken symmetries the electromagnetic excitations can acquire fractional winding numbers, revealing analogies between light scattering from planar nanostructures and anyon matter.

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A recent experimental study of optical interactions with non-magnetic metal planar chiral nanostructures consisting of arrays of four-fold chiral gammadions provides intriguing evidence of broken time reversal (\mathcal{T}) symmetry [1, 2]. Planar chiral objects are characterized by the fact that they cannot be brought into congruence with their enantiomeric or mirror image form without lifting the object from the plane. Among the intriguing properties of chiral arrays is that they display non-reciprocal polarization effects in diffraction which resemble the well-known non-reciprocity of the Faraday effect [3]. In the experiment of Ref. [1] chiral nanostructures whose topographies were mirror images of each other were found to lose there mirror symmetry when viewed in polarized light, an effect that also occurred for individual chiral gammadions. It has been proposed that these unusual symmetry properties result from the chiral optical response associated with plasmon and volume modes in the grooves of the structure [1].

In this paper we theoretically show that electromagnetic (EM) excitations in planar chiral nanostructures can exhibit broken \mathcal{T} and parity (\mathcal{P}) symmetries, and a physical argument is given for how these broken symmetries can be manifested in the properties of light scattered from the nanostructures. We also point to analogies between light scattering from planar chiral nanostructures and from anyon matter [4].

To describe the experiment of Ref. [1] we consider a thin metallic (gold-titanium) film with an array of four-fold chiral gammadions carved in it. We neglect coupling between individual gammadions based on the high losses suffered by propagating surface plasmon-polariton waves propagating between gammadions due to the titanium layer covering the structure. These losses, however, are negligible for EM excitations confined in the grooves of individual gammadions as considered here, since the titanium layer is absent in the grooves. We thus consider an individual four-fold chiral gammadion centered on the origin, an example of which is shown in Fig. 1. Here the gammadion lies in the (x, y) plane and we have chosen

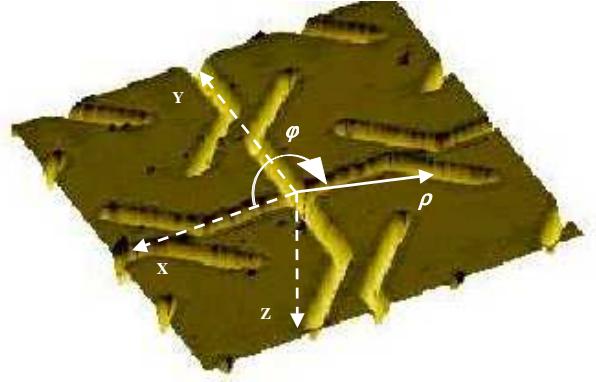


FIG. 1: [colors online] An AFM image of a fragment of the planar chiral structure showing one gammadion carved in a thin gold/titanium film, as described in Ref. [2]. The electromagnetic excitations considered in the present paper are concentrated in the grooves of the structure that form a single gammadion element. The characteristic sizes of the gammadion's elements, their width and depth are $1.5 \mu\text{m}$, 700 nm , and 120 nm , respectively.

to align the x -axis with the central portion of one gammadion groove and the y -axis with another, the z -axis being perpendicular to the plane of the gammadion. For future use we also introduce a cylindrical coordinate system for which points in the plane of the gammadion are represented by the radial coordinate ρ and azimuthal angle ϕ as indicated in Fig. 1. Then, for an EM field of frequency ω the dielectric tensor around the gammadion structure has elements $\epsilon_{\mu\nu}(\vec{r})$, with $\mu, \nu = x, y, z$, where we omit the explicit ω dependence for simplicity in notation. We approximate the tensor as Hermitian and its diagonal elements as real by the virtue of the fact that for the optical frequencies of interest in gold $|Re(\epsilon_{\mu\mu})| \gg |Im(\epsilon_{\mu\mu})|$, and the four-fold gammadion symmetry implies $\epsilon_{xx} = \epsilon_{yy}$. The generally complex off-diagonal tensor element $\epsilon_{xy} = \epsilon_{yx}^*$ describes polarization coupling in the (x, y) plane due to the chiral gammadion patterning, and we assume that this is the domi-

nant source of polarization coupling and hereafter set all other off-diagonal tensor elements to zero. To proceed we expand the tensor elements $\epsilon_{xy}(\vec{r})$ into azimuthal basis functions

$$\epsilon_{xy}(\vec{r}) = \sum_{\ell=-\infty}^{\infty} \epsilon_{xy}^{(\ell)}(\rho, z) e^{i\ell\phi}. \quad (1)$$

