OVERVIEW OF THE STATE-OF-THE-ART AND MOST PROMISING MEASUREMENT TECHNIQUES

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Summary

Most measurements of metamaterials consist of transmission and reflection, often followed by a transformation to retrieve the real and imaginary parts of *epsilon* and *mu*. Because the phase information can be more difficult to obtain than in say S parameter measurements in the microwave region, alternative schemes have been developed. Similarly, a wedge-shaped metamaterial in the microwave region can straightforwardly demonstrate negative refraction – which becomes more difficult in the NIR and visible, with the need to obtain large area samples with several layers. For some experiments, simulations of the structures are essential for the ultimate goal of the retrieval of *epsilon* and *mu*. The normalised S parameters retrieved from the simulations are used to extract *epsilon* and *mu* via the Fresnel equations,[i] often with simplifying assumptions. Techniques for retrieving the phase include the use of phase masks[ii], angular resolved measurements[iii], ellipsometry[iv] where the ratio of TM-to-TE polarized light is plotted as a function of frequency for oblique incidence - and femto-second laser interferometry,[v] where the group and phase velocities are obtained from interferograms. A detailed account of the main issues at optical wavelengths is given below.

This overview is restricted to the plane-wave characterization of nanostructures. There exists a number of other methods including near-field optical measurements, the optical Fourier spectroscopy, and the measurement of the optical transfer function form, which will be considered at a later stage.

Characterization of metamaterials at optical wavelengths - Martin Wegener

1. Introduction

The notion “optical wavelengths” is generally not strictly defined and has often been misused, aiming at suggesting to laymen wavelengths in the visible part of the electromagnetic spectrum. Here, in agreement with a bulk of literature, we define “optical wavelengths” as wavelengths of just a few micrometers, equivalent to about 100 THz frequency of light and above. At yet smaller frequencies, approaching the far-infrared (or “THz regime”), the issues of sample characterization tend be somewhat different from the optical regime.

Before directly addressing optical characterization issues, it is instructive to briefly recall how today’s state-of-the-art metamaterials at optical frequencies look like – as this aspect poses relevant boundary conditions to the characterization process.

The vast majority of metamaterial structures have been made via serial, hence time-consuming, lithographic approaches (e.g., electron-beam lithography, focused-ion-beam lithography, or direct laser writing). As a result, typical sample footprints are only on the order of $(100\ \mu m)^2$. Rather recent short [1,2] and extensive [3] reviews on corresponding magnetic and/or negative-index metamaterials can be found in the literature. Examples of notable exceptions are metamaterial samples made via holographic lithography [4,5] or via nano-imprint techniques [6]. The footprint of all these is on the order of square-centimetres. Yet much larger footprints can be realized along these lines in the future. Furthermore, the vast majority of metamaterials at optical wavelengths demonstrated experimentally thus far contain only a single functional layer [1-3] (which can mean more than just one actual layer). Notable recent experimental exceptions are a three-functional-layer negative-index metamaterial at 1.4-µm wavelength [7], a four-functional-layer magnetic metamaterial at 3.7-µm wavelength [8], and a ten-functional-layer negative-index metamaterial at 1.8-µm wavelength [9]. All of these have a total thickness that is significantly less than one wavelength of light. Interesting metal-insulator-metal slot waveguide structures supporting backward waves over many wavelengths of light along the propagation direction in the waveguide plane have also been reported [10]. However, we will refrain from discussing their optical characterization here as they are not “metamaterials” in the sense used here. All of the actually fabricated published metamaterial structures are anisotropic, often they are even uniaxial. Also, the low symmetry of most metamaterial structures generally allows for strong polarization dependencies. Furthermore, essentially all metamaterial structures are mechanically supported by some sort of a dielectric substrate – an aspect, which has to be accounted for in the optical characterization process as well.

2. Linear optical characterization of metamaterials

2.1. The ideal measurement: The conceptually perfect experiment on a periodic metamaterial with sub-wavelength period can do nothing more than measure the frequency-dependent complex reflectance and the complex transmittance of the sample for all angles of incidence and for all incident polarizations of the impinging ideal monochromatic plane wave. Furthermore, this ideal measurement should comprise analysis of the (generally elliptical) polarization state of light emerging from the sample. Clearly, in linear optics, frequency-domain information can equivalently be expressed in the time-domain, where “complex” translates into amplitude and phase of the wave. For imperfect or for (intentionally) non-periodic metamaterial samples, also scattering of light into the entire solid angle can occur because of the lack of translational invariance. This scattering would also have to be characterized completely in an experiment.

