Determining the complex Jones matrix elements of a chiral 3D optical metamaterial

Cédric Kilchoer, Narjes Abdollahi, Ullrich Steiner, Ilja Gunkel, and Bodo D. Wilts

Submitted: 09 September 2019. Accepted: 27 November 2019. Published Online: 17 December 2019
Determining the complex Jones matrix elements of a chiral 3D optical metamaterial

Cédric Kilchoer, Narjes Abdollahi, Ullrich Steiner, Ilja Gunkel, and Bodo D. Wilts

AFFILIATIONS
Adolphe Merkle Institute, University of Fribourg, Chemin des Verdiers 4, 1700 Fribourg, Switzerland

bodo.wilts@unifr.ch

ABSTRACT

Due to their strong optical activity, chiral metamaterials are attractive optical elements for the control of the polarization of light. Efficient broadband circular polarizers can be implemented through chiral nanostructures that are periodic and possess certain spatial symmetries. Here, we demonstrate a new method to fully characterize any generalized chiral medium without the use of optical phase-retarding elements, such as quarter-wave plates. Using the advantage of symmetry considerations, all parameters of the complex Jones matrix associated with the metamaterial were determined by two linear-polarization experiments. A coordinate transformation then enabled the calculation of the gyro-optical response of the sample, i.e., its circular dichroism and circular polarization conversion, which is shown to be in good agreement with direct measurements. This approach is versatile, allowing to calculate the optical response in intensity and phase of any generalized chiral metamaterial upon linear, circular, or elliptical polarized illumination.

I. INTRODUCTION

Metamaterials, various optical materials including plasmonic metasurfaces and three-dimensional (3D), structured plasmonic arrays, have recently attracted attention as they are extremely efficient in tailoring a material’s optical response and controlling the polarization of incident light. Many 3D chiral metamaterials have emerged as novel optical materials, e.g., efficient broadband circular polarizers at infrared and optical wavelengths. The performance of these materials is often characterized based on the circular dichroism (CD), the difference in transmission of right-handed (RH) and left-handed (LH) circularly polarized light. This characterization requires expensive optical components, such as quarter-wave plates with minimal retardation errors over the wavelength range of interest. These metamaterials can additionally exhibit a strong optical activity, which is defined as the effect of the rotation of linearly polarized light propagating through a chiral medium. Both CD and optical activity are generally pronounced in chiral metamaterials.

Periodically arranged 3D spirals are the typical example of a chiral metamaterial in which the handedness of the helices dictates the sign of the CD. For example, Gansel et al. demonstrated periodic gold helices exhibiting a strong CD at mid-infrared wavelengths in transmission, which were manufactured by direct laser writing followed by gold electrodeposition. These helical structures and their circular dichroism can be further optimized by increasing the number of intertwined helices within the unit cell that makes up the metamaterial. Employing focused ion beam-induced deposition techniques, helices with a smaller radius were fabricated, exhibiting a strong CD between 500 and 1000 nm. By carefully controlling the deposition angle, helices can be grown from gold nanoparticle seeds, yielding sub-100 nm gold helices with material-based tuneable CD at visible frequencies. In general, the intensity and the polarization state of light after traveling through a chiral medium strongly depend on the initial polarization state of the light.

In 1941, Jones introduced a matrix-based method to describe the polarization of a propagating electromagnetic wave inside complex optical systems. The intensity of light and its polarization are affected when passing through an optical element. Jones’ calculus determines the intensity and the polarization of light after passing through a complex optical system, i.e., a series of optical elements, using wavelength-dependent $2 \times 2$ matrices for each optical component. A serial multiplication of each optical
component allows the derivation of the intensity and polarization of exiting waves passing through the complex optical system upon the illumination by polarized light.

Jones’ calculus is broadly used for standard optical elements and birefringent crystals. A well-known example are linear polarizers, allowing only the transmission of light with the electric field component oscillating along the axis of the polarizer. Phase retarders, such as quarter-wave and half-wave plates, alter the state of polarization by rotating the propagating electromagnetic field. The influence of these optical elements on the state of polarization is well described by a wavelength-dependent $2 \times 2$ Jones matrix. While not limited to these simple optical elements, more complex materials can also be described by the Jones formalism and its use for optical metamaterials is slowly emerging. More complicated descriptions are also possible, for example, using the $4 \times 4$ Berreman or Müller matrices. We here focus on the Jones formalism due to its rather simple approach and strong descriptive power.

