# RECOMMENDATIONS ON HOW TO CHOOSE THE APPROPRIATE MEASUREMENT TECHNIQUE FOR EM NSM CHARACTERIZATION AND THE PARAMETERS' EVALUATION

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# Introduction

The constitutive relations are used in macroscopic electromagnetics to describe interactions of electromagnetic fields and waves with medium. The fundamental parameters such as dielectric permittivity $\varepsilon$ and magnetic permeability $\mu$ are usually employed to characterise the medium properties. However these parameters cannot be measured directly, and their evaluation is based on the retrieval from the measurable quantities. However, there is still no consensus on interpretation of the measurement data for nano-structured materials and metamaterials especially when their intrinsic electric and magnetic resonances fall within the measurement frequency range. Also, the effective material parameters (EMP), deduced from the measurement data, often depend not only on the material internal microscopic structure but also on the size and shape of the measured sample as well as on the surrounding medium. Therefore experimental characterisation of nano-structured materials and metamaterials, and establishing the relationship between the measured electromagnetic response and the material composition, internal structure and sample shape poses tremendous challenges.

As outlined in [1], the material characterisation is based upon retrieval of the material parameters from the obtained experimental data. Therefore EMP evaluation is inherently dependent on the retrieval models, which determine the relationships between the measurable quantities and the description of the properties of nano-structured materials. It is also necessary to note that because different physical properties may have a dominant role in different frequency ranges and specific applications, the measurement techniques for evaluating the material performance should be tailored respectively. Thus the initial information about the physical properties of the material specimens plays a pivotal role in determining suitable measurement techniques.

While microscopy and chemical analysis are employed to determine the basic structural characteristics of nano-structured materials, the electromagnetic characterisation of metamaterials involves transmission and reflection measurements, followed by the retrieval procedure to obtain the real and imaginary parts of $\varepsilon$ and $\mu$, or refractive index *n* and characteristic impedance *Z* of the medium. In optical range, the phase information of the scattered fields is more difficult to obtain than in S-parameter measurements at the microwave frequencies. Therefore alternative schemes have been used in the infrared and visible ranges with the need to obtain large area samples containing several layers. For some experiments, simulations of the structures are essential to assist the EMP retrieval. The normalised S-parameters de-embedded from the simulations are then used to extract $\varepsilon$ and $\mu$ via the Fresnel equations [2], often with simplifying assumptions. The techniques for retrieving the phase include

- the use of phase masks [3],
- angular resolved measurements [4],
- ellipsometry [5] where the ratio of TM-to-TE polarized light is plotted as a function of frequency for oblique incidence,
- and femto-second laser interferometry [6], where the group and phase velocities are deduced from interferograms.

A detailed account of the main issues is given below.

# Measurement techniques for optical range

## General issues

The notion “optical range” is generally not strictly defined and has often been misused, referring primarily to the visible part of the electromagnetic spectrum. Here, in agreement with a bulk of literature, the “optical range” is defined as wavelengths of a few micrometers, equivalent to about 100 THz frequency of light and above.

Before addressing the issues of optical characterisation, it is necessary to briefly outline metamaterial features at optical frequencies that impose certain constraints on the measurements and characterisation process.

The vast majority of metamaterial structures have been manufactured by lithographic techniques with the typical sample footprints on the order of 100 $\mu m^2$ [7]-[9]. Examples of notable exceptions are metamaterial samples with the footprint on the order of square-centimetres, made via holographic lithography [10], [11] or via nano-imprint techniques [12].

Another important aspect is that the vast majority of metamaterials at optical wavelengths demonstrated experimentally contain only a single functional layer [7]-[9]. The recent experimental exceptions are a three-functional-layer negative-index metamaterial at 1.4-μm wavelength [13], a four-functional-layer magnetic metamaterial for 3.7-μm wavelength [14], and a ten-functional-layer negative-index metamaterial at 1.8-μm wavelength [15]. However, all these samples have a total thickness significantly less than one wavelength of light in the free space.

All of the actually fabricated metamaterial structures reported in the literature are anisotropic, often they are even uniaxial. Also, the low symmetry of most metamaterial structures generally allows for strong polarization dependencies.

Finally, essentially all metamaterial structures are mechanically supported by some sort of a dielectric substrate – an aspect, which has to be taken into account in the optical characterisation of nano-structured metamaterials.

## Optical measurement of metamaterials

*The ideal measurements:* The conceptually perfect experiment on a periodic metamaterial with sub-wavelength period consists of measuring the frequency-dependent complex reflectance and the complex transmittance of the sample for all angles of incidence and for all incident polarisations of the impinging perfect monochromatic plane wave. Furthermore, this ideal measurement should comprise analysis of the (generally elliptical) polarization state of light emerging from the sample. In linear optics, frequency-domain information can equivalently be represented in the time-domain, where "complex" translates into amplitude and phase of the wave. For imperfect or for 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 analysed completely in an experiment.

Retrieval of EMP such as refractive index, impedance, electric permittivity, magnetic permeability, bi-anisotropy parameter, etc. is *not* subject of the experimental measurements but it is subject of *interpretation* (!) and post-processing of the acquired experimental data. This distinct step, which is closely related to and interlinked with the retrieval of these parameters from theoretical calculations, is discussed in [1].

