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Robust Extraction of Hyperbolic Metamaterial Permittivity using Total Internal Reflection Ellipsometry

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Abstract

Hyperbolic metamaterials are optical materials characterized by highly anisotropic effective permittivity tensor components having opposite signs along orthogonal directions. The techniques currently employed for characterizing the optical properties of hyperbolic metamaterials are limited in their capability for robust extraction of the complex permittivity tensor. Here we demonstrate how an ellipsometry technique based on total internal reflection can be leveraged to extract the permittivity of hyperbolic metamaterials with improved robustness and accuracy. By enhancing the interaction of light with the metamaterial stacks, improved ellipsometric sensitivity for subsequent permittivity extraction is obtained. The technique does not require any modification of the hyperbolic metamaterial sample or sophisticated ellipsometry set-up, and could therefore serve as a reliable and easy-to-adopt technique for characterization of a broad class of anisotropic metamaterials.

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Author Contributions:
The sample was fabricated and characterized by C. Zhang, C. Ji, W. Zhu, and Z. Zhang. The ellipsometry measurement was performed by N. Hong, T. E. Tiwald, S. Schoeche, and J. N. Hilfiker. Data was analyzed by C. Zhang, N. Hong, W. Zhu, X. Chen, A. Agrawal, L. J. Guo, and H. J. Lezec. The manuscript was written through contributions of all authors.

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website.

Measured permittivity of Cu-doped Ag and Ta₂O₅; X-ray diffraction (XRD) characterization of Cu-doped Ag and Ta₂O₅; Definition of the mean-squared-error (MSE); Isotropic multi-layer modeling of HMMs; Effective in-plane and out-of-plane permittivity components calculated by EMT; Uncertainty values of extracted parameters; Uniqueness test of fitting parameters

The authors declare no competing financial interest.

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Keywords

spectroscopic ellipsometry; total internal reflection ellipsometry; hyperbolic metamaterial; anisotropic metamaterial

Hyperbolic metamaterials (HMMs) are highly anisotropic structures that exhibit metallic ($\text{Re}(\epsilon) < 0$) and dielectric ($\text{Re}(\epsilon) > 0$) responses along orthogonal directions.^{1, 2} They have been utilized to demonstrate various phenomena, including broadband light absorption,^{3, 4} enhanced spontaneous emission,⁵⁻⁷ asymmetric light transmission,⁸ engineered thermal radiation,^{9, 10} and sub-diffraction imaging.¹¹⁻¹³ For an HMM formed by a planar stack of alternating metal and dielectric layers, the optical response is described by an effective magnetic permeability equal to the value of free-space, and a complex effective relative electric permittivity tensor of the form:

$$\epsilon_{eff} = \begin{pmatrix} \epsilon_{\parallel} & 0 & 0 \\ 0 & \epsilon_{\parallel} & 0 \\ 0 & 0 & \epsilon_{\perp} \end{pmatrix} \quad 1$$

Here, the subscripts $\parallel$ and $\perp$ indicate permittivity components for electric field orientation parallel and perpendicular to the plane of layers, respectively. The complex effective permittivities $\epsilon_{\parallel} = \epsilon'_{\parallel} + i\epsilon''_{\parallel}$ and $\epsilon_{\perp} = \epsilon'_{\perp} + i\epsilon''_{\perp}$ can be calculated using the Maxwell-Garnett effective medium theory (EMT):¹⁴

$$\begin{cases} \epsilon_{\parallel} = \rho \cdot \epsilon_m + (1 - \rho) \cdot \epsilon_d \\ \frac{1}{\epsilon_{\perp}} = \frac{\rho}{\epsilon_m} + \frac{1 - \rho}{\epsilon_d} \end{cases} \quad 2$$

where ρ is the volumetric fraction of the constituent metal layers, and ϵ_m and ϵ_d are the local complex permittivities of the constituent metal and dielectric substances, respectively.