Jones matrices describing metamaterials have been determined experimentally26–28 or by analytic modeling.29,30 While the absolute values of the Jones matrix parameters are usually easily derived for most systems when characterized under linearly polarized illumination,21,28–30 determining the complex elements of the Jones matrix is demanding and requires a careful analysis of the optical response under a range of illumination conditions. Pfeiffer et al.31 experimentally determined the transmission coefficients of a manufactured 2D metasurface to confirm their analytical model. The transmission intensity was recorded under illumination of linearly polarized light at different angles at infrared wavelengths. The intensities in copolarization and cross-polarization were recorded and fitted to determine the four parameters of the related Jones matrix. This procedure can characterize any sample but has the inherent drawback that it does not provide the accurate phase information. In this specific analysis,32 the phase information is not important and does not influence the efficiency of the device. In general, however, the phase information is important to understand a polarization conversion caused by complex optical elements.33

Determining the complex parameters of $2 \times 2$ Jones matrices is particularly challenging for 3D metamaterials since all four matrix elements can be complex. Symmetry considerations of periodic nanostructures allow, however, the simplification of some parameters. Menzel et al.34 have introduced a classification based on the symmetry of anisotropic and chiral metamaterials. Periodic metamaterials can be sorted into five different groups, each group associated with a specific symmetry that simplifies the four complex Jones matrix elements. For example, for a mirror symmetry plane parallel to the propagation direction of incident light, the Jones matrix is diagonal with the off-diagonal terms being zero. Considering rotational symmetries allows classifying chiral periodic metamaterials, which can be assigned to three different groups. In the first group, structures without any symmetry, all elements of the Jones matrix are different. Notwithstanding, the four parameters of the related Jones matrices can still be determined experimentally. The presence of rotational symmetry, however, simplifies the Jones matrix elements, allowing the determination of the phase relation between incoming and outgoing polarizations.

In this manuscript, the focus lies on generalized chiral media that by definition have a $C_3$ or above rotational symmetry with respect to the $z$-axis as can be seen in stacked twisted resonators, oligomers, or 3D spirals. The influence of the rotational symmetry on the optical properties was studied in detail for gold helix metamaterials, where a $C_3$ or above rotational symmetry is desirable to avoid circular polarization conversion and to maximize the CD.24–26 Here, we describe a method to determine the complex Jones matrix of any generalized chiral medium at visible wavelengths, exemplified by 3D chiral optical metamaterials with different handednesses. We show that the CD can be directly obtained from the complex Jones matrix, which is derived under linearly polarized illumination.

II. THEORETICAL CONSIDERATIONS AND EXPERIMENTAL PRACTICE

In all generality, a Jones matrix has four elements, all of which can be complex. As described above, symmetry considerations enable a considerable simplification of the elements. The single gyroid morphology investigated here possesses a body-centered cubic symmetry with an inherent chirality.36–38 By definition, the single gyroid can be left- or right-handed with the symmetry properties of the $O$ point group.39 Block copolymer self-assembly allows the replication of periodic single gyroid nanostructures with the (110) out-of-plane orientation.39–41 This out-of-plane orientation is required to fulfill the symmetry considerations of generalized chiral media with a $C_3$ rotational symmetry with respect to a linear axis. This sample is an ideal test case for the determination of the Jones matrix of a chiral metamaterial.

In transmission, the Jones matrix of a generalized chiral medium has the following form in a Cartesian base:

$$T_{\text{C3}} = \begin{pmatrix} T_{xx} & T_{xy} \\ T_{yx} & T_{yy} \end{pmatrix} = \begin{pmatrix} A & B \\ -B & D \end{pmatrix} = \begin{pmatrix} |A|e^{i\beta} \\ -|B|e^{i\delta} \end{pmatrix}. (1)$$

where the (nonzero) complex coefficients $A$, $B$, and $D$ can be expressed using Euler’s formula with the exponents $\beta$ and $\delta$, where $|\cdot|$ denotes the absolute value. Due to the freedom to choose the initial phase, $A$ is chosen to be real. Furthermore, all the complex parameters of the Jones matrix and the equations shown in this work are wavelength-dependent. For clarity, we do not show this as an additional index. Using a circular base to describe the polarization state, this matrix can be rewritten as

$$T_{\text{C3,circ}} = \begin{pmatrix} T_{RR} & T_{RL} \\ T_{LR} & T_{LL} \end{pmatrix} = \frac{1}{2} \begin{pmatrix} A + D + 2iB & A - D \\ A - D & A + D - 2iB \end{pmatrix}. (2)$$

where $T_{RR}$, $T_{RL}$, $T_{LR}$, and $T_{LL}$ are the Jones matrix coefficients in the circular base. The CD of the chiral material is given by the difference between the main-diagonal elements $T_{RR} - T_{LL} = 2iB$. Since the off-diagonal terms $T_{RL}$ and $T_{LR}$ have nonzero values, a polarization conversion between the two circular states is generally expected.

