

The Interaction of Electromagnetic Radiation with Composite Materials and Metasurfaces

Habilitation Thesis Summary

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Summary

The thesis presents my work in the study of the interaction between electromagnetic radiation and material media such as liquid crystals, liquid crystal composites, thin films, metasurfaces and photovoltaic structures, spanning a twelve year research effort since finishing my Ph. D. The Thesis is structured into five chapters, as follows:

- Chapter 1 gives a detailed context of the work and the motivation to study the effects arising from the interaction between the electromagnetic field and the above-mentioned media, casting a birds-eye view on the current state-of-the art in the two complementary fields.
- Chapter 2 details the work conducted in the characterization of the optical properties of composite liquid crystal-based materials under the effects of external electric and magnetic fields, and highlights both experimental studies and theoretical modelling performed in order to gain a deeper understanding of the dynamics of complex molecular systems undergoing external actions.
- Chapter 3 details the work conducted in the theoretical modelling of negative-index materials on one hand, and numerical analyses of metasurface architectures that are designed to perform certain optical functions (selective absorption, polarization and phase control, active phase switching, remote sensing by means of triggerable materials and so on). The work presented here represents the bulk of my expertise, and has been at the center of my research for more than six years.
- Chapter 4 details the history of my activities in all fields of academic work and presents the development perspectives in the two main research directions presented in chapters 2 and 3, from a management point of view.
- Chapter 5 concludes the thesis, by giving a brief summary of the information presented throughout, and highlighting the main achievements realized until now.

The information presented in full detail in the Thesis will be summarized in the following sections.

Context of the work

The interaction between radiation and matter is the fundamental process behind almost every modern invention that deals with information processing and computing. The collection of phenomena resulting from the interaction of electromagnetic radiation and material media is generally known as *optics*. The response of matter in the wake of its interaction may constitute full absorption, followed by energy conversion, or partial absorption, with the remainder being reflected and refracted outwards. The process behind reflection and refraction of the field as a result of interaction is known as *scattering*. Depending on the phase profile of the interface, scattering can be *specular* (for a quasi-constant phase profile) or *diffuse*, for a rapidly spatially-varying phase profile. While theoretically the interface does not have volume, scattering of the input radiation field has to take into account the material elements that act as scatterers. Therefore, in the case of reflection, the interaction volume has to take into account the so-called *skin-depth*, while for transmission, the interaction has to take into account the entire volume of the interaction medium. To achieve control over the electromagnetic properties of the output radiation field, a multitude of optically-responsive materials can be used. Whether happening in reflection or transmission, linear scattering affects the amplitude, polarization and phase of an incident field, the control of which can serve as a means of information transmission and interpretation. Nonlinear scattering serves as a means of radiation control by means of radiation, wherein the frequency of the output optical field is controlled by means of the input field and material properties. When discussing scattering materials, the majority of these are either dielectrics, which can be used in bulk or thin-film versions, the latter being exceptionally useful in the creation of selective layering with respect to the desired output response. Applications of such layering constitute optical filtering as functions of either frequency or input polarization. If the materials possess externally-addressable properties, the scattering process can be made *adaptive*, with direct applications in optical switching. A class of optically-adaptive media is represented by *liquid crystals*, which are dielectric media exhibiting no translational order but high orientational order. Liquid crystals owe their molecular orientation properties to their chemical composition, which allows intermediate matter phases that span several tens of degrees around normal room temperature. When in their transitional phases, the rotational degrees of freedom attained by the liquid crystal molecules can be controlled by means of external electric and/or magnetic fields. By combining this advantage with a convenient layering scheme, the liquid crystal-endowed optical layering can benefit from their adaptive properties, and lead to the realization of polarization-based switches and controllers. When used in conjunction with a host medium, such as a polymer matrix, the resulting architecture, known as a polymer-dispersed liquid crystal acts as an on-demand optical switch between specular and diffuse scattering, based on the variation in the refractive index mismatch as a function of the molecular orientation angle with respect to the interface normal established by the polymer matrix. The angular velocity with which the liquid crystal is allowed to rotate is directly linked to the chemical composition of the liquid crystal and the ambient temperature at which it is operated, and is usually a material constant. To enhance the temporal dynamics of switching, a molecule of liquid crystal may be surrounded by microscopic "enhancers", which are essentially nanoparticles that respond to electric or magnetic fields faster than the liquid crystal molecule. When fixed at the ends of the liquid crystal molecule, usually by adhesive forces, and subjected to external fields, the nanoparticles develop an angular momentum greater than that of the liquid crystal molecule, which forces a rotation with an increased angular velocity. This enhancement reduces the switching times between user-defined "on" and "off" states, from milliseconds to, in some cases, tens of microseconds. The adaptiveness of liquid crystal-based optical devices comes with a few drawbacks, one of which is the relatively-long transient regime, in which the quasi-chaotic rotation of the liquid crystal molecules can be challenging for stability. Also, when adding nanoparticles to the system, both internal and external forces make the nanoparticles shift from their initial position, making them aggregate in some region of the system. The resulting segregation causes loss of the enhanced properties, unless repetitive agitation under ultrasound is per-