The key to the array of rich phenomena enabled by HMMs is their highly anisotropic permittivity. HMMs reported to date are often described by numerically calculated permittivity tensors based on EMT, which utilizes constituent metal and dielectric permittivities reported in the literature or measured by spectroscopic ellipsometry.^{7, 15-17} However, the accuracy of this calculation is limited by the known precision of experimental values of layer thicknesses and local permittivities, as well as non-modeled effects such as layer roughness, strain, and interlayer diffusion. In contrast, spectroscopic ellipsometry provides a more direct path to determine the optical properties of as-fabricated structures.¹⁸⁻²¹ Recently, spectroscopic ellipsometry has been utilized to extract the effective complex permittivity tensor of HMMs, which are treated as homogenous, uniaxial materials in the ellipsometry modeling procedure.²² Though a reasonable correspondence between ellipsometry-extracted and EMT-calculated in-plane permittivity components is obtained, the respective out-of-plane permittivity components display a non-negligible discrepancy.

In this work, we demonstrate how both the in-plane and out-of-plane effective permittivities of an HMM operating at ultraviolet, visible, and near-infrared frequencies can be accurately extracted using a coupling-prism-enabled spectroscopic ellipsometry technique based on total internal reflection (TIR). For reference, this technique is compared to two other spectroscopic ellipsometry methods commonly used to date for optically transmissive HMM characterization, namely (1) interference enhancement (IE),^{23, 24} in which reflection-mode spectroscopic ellipsometry exploits a substrate coated with a silicon oxide layer to enhance light-HMM interaction, and (2) reflection-mode spectroscopic ellipsometry plus transmission (SE+T),²⁵ which adds normal-incidence transmittance spectroscopy to standard reflection-mode ellipsometry. Although both IE and SE+T techniques have been successfully used for characterizing isotropic thin absorbing films, we show here that neither method is able to robustly extract the HMM out-of-plane effective permittivity. In contrast, the TIR method is demonstrated to provide robust extraction of the entire permittivity tensor having well-converged fitting parameters. In particular, measurement sensitivity to the out-of-plane permittivity is improved compared to both the IE and SE+T cases, via prism-mediated enhancement of the out-of-plane electric field inside the HMM. The TIR technique requires neither modification of the HMM sample itself nor substantial re-configuration of a standard ellipsometer, and can therefore serve as a reliable and easy-to-adopt technique for the characterization of both HMMs and a variety of other anisotropic metamaterials.

Results

Implementation of the hyperbolic metamaterial

The HMM studied here (Figure 1a), designed to exhibit a type-II hyperbolic dispersion ($\text{Re}(\epsilon_{\parallel}) < 0$ and $\text{Re}(\epsilon_{\perp}) > 0$) in the wavelength regime longer than 600 nm, is based on alternating layers of Cu-doped Ag (nominal thickness: 8 nm) and Ta₂O₅ (nominal thickness: 20 nm) to a total number of 4 and 3, respectively. The HMM is terminated with an additional half layer of Ta₂O₅ (nominal thickness: 10 nm) on each side, yielding a total nominal thickness of 112 nm. Ag is chosen for its low optical loss from the mid-ultraviolet to near-infrared and Cu doping is employed to enable formation of ultra-thin and smooth films with optical properties similar to that of pure Ag.^{26–28} The deposited Ta₂O₅ film is nominally stoichiometric. The measured permittivity as well as x-ray diffraction (XRD) characterization of Cu-doped Ag (poly-crystalline) and Ta₂O₅ (amorphous) films are listed in Section I and II, Supporting Information. The choice of 4 pairs of metal and dielectric films, where each film has a deep subwavelength thickness and roughness, enables modeling of the HMM as a homogenous effective medium.^{29–31} HMM samples are deposited on two types of substrates: a single-side polished, silicon substrate coated with a 300 nm thick thermal oxide layer for the IE measurement (Figure 1b), and a double-side polished, 500 μm thick fused silica substrate for both the SE+T and TIR measurement (Figure 1c and 1d, respectively).