To measure the phases, multiple measurements need to be carried out under an in-plane sample rotation at different angles $\theta$. Upon rotation of an optical element by an angle $\theta$, the associated Jones matrix becomes $T(\theta) = R(\theta)TR(\theta)$, where the rotation matrix $R(\theta)$ is defined by

$$R(\theta) = \begin{pmatrix} \cos(\theta) & \sin(\theta) \\ -\sin(\theta) & \cos(\theta) \end{pmatrix}. (3)$$
For an arbitrary angle \( \theta \), the relation \( R(-\theta)T_{C_{x,y}, 0}R(\theta) \) can be expanded

\[
T_{C_{x,y}, 0} = \begin{pmatrix} T_{xx}(\theta) & T_{xy}(\theta) \\ T_{yx}(\theta) & T_{yy}(\theta) \end{pmatrix} = \begin{pmatrix} \cos(-\theta) & \sin(-\theta) \\ -\sin(-\theta) & \cos(-\theta) \end{pmatrix} \begin{pmatrix} A & B \\ -B & D \end{pmatrix} \begin{pmatrix} \cos(\theta) & \sin(\theta) \\ -\sin(\theta) & \cos(\theta) \end{pmatrix} = \begin{pmatrix} A\cos^2(\theta) + D\sin^2(\theta) & (A - D)\cos(\theta)\sin(\theta) + B \\ (A - D)\cos(\theta)\sin(\theta) - B & D\cos^2(\theta) + A\sin^2(\theta) \end{pmatrix}.
\] (4)

where \( t_x(\theta) \) and \( t_y(\theta) \) are the transmitted amplitudes for the two polarizations (Jones vector).

All matrix elements are a function of the rotation angle \( \theta \). For the Jones matrix to be expressed in the form of Eq. (1), its principal coordinate axis needs to be aligned to the sample symmetry axis.\(^{11}\) Since the in-plane orientation of the sample is unknown at the time of a measurement, an additional angular offset \( \theta_0 \), the difference between the sample and the polarizer axis, needs to be included as an additional free fit parameter. For the fitting procedure, it is convenient to set \( A \) to be purely real at the specific angle \( \theta \). The second Jones element, \( D \), remains complex and is expressed in the form \( |D|e^{i\delta} \). With linearly polarized incident light along the \( x \)-axis, Eq. (5) becomes

\[
\begin{pmatrix} t_x(\theta) \\ t_y(\theta) \end{pmatrix} = \begin{pmatrix} A\cos^2(\theta) + D\sin^2(\theta) \\ 0 \end{pmatrix} = \begin{pmatrix} |A|\cos^2(\theta - \theta_0) + |D|e^{i\delta}\sin^2(\theta - \theta_0) \\ 0 \end{pmatrix}.
\] (6)

To experimentally determine the four parameters of the Jones matrix, the transmitted intensity is recorded while rotating the sample through a range of \( \theta \) angles. More specifically, a combination of measurements in co- and cross-polarization allow determining the Jones parameters in Eq. (4) based on a two-step fitting procedure of the recorded intensities, as detailed below.

A. Copolarization

In a first measurement series, two linear polarizers are coaligned along the \( x \)-axis while the sample is rotated through different \( \theta \) angles. This allows the extraction of the \( A \) and \( D \) parameters of the Jones matrix in Eq. (4),

\[
\begin{pmatrix} t_x(\theta) \\ t_y(\theta) \end{pmatrix} = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \begin{pmatrix} A\cos^2(\theta) + D\sin^2(\theta) \\ (A - D)\cos(\theta)\sin(\theta) - B \end{pmatrix} = \begin{pmatrix} 0 \\ 1 \end{pmatrix} = \begin{pmatrix} A\cos^2(\theta) + D\sin^2(\theta) \\ 0 \end{pmatrix}.
\] (5)

The relative intensity collected after the analyzer is the sum of the squares of the absolute values of the two components of the Jones vector,

\[
I(\theta) = (t_x^* t_x + t_y^* t_y) = t_x^* t_x + t_y^* t_y.
\] (7)

B. Cross-polarization

In cross-polarization, the off-axis elements become important. When the axis of the second polarizer (analyzer) is rotated by 90° compared to the axis of the first linear polarizer (crossed polarizers), Eq. (5) becomes

\[
\begin{pmatrix} t_x(\theta) \\ t_y(\theta) \end{pmatrix} = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} \begin{pmatrix} A\cos^2(\theta) + D\sin^2(\theta) \\ (A - D)\cos(\theta)\sin(\theta) + B \end{pmatrix} = \begin{pmatrix} 0 \\ A - D \cos(\theta)\sin(\theta) - B \end{pmatrix} = \begin{pmatrix} 0 \\ (A - D)\cos(\theta)\sin(\theta) - B \end{pmatrix}.
\] (8)

In this way, all elements of the (complex) Jones matrix for this metamaterial can be determined based on fitting measured copolarized and cross-polarized intensities to Eqs. (6), (7), and (9).