Anything beyond this, e.g., retrieval of whatever effective optical parameters (refractive index, impedance, electric permittivity, magnetic permeability, bi-anisotropy parameter, etc.), is *not* subject of the experimental optical characterization process itself but it is subject of *interpretation* (!) of the acquired experimental data. This distinct step – which is closely related to and interlinked with the retrieval of these parameters from theoretical calculations – will be discussed in more detail below.

2.2. Instrumental limitations: At optical frequencies, it is very difficult or even impossible with current technology to measure the electric- and magnetic-field components of the electromagnetic light wave directly versus real time. In contrast to this, frequency-domain techniques are very common. Usually, grating spectrometers or Fourier-transform spectrometers merely deliver the intensity of light versus wavelength or versus frequency, i.e., all phase information is lost. Using interferometric techniques, phase information can be (partially) recovered. Commercially available ellipsometers promise to deliver optical constants of thin-film samples by means of angle- and polarization resolved reflectance measurements. Extreme caution is indicated at this point, because the underlying commercial software for analyzing (or, more precisely, *interpreting*) these data is well-prepared for dielectric material responses and layered systems, but it is usually not prepared at all to deal with magnetic responses (or negative refractive indices). Also, the low symmetry of metamaterial samples can be problematic.

Due to the limited lateral footprint of typical samples (see 1.), the incident light wave has to be focused onto the metamaterial (and/or the sample has to be imaged). This clearly introduces an undesired spread of the incident wave vector components of light, i.e., the experiment effectively averages over a certain spread of incident angles, obviously leading to obscured data. The impact of that averaging process depends on the specific metamaterial under investigation. For example, it is quite common to image the samples by means of a microscope. As often large spectral bandwidths have to be investigated, reflective microscope objectives are mandatory in order to avoid chromatic aberrations that would otherwise occur for glass-based lenses. It is well known that such Cassegrain objective lenses essentially cut out the normal incidence contribution and average over a cone of angles of incidence (e.g., between 15 and 30 degrees with respect to the surface normal for a numerical aperture of NA=0.5). Again, the relevance of these “artifacts” needs to be evaluated for each metamaterial structure separately.

2.3. State-of-the-art: All experiments that have been published until today are very far away from the conceptual ideal described above (2.1.).

2.3.1. Suppose that the metamaterial structure exhibits *inversion symmetry* along the surface normal. In this case, the normal-incidence reflectance and transmittance spectra do *not* depend on from which side of the sample they are taken. A quite common procedure is to measure intensity transmittance and/or reflectance spectra of a metamaterial slab of thickness *l* for normal incidence of light and for two relevant (i.e., linearly independent) incident polarizations, either linear or circular. Clearly, these data composed of two quantities for each wavelength are insufficient to retrieve the optical parameters, e.g., the complex refractive index and the complex impedance of the slab for each wavelength, in an unambiguous fashion. Thus, usually, the experimental data are compared with theoretical calculations based on the designed structure and based on additional information regarding geometrical parameters obtained from optical and/or electron micrographs of the metamaterial. If sufficiently good agreement between experiment and theory is obtained, one may take the theory to evaluate missing information.

One way of further *analyzing/interpreting* the data is to construct in the computer a fictitious slab of thickness *l* (with or without substrate) that has strictly the same complex reflectance and transmittance spectra as those of the metamaterial. This "retrieval" procedure (see, e.g., the review [3] for selection of the various branches occurring in this process) delivers the two complex quantities refractive index and impedance, or, equivalently, the complex electric permittivity and the complex magnetic permeability of the slab. While this procedure is absolutely well defined and very well developed in many laboratories around the world, one should be cautious in *interpreting* these retrieved quantities. They do reflect the optical properties of the metamaterial slab with thickness *l* – yet they are not necessarily "material" properties in the usual sense: One might be tempted to take the knowledge from normal optical materials and transfer that to metamaterials. For example, if one followed the retrieval procedure described above for a thin film of silica (SiO_2) of thickness *l*, it is clear that the analysis of a corresponding film of thickness 2*l* would deliver very nearly the identical material parameters. This is *not* (!) necessarily the case for metamaterials. Generally, (near-field optical) interaction effects among the different functional layers of the metamaterial can modify the "material parameters". Whether or not that is a significant effect needs to be evaluated for each metamaterial structure under investigation – there is simply no generic answer. Two published experiments at optical frequencies that have addressed this aspect [7,9], and have come to the conclusion that these interaction effects are not too strong for their conditions (both are several layers of double fishnet negative-index metamaterials). A striking counter-example is Ref. [8], where the strong coupling between adjacent layers of split-ring resonators has tremendously distorted the properties of a single layer. Yet, the answer to this question also depends on how closely one evaluates the data. For example, it is known from (dielectric) photonic crystals that, for certain aspects (e.g., slow group velocities), even more than 100 lattice constants may not yet reproduce the behaviour anticipated from band structure calculations – which obviously address the fictitious infinite "material".