Measurement procedure

Optical probing during reflection-mode spectroscopic ellipsometry is performed at discrete angles θ_i with respect to the normal to the plane of the HMM layers, under two fundamentally different configurations, with light incident upon (1) the HMM-free space

interface for IE ($\theta_{i=1,2,3} = 55^\circ, 65^\circ, 75^\circ$), and SE+T ($\theta_{i=1,2,3} = 50^\circ, 60^\circ, 70^\circ$) measurements (Figures 1b and 1c, left panel), and (2) the HMM-fused silica interface for the TIR ($\theta_{i=1} = 60^\circ$) measurement, by means of an equilateral coupling prism in optical contact with the silica substrate (Figure 1d). The optical contact is achieved by using an index matching liquid, which provides close index matching to the fused silica substrate over the entire wavelength range of this study. The complex electric-field reflection coefficients for each polarization, r_p and r_s , are recorded at each incident angle as a function of frequency f over an ultraviolet to near-IR frequency range of 1500 THz to 176 THz (corresponding to free-space wavelength λ_0 between 200 nm to 1700 nm). Here, $p(s)$ refers to a polarization state whose electric field oscillates parallel (perpendicular) to the plane of incidence. Ψ and Δ functions Ψ_{meas} and Δ_{meas} for each measurement method are then determined from the relation

$$\tan(\Psi_{meas}(f, \theta_i)) \cdot e^{i \cdot \Delta_{meas}(f, \theta_i)} = \frac{r_p(f, \theta_i)}{r_s(f, \theta_i)} \quad 3$$

The SE+T procedure is completed by acquisition (Figure 1c, right panel) of the power transmission coefficient at normal incidence, $T(f)$.

Iterative Modeling Procedure

Here we treat the HMM structure as a homogeneous, uniaxial medium and extract effective permittivity tensor components $\epsilon_{\parallel}(f)$ and $\epsilon_{\perp}(f)$ through iterative comparison of the outputs of an electromagnetic transfer-matrix-method (TMM)³² calculation, using a free-parameter model for the permittivity tensor, to the experimental outcomes of each of the three ellipsometry measurement techniques.

For each of the IE, SE+T, and TIR schemes, respectively, the modeled values of Ψ and Δ at a specific frequency f and angle of incidence θ_i , $\Psi_{model}(f, \theta_i)$ and $\Delta_{model}(f, \theta_i)$, are obtained via a two-stage iterative process, which treats the HMM as a homogeneous, uniaxial medium, defined by a complex effective permittivity tensor characterized by in-plane and out-of-plane components $\epsilon_{\parallel}$ and $\epsilon_{\perp}$. The non-oscillatory in-plane permittivity function $\text{Im}(\epsilon_{\parallel}(f))$ is modeled by a B-spline curve;³³ $\text{Re}(\epsilon_{\parallel}(f))$ is then derived by applying the Kramer-Kronig rule to the modeled function $\text{Im}(\epsilon_{\perp}(f))$. The out-of-plane permittivity function $\text{Im}(\epsilon_{\perp}(f))$, characterized by two spectral peaks, is described by a two-oscillator model $\text{Im}(\epsilon_{\perp}) = \text{Im}(\epsilon_{Lorentz}) + \epsilon_{Tauc-Lorentz, \text{Im}}$, consisting of the sum of the imaginary part of a Lorentz oscillator function matching the lower frequency peak, given by

$$\epsilon_{Lorentz}(f) = A_1 \frac{f_{1,r} f_{1,c}}{f_{1,c}^2 - f^2 - i f_{1,r} f} \quad 4$$

and a Tauc-Lorentz oscillator function^{34, 35} matching the higher frequency peak, given by

$$\varepsilon_{\text{Tauc-Lorentz,Im}}(f) = \begin{cases} \frac{A_2}{f} \frac{f_{2,c} f_{2,r} (f_g - f)^2}{(f_{2,c}^2 - f^2)^2 + f_{2,r}^2 f}, & f \geq f_g \\ 0, & f < f_g \end{cases} \quad 5$$

where A_n is the unit-less amplitude of the oscillator, $f_{n,r}$ and $f_{n,c}$ represent the broadening and central frequency of the oscillator, respectively, for the Lorentz ($n=1$) and Tauc-Lorentz ($n=2$) models, and f_g represents the band-edge frequency of the Tauc-Lorentz oscillator.