III. RESULTS AND DISCUSSION

To demonstrate the ability to experimentally determine all Jones matrix parameters for a chiral metamaterial, the optical
response of a silver single gyroid metamaterial was measured and analyzed [Figs. 1(b) and 1(c)]. The silver gyroid sample was mounted onto a manual rotation stage that was placed between two linear polarizers as depicted in Fig. 1(a), allowing to manually rotate the sample through a $360^\circ$ range. The optical transmission in the co- and cross-polarization configuration was measured with a custom-built microspectrophotometer. Linearly polarized light (defining $0^\circ$ and the $x$-axis in the here used coordinate system) propagated through the sample that was rotated at an angle $\theta$. In copolarization, the axes of the two polarizers were coaligned, resulting in the spectra shown in Fig. 1(d), in which the transmitted intensity is collected in $10^\circ$ angular steps. The transmittance spectrum exhibits a wavelength-dependent $180^\circ$-periodicity. The corresponding cross-polarization measurements, in which the analyzer is aligned at $90^\circ$ with respect to the $x$-axis, show a $90^\circ$ periodicity, in which the maxima of every second peak have a 100-nm wavelength offset. As explained in Sec. II, both co- and cross-polarization measurements are needed to determine the full set of complex parameters of the Jones matrix.

A. Parameter fitting

First, the intensity recorded in copolarization was fitted to Eqs. (6) and (7) for the entire spectral range. In this way, the parameters $|A|$, $|D|$, and $\delta$ were extracted, as well as the angular offset $\theta_0$. The offset angle $\theta_0$ was constant within a $\pm 2.5^\circ$ margin across the entire spectral range (Fig. S1 of the supplementary material). Figure 2 shows the fits for three different wavelengths (500 nm, 600 nm, and 700 nm), all showing a good agreement with the corresponding experimental data. Experimental copolarization peaks and plateaus are well-fitted by the theoretical model, allowing the reliable extraction of $|A|$, $|D|$, $\delta$, and $\theta_0$.

In a second step, the Jones parameters $B$ and $\beta$ were determined by fitting Eqs. (7) and (9) to the spectrum in Fig. 1(e), using the previously extracted parameters. Figure 2(b) shows the fit together with the measured spectra for three selected wavelengths. The close agreement shows the robustness of the extraction of the Jones parameters $B$ and $\beta$.

FIG. 1. Co- and cross-polarized transmittance measurements as a function of the in-plane sample rotation angle $\theta$. (a) Schematic of the experimental setup. Linearly polarized light passes through the sample, which is rotated through $\theta$. The $\lambda/4$-plates were only used in the measurements of Sec. III B. The transmittance was recorded in either copolarization (second linear polarizer or analyzer at $0^\circ$) or cross-polarization ($90^\circ$). (b) 3D representation of the silver single gyroid optical metamaterial used in this study. (c) Scanning electron micrograph of the surface of a silver gyroid metamaterial oriented with the $(110)$ out-of-plane orientation. Scale bar: 500 nm. $\theta$- and wavelength-resolved transmittance of a silver gyroid metamaterial in (d) copolarization and (e) cross-polarization.
FIG. 2. Experimental data (symbols) and fits (solid lines) for three different wavelengths. (a) Equations (6) and (7) were fitted to the copolarization spectra, allowing the determination of the Jones parameters $|A|$, $|D|$, $\delta$, and $\theta_0$. (b) The parameters $|B|$ and $\beta$ were then determined by fitting Eqs. (7) and (9) to the cross-polarization spectra.

Figure 3 shows the spectral variation of all measured parameters of the complex Jones matrix extracted from the two angularly resolved linear polarization transmission experiments of the 3D chiral silver gyroid metamaterial sample.

B. Jones matrix in circular base

To validate the reliability of our method, we calculated the Jones’ matrix elements in the circular base from those in the linear base using Eq. (2). Since the single gyroid is a chiral morphology, we expect a significant CD. To test this, the gyro-optical response of the gyroid was measured and compared to predictions derived from the analysis above. To this end, the optical setup was modified by inserting quarter-wave plates between the two linear polarizers and the sample, as indicated by the dashed lines in Fig. 1(a). This provides right- and left-handed circularly polarized (RCP and LCP, respectively) illumination of the sample and the analysis of the circular polarization of the transmitted light. Depending on the setting of the two quarter-wave plates, the RCP transmitted intensity $I_{RR}$ upon RCP illumination was measured. Similarly, $I_{LL}$, $I_{RL}$, and $I_{LR}$ were also recorded as a function of wavelength.