*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 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. However all phase information is lost here. 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 polarisation resolved reflectance measurements. Extreme caution should be exercised at this point, because the built-in commercial software for processing (or, more precisely, *interpreting*) the acquired data is devised for the basic dielectric material responses and layered systems. But it is usually not suitable to deal with magnetic responses or unconventional properties of the materials. Also, the low symmetry of metamaterial samples can be problematic.

Due to the limited lateral footprint of typical samples, the incident light wave has to be focused onto the metamaterial sample. This clearly introduces an undesired spread of the incident wave vector components of light, i.e., the experimental data effectively averages over a certain range of incidence 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 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 assessed for each metamaterial structure separately.

<u>*State-of-the-art procedures:*</u> All experiments that have been published to date are very far away from the conceptual ideal setup described above.

For the metamaterial structure with *inversion symmetry* along the surface normal, the normal-incidence reflectance and transmittance spectra do *not* depend on from which side of the sample they are taken. A 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 unambiguously the optical parameters, e.g., the complex refractive index $n$ and the complex impedance $Z$ of the slab at each wavelength. Thus, usually, the experimental data are compared with theoretical calculations based on the designed structure and additional information about geometrical parameters obtained from optical and/or electron microscopy of the measured metamaterial samples. If sufficiently good agreement between experiment and theory is achieved, one may use the theory to provide missing information.

The situation is more complex in the metamaterial structure which 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 differ. 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. Similarly to the preceding case discussed above, additional theoretical input is required for the EMP retrieval. One option is to obtain 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 in [16]. Needless to say that the meaning of these quantities underlies the same restrictions already outlined above, 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 known thickness $L$.

Additional information can alternatively be obtained from normal-incidence *interferometric experiments* that partly recover the missing phase information as discussed above, see also [6], [10], [17], [18]. 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 of the parameter interpretation raised earlier.

The situation becomes ever more complex at *oblique incidence of light* onto the metamaterial slab. 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 [19], [20]. Transmittance intensity and/or reflectance spectra for various angles with respect to the surface normal as well as for various different azimuth angles have been obtained, but these measurements have not been translated into effective “material” parameters. The relevant procedures for the EPM retrieval were suggested in [21], [22].

In wedge-type metamaterial samples (in contrast to the slabs with parallel interfaces discussed so far), the transmitted wave generally deflected due to refraction. Measuring the corresponding angles allows the effective refractive index *n* to be inferred by using Snell’s law. Such obtained *n* is generally distinct from the refractive index for conventional dielectrics [23]. Early experiments addressing the change in the direction of the Poynting vector (energy flow) have been presented in [15]. Retrieval of EMP from such refraction experiments again requires additional information which can be obtained from the theoretical models and/or complementary measurements.

*Chiral structures* are a subclass of bi-anisotropic materials rotating any incident linear polarization of light, i.e., exhibiting optical activity. Notably it was found that in contrast to the general bi-anisotropic case, reflectance and absorbance here do not depend on which side of the sample they are taken under normal incidence.

Provided that phase information is completely obtained, measurements can be performed equivalently either with linear or with circular polarization of the incident light. Then phase-sensitive transmittance and reflectance spectra can be obtained with linear polarization of the incident light [24] or phase sensitive time-resolved data can be Fourier transformed. In both cases, data have to be taken for different linear polarizations for retrieval of three complex parameters (electric permittivity, magnetic permeability, and chirality parameter shall be retrieved). If no phase information is acquired (i.e., only intensity measurements combined with theory), a minimum requirement for meaningful parameter retrieval is that one checks that the two incident circular polarisations of light stay circular (with the same handedness) upon transmission. Furthermore, in accordance with the generalized Fresnel coefficients, left-handed circular polarization has to turn into right-handed circular polarisation and vice versa in reflection. In other words, circular polarisation conversion needs to be small.

Actually measuring broadband intensity transmittance and reflectance spectra with incident circular polarization of light is far from trivial. Broadband linear polarizers are readily available in most spectral regimes and a quarter-wave plate can turn this linear polarization into circular polarization. However, a usual quarter-wave plate has obvious inherent wavelength dependence. At optical and near-infrared frequencies, so-called super-achromatic quarter-wave plates are commercially available. At mid-infrared frequencies, such super-achromatic quarter-wave plates have to be custom-made. Also, these structures are difficult to integrate into existing (commercial) instrumentation. At THz and microwave frequencies, such super-achromatic quarter-wave plates are presently not available.

# Concluding Remarks

The macroscopic (phenomenological) electromagnetic parameters of nano-structured materials cannot be measured directly. Therefore their evaluation is the result of retrieval from the measured data. The transmission and reflection measurements represent the main techniques used for the electromagnetic characterisation of nano-structured materials and metamaterials in optical range. The measurements and characterisation of metamaterials at optical frequencies are usually limited by the sample small size and thickness, which is significantly less than wavelength of light. All of the actually fabricated metamaterial structures reported in the literature are anisotropic, often they also are uniaxial or chiral. These features impose additional requirements on the data acquired from the measurements. Several measurements techniques used for the characterisation metamaterials at optical frequencies have been presented, and their merits and shortcomings have been discussed for a few nano-structured metamaterials.

It is important to emphasise that the effective material parameters are retrieved from the measurement data using the specific models of materials. Physical meaning of the retrieved parameters is frequently a subject of interpretation and depends on the quality of the models used. Therefore further advances in characterisation of metamaterials will depend on the progress in developing a better understanding of interrelation between the measured and descriptive parameters of the metamaterials along with modelling of the practical material specimens and the experimental setups employed for the measurements and data acquisition.

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