In the first stage of a given iteration, B-spline free parameters are set to yield function $\text{Im}(\varepsilon_{\parallel}(f))$, and free parameters A_n , $f_{n,r}$, $f_{n,c}$, and f_g are set to yield $\text{Im}(\varepsilon_{\perp}(f))$. $\text{Re}(\varepsilon_{\parallel}(f))$ and $\text{Re}(\varepsilon_{\perp}(f))$ are then calculated based on the Kramers-Kronig relation.

In the second stage of the iteration, $\Psi_{\text{model}}(f, \theta_i)$ and $\varepsilon_{\text{model}}(f, \theta_i)$ are computed using the TMM, based on first-stage estimates for $\varepsilon_{\parallel}(f)$ and $\varepsilon_{\perp}(f)$, as well as for the total HMM thickness d (also treated as a free parameter in the iteration).

After each two-stage iteration, a regression-analysis (Levenberg-Marquardt algorithm) comparison of the modeled curves, $\Psi_{\text{model}}(f, \theta_i)$ and $\varepsilon_{\text{model}}(f, \theta_i)$, to the experimentally acquired set of curves for a given measurement scheme, $\Psi_{\text{meas}}(f, \theta_i)$ and $\varepsilon_{\text{meas}}(f, \theta_i)$, yields a net mean-squared-error (MSE) averaged over the pairwise model-to-measurement fits for each value of θ_i . The definition of MSE is provided in Section III, Supporting Information. In the case of SE+T scheme, measured and TMM-calculated normal-incidence transmission functions, $T_{\text{meas}}(f)$ and $T_{\text{model}}(f)$, respectively, are additionally compared. In the SE+T analysis, the T data is given a weight which is twice of that of the SE data. Minimization of the MSE leads to termination of the iteration process.³⁶ If the MSE is not minimized, a new set of free parameters is generated, and the two-stage iterative loop is repeated.

Computation of Free-Parameter Starting Values for Iterative Modeling

Initial guesses for HMM thickness d and HMM complex permittivity functions $\varepsilon_{\parallel}(f)$ and $\varepsilon_{\perp}(f)$ are used in the two-stage iterative process to facilitate convergence. The starting value for d is obtained through TMM of the HMM to match the set of experimentally acquired curves for the SE+T measurement scheme, $\Psi_{\text{meas}}(f, \theta_i)$ and $\varepsilon_{\text{meas}}(f, \theta_i)$ (Section IV, Supporting Information). The multi-layer modeling procedure predicts individual layer thicknesses $d_{\text{Ag}} = 7.31$ nm and $d_{\text{Ta}_2\text{O}_5} = 18.54$ nm, for Ag and Ta₂O₅ layers, respectively (which are both close to nominal deposited-layer values of 8 and 20 nm, respectively), yielding $d = 103.4$ nm. The initial guesses for functions $\varepsilon_{\parallel}(f)$ and $\varepsilon_{\perp}(f)$ are provided by EMT (Equation 2) based on metal volumetric fraction given by $\rho = d_{\text{Ag}} / (d_{\text{Ag}} + d_{\text{Ta}_2\text{O}_5}) = 28.28\%$ and constituent-layer permittivities ε_{Ag} and $\varepsilon_{\text{Ta}_2\text{O}_5}$ directly obtained from ellipsometric characterization of reference thin films (Section I, Supporting Information).