The measured gyro-optical intensities were then compared to the Jones matrix predictions of Eq. (2). For example, the RCP transmitted amplitude upon RCP illumination is given by

$$
I_{RR} = \left( \begin{array}{c} 1 \\ 0 \end{array} \right) \left( \begin{array}{cccc} T_{RR} & T_{RL} \\ T_{LR} & T_{LL} \end{array} \right) \left( \begin{array}{c} 1 \\ 0 \end{array} \right).
$$

Using the expression of the Jones matrix from Eqs. (2) and (7), $I_{RR}$ can be calculated from the values $|A|$, $|B|$, $|D|$, $\beta$, and $\delta$ determined above. The other three gyro-optical intensities can be

FIG. 3. Jones matrix parameters for a silver gyroid optical metamaterial. (a) Extracted $|A|$, $|B|$, and $|D|$ parameters from co- and cross-polarization measurements. (b) Extracted $\beta$ and $\delta$ parameters. The shaded areas show the 95% confidence interval of the fits.
FIG. 4. Transmission of circularly polarized light through a silver gyroid optical metamaterial. Comparison between experimental data and calculated spectra from Eqs. (2) and (7), using the fitted values of $|A|$, $|B|$, $|D|$, $\beta$, and $\delta$ from the linear polarization experiments. Experimental data were acquired using two sets of a linear polarizer and a quarter-wave plate, as indicated in Fig. 1. $I_{RR}$ ($I_{LL}$) is the RCP (LCP) transmission upon RCP (LCP) illumination. $I_{RL}$ and $I_{LR}$ are related to polarization conversion from one handedness to the other.

determined in an analogous fashion. Figure 4 compares the measured gyro-optical intensities with these predictions. Clearly, the calculated gyro-optical transmittance spectra, which are based on the analysis of linearly polarized experiments, match the experimental curves very well.

Figures 5(b) and 5(c) show microscopy images corresponding to the $I_{RR}$ and $I_{LL}$ settings of the quarter-wave plates [dashed lines in Fig. 1(a)]. Two types of domains in the silver gyroid are clearly visible, corresponding to positive CD ($I_{RR} > I_{LL}$) and negative CD ($I_{RR} < I_{LL}$), which are indicative of different orientations of the chiral axes of the gyroid. Figure 5(a) compares the Euler exponents of the Jones matrix parameters of the right-handed (RH) and left-handed (LH) single gyroid domains marked in Figs. 5(b) and 5(c). Only the off-axis phase $\beta$ is significantly affected by the handedness of the domains. Across the entire spectral range, the $\beta$-values corresponding to RH and LH single gyroids, $\beta_{RH}$, $\beta_{LH}$, respectively, differ by $\approx \pi$ for the two handednesses.

This is in agreement with the relation between the diagonal elements of the Jones matrix $T_{RR} - T_{LL} = 2iB = 2i|B|e^{i\delta}$ from Eq. (2). Both the real and the imaginary part of the off-diagonal Jones matrix elements, $B$ and $-B$, in Eq. (1) control the sign and strength of the CD of the chiral sample. Figures 5(d) and 5(e) show the scalar reconstructions at the wavelength exhibiting a maximum CD (510 nm). A peak in the CD is reached when the phase difference between the vectors $B$ and $A + D$ is around $\pm \pi/2$, with the sign indicating whether the sample transmits more RCP or more LCP light. Note also the substantial magnitude of $I_{RL}$ in Fig. 4, which is indicative of a polarization conversion of the gyroid metamaterial converting the polarization from RCP to LCP light, and vice versa for $I_{LR}$.

Our approach demonstrates the versatility of the base transformation (Fig. 4), which is enabled by the complex notation of the Jones matrix, preserving the phase information upon all mathematical operations. The phase information is crucial to determine the strength of the CD and its handedness. An analysis of the eigenpolarizations of the retrieved Jones matrix is shown in Fig. S2 of the supplementary material, where the wavelength-dependent polarization eigenstates are displayed on the Poincaré sphere. These states are elliptically counter-rotating, as expected for all generalized chiral media. This shows that the Jones matrix characterization allows the complete understanding of the metamaterial optical properties without the use of more complex analytical approaches, such as the Berreman or Müller matrices.
The main limiting factor of this method arises from the required symmetry properties of the samples. For a metamaterial that does not possess any symmetry, i.e., it is part of the arbitrary complex media group, all four parameters of the Jones matrix are distinct. In this case, a copolarization measurement depends on all four complex matrix parameters and assessing them based on fits to the data is no longer possible. However, in practice, most metamaterials used as circular polarizers feature different symmetries, e.g., a $C_3$ or $C_4$ rotational symmetry. These materials have purely circular eigenstates, avoid polarization conversion, and therefore maximize the CD.\textsuperscript{31,42}

Co- and cross-polarization measurements using only linear polarizers enable a complete broadband optical characterization, without the need for quarter-wave plates or other phase retarding components. Commercially available quarter-wave plates are often expensive and have a limited spectral range, particularly at short wavelengths, making the direct gyro-optical characterization cumbersome.

