Nanostructured Metamaterials

Exchange between experts in electromagnetics and material science
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Exchange between experts in electromagnetics and material science

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MAGNONICS:

Project Title: Magnonics: Mastering Magnons in Magnetic Meta-Materials

Start and End Dates: 15/09/2009 till 14/09/2012
EU Contribution: EUR 3 499 820

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MAGNONICS aims to realise, on one hand, new nanotechnologies and, on the other hand, a new class of metamaterials, i.e., magnonic metamaterials, and thereby to prove the concept of magnonics. In other words, the consortium aims to explore metamaterials that can be viewed as obtained by integration of magnetic materials into conventional metamaterial structures and by a full exploitation of scientific and technological opportunities resulting from the tailored magnonic band spectrum.

This project produces exploitable intellectual property concerning:

- Top-down and bottom-up nanotechnologies for fabrication of periodic magnetic nanostructures.
- Advanced experimental and theoretical techniques for characterisation of magnonic and electromagnetic properties of magnonic metamaterials.
- Functional nanomaterials for and concepts of non-volatile logic architectures and devices for microwave signal processing.

EXPLOITABLE RESULTS

- Magnonic logic architectures responsive to/driven by free space microwaves
- 3-dimensional periodic magnetic nanostructures produced by protein crystallization
- Magnonic logic architectures interfaced with magneto-electronic (spintronic) devices
- Microwave signal processing using magnonic crystals
- Micromagnetic modeling of static and dynamic properties of devices containing magnetic components
- Magnonic metamaterials with tailored effectively continuous electromagnetic properties
- Magnonic metamaterials with tailored band gap and effectively continuous magnonic (spin wave) properties
The objective of METACHEM is to use the extreme versatility of nanochemistry to design and manufacture bulk metamaterials exhibiting non-conventional electromagnetic properties in the range of visible light. This spectral domain requires nano-scale patterns, typically around 50 nm in size or less. The strategy consists in designing and synthesizing ad-hoc nano particles as optical plasmonic nano-resonators and organising them through self-assembly methods in 2 or 3 dimensional networks in order to produce dense highly ordered structures at a nano-scale level. Several subprojects corresponding to different routes are proposed, all of them based on existing state-of-the-art chemical and self assembly methods. In addition, the important issue of losses inherent to the plasmonic response of the nano-objects is addressed by the adjunction of loss-compensating active gain media.

Main goals: Design and synthesize optically isotropic meta-materials with exotic and extreme properties realized by simple and cheap chemical methods.

Targeted properties: artificial optical magnetic and dielectric properties, optical left-handed materials, near-zero permittivity/permeability; negative index materials, low-loss plasmonic structures.

METACHEM
METACHEM FP7-NMP-SMALL-2009-228762

Project Title: Nanochemistry and self-assembly routes to metamaterials for visible light

Start and End Dates: 15/09/2009 till 14/09/2013
EU Contribution: EUR 3,699,990

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EXPLOITABLE RESULTS

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The NANOGOLD project aims at evaluating the potential of metallic nanoparticles (NPs) for the bottom-up fabrication of optical metamaterials, from design to fabrication. The objective is to use electromagnetic effects (i.e., plasmon resonance in metal particles, interference in layers, and scattering of clusters) on different length scales to create materials with non-conventional electromagnetic properties.

The approach is interdisciplinary and combines inorganic chemistry, organic macromolecular synthesis, physics of electromagnetic resonances and liquid-crystal technology. In a bottom-up approach, the metallic nanoparticles (resonant entities) are organized via self-organization on the molecular scale. Self-organization of composite materials is a unique approach that overcomes limitations of conventional planar fabrication technology, which is, at present, nearly exclusively used for the fabrication of metamaterials. This research will help closing the technological gap between a bottom-up nanostructure fabrication and real-world applications.
The aim of NIM_NIL was the development of a production process for 3D Negative Index Materials (NIMs) in the visible regime combining UV-based nanoimprint lithography (UV-NIL) on wafer scale using the new material graphene and innovative geometrical designs.

This project demonstrated results going beyond state-of-the-art in three important topics regarding NIMs: the design, the fabrication using nanoimprint lithography (NIL) and the optical characterisation by ellipsometry. At the end of the project a micro-optical prisms made from NIM were fabricated to directly verify and demonstrate the negative refractive index.