The corresponding real and imaginary parts of the initial-guess values of in-plane and out-of-plane permittivities, $\epsilon_{\parallel, \text{EMT}}(f)$ and $\epsilon_{\perp, \text{EMT}}(f)$, respectively, are displayed in Figure SI-4, Section V, Supporting Information. As expected for an HMM incorporating a Drude metal such as Ag, the EMT-predicted permittivity tensor varies significantly as a function of f over the near-infrared (near-IR), visible and near-, mid- and deep-ultraviolet (UV) ranges.

As the frequency is continuously increased from the near-IR (starting at $f = 250$ THz, corresponding to $\lambda_0 = 1200$ nm), through the visible ($f = 384$ THz to 789 THz, corresponding to the interval $\lambda_0 = 780$ nm to 380 nm), and into the deep-UV (reaching $f = 1600$ THz, corresponding to $\lambda_0 = 187$ nm), the function $\text{Re}(\epsilon_{\parallel, \text{EMT}}(f))$ (Figure SI-4a) increases monotonically from large negative to small positive values, mimicking the behavior of a Drude metal. The zero crossing at $f = 500$ THz corresponds to the epsilon-near-zero condition ($\text{Re}(\epsilon_m) = (\rho - 1) \epsilon_d / \rho$). Simultaneously, $\text{Im}(\epsilon_{\parallel, \text{EMT}}(f))$ (Figure SI-4b) rapidly decreases from high values in the near-IR due to free-electron damping in the Ag, to relatively low values throughout the visible range and near-UV (i.e., up to a frequency $f \approx 950$ THz, corresponding to $\lambda_0 \approx 316$ nm). As f increases into the mid- and deep-UV, $\text{Im}(\epsilon_{\parallel, \text{EMT}}(f))$ increases again due to the onset of significant absorption in the as-deposited Ta_2O_5 dielectric film.

In contrast, the real and imaginary parts of the out-of-plane permittivity $\epsilon_{\perp, \text{EMT}}(f)$ (Figures SI-3c and 3d, respectively) exhibit pronounced oscillatory behavior as a function of frequency from the mid-visible ($f \approx 600$ THz corresponding to $\lambda_0 \approx 500$ nm) to the mid-deep-UV ($f \approx 1200$ THz, corresponding to $\lambda_0 \approx 250$ nm). $\text{Re}(\epsilon_{\perp, \text{EMT}}(f))$ and $\text{Im}(\epsilon_{\perp, \text{EMT}}(f))$ each display two spectral peaks in this range, where the lower-frequency peak is related to the epsilon-near-zero condition for the in-plane permittivity and the higher frequency peak is related to absorption in the constituent Ta_2O_5 dielectric material.

Parameter extraction results

The best-match modeled *Psi* and *Delta* curves $\Psi_{\text{model}}(f)$ and $\Delta_{\text{model}}(f)$ resulting from iterative modeling of the experimentally acquired spectroscopic ellipsometer data for the HMM are plotted in Figure 2, for each of the explored measurement configurations: IE, SE +T, and TIR. Experimental curves $\Psi_{\text{meas}}(f, \theta_i)$ and $\Delta_{\text{meas}}(f, \theta_i)$ are also displayed for comparison, restricted here, for sake of graphical clarity, to a single angle in the IE ($\theta_i = 65^\circ$) and SE+T ($\theta_i = 60^\circ$) cases (the TIR measurement being performed only at the single, displayed angle of $\theta_i = 60^\circ$).