The basis functions $\exp(i\ell\phi)$ are useful for characterizing the chirality of the structure as each has an associated integer winding number ℓ such that the phase varies from zero to $2\pi\ell$ upon circling around the z -axis. The reality of the diagonal dielectric tensor elements implies that they have equal weighting of the winding numbers $\pm\ell$, $\epsilon_{\mu\mu}^{(\ell)} = (\epsilon_{\mu\mu}^{(-\ell)})^*$, meaning that the chirality is not manifest in the diagonal elements. In contrast, the generally complex off-diagonal tensor elements can manifest the chirality of the gammadion. For illustrative purposes we consider the simple case that the off-diagonal elements are dominated by a single winding number ℓ , the contribution from $-\ell$ being set to zero, for example $\ell = 4$ for a gammadion with four-fold symmetry and $\ell = -4$ for the corresponding enantiomeric form, and we write the off-diagonal elements as $\epsilon_{xy}(\vec{r}) = (\epsilon_{yx}(\vec{r}))^* = \kappa(\rho, z) e^{i\ell\phi}$. Finally, for the diagonal elements we ignore, to lowest-order, the patterning of the gammadion, so that the diagonal elements $\epsilon_{\mu\mu}(z)$ are functions of z only, so the dielectric tensor becomes

$$\epsilon(\vec{r}) = \begin{pmatrix} \epsilon_{xx}(z) & \kappa(\rho, z) e^{i\ell\phi} & 0 \\ \kappa(\rho, z) e^{-i\ell\phi} & \epsilon_{xx}(z) & 0 \\ 0 & 0 & \epsilon_{zz}(z) \end{pmatrix}. \quad (2)$$

The dielectric tensor (2) describes essentially a non-local medium for which the off-diagonal tensor elements are space-dependent through the chiral terms $\exp(\pm i\ell\phi)$, and we have taken the function $\kappa(\rho, z)$ to be real so as to highlight the effects of the chirality. We further assume that $|\epsilon_{\mu\mu}| \gg |\kappa|$.

To proceed we examine EM excitations that are bound to an individual chiral gammadion or waveguide. These EM waves are time-harmonic solutions of the Maxwell equations in the presence of the gammadion waveguide with electric fields of the form $\vec{E}(\vec{r}, t) = [\vec{E}(\vec{r}, \omega, t) e^{-i\omega t} + c.c.]/2$. In the slowly-varying envelope approximation when the vector electric field envelope $\vec{E}(\vec{r}, \omega, t)$ changes slowly on the time-scale $1/\omega$ and we neglect second-order time derivatives, Maxwell's equations lead to

$$-\nabla \times \nabla \times \vec{E} + 2i\omega\mu_0\epsilon(\vec{r}) \frac{\partial \vec{E}}{\partial t} + \mu_0\omega^2\epsilon(\vec{r})\vec{E}(\vec{r}) = 0, \quad (3)$$

with the generally complex dielectric tensor $\epsilon(\vec{r})$ given by Eq. (2). For transverse electric (TE) waves whose polarization lies in the (x, y) plane we find, using the dielectric tensor (2) and neglecting the off-diagonal elements to dominant order, that $\nabla \cdot \vec{D} = \nabla \cdot (\epsilon(\vec{r})\vec{E}) =$