The outcome of the NIM_NIL project are highlighted in the following pages:

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The use of wave phenomena to process information is an established practice in the communication industry. E.g., magnetostatic spin waves propagating in ferrite materials have played an important role in radio frequency communications. It has been recognised recently that the frequencies of short wavelength dipole-exchange spin waves propagating in carefully tailored magnetic waveguides could match those of the microwave carrier signal in the next generation communication standards. Combined with prospects for the device miniaturisation facilitated by the naturally short wavelength of spin waves, this has triggered a remarkable wave of research into spin wave based (“magnonic”) signal processing and logical computation and generally into magnonics as the study of spin waves and associated technologies.

The delivery of the microwave signal to nanoscale magnonic circuits and devices is however challenging. In part, this is due to the architectural challenge of dealing with multiple impedance matched electrical current leads conventionally used for the excitation of spin waves. A wireless delivery of the microwave signal to required locations for subsequent magnonic processing could present an attractive alternative but has remained impractical due to the direct microwave to spin wave signal conversion being forbidden by the linear momentum conservation law.

Within the FP7 MAGNONICS project, we have developed a novel concept of signal conversion and the associated magnonic architecture, as illustrated in Figure 2. It stems from the widely known phenomenon of resonant field enhancement in the vicinity of metallic nanostructures at frequencies around the surface plasmon-polariton resonance. The linear momentum conservation limitation is circumvented by artificially breaking the translational symmetry by introducing special microwave-to-spin-wave antennas into the circuitry.
3-DIMENSIONAL PERIODIC MAGNETIC NANOSTRUCTURES PRODUCED BY PROTEIN CRYSTALLIZATION

List of partners involved in the specific result

- UNIVBRIS (University of Bristol, UK)

EXPLOITABLE RESULTS

Periodic magnetic nanostructures have a range of applications, including that focused on here: magnonic metamaterials. In contrast to the many top-down physical methods available to fabricate such nanostructures, we have developed a biochemical approach using a cage-shaped protein (for example, ferritin) as a template. The procedure is illustrated in Figure 1 for ferritin. A magnetic nanoparticle, for example pure magnetite $\text{Fe}_3\text{O}_4$, is synthesized chemically in the protein's central cavity with reagents entering via the natural channels in the protein (Figure 1a). The nanoparticles are then assembled to form a periodic array by crystallizing the host protein as shown in Figure 1b. In order to ensure that each protein contains a magnetic nanoparticle, a form of magnetic separation chromatography has been developed. This serves not only to reject unfilled protein molecules but also makes the nanoparticle size distribution even more monodisperse (typically $7.9 \pm 0.8$ nm for magnetite templated by ferritin) as shown in Figure 2. By these means it is possible to fabricate face-centred cubic arrays of magnetic nanoparticles where each dimension of the array can be as large as $\sim 500 \mu$m. Such structures would be very difficult to make by any other means.

![Figure 1](image1.png)

![Figure 2](image2.png)
Magnonic logic architectures interfaced with magneto-electronic (spintronic) devices

List of partners involved in the specific result

- CNISM, Perugia (Consorzio Nazionale Interuniversitario per le Scienze Fisiche, Unità di Perugia, IT)

Micro-focused Brillouin Light Scattering technique is a very powerful tool for mapping the spin intensity with sub-micrometric lateral resolution of about 250 nm. Here we have directly measured the spin waves coherently excited in a magnetic film by injection of a dc spin-polarized current through a nano-sized electrical contact. We showed that spin waves with tunable frequencies can propagate for several micrometres away from a perpendicularly magnetized nanocontact. The possibility to generate spin waves through point contact is attractive to design a new generation of magnonic devices, where spin wave are used as information carrier. In fact, one can easily foresee electrical and magnetic field control, broadband operation, fast spin-wave frequency modulation.

Left: Schematic sample layout. Cross-section of the sample, revealing the layers of the spin valve mesa and the active area of the STO device. An aluminium coplanar waveguide is deposited onto the spin valve mesa, and an optical window is etched into the central conductor of the waveguide close to the nanocontact. Right: Integrated intensity of the spin-wave excitations detected using micro-focused BLS as a function of distance from the centre of the point contact.
Microwave signal processing based on short-wavelength spin waves allows one to miniaturize microwave devices and components to the micro- and nanoscale. This is similar to devices such as filters and delay lines based on surface acoustic waves (SAWs) being an integrated part of today’s telecommunication technology. The partners TUM, CNISM, UNEXE and AMU of the FP7 project MAGNONICS have discovered that the signal processing based on spin waves and magnonic crystals (see figure) provides novel functionality. Using external magnetic fields of different orientation we have shown that spin wave transmission is switched on and off on purpose. Different magnetic states created in one and the same magnonic crystal have allowed us to even reprogram the filter and signal processing characteristics during operation and in the remanent state. This goes beyond SAWs and photonic crystals used in communication technology so far. Our results suggest a new class of multi-functional components for microwave technology.