IV. CONCLUSIONS

We have developed a Jones matrix method for the optical characterization of any generalized chiral medium by experimentally determining the complex Jones matrix parameters. This method enables the derivation of the gyro-optical response of a chiral sample (i.e., its CD) through co- and cross-polarized transmittance measurements using only linear polarizers. Using symmetry properties of chiral media, the complex parameters of the Jones matrix were determined by a fitting routine. The full characterization of the Jones matrix provides a complete understanding of the polarization state of the light traveling through the chiral metamaterial, for any incoming light polarization and any in-plane rotation of the metamaterial. Based on a coordinate transformation, the optical response of the sample was calculated upon circular polarized illumination, in good agreement with the experimental measurements, demonstrating the analysis of a sample’s CD without employing quarter-wave plates. This method is efficient for a wide spectral range, enabling characterization of chiral metamaterials with relatively simple optical equipment, such as an optical microscope equipped with linear polarizers.

V. MATERIALS AND METHODS

A. Sample fabrication

The fabrication of the silver single gyroid metamaterial sample followed the procedure described in Ref. 40. In brief, a 10% solution of polyisoprene-$h$-polystyrene-$h$-poly(ethylene oxide) (ISO) triblock terpolymer ($80 \text{ kg/mol}$, $f_{SI} = 0.30$, $f_{PS} = 0.53$, $f_{PEO} = 0.17$) in anhydrous anisole (Sigma-Aldrich) was spin coated for $60 \text{ s}$ at $1200 \text{ rpm}$ onto a fluorine-doped tin oxide coated glass substrate (FTO glass, Sigma-Aldrich). Prior to film deposition, the FTO glass was immersed in a Piranha bath and then silanized by immersion in a $0.2\%$ solution of octyltrichlorosilane (Sigma-Aldrich) in cyclohexane for $15 \text{ s}$.

The as-spun samples were annealed in a controlled solvent vapor atmosphere, as described in Ref. 39. Annealed films were illuminated by UV light (Mineralight XX-15S, $254 \text{ nm}$) for $15 \text{ min}$ and washed with ethanol for $30 \text{ min}$ to remove the PI from the matrix, thereby obtaining a voided gyroid network. The sample was backfilled with silver by electrodeposition. The electrodeposition was carried out with a Metrohm AutoLab PGSTAT302N potentiostat using a MetSil 500CNF (Metalor) silver solution. Ag/AgCl with KCl (Metrohm) and platinum electrode tip (Metrohm) were used as the reference and counter electrodes, respectively.

B. Optical characterization

A Zeiss Axio Scope.A1 (Zeiss AG, Oberkochen, Germany) polarized light microscope and a xenon light source (Thorlabs SLS401; Thorlabs GmbH, Dachau, Germany) were used for all optical experiments. For spectroscopic measurements, the transmitted and reflected light were collected by an optical fiber (QP230-2-XSR, $230 \mu\text{m}$ core) with a measurement spot size of $<20 \mu\text{m}$. The spectra were recorded with a spectrometer (Ocean Optics Maya2000 Pro; Ocean Optics, Dunedin, FL, USA). Measurement errors are below $0.05\%$ for the selected wavelength range. Light microscopy images were acquired with a Point Grey GS3-U3-2855C-C (FLIR Integrated Imaging Solutions, Inc., Richmond, Canada) CCD camera. The linear polarizers (Thorlabs WP25M-UB, Thorlabs) and quarter-wave plates (Halle, Germany) were both superachromatic, allowing an effective measurement range of $400-850 \text{ nm}$. Slight deviations of the chromatic retardance of the quarter-wave plates have no significant impact on the measured transmittance.

C. Scanning electron microscopy (SEM)

Samples were imaged using a TESCAN (TESCAN, a.s., Brno, Czech Republic) MIRA3 field emission scanning electron microscope.

D. Fitting method

The model was fitted to the experimental data using a custom MATLAB code. To extract the Jones matrix parameters, the theoretical model fitted the angle-dependent co- and cross-polarization spectra using the Levenberg-Marquardt nonlinear least squares algorithm for each wavelength in the wavelength-region of interest.

SUPPLEMENTARY MATERIAL

See the supplementary material for supplementary movies and details of the presented method. Additional data related to this publication is available at Zenodo data repository (http://dx.doi.org/10.5281/zenodo.3471335).

ACKNOWLEDGMENTS

We thank Matthias Saba for helpful comments on this manuscript and Yibei Gu and Ulrich Wiesner for the provision of the triblock terpolymer. This research was supported by the Swiss National Science Foundation through Grant No. 163220, the Ambizione program (Grant No. 168223 to B.D.W.), and the National Centre of Competence in Research “Bio-Inspired Materials.”