formed, and the system is reset. Also, a region of liquid crystal composite that is exposed to high intensity radiation can absorb some part of that radiation and increase its local temperature above the phase transition, rendering the optical properties of the system null. To bypass these disadvantages, liquid crystals have been replaced with thin films of various metals and dielectrics, deposited in the layer stack configuration on a substrate. These optical structures have much lower transient regimes, as well as a well defined effective response, but lose their adaptiveness due to the lack of rotational degree of freedom of the molecules. Also, the tuning possibility of the scattering response is almost null, due to the fact that the homogeneity of each layer ensures that any external excitation will trigger the same type of response across the full bandwidth of the input field. This property is called locality, and it is associated to all conventional materials. To circumvent locality and explore the optical effects produced by nonlocality, instead of depositing a single material per layer, a solution based on imprinting nanometer-size elements of various metals and dielectrics has been employed. The materials that are used have to be different than that of the substrate they are deposited on, and their sizes have to be only a small fraction of the operating wavelength in order to produce sharp variations in the phase profile across the surface. These structures have been known to oscillate at other frequencies than that of the plasma frequencies corresponding to each material component, and therefore, are called *meta-response materials*, or in short *metamaterials*, with the two-dimensional configurations being known as *metasurfaces*. Due to the fact that the sizes of the metasurface elements are fractions of the operating wavelength, the metasurface response is typically designed to operate for one specific wavelength, offering a monochromatic designer response. This design consideration is usually known as *the wavelength condition*. To establish a broadband response, the elements that are deposited on the substrate follow a more complex pattern, containing multiple elements of various sizes (within the metasurface wavelength condition), that apply the wavelength condition independently. Also, by using externally-addressable materials in their architecture, metasurfaces can be made readily-tunable to a specific working frequency, or can be made extremely sensitive with respect to the external excitation. This extended set of properties constitutes a basis for realizing optical controllers or optical sensors. Adding to these properties the fact that metasurfaces can be very easily and readily integrated on semiconductor wafer substrates, metasurface-based optical control can be obtained for optical chips and miniature optical sensor systems.

Work Conducted and Main Achievements

Given the above context, the paper presents the work I conducted in studying the interaction between optical fields and optical materials, with a focus on liquid crystal composites and metasurfaces. The work presented in the paper began after receiving my Ph. D. degree in 2012, and continues in present day. My activity has been divided into two major directions, detailed below:

1. The study and characterization of the electric, electro-optic, magnetic and magneto-optic properties of composite materials - liquid crystals in conjunction with micro- and nanoparticles. This activity has been performed in the Condensed Matter Laboratory in the National University of Science and Technology Politehnica Bucharest, from 2014 to present time. The work consisted mostly in the experimental validation of theoretical models that describe the optical behavior of such systems under the influence of external fields and various thermal conditions. The studies carried out focused mainly on determining the optical properties of liquid crystal composites under the action of external electric and magnetic fields. The studies include: (a) the determination of the critical field value associated to the Freedericksz transition, which is defined as the field value at which the liquid crystal molecules start to rotate effectively parallel to the field direction; (b) the characterization of the directional scattering properties of polymer-dispersed liquid crystal composites, together with the on-off and off-on switching times, optical

transmission of the samples, and dynamics of relaxation in the case of pure nematic liquid crystals. The work conducted was coordinated by prof. D. Mănăilă-Maximean, and was performed in close collaboration with prof. E. Petrescu and assoc. prof. C. Cîrtoaje.

2. The study and characterization of organic photovoltaic composites, with a focus on improving both the energetic photon-to-exciton conversion efficiency and fill-factor, by convenient doping with certain fluorescent dyes. This activity was conducted in the Condensed Matter Laboratory in the National University of Science and Technology Politehnica Bucharest, from 2014 to present day. The activity revolved around the fabrication of new organic solar cells doped with specific dyes, which enhance the emission around the highest spectral absorption window of the solar cells. The resulting system was then characterized as from an electrical point of view, with the extraction of relevant electrical and energy parameters. The work conducted was coordinated by prof. D. Mănăilă-Maximean, and was performed in close collaboration with dr. A. Bărar.
3. The study and characterization of metasurface architectures interacting with optical and terahertz incident fields. The work started in 2020 and continues in present day. The work consists mainly of theoretical modeling and numerical analyses of the various types of optical response of metasurface architectures that were created to serve a specific application: selective absorption, optical switching, adaptive phase control, generalized reflection and refraction, broadband absorption and optical confinement and energy conversion for remote sensing. The work conducted was performed under my coordination and it involved close collaboration with prof. D. Mănăilă-Maximean and dr. A. Bărar. The work done in the characterization of metasurfaces for remote sensing has earned me a Fulbright Visiting Scholar Grant to the City University of New York, where I collaborated with prof. B. A. Gross and prof. F. Moshary to the numerical analysis of a proof-of-principle version of a highly sensitive CO₂ sensor that leverages humidity, by means of incorporating new CO₂ - triggerable hydrophilic polymeric elements in the architecture. During this time, I collaborated with dr. A. Bărar, S. A. Maclean and A. Taylor in New York University to establish a new „fractal”-type metasurface that is able to provide large bandwidth optical confinement, by using elements of varying sizes displaced in a unit cell.