For all techniques, the measured and best-match modeled *Psi* and *Delta* curves exhibit a close correspondence, with TIR technique yielding data with the best fit, as evidenced by a MSE of 11.27 which is smaller than those of the IE and SE+T techniques (24.02 and 19.73 respectively). The corresponding iterative-modeling fitted functions $\text{Im}(\epsilon_{\perp}(f))$, are displayed for each characterization technique in Figure 3a-c, along with the two component oscillator functions $\text{Im}(\epsilon_{\text{Lorentz}}(f))$ and $\epsilon_{\text{Tauc-Lorentz}, \text{Im}}(f)$ (for which the extracted free fitting parameters are listed in Table 1. The uncertainty value represents the figure of merit (FOM), which is a product of the standard 90 % confidence limit of the extracted free fitting parameter and the MSE (Section VI, Supporting Information). The function $\text{Im}(\epsilon_{\perp}(f))$ predicted from EMT is also plotted in Figure 3a-c for reference. Although the fitted *Psi* and

Delta curves show relatively low MSEs for both IE and SE+T measurements, the two corresponding derived functions $\text{Im}(\epsilon_{\perp}(f))$ both exhibit significantly different characteristics compared to those predicted by EMT (Figures 3a-b). In contrast, only the TIR method produces an extraction of $\text{Im}(\epsilon_{\perp}(f))$ that closely matches the EMT prediction (Figure 3c), hinting at a physically sound outcome.

To further evaluate the robustness of the TIR measurement and modeling procedure applied to a type-II HMM, along with the soundness of extracted permittivity values relative to those predicted by the standard IE and SE+T methods, we perform a number of parameter uniqueness tests. We first choose a model parameter of interest, define a set of test values around its best-fit value, and compute the corresponding regression-analysis-fitting MSE. During the computation, the chosen parameter is fixed at each test value, while all other model parameters are allowed to vary, and the resulting MSE is recorded. The result of this uniqueness test is a plot of the MSE versus the pre-defined test parameter values. If the MSE increases rapidly as the test parameter deviates from its best-fit value, we can infer that the model has a strong sensitivity to this parameter, and the extraction of this parameter is uniquely defined, since no other combination of the remaining fit parameters is able to produce a similar MSE. Otherwise, if the MSE is relatively insensitive to change of the test parameter, we can infer that this parameter cannot be uniquely extracted, and the analytical permittivity modeling is not robust. Such lack of robustness can be caused by the limited sensitivity of the measurement method to the specific permittivity tensor component.

The uniqueness test performed here uses the three Lorentz oscillator free parameters (amplitude A_1 , broadening $f_{1,r}$, and central frequency $f_{1,c}$). Figures 3d to 3f show the results of a parameter uniqueness test for the three characterization techniques. Here we plot the result of the uniqueness test of A_1 for each technique, where this parameter is varied over 20 different values around the best-fit value obtained from earlier parameter extraction. For both IE and SE+T schemes, the resulting curves $\text{MSE}(A_1)$ exhibit a flat appearance with no clear minimum. This confirms that the fitting process is not able to find a uniquely defined solution for A_1 , and the extraction of optical permittivity is not robust. In contrast, for the TIR measurement, the curve of $\text{MSE}(A_1)$ displays a well-defined minimum about the best-fit value of A_1 . Similar conclusions concerning the relative robustness of the TIR method, compared to that of the IE and SE+T methods, are obtained for uniqueness tests of the broadening $f_{1,r}$ and central frequency $f_{1,c}$ (Section VII, Supporting Information).

The superiority of the TIR technique in effective-medium parameter extraction for the studied type-II HMM compared to the other two spectroscopic ellipsometry techniques is further confirmed by comparison of extracted permittivity curves plotted as a function of free-space wavelength λ_0 (Figure 4). Considerable discrepancy between the EMT-predicted and IE-extracted or SE+T-extracted permittivity curves is evident. In particular, the extracted out-of-plane permittivity functions $\text{Re}(\epsilon_{\perp}(f))$ and $\text{Im}(\epsilon_{\perp}(f))$ (Figures 4c and d) fail to follow the oscillatory features predicted by EMT (Figure 4c and d insets). In contrast, good correspondence between the EMT-predicted and TIR-extracted permittivity curves is obtained.