In other words, we very often deal with the situation when the effective parameters retrieved from the reflection/transmission measurements are not the usual (local) parameters, and they do not adequately describe the electric and magnetic polarization responses [21]. Even in structures where near-field interactions between inclusions are negligible, the retrieved parameters are non-local beyond the quasi-static limit (the difference becomes significant for frequencies approaching to the resonance band of inclusions) [22]. The main reason of the difference between the retrieved and local electromagnetic is the nonzero phase shift of the wave per unit cell of the MTM lattice. That leads to a not negligible Drude transition effect at the sample interfaces [23].

2.3.2. The situation is yet more complex if the metamaterial structure has *no centre of inversion* along the propagation direction of light – still restricting ourselves to *normal incidence of light* onto the metamaterial slab. In this case, the complex transmittance and reflectance spectra are no longer the same for the two possible directions of incidence. In other words: For each wavelength one has eight generally different quantities to be measured – provided that the polarization state of the incident light is conserved in both reflectance and transmittance. Otherwise, the number of independent optical quantities generally doubles. In case of reciprocal structures (i.e., no static magnetic field and no absorption), the two complex transmittances are strictly identical, while the two complex reflectances are not. This results in six parameters for each wavelength. Again, published experiments have not measured field coefficients but rather the three corresponding intensity coefficients, and, as a result, the problem is underdetermined. In close analogy to our discussion in 2.3.1., additional theoretical input is required if effective optical parameters shall be extracted. One option is to retrieve the two complex impedances for the two sides of incidence and the single complex refractive index. Another equivalent option is to retrieve the complex permittivity and permeability as well as the bi-anisotropy parameter. Experimentally, this has only been done once in the literature so far [11]. Needless to say that the meaning of these quantities underlies the same restrictions already outlined in section 2.3.1., i.e., caution has to be exercised in interpreting these quantities as "material" parameters. They do, however, have a well defined meaning for the film of thickness l under investigation. A variation of the second option is to replace the bi-anisotropy parameter by a wave-vector dependence of permittivity and/or permeability [12].

2.3.3. Additional information can be obtained from normal-incidence *interferometric experiments* that – at least partly – recover the missing phase information addressed above. Corresponding publications include Refs. [4,13-15]. These additional inputs provide further sensitive tests of the level of agreement between experiment and theory. In that sense, they are very important. However, these additional experimental data do not at all change the conceptual questions raised in 2.3.1.

2.3.4. The situation becomes quite a bit more complex if *oblique incidence of light* onto the metamaterial slab is considered. For usual optical materials, generally all optical quantities become tensors of rank three. Only very few experiments on metamaterials at optical frequencies have addressed oblique incidence of light [16,17]. These papers have just reported intensity transmittance and/or reflectance spectra for various angles with respect to the surface normal as well as for various different azimuth angles, but they have completely refrained from translating these measurements into effective "material" parameters. Theoretical publications addressing possible retrieval procedures are, however, available in the literature [18,19], but these studies are incomplete and not confirmed experimentally. On the other hand, only oblique incidence probing allows one to determine full set of components of material parameter tensors for anisotropic structures.

2.3.5. For wedge-type metamaterial samples (rather than the slabs discussed so far), the direction of the transmitted light wave generally changes due to refraction. Measuring the corresponding angles according to Snell's law allows for inferring the refractive index in Snell's law – which, however, is generally distinct from the refractive index n that one refers to when stating that the material phase velocity of light, c, is slower by factor n than the vacuum speed of light, c_0, i.e., $c=c_0/n$. A brief discussion of this aspect can, e.g., be found in Ref. [20]. Early experiments addressing the mentioned change in the direction of the Poynting vector (energy flow) have been published [9]. Inferring other optical parameters from such refraction experiments again requires making reference to some sort of theoretical modeling.

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4. Concluding Remarks

Optical metamaterials, because of their typically limited area and number of functional layers, are more easily understood as finite structures with interfaces that define their optical behaviour. This definition is in contrast to a conventional material in which their bulk properties define their optical properties. The retrieved 'material' parameters are the subject of interpretation unless they sufficiently extended in the measurement direction to remove interface effects. Also the phase information is harder to obtain giving the need for either more sophisticated measuring tools, such as time domain or interference methods, or alternatively, more complex structures ie phase masks made with the metamaterial.

5. Summary References

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