$\epsilon_{xx}(z)\nabla \cdot \vec{E} = 0$, which yields $\nabla \cdot \vec{E} = 0$. This restriction to TE polarization is justified if we assume that the diagonal dielectric tensor element $\epsilon_{xx}(z)$ provides sufficient confinement along the z -axis so that the usual designation of TE polarization (electric field perpendicular to z) and transverse magnetic (TM) polarization (magnetic field perpendicular to z) for waveguides applies [5]. For strong confinement along the z -axis we assume that the field structure along the z -direction is dominated by the normalized fundamental mode $u(z)$ determined from $[d^2/dz^2 + \mu_0\omega^2\epsilon_{xx}(z)]u(z) = \beta^2 u(z)$, with β^2 the corresponding eigenvalue. Then substituting $\vec{E}(\vec{r}, \omega, t) = u(z)[\vec{e}_x\mathcal{E}_x(\rho, z, t) + \vec{e}_y\mathcal{E}_y(\rho, z, t)]$ in Eq. (3), with $\vec{e}_{x,y}$ denoting unit vectors along the respective directions, projecting out the mode structure along z , and introducing the spinor notation $\bar{\mathcal{E}}(\rho, \phi, t) = \text{col}(\mathcal{E}_x(\rho, \phi, t), \mathcal{E}_y(\rho, \phi, t))$, Eq. (3) reduces to the following effective planar equations for the EM excitations coupled to the chiral waveguide

$$2i \left(\frac{\beta^2}{\omega} \right) \frac{\partial \bar{\mathcal{E}}}{\partial t} = \hat{H}\bar{\mathcal{E}}, \quad (4)$$

where the Hermitian Hamiltonian is given by

$$\hat{H} = \hat{H}_D - \begin{pmatrix} 0 & \mu_0\omega^2\bar{\kappa}(\rho)e^{i\ell\phi} \\ \mu_0\omega^2\bar{\kappa}(\rho)e^{-i\ell\phi} & 0 \end{pmatrix}, \quad (5)$$

with diagonal part $\hat{H}_D = -(\nabla_{\perp}^2 + \beta^2)I$, I being the unit (2×2) matrix and ∇_{\perp}^2 the transverse Laplacian acting in the (x, y) plane, and $\bar{\kappa}(\rho) = \int dz |u(z)|^2 \kappa(\rho, z)$. We remark that by projecting out the mode structure $u(z)$ along z we have rendered the system effectively two-dimensional (2D). However, the physical system as described by Eqs. (4) is still sensitive to the selection of the direction of the z -axis since the gammadion winding number ℓ is defined with respect to the angular direction of phase advance around the z -axis of the chiral term $\exp(i\ell\phi)$. The key ingredient for the following discussion is that we are considering TE waves for which the electric field has only two vector components instead of three, and this allows for our two component spinor approach. This 2D planar approximation for TE waves is valid as long as the structure is tightly confined and single-mode along the z -axis. We note that Eqs. (4) are analogous to the Bogoliubov equations for electrons in a superconductor [6], with $\kappa(\rho, z)$ playing the role of the pair potential.

We next examine the time-reversal properties of the spinor wave Eq. (4). For our system a suitable antilinear time-reversal operator is $\mathcal{T} = K_0 I$, where K_0 is the complex conjugation operator [7]. By operating \mathcal{T} on Eq. (4) we obtain the time-reversed equation

$$2i \left(\frac{\beta^2}{\omega} \right) \frac{\partial}{\partial(-t)}(\mathcal{T}\bar{\mathcal{E}}) = \mathcal{T}\hat{H}\mathcal{T}^{-1}(\mathcal{T}\bar{\mathcal{E}}), \quad (6)$$

and for \hat{H} given in Eq. (5) we find

$$\mathcal{T}\hat{H}\mathcal{T}^{-1} = \hat{H}^* \equiv \mathcal{P}\hat{H}\mathcal{P}^{-1}. \quad (7)$$

For our 2D reduced system described by the spinor Eq. (4) the linear parity operator \mathcal{P} has the effect of reversing $\phi \rightarrow -\phi$, which can be implemented for the Hamiltonian (5) using $\mathcal{P} = \sigma_x$, with σ_μ being the Pauli-spin matrices. Since a structure and its corresponding enantiomeric form are interconverted by space inversion [8], Eq. (7) tells us that time-reversal is equivalent to spatial inversion. This feature was noted in Ref. [1] and simply means that under time-reversal incident EM waves approach the planar structure from the opposite direction and perceive it as the enantiomeric structure of opposite chirality. In the absence of chirality $\ell = 0$, when the Hamiltonian in Eq. (5) is purely real, the time-reversed and space inverted Hamiltonians in Eq. (7) are both equal to \hat{H} meaning that both \mathcal{T} and \mathcal{P} symmetries are intact. In contrast, in the presence of chirality \mathcal{T} and \mathcal{P} symmetries are simultaneously broken for the planar chiral structure. However, Eq. (7) implies that $(\mathcal{PT})\hat{H}(\mathcal{PT})^{-1} = \hat{H}$, so that the system is always invariant under the combined action of \mathcal{PT} symmetry [4].