The authors declare no conflict of interest.
REFERENCES

1. J. Hao, Y. Yuan, L. Ran, T. Jiang, J. A. Kong, C. T. Chan, and L. Zhou, “Manipulating electromagnetic wave polarizations by anisotropic metamaterials,” Phys. Rev. Lett. 99, 063908 (2007).

2. Y. Ye and S. He, “90° polarization rotator using a bilayered chiral metamaterial with giant optical activity,” Appl. Phys. Lett. 96, 203501 (2010).

3. J. Kaschke, L. Blume, L. Wu, M. Thiel, K. Bade, Z. Yang, and M. Wegener, “A helical metamaterial for broadband circular polarization conversion,” Adv. Opt. Mater. 3, 1411–1417 (2015).

4. Z. Wang, F. Cheng, T. Winor, and Y. Liu, “Optical chiral metamaterials: A review of the fundamentals, fabrication methods and applications,” Nanotechnol. 27, 412001 (2016).

5. M. Decker, R. Zhao, C. M. Soukoulis, S. Linden, and M. Wegener, “Twisted splitting-ring-resonator photonic metamaterial with huge optical activity,” Opt. Lett. 35, 1593 (2010).

6. O. Arteaga, J. Sancho-Parramon, S. Nichols, B. M. Moxz, A. Canillas, S. Bosch, G. Markovich, and B. Kahr, “Relation between 2D/3D chirality and the appearance of chiroptical effects in real nanostructures,” Opt. Express 24, 2242–2252 (2016).

7. J. K. Gansel, M. Thiel, M. S. Rill, M. Decker, K. Bade, V. Saile, G. Von Freymann, S. Linden, and M. Wegener, “Gold helix photonic metamaterial as broadband circular polarizer,” Science 325, 1513–1519 (2009).

8. J. Kaschke, J. K. Gansel, and M. Wegener, “On metamaterial circular polarizers based on metal N-helices,” Opt. Express 20, 26012 (2012).

9. M. Esposito, V. Tasco, M. Cuscinà, F. Todisco, A. Benedettì, I. Tarantini, M. D. Giorgi, D. Sanvitto, and A. Passaseo, “Nanoscale 3D chiral plasmonic helices with circular dichroism at visible frequencies,” ACS Photonics 2, 105–115 (2015).

10. J. Kaschke and M. Wegener, “Gold triple-helix mid-infrared metamaterial by STED-inspired laser lithography,” Opt. Lett. 40, 3986 (2015).

11. Z. Y. Yang, M. Zhao, P. X. Lu, and Y. F. Lu, “Ultrabroadband optical circular polarizers consisting of double-helical nanostructure stacks,” Opt. Lett. 35, 2588 (2010).

12. J. Kaschke, M. Blome, S. Burger, and M. Wegener, “Tapered N-helical metamaterials with three-fold rotational symmetry as improved circular polarizers,” Opt. Express 22, 19936 (2014).

13. M. Esposito, V. Tasco, F. Todisco, M. Cuscinà, A. Benedettì, D. Sanvitto, and A. Passaseo, “Triple-helical nanostructures by topographical rotary growth for chiral photonics,” Nat. Commun. 6, 6484 (2015).

14. A. G. Mark, J. G. Gibbs, T.-C. Lee, and P. Fischer, “Hybrid nanocolloids with programmed three-dimensional shape and material composition,” Nat. Mater. 12, 802–807 (2013).

15. H. Singh, G. Nair, A. Ghosh, and A. Ghosh, “Wafer scale fabrication of porous three-dimensional plasmonic metamaterials for the visible region: Chiral and beyond,” Nanoscale 5, 7224 (2013).

16. R. Jones, “A new calculus for the treatment of optical systems I. Description and discussion of the calculation,” J. Opt. Soc. Am. 31, 488 (1941).

17. P. Yeh, “Extended Jones matrix method,” J. Opt. Soc. Am. 72, 507 (1982).

18. D. Goldstein, Polarized Light (CRC Press, 2010).

19. A. Gerrard and J. Burch, Introduction to Matrix Methods in Optics, 1st ed. (Dover Publications, 2012).

20. S.-Y. Lu and R. A. Chipman, “Homogeneous and inhomogeneous Jones matrices,” J. Opt. Soc. Am. A 11, 766 (1994).

21. D. G. Stavenga, H. L. Leertouwer, and B. D. Wilts, “Quantifying the refractive index dispersion of a pigmented biological tissue using Jamin–Lebedeff interference microscopy,” Light: Sci. Appl. 2, e100 (2013).

22. D. W. Berreman, “Optics in stratified and anisotropic media: 4 × 4 matrix formulation,” J. Opt. Soc. Am. 62, 502–510 (1972).

23. W. S. Bickel and W. M. Bailey, “Stokes vectors, Mueller matrices, and polarized scattered light,” Am. Phys. Soc. 53, 468–478 (1985).