Throughout my activities, apart from collaboration with colleagues, I have coordinated a number of students in finishing their undergraduate and postgraduate studies in the field of materials characterization. Recently, based on my expertise, I have been selected to assist in the coordination of multiple Ph. D. students, as part of the coordination team. In terms of impact, the work conducted up to this point has resulted in the publication of multiple ISI-indexed articles, most of which are in the Q1/Q2 quartiles. Excluding self-citations, the work has achieved more than 330 citations according Clarivate Web of Science, and generated a Hirsch index of 11 (Web of Science). The expertise gained throughout the work also resulted in the winning of a national grant in the Extreme Light Infrastructure framework, in which I have proposed the incorporation of new architectures and materials for deposition in order to create custom interactions between ultrashort pulses and two-dimensional structures such as thin film layers and metasurfaces. The aim of the project is to produce flat surfaces with designer phase profiles and with controlled thermal conductivities, that may increase the damage threshold or increase the nonlinear refractive index. The successful design of such optical elements may establish new state-of-the art in optical mirrors and gratings design for ultrashort pulse laser systems.

Going Forward - Outline of the Managerial Plan

As a Ph. D. coordinator, my tentative research group will continue and expand the work that has been performed up until now, focusing mainly on the characterization of the effects resulting from

the interaction of electromagnetic radiation and thin material media (i.e. thin films and metasurfaces), with direct applications in various fields of optics. The activity will center around the numerical analysis and experimental validation of the optical properties of the envisioned architecture. The application directions currently considered are:

1. The conception and numerical analysis of composite liquid crystal films and/or adaptive metasurfaces able to respond to external fields.
2. The characterization of composite materials under high-intensity level fields, with a strong focus on nonlinear effects.
3. The conception and characterization of liquid crystal composite-based sensors and actuators, in terms of responsivity, sensitivity, duty cycles and temporal dynamics. A special attention can be offered to the characterization of the so-called *asymmetric resonance sensors*, by leveraging Fano resonance effects.
4. The conception and characterization of composite liquid crystal/metasurface layers that are able to realize optical confinement long enough for the energy to be transferred to photovoltaic structures, with the direct effect of energy efficiency enhancement.
5. The conception and characterization of negative index metasurfaces, with a strong focus on highly nonlinear processes, directional control, chirality and two-photon absorption.
6. The conception and characterization of adaptive metasurfaces, that incorporate externally addressable materials to change their response.
7. The conception and characterization of composite liquid crystal-based materials and metasurfaces incorporated in fiber optics: isolators, circulators, fiber gratings.
8. The conception and characterization of ultrafast processes in materials, with a direct focus on the realization of integrated saturable absorbers, essential to the generation of femtosecond-duration optical pulses.
9. The conception and characterization of second-order nonlinear metasurfaces that exhibit application-specific properties, with a direct use in ultrashort pulse diagnostics.
10. The conception and characterization of metasurface-based chirped mirrors, that are able to impart custom wavelength-specific optical paths, which may also be externally-controllable.
11. The conception and characterization of liquid crystal composites and/or metasurfaces that provide a customized temporal profile for ultrashort pulses.
12. The conception and characterization of metasurface-based linear devices, in reflection and transmission, with a strong focus on chiral effects.
13. The conception and characterization of composite materials and/or metasurfaces for integrated atmospheric RADAR/LIDAR devices.

At present time, I am activating as an associate professor in the National University Politehnica of Bucharest and as a III-rd degree researcher in the Laser System Department of ELI-NP. Given this exposure to academic infrastructures, the numerical analysis and modelling can be performed in the Condensed Matter Laboratory in the National University Politehnica Bucharest and within the ELI-NP infrastructure, the manufacturing of the samples can be realized under collaboration with the Solid Target Laboratory of ELI-NP, and the testing of the optical properties can be performed by in the Optics Laboratory of ELI-NP, or under collaboration with the Optical Characterizations

Laboratory in the National Institute of Plasma and Laser Radiation in Măgurele, Romania. Going forward, all experimental characterization will be able to be performed in the Center for High Power Optics that will be constructed under the umbrella of the ELI-NP infrastructure, and which will be operational starting with 2028. The ongoing research grant on which I serve as Project Director (ELI-RO/OMP/2025/001 - acronym „DIOEMT”) focuses on integrating the above-mentioned research directions into the scope of the Center, with the private sector being actively involved in these activities (Okamoto Optics - Japan, Beneq - Finland, NTG - Germany and Plymouth Gratings - UK). Also, I aim to continue to write national and international grant proposals to establish funding.