Discussion

Type-II HMMs present a particular challenge for optical characterization, in that light incident from any angle upon a flat surface of such a metamaterial facing free space is, by design, inhibited, from crossing the surface (beyond evanescent penetration) and coupling to propagating modes within the metamaterial (which take the form of high-wavevector modes propagating only at oblique angles with respect to the normal to the layers). Whereas all three explored methods, TIR, IE, and SE+T, yield comparable outcomes for extraction of the parallel complex permittivity component, $\epsilon_{\parallel}(f)$, accurate extraction of the out-of-plane permittivity component, $\epsilon_{\perp}(f)$, benefits from enhanced interaction of the z-component of *p*-polarized light with the bulk of the metamaterial, such as is intentionally achieved with the TIR configuration.

In all cases, resolution of $\epsilon_{\perp}$ requires probing with incident light having an electric field component E_z oriented normal to the plane of the constituent layers of the metamaterial (referring to the coordinate system of Figure 1a), in other words with *p*-polarized light. To explore the efficiency under which this field component can be generated inside the metamaterial, and therefore the efficiency with which it can sample $\epsilon_{\perp}$, its magnitude under *p*-polarized illumination, $|E_z(z, \lambda_0)|$, is computed using TMM, under respective experimental configurations used for IE, SE+T and TIR schemes (with the angle of incidence set to 60° in all three cases). Calculation results, plotted in each case (Figures 5a-c) as function of depth *z* from the surface and free-space wavelength λ_0 , reveal that highest out-of-plane field magnitudes within the bulk of the metamaterial are obtained for illumination in the TIR configuration, compared to the IE and SE+T cases, suggesting that the TIR coupling prism scheme (Figure 1d) is effective in boosting $|E_z|$ to adequate levels for robust retrieval of $\epsilon_{\perp}$.

A plot of the integral of $|E_z|$ across the thickness *d* of the HMM, $\int_0^{-d} |E_z(z)| dz$, confirms that the normal field profile for TIR configuration exhibits, compared to the other two methods, overall stronger values of $|E_z|$ within the metamaterial, over a significant fraction of the explored wavelength range, namely from the near-UV ($\lambda_0 \approx 320$ nm) to the red end of the visible ($\lambda_0 \approx 750$ nm). Thus, by purposely enhancing the interaction between E_z and the HMM, the TIR configuration yields the most accurate extraction of both in-plane and out-of-plane permittivities compared to the two other methods.

Note that modeling and fitting permittivity data over a wavelength range encompassing not only the hyperbolic regime (600 nm and above), where both the in-plane and out-of-plane permittivity components are featureless and vary monotonically, but also the short wavelength regime (below 600 nm), where these components display strong characteristic fluctuations, provides a rich variety of spectral features to match, yielding a physically sound parameter extraction. In particular, this leads to a permittivity fit in the hyperbolic region that is more accurate than would be obtained using experimental data only from the relatively featureless hyperbolic spectral region. Also, extending the fit wavelength range to the short wavelength increases the confidence of parameter extraction over the entire spectral range, as the out-of-plane absorption in the short wavelength regime is intrinsically related to the hyperbolic dispersion at the longer wavelength regime for a HMM made of Drude metals (e.g., Ag or Au).²²

Conclusion

We demonstrate a new characterization technique to robustly extract the permittivity of hyperbolic metamaterials using total internal reflection (TIR) ellipsometry. During the TIR measurement, the interaction of *p*-polarized probe light with the HMM is significantly enhanced, thus providing sufficient sensitivity for accurate extraction of both in-plane and out-of-plane components of the effective complex permittivity tensor. The TIR ellipsometry technique does not require any modification of the HMM sample itself or of the ellipsometry system, making it an easily adoptable technique to characterize a broad range of anisotropic metamaterials.

Methods:

The HMMs were fabricated by sequential sputter deposition of metal and dielectric layers. The chamber base pressure was pumped down to about 0.13 mPa before film deposition. During deposition, the Ar gas pressure was 0.6 Pa and the substrate holder was rotated at the speed of 10 min⁻¹. For the deposition of Cu-doped Ag, the deposition rate of Ag and Cu was 1.109 nm/s and 0.19 nm/s, respectively.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Figure 1.