24. G. Kenanakis, A. Xomalis, A. Selimus, M. Varvokakai, M. Farsari, M. Kafesaki, C. M. Soukoulis, and E. N. Economou, “Three-dimensional infrared metamaterial with asymmetric transmission,” ACS Photonics 2, 287–294 (2015).

25. M. Kang, J. Chen, H.-X. Cui, Y. Li, and H.-T. Wang, “Asymmetric transmission for linearly polarized electromagnetic radiation,” Opt. Express 19, 8347 (2011).

26. J. Sperrhake, M. Decker, M. Falkner, S. Fasold, T. Kaiser, I. Staude, and T. Pertsch, “Analyzing the polarization response of a chiral metamaterial stack by semi-analytic modeling,” Opt. Express 27, 1236 (2019).

27. C. Pfeiffer, C. Zhang, V. Ray, L. Jay Guo, and A. Grbic, “Polarization rotation with ultra-thin bianisotropic metasurfaces,” Optica 3, 427 (2016).

28. Y. Cheng, Y. Nie, X. Wang, and R. Gong, “An ultrathin transparent metamaterial polarization transformer based on a twist-split-ring resonator,” Appl. Phys. A 111, 209–215 (2013).

29. Z. Li, M. Gokkavas, and E. Ozbay, “Manipulation of asymmetric transmission in planar chiral nanostructures by anisotropic loss,” Adv. Opt. Mater. 1, 482–488 (2013).

30. D. Liu, Z. Xiao, X. Ma, L. Wang, K. Xu, J. Tang, and Z. Wang, “Dual-band asymmetric transmission of chiral metamaterial based on complementary u-shaped structure,” Appl. Phys. A 118, 787–791 (2015).

31. C. Menzel, C. Rockstuhl, and F. Lederer, “Advanced Jones calculus for the classification of periodic metamaterials,” Phys. Rev. A 82, 053811 (2010).

32. C. Menzel, C. Helgert, C. Rockstuhl, R. E. Kley, A. Tünnemann, T. Pertsch, and F. Lederer, “Asymmetric transmission of linearly polarized light at optical metamaterials,” Phys. Rev. Lett. 104, 253902 (2010).

33. L. Wu, Z. Yang, Y. Cheng, Z. Lu, P. Zhang, M. Zhao, R. Gong, X. Yuan, Y. Zheng, and J. Duan, “Electromagnetic manifestation of chirality in layer-by-layer chiral metamaterials,” Opt. Express 21, 5239 (2013).

34. M. Hentschel, M. Schäferling, T. Weiss, N. Liu, and H. Giessen, “Three-dimensional chiral plasmonic oligomers,” Nano Lett. 12, 2542–2547 (2012).

35. J. K. Gansel, M. Wegener, S. Burger, and S. Linden, “Gold helix photonic metamaterials: A numerical parameter study,” Opt. Express 18, 1059 (2010).

36. S. S. Oh, A. Demetriadou, S. Wuestner, and O. Hess, “On the origin of chirality in nanoplasmonic gyroid metamaterials,” Adv. Mater. 25, 612–617 (2013).

37. A. Dolan, B. D. Wilts, S. Vignolmi, J. J. Baumberg, U. Steiner, and T. D. Wilkinson, “Optical properties of gyroid structured materials: From photonic crystals to metamaterials,” Adv. Opt. Mater. 3, 12–32 (2015).

38. A. Schoen, “Infinite periodic minimal surfaces without self-intersections,” NASA Technical Report No. 1–92, 1970.

39. R. Dehmel, J. A. Dolan, Y. Gu, U. Wiesner, T. D. Wilkinson, J. J. Baumberg, U. Steiner, B. D. Wilts, and I. Gunkel, “Optical imaging of large gyroid crystals in block copolymer templates by confined crystallization,” Macromolecules 50, 6255–6262 (2017).

40. A. Dolan, R. Dehmel, A. Demetriadou, Y. Gu, U. Wiesner, T. D. Wilkinson, I. Gunkel, O. Hess, J. J. Baumberg, U. Steiner et al., “Metasurfaces atop metamaterials: Surface morphology induces linear dichroism in chiral optical metamaterials,” Adv. Mater. 31, 1803478 (2019).

41. J. A. Dolan, K. Korzeb, R. Dehmel, K. C. Gödel, M. Stelik, U. Wiesner, T. D. Wilkinson, J. J. Baumberg, B. D. Wilts, U. Steiner, and I. Gunkel, “Controlling self-assembly in gyroid terpolymer films by solvent vapor annealing,” Small 14, 1802401 (2018).

42. J. Kaschke and M. Wegener, “Optical and infrared helical metamaterials,” Nanophotonics 5, 510–523 (2016).