More generally, the off-diagonal matrix element $\epsilon_{xy}(\vec{r})$ of the dielectric tensor may expressed as a sum over winding numbers ℓ as in Eq. (1), a special example of which is the single dominant winding number case considered above. Then, unless $\epsilon_{xy}^{(\ell)} = (\epsilon_{xy}^{(-\ell)})^*$ for all ℓ and $\epsilon(\vec{r})$ is real, both \mathcal{T} and \mathcal{P} symmetries are simultaneously broken. Essentially the time reversal and parity asymmetries in planar chiral structures are underpinned by the non-locality of the gammadion response, meaning that excitation of the extended gammadion as a whole is crucial. A planar structure consisting of individually radiating atoms could not produce such a symmetry breaking interaction.

We are now in a position to discuss how these new results relate to the experiment of Ref. [1] which provided strong experimental evidence of broken \mathcal{T} and \mathcal{P} symmetries. Distinct polarization phenomena for media where \mathcal{T} and \mathcal{P} symmetries are simultaneously broken have previously been discussed in relation to anyon matter in high-temperature superconductors, initially by Wen and Zee [9] and later by Canright and Rojo [4]. For the purpose of analyzing how the broken symmetries applicable to TE excitations may lead to observable effects, it is important to realize that an incident EM field can couple energy into the planar TE waves of individual chiral waveguides as strict momentum conservation is relaxed at the edges of the grooves of the gammadiions, and by the same argument EM energy can be out-coupled from individual chiral waveguides. Furthermore, previously the in-plane patterning of the diagonal dielectric tensor element $\epsilon_{xx}(z)$ was neglected. At next order the radial dependence of $\epsilon_{xx}(\rho, \phi, z)$ provides a grating structure that can in-couple and out-couple energy to radiation modes analogous to the emission mechanism of surface emitting circular grating lasers [15]. This coupling between the incident, out-coupled, and TE waves of the

chiral waveguides, alongside the losses present in real systems, give the TE waves a finite lifetime. Furthermore, these mechanisms offer an explanation of why the fields scattered from chiral planar nanostructures can manifest the broken \mathcal{T} and \mathcal{P} symmetries applicable to the bound TE waves, since the diffracted fields from the nanostructures have their origin in energy out-coupled from the TE waves excited by the incident field. Thus, it is physically reasonable that the scattered EM fields will inherit the broken \mathcal{T} and \mathcal{P} symmetries exhibited by the planar TE waves.

We have also found that concomitantly with the breaking of the \mathcal{T} and \mathcal{P} symmetries the corresponding TE excitations bound to chiral waveguides can acquire fractional winding numbers and we now demonstrate this interesting property which also elucidates the vector nature of the TE waves. Inspection shows that consistent stationary solutions of Eq. (4), corresponding to time-harmonic solutions at frequency ω , assume the form

$$\bar{\mathcal{E}}(\rho, \phi, t) = \begin{pmatrix} a_x(\rho)e^{i(m+\ell)\phi} \\ a_y(\rho)e^{im\phi} \end{pmatrix}, \quad (8)$$

where the winding number of the x-polarized component of the field is $(m + \ell)$, and m is that for the y-polarized component, $m = 0, \pm 1, \pm 2, \dots$ being an integer that we use to label the solutions. This form of solution ensures consistency between the azimuthal variation of the electric field polarization components in the presence of the phase dependent coupling terms $\exp(\pm i\ell\phi)$ in the dielectric tensor. We introduce the average winding number for a TE vector field as

$$\langle W_{EM} \rangle = \frac{\int_0^{2\pi} d\phi \int_0^\infty \rho dp \bar{\mathcal{E}}^\dagger(\rho, \phi, t) \hat{W}_{EM} \bar{\mathcal{E}}(\rho, \phi, t)}{\int_0^{2\pi} d\phi \int_0^\infty \rho dp \bar{\mathcal{E}}^\dagger(\rho, \phi, t) \bar{\mathcal{E}}(\rho, \phi, t)}, \quad (9)$$