Microwave device consisting of two coplanar wave guides (yellow color) integrated to a magnonic crystal formed by periodic 120 nm diameter holes in a ferromagnetic thin film (gray color). The lattice constant is 300 nm. The enlarged colorful image (simulation) illustrates signal transmission at 2.9 GHz via channels of large spin precession amplitude (red color) along the holes edges. The transmission can be controlled by an in-plane magnetic field. The substrate is green.
Production of 3-D nanostructured assemblies of plasmonic resonators

List of partners involved in the specific result
- CRPP Bordeaux (Centre de Recherche Paul Pascal, FR)
- CNR-IPCF Bari (Consiglio Nazionale delle Ricerche, Istituto per i Processi Chimico-Fisici, IT)

One of the objectives in METACHEM is to obtain metamaterials by controlled stacking and self-assembly of nanoparticles under the sole effect of interparticle forces, resulting in dense 2D or 3D "superlattices". Fabrication is planned along different main routes:
- Direct ordering (2D self assembly + 1D directed) by using Langmuir Blodgett and layer by layer methods to fabricate nanostructured materials
- Spontaneous 3D self-organization by physical chemical routes, such as solvent evaporation

Results:
- Control of the Langmuir-Blodgett and Langmuir-Schaefer assemblies of silica nanoparticles and core-shell silica@silver nanoparticles of subwavelength size has been successfully achieved along with the transfer of the obtained multilayer films
- Self-assembly of 15 nm size Au nanoparticles achieved by solvent evaporation by controlling solvent composition, temperature and nanoparticle size distribution

(A) And (B) ESEM micrographs of 1 and 3 layers of core-shell Ag@SiO₂ nanoparticles deposited on a silicon substrate by the modified Langmuir-Schaefer technique. (C) Macroscopic pictures show the uniform surface coverage with no cracks. (D) SEM image of self-assembled 15 nm Au nanoparticles from a nonane solution onto a plasma treated silicon surface (T=40 °C) (E) TEM images and statistical analysis of Au nanoparticles from size selected fraction.
The nanochemist partners of the METACHEM project have developed and optimized the fabrication of a different number of plasmonic nanoparticles and nanoclusters in significant quantities. Moreover, these nanoparticles have been loaded with different surface functionalities. The Au and Ag nanoparticles available on this consortium are monodispersed and with precise sizes. Additionally, these nanoparticles can be fabricated and dispersed in water as solvent.

To summarize we list below the particles and clusters available:

**Particles:**
- Spherical Au (10-60 nm), Ag (12-30 nm)
- Triangles Ag (35 nm)
- Surface functionalization (citrate, CTAB, PSS, PAH, PDDA and SiO$_2$)
- Dielectric shell can be adjusted between 5-180 nm

**Clusters:**
- Core-shell clusters with a core of Au(60 nm), SiO$_2$-coated Au(60nm), and with a shell composed of Au(15 nm)
- Core-shell clusters with a core of SiO$_2$ (65 nm) and with a shell composed of spheres Ag(27 nm) and triangles of Ag(35 nm)
- Core-shell clusters with a core of SiO$_2$ (65 nm) covered with a shell of 6 polystyrene particles

Transmission electron microscopy image of a central 60 nm Au nanoparticle surrounded by 15 nm Au nanoparticles.
We use a microfluidic technique (microevaporation) in order to induce the formation of dense states of functional nanoparticles (NPs). The goal is to generate and shape up crystals of NPs working as metamaterials in the visible range. Microevaporation allows the concentration of any solute in a microfluidic device. The device is made of a microchannel molded in a silicon elastomer in contact with a thin membrane permeable to water. As water evaporates, NPs are continuously concentrated and may form very concentrated states. The figure below illustrates the geometry and gives a series of examples of the structures we obtained.
We use self-assembly of templating polymer structures to fabricate anisotropic metal/dielectric hybrid nanocomposites with characteristic sizes 10-100 nm. Nanocomposites of polymers and gold nanoparticles are produced, both disordered (with simple polymers) and ordered (with block copolymers) with nanoparticles organized in alternating layers of a lamellar structure. This can be achieved with different particle sizes and concentrations.