(a) Schematic cross-sectional view of the type-II hyperbolic metamaterial (HMM) characterized in this study. (b-d) Spectroscopic ellipsometry configurations under the IE (b), SE+T (c), and TIR (d) measurement schemes. E_s and E_p represent the electric field of the incident probe beam, under s- and p-polarization orientation, respectively. The complex field reflection coefficient in each case is defined as r_s and r_p , respectively.

Figure 2.

Measured and best-match modeled curves ψ (f) (a-c) and Δ (f) (d-f) using (a, d) IE scheme at 65° angle of incidence; (b, e) SE+T scheme at 60° angle of incidence; (c, f) TIR scheme at 60° angle of incidence. The fitting mean-squared-error (MSE) is also listed for each scheme. Legend in figure 2a applies to figure 2b and 2c. Legend in figure 2d applies to figure 2e and 2f.

Figure 3.

(a-c) EMT-predicted imaginary part of the out-of-plane permittivity $\text{Im}(\epsilon_{\perp})$ (orange solid line), extracted $\text{Im}(\epsilon_{\perp})$ (green solid line), and its corresponding two oscillator functions (red and blue dashed lines) for the IE (a), SE+T (b), and TIR schemes (c). Legend in figure 3a applies to figure 3b and 3c. (d-f) Parameter uniqueness test of the amplitude A_1 of the Lorentz oscillator for the IE (d), SE+T (e), and TIR schemes (f). The dashed line denotes the best-fit value of the amplitude A_1 (used in Figure 3a to c).

Figure 4.

EMT-calculated (dashed lines) and extracted (solid lines) complex permittivity components of the fabricated HMM: (a) real part of the in-plane permittivity, $\text{Re}(\epsilon_{||})$; (b) imaginary part of the in-plane permittivity, $\text{Im}(\epsilon_{||})$; (c) real part of the out-of-plane permittivity, $\text{Re}(\epsilon_{\perp})$; (d) imaginary part of the out-of-plane permittivity, $\text{Im}(\epsilon_{\perp})$. The insets in c and d display magnified views over wavelength range from 200 nm to 600 nm. Legend in figure 4a applies to figure 4b to 4d.

Figure 5.

(a-c) TMM-calculated distribution of the magnitude of the out-of-plane component of the electric field $|E_z|$ inside the HMM, plotted as a function of depth from the surface and free-space wavelength, under IE (a), SE+T (b), and TIR (c) ellipsometry configurations, under p -polarized illumination at $\theta_i = 60^\circ$ and identical incident intensities at the HMM surface. (d) Integrated field magnitude over full thickness of HMM vs. wavelength, for each of the three measurement configurations.

Table 1.

Parameters of the two oscillator functions for the three ellipsometry methods

	Lorentz Oscillator			Tauc-Lorentz Oscillator				Thickness (nm)
	A_1	$f_{L,r}$ (THz)	$f_{L,e}$ (THz)	A_2	$f_{2,r}$ (THz)	$f_{2,e}$ (THz)	f_g (THz)	
IE	4.80 ± 1.29	2237.15 ± 1189.90	2099.78 ± 779.37	3045.58 ± 1666.13	22.73 ± 12.24	746.68 ± 5.42	1020.63 ± 2417747.55^a	112.81 ± 0.73
SE+T	22.38 ± 2417747.55	0 ± 21204.96	0 ± 21207.27	2214.67 ± 2018.65	69.15 ± 63.45	747.88 ± 30.93	862.74 ± 2417747.55^a	107.71 ± 0.76
TIR	17.20 ± 0.39	103.10 ± 1.22	831.30 ± 2.22	296.34 ± 27.61	271.30 ± 19.44	1064.16 ± 6.29	902.88 ± 5.54	106.61 ± 0.43

^aWeak sensitivity to out-of-plane absorption leads to a physically implausible f_g value which is greater than f_c .