Characterizing metamaterials using spectroscopic ellipsometry

T. W. H. Oates¹, B. Dastmalchi², K. Hingerl², I. Bergmair³, C. Helgert,⁴ T. Pertsch,⁴ K. Hinrichs¹

¹Leibniz – Institut für Analytische Wissenschaften – ISAS – e.V., Albert-Einstein-Strasse 9, 12489 Berlin, Germany

² Center for Surface and Nanoanalytics, Johannes Kepler University, Altenberger Str. 69, 4040 Linz, Austria.

³ Functional Surfaces and Nanostructures, PROFACTOR GmbH, Im Stadtgut A2, 4407 Steyr-Gleink, Austria

⁴ Institute of Applied Physics, Friedrich-Schiller-Universität Jena, Max-Wien-Platz 1, 07743, Jena, Germany

Abstract

Traditionally ellipsometry is the method of choice for determining the dielectric function of isotropic bulk and composite materials. In recent years, the extension to generalized and Mueller matrix ellipsometry has facilitated the determination of the dielectric tensors of uniaxial and biaxial anisotropic materials. In this paper we investigate how the dielectric and permeability tensors of metamaterials may be characterized using spectroscopic ellipsometry. Fishnet metamaterials are measured in the IR range from 2500 - 9500 cm⁻¹ and the optical parameters are extracted by inverting the measured data in a three layer model.

1. Introduction

Ellipsometry is the characterisation technique of choice for determining the permittivity of bulk materials. As with all spectroscopic methods, an ellipsometric measurement cannot directly deliver the dielectric properties, but must be combined with an appropriate model to retrieve the real and imaginary parts of the complex permittivity, ε_1 , ε_2 (or complex refractive index, *n*, *k*). For bulk isotropic materials the model is well established, originating with the work of Drude over 100 years ago. Thanks to advances in automation and computational power ellipsometry is increasingly utilised to determine effective dielectric functions of nanostructured composites [1], anisotropic crystals and composites [2]) and optically and magneto-optically active materials [3].

The extension of ellipsometric methods to materials with artificial magnetism is a logical one, however the substantial increase in the number of variables in the model requires careful consideration. To characterize the anisotropic permittivity, ε , and permeability, μ , of metamaterials with biaxial anisotropy requires the determination of two orthorhombic rank-2 complex tensors. Experimentally this has not been performed previously and it is not yet clear which models can uniquely determine μ . In this paper we present spectroscopic ellipsometric measurements of fishnet materials designed to display magnetic resonances at normal incidence in the near infrared. After identifying the source of the features in the data we retrieve the components of ε and μ . By sequentially increasing the number of fit parameters in the optical model we check the stability and goodness-of-fit of the model. By coupling measurements at multiple incidence and rotational angles, and introducing generalized ellipsometric measurements [3] we can further constrain the fit parameters. The differences in the data are discussed and the influence of spatial dispersion is considered.

2. Experimental

Ellipsometry measures the change in polarization state of light reflected from a material. The advantage of ellipsometry is the simultaneous measurement of both the amplitude ratio and phase difference of orthogonally polarized light. This provides two measured parameters with which to calculate the real and imaginary parts of the dielectric function. In the Jones formalism one defines the polarization states as orthogonal electric field components, E_p and E_s . Reflection from a surface of a propagating light ray is expressed by the Jones matrix

$$\begin{bmatrix} E_{rp} \\ E_{rs} \end{bmatrix} = \begin{bmatrix} r_{pp} & r_{ps} \\ r_{sp} & r_{ss} \end{bmatrix} \begin{bmatrix} E_{ip} \\ E_{is} \end{bmatrix}$$

where matrix elements are the reflection coefficients and the subscripts *r* and *i* denote the reflected and incident rays, respectively. In isotropic materials (or anisotropic materials measured parallel to the optical axes) the off diagonal elements, r_{ps} and r_{sp} , are zero. The diagonal elements r_{pp} and r_{ss} may then be simply written r_p and r_s . This allows one to define the ellipsometric parameters, Ψ and Δ , as the ratio of the reflection coefficient $\rho \equiv r_p / r_s = \tan \Psi e^{-i\Delta}$. The angles Ψ and Δ correspond to the amplitude ratio and the phase difference of the reflectances, respectively.