Three different methodologies can be used to formulate the nanocomposites.

a/ neutral solvent.
Nanoparticles and block copolymer macromolecules are first dispersed in a neutral solvent for all species, and then slowly dried. The organization occurs spontaneously upon drying, provided the nanoparticles have a strong affinity for one of the block of the copolymers.

b/ selective solvent.
The lamellar structure is first obtained with an amphiphilic copolymer without particles. The structure is then swollen with an aqueous sol of nanoparticles, which swells selectively one of the domains only, preserves the lamellar long range order and allows the introduction of the nanoparticles.

c/ in-situ reduction of gold
Gold salts are introduced in the ordered copolymer matrices, in such a way that they are selectively confined within one of the domains. They are then reduced in-situ using either a UV, a chemical or a temperature trigger.

Main result:
Control of the lamellar organization of nanoparticles of size between 3 and 8 nm in different self-assembled block copolymer matrices, with lamellar period between 30 and 120 nm.
Liquid Crystal Materials with Coupled Resonant Entities

List of partners involved in the specific result

- UHULL (University of Hull, UK)
- USFD (University of Sheffield, UK)
- UPAT (University of Patras, GR)

Research into nanoparticles functionalized to exhibit self-assembling properties is currently receiving increasing attention due to recent predictions, suggesting that such systems could provide the materials base for metamaterials, with interesting properties including negative refractive index properties. This is especially interesting as theory has shown that to obtain negative dielectric permittivity the self-assembling particles can be much smaller than the wavelength of visible light. Thus the use of bottom-up approaches using the small nanoparticles 1.5-10 nm in diameter functionalized with organic liquid crystal groups, promoting self-assembly in the solid state is a very promising approach. [1,2] The organic material can be designed so that 2D or 3D organisation of the particles is dialled in, the distances between of the nanoparticles can be controlled to address plasmonic interactions. Moreover for the preparation of nanoparticle coatings on surfaces, established deposition techniques and know-how from LC manufacturing can be applied, minimizing thus technological risk. The use of LC polymer technology gives the self-organized particle films mechanical self healing properties and allows for further post deposition processing, such as photo-alignment or photo-curing. This approach is only as its infancy, as the size and shape of the nanoparticles as well the chemistry of the organic groups can be varied extremely widely.

Schematic representation of the Au NPs covered with LC groups and hydrocarbon chains.

TEM micrograph of such a LC nanoparticle film.

Owing to their subwavelength dimensions and strong plasmonic resonances, metallic nanoparticles are currently considered as promising candidates for the bottom-up fabrication of optical metamaterials. A major challenge in this respect is to control the organization of the NP building blocks into specific arrangements so as to tailor the resulting macroscopic optical response (e.g., permittivity and/or permeability). Complex architectures with 2D or 3D dimensionalities can be obtained by bottom-up nanofabrication techniques using polymer as a host template. NP-polymer multilayers can be prepared from solution by successive spincoating depositions allowing to control the thickness of the different layers and consequently the optical response of such 1-D hybrid photonic crystal, due to the interplay between the Bragg mode of the multilayer structure and the NP plasmon resonance. In another approach, polymers are used as surfactant to form dense spherical NP clusters following an oil-in-water emulsion process [1]. Following the emulsification of a NP-enriched oil phase in water, the formation polymer capsules allow to confine a discrete number of NPs, which aggregation into clusters is triggered in a second step by the addition of a molecular linker.

Fabrication of multidimensional functional assemblies of resonant nanoparticles

List of partners involved in the specific result
- UNIGE (Université de Genève, CH)
- JENA (Institute of Condensed Matter Theory and Solid State Optics, Abbe Center of Photonics, Friedrich-Schiller-Universität, DE)
- VI (Virtual Institute for Artificial Electromagnetic Materials and Metamaterials METAMORPHOSE VI AISBL, FI)

Exploitable Results

The rapidly increasing interest shown in metamaterials over the previous decade has been largely driven by the desire to control these properties in the visible region of the electromagnetic spectrum. Where previously the effects, such as negative refractive indices and cloaking, have been principally confined to longer wavelength domains, down-scaling of the structures used would, in some cases, be sufficient to make optical observations. The bottom-up organisation of colloidal metallic nanoparticles (MNPs), which support a localised surface plasmon resonance, offer one exciting route to achieving this as well as many advantages over more traditional top-down methods. Bottom-up approaches also offer solutions to one of the other principal challenges faced, namely the extension into the third dimension in order to produce bulk materials with effective medium parameters.