where we have defined the EM winding number operator $\hat{W}_{EM} = -i(\partial/\partial\phi)I$. For the EM wave solutions in Eq. (8) we find

$$\langle W_{EM} \rangle = m + \ell(1 + \bar{s}_1)/2, \quad (10)$$

where

$$\bar{s}_1 = \frac{\int_0^{2\pi} d\phi \int_0^\infty \rho dp [|a_x(\rho)|^2 - |a_y(\rho)|^2]}{\int_0^{2\pi} d\phi \int_0^\infty \rho dp [|a_x(\rho)|^2 + |a_y(\rho)|^2]}. \quad (11)$$

is the spatially averaged and normalized value of the element $s_1 = (|\mathcal{E}_x|^2 - |\mathcal{E}_y|^2)$ of the Stokes vector $\vec{s} = (s_1, s_2, s_3)$ [10] over the plane of the individual waveguide. In general, $-1 < \bar{s}_1 < 1$, so that the EM winding number in Eq. (10) can be non-integer, that is, fractional. The average winding number of the field is associated with its orbital angular momentum that has its origin in the spatial structure of the fields [11], as distinct from the spin angular momentum associated with the polarization state of the field. The average spin angular momentum is proportional to the spatially averaged

value of the $s_3 = 2Im(\mathcal{E}_x(\rho, \phi, t)^*\mathcal{E}_y(\rho, \phi, t))$ component of the Stokes vector [10], which for $\ell \neq 0$ yields $\bar{s}_3 = 0$ for the EM solutions in Eq. (8) by virtue of the azimuthal integrals over ϕ . This implies that in the presence of planar chirality the state of polarization of the vector field components varies over the plane in such a way that the average spin angular momentum is zero, and the average angular momentum of the coupled fields is equal to the orbital angular momentum associated with the fractional modal winding number. To see the consequence of this we realize that here we are discussing the classical TE vector modes of the structure, and we anticipate that upon quantization of the field we can add photons to each of these vector modes [12]. Associated with each photon added to vector TE mode of index m there will be an average angular momentum directed along the z-axis $\langle J_z \rangle = \hbar \langle W_{EM} \rangle = \hbar(m + \ell(1 + \bar{s}_1)/2)$, that is, the average angular momentum of the quantized vector modes can be fractional. The average winding number for the EM excitation can be fractional since the deviation from the integer values expected for a bosonic field is fully accounted for by the angular momentum associated with the chiral waveguide [13, 14].

It is worthwhile commenting on the nature of the EM mode interaction with the chiral gammadiion. As noted earlier, polarization is associated with the spin of the EM field, whereas the spatial variation of the EM field describes orbital angular momentum, for example, a Laguerre-Gaussian laser field [11]. From Eq. (8) we see that for $\ell \neq 0$ the relative phase between the two vector field components varies with azimuthal angle ϕ , meaning that the polarization state of the field is not spatially homogeneous, as is obvious from the polarized images provided in Ref. [1]. Furthermore, since the two vector field components have different winding numbers they carry different orbital angular momenta. Thus, the chiral waveguide intertwines the spin and orbital angular momenta for the EM modes in such a way that they cannot be separated, in contrast to optical activity, for example, in which the molecular chirality causes polarization rotation but otherwise leaves the orbital angular momentum unchanged.

In summary, we have elucidated the physical mechanisms by which EM waves coupled to chiral waveguides can display broken \mathcal{T} and \mathcal{P} symmetries. Symmetry is recovered in the form of enantiomeric reversibility \mathcal{PT} based on the equality of excitations in the forward and

time-reversed processes involving left- and right-handed structures, respectively. We have argued that these broken symmetries should also be manifest in the fields scattered from the structures as reported in Ref. [1]. Furthermore, we have shown that the TE excitations bound to chiral waveguides can acquire fractional winding numbers. We finish by drawing attention to the analogy between the light scattering from anyon matter and chiral nanostructures, since both involve broken \mathcal{T} and \mathcal{P} symmetries and excitations with fractional angular momentum [13, 16].

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* Electronic address: ewan.wright@optics.Arizona.EDU

† Electronic address: n.i.zheludev@soton.ac.uk

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