Fig. 1: Measured ellipsometric parameters for a fishnet metamaterial.

We investigate a fishnet metamaterial made by nano-imprint lithography (NIL). The sample is a three layer (gold, MgO, gold - 30 nm each) construction with rectangular holes (x= 139 nm, y = 373 nm) and periodic unit cell (x = 500 nm, y = 600 nm). The ellipsometric parameters measured in the yzplane at multiple oblique incidence angles are shown in Fig. 1. There are three regions of interest in the data. In the long wavelength region (below 4000 cm⁻¹) the material may be described by an effective dielectric function, and we expect little or no magnetic effects. At the other end of the spectrum (above 8000 cm⁻¹) the periodic structure causes diffraction effects, observed as the Wood/Rayleigh anomalies near 8500 cm⁻¹. In the intermediate region there are two resonances associated with the symmetric (7000 cm⁻¹) and anti-symmetric (4500 cm⁻¹) coupling of the localized plasmon resonances in the gold layers. The anti-symmetric resonance is designed to result in a negative effective μ and a negative refractive index.

3. Retrieval of the effective parameters

By constructing a three-layer optical model (ambient/film/substrate) we investigate the extraction of the optical parameters from the measured data. We fix the optical properties of the semi-infinite ambient and substrate layers and also the thickness of the film layer to 92 nm (as determined by AFM measurements). This model homogenizes the three layers of the metamaterial into a single layer. As fit parameters we compare two models; the standard ellipsometry form where the permeability $\mu = 1$ and the real and imaginary parts of ε (ε_1 and ε_2) are variables, and a modified form where the real and imaginary parts of μ (μ_1 and μ_2) are also variables. The results of the two models are shown in Fig. 2.



Fig. 2: The results of the retrieval procedure to determine the effective ε and μ from the measured data. The left graph shows the results for the model where only complex ε is fit, and the right graph shows where both complex ε and μ are fit parameters.

The results show that an isotropic permittivity gives a reasonable approximation for the effective dielectric properties of the metamaterial. The real part, ε_l , shows a transition from an insulator to a conductor at the position of the symmetric resonance (the effective plasma frequency). However, the negative values of ε_2 are unphysical and imply that the material is active. The high frequency region of negative ε_2 may be associated with the Wood's anomaly while the low frequency region occurs around the anti-symmetric resonance, implying that the effect is associated with spatial dispersion. If a permeability is included in the fit then ε_2 is negative over almost the entire range. This is caused by the fact that the real part of the permeability is close to zero. The permeability shows resonances at both the symmetric and anti-symmetric resonances. The difference between the measured and fitted values is significantly improved with the addition of a permeability, however the number of measured parameters must be increased to determine the anisotropic tensor elements which correctly define the material optical properties.

4. Conclusion

Metamaterial fishnets constructed using NIL were measured using spectroscopic ellipsometry and the effective parameters extracted by applying a three layer model. The general features of the dielectric functions are correctly determined by this simple model. To properly characterize the effective permittivity and permeability of these biaxial materials requires more measured parameters to unambiguously determine the values of the complex tensor elements. Measurements using generalized ellipsometry will be performed and the number of variables increased to improve the optical modelling. This work was financially supported by the EU under the FP7 project NIMNIL **References**

^{1.} T. W. H. Oates, and E. Christalle, "Real-time spectroscopic ellipsometry of silver nanoparticle formation in poly(vinyl alcohol) thin films," J. Phys. Chem. C **111**, 182-187 (2007).

^{2.} T. W. H. Oates, M. Ranjan, S. Facsko, and H. Arwin, "Highly anisotropic effective dielectric functions of silver nanoparticle arrays," Opt. Express in press (2011).

^{3.} M. Schubert, "Generalized ellipsometry and complex optical systems," Thin Solid Films **313**, 323-332 (1998).