Layered MNP arrays can be produced using bottom-up nanofabrication techniques. The particles, grown in solution, adsorb onto modified substrates designed to provide an electrostatic attraction between the two. The control of array spacing, and therefore optical properties, is achieved through the build-up of individual polyelectrolyte layers between the metallic nanoparticle arrays in a process known as layer-by-layer (LbL) assembly. [1] There are no limits to the number of either MNP or polymer layers that can be deposited giving this method an inherent flexibility and allowing truly three dimensional structures to be fabricated. Such systems have been tested as potential SERS substrates.

Fabrication technology for 3D NIM materials and etching processes for micro-optical NIM prisms

List of partners involved in the specific result
- PRO (PROFACTOR GmbH, AT)
- JENA (Institute of Applied Physics, Abbe Center of Photonics, Friedrich-Schiller-Universität Jena, DE)
- SEN (Sentech Instruments GmbH, DE)
- MRT (micro resist technology GmbH, DE)

EXPLOITABLE RESULTS

During the project a complete fabrication process for 3D NIM-materials by using nanoimprint lithography (NIL) has been developed. For this specific stamps, nanoimprint resists and etch recipes were developed and multilayerd NIMs were manufactured.

In order to demonstrate the applicability of the NIM-multilayers additionally a resist prism was formed on top of these layers and a special etching process was adapted, in order to transfer the prism into the NIM layers.

The developed processes can clearly be extended to larger areas by e.g. a step and repeat process.

These 3D-negative index metamaterials have the potential to be employed in various electromagnetic components and devices. For microwave NIMs there are applications as Metamaterial antennas, microwave radar absorbers, electrically small resonators, waveguides, phase compensators and other advanced devices (e.g. microwave lens)

At optical wavelength the NIMs can be employed to develop a superlens which can be used for imaging below the diffraction limit. Other potential applications for negative index metamaterials are optical nanolithography, nanotechnology circuitry.

Sideview of multilayer NIM.

SEM image of micro-optical prisms etched into NIM layers.
Graphene production and applications

Graphene is a layer of sp2 carbon atoms arranged in a hexagonal lattice. Samples of single- and few-layer graphene with areas of square centimetres can be manufactured with a chemical vapour deposition (CVD) technique, and transferred to other substrates. We have developed prototype and scalable CVD reactors for graphene. The CVD approach to producing graphene relies on decomposing carbon, e.g. from CH$_4$, onto the nickel and copper catalyst substrates. The thickness and crystalline ordering of graphene are controlled, beside the catalyst, by the type and concentration of the carbonaceous precursor, by the temperature, by the H$_2$ dilution and by the cooling rate. After a chemical etching of the nickel and copper, the graphene layer detaches and can be transferred to another substrate, including Silicon, SiC, SiO$_2$, Al$_2$O$_3$, glass and plastics. p-type doping of graphene is also achieved using both gold nanoparticles and nitric acid treatment. The demonstration of large-area graphene from CVD is a promising step towards the industrial production of graphene for applications such as flexible and transparent conductive electrodes for displays, light emitting diodes and solar cells.

List of partners involved in the specific result

- CNR-IMIP, Bari (Consiglio Nazionale delle Ricerche, Instituto di metodologie inorganiche e dei plasmi, IT)
In contrast to thin gold or silver films, the carrier density in graphene can be tuned by “electric field doping”. This, combined with graphene’s inherent mechanical robustness, chemical stability and absence of roughness, makes micro and nano-patterned graphene an interesting ground for future photonic applications. Electrically doped graphene will be used in various photonic devices that rely on tunable plasmonic resonances, while many of those applications will require large areas (~cm²) of micro and nanostructured graphene. Within NIM_NIL, we have developed a process for micro and nano-patterning up to 2x2 cm² area graphene by means of UV-Nanoimprint lithography. An imprint resist (UVCur06) on top of a transfer layer (LOR1A) was structured using UV-NIL on top of chemical vapor deposited graphene transferred to a SiO₂ substrate (a). The imprinted structures in (b) were transferred into graphene using oxygen plasma etching. Afterwards the LOR layer was lifted off in a developer such that the patterned graphene was left on top of the SiO₂ substrate (c). Features down to 20 nm have been demonstrated.

(a) transferred CVD graphene on SiO₂ with size of 2x2 cm². (b) Imprint on 2x2 cm² graphene (c) Sample after etching through the graphene and lift-off of resists, SEM figures show a successful large area patterning of graphene for 3 µm and 2.5 µm period grating. (d) Raman mapping of graphene lines.
MILLING TECHNOLOGY FOR MICRO OPTICAL DEVICES AND STAMPS FOR NANOIMPRINT LITHOGRAPHY

List of partners involved in the specific result
   › JPS (Jenoptik Polymer Systems GmbH, DE)

We evaluated existing technologies for creating micrometer structures for NIM stamps, i.e. electron beam and grey scale lithography as well as ultra precision diamond turning and milling technologies
   › Evaluation of usable materials for the NIM stamps, like aluminium, nickel and PMMA

Fabrication of stamps for nanoimprint lithography following the structure requirements depending on the optical calculation of micro and nanometer structures
   › Evaluation of structure quality with respect to dimensions, structure angles, surface quality, i.e. roughness, form deviation, tool abrasion and tool life time
Nanoimprint lithography materials and process for fabrication of micro optical devices

List of partners involved in the specific result
- MRT (Micro Resist Technology GmbH, DE)
- PRO (PROFACTOR GmbH, AT)

Exploitable Results

The library of 3D surface topographies which can be generated by NIL is extending daily. The hybrid polymer materials which are developed by the NIM_NIL partners are excellent candidates which exhibit simultaneously very good imprint characteristics and optical properties. The material OrmoComp in this sense has superior characteristics to be applied as a thin planarization layer to cover the generated metallic NIM arrays to allow multi-stacking. Besides, it delivers thick layers to replicate the desired prismatic micro-structures on top of the NIM stack. This enables the fabrication of the planned NIM prism demonstrator. The application field of OrmoComp was extended within this project from an optical NIL material to an optically active etch mask for hybrid processes. Simulations show that prisms with different slopes show different negative index behavior. These different prisms may contribute to the general functionality of a unit cell. One can implement new 3D stamp fabrication techniques and versatile designs to cover e.g. negative index behavior on a wide range of wavelengths or to generate NIM diffuser cells, etc.

SEM images of first etching results for NIM prism into 3D NIM material.
A) FIB cut into etched prisms.
B) Higher magnification of cross section.

Diverse prism structures with different inclination and orientation can be simultaneously replicated by NIL into optically active materials to generate unit cells overcoming wavelength dependence of negative index properties.

Hybrid topographies with different optical functionality can be imprinted and subsequently etched in NIMs to achieve defined negative index properties.
Fabrication of metallic nanostructures using nanoimprint lithography

List of partners involved in the specific result

- PRO (PROFACTOR GmbH, AT)
- MRT (micro resist technology GmbH, DE)
- KU (Korea University, KR)
- JENA (Institute of Applied Physics, Abbe Center of Photonics, Friedrich-Schiller-Universität Jena, DE)

We developed an UV-Nanoimprint lithography based lift-off process for the fabrication of metallic nanostructures. Ormoceres or sol-gels on top of a transfer layer e.g. LOR1A are structured using UV-NIL (a), (d). Recessed sidewalls are achieved by a simple oxygen plasma etching due to different etching rates of both resists (b), (e). After deposition of the necessary metal and dielectric layers (f) - in our case 40 nm Ag / 20 nm SiO$_2$ / 40 nm Ag for fabrication of Negative Index Materials - the resist is lifted off and the metallic nanostructures remain on the substrate (c), (g). The smallest feature sizes demonstrated by this process are 50 nm line width and an aspect ratio (i.e. ratio of height to width) of up to three. This process suitable for mass production can be used to fabricate metallic or dielectric nanostructures on various substrates finding applications in biophysics, photonics and electronics.

(a) Nanostructured Ormocere on top of transfer layer (LOR1A)
(b) Recessed sidewalls due to faster etching of LOR1A in comparison to Ormocere in Oxygen plasma
(c) Cross section of metallic structure (stack of silver/SiO$_2$/silver after lift-off)
(d)-(g) shows a schematic drawing of process steps (a) - (c).
PROCESSING AND PASSIVATION OF METALLIC NANOSTRUCTURES

An intense effort is underway to find processing and/or coatings that inhibit metal nanostructure oxidation and/or tarnishing. Oxidation typically is inhibited by introducing a stable protective coating, which, however, have limitations, e.g., for optical applications, it may alter the transparency or optical resonances of the tailored metallic nanostructures. As an innovative approach, we have developed remote H\textsubscript{2} plasma processing of metallic (silver and gold) periodic nanostructures effective in inhibiting metal oxidation and stabilize surface plasmon resonances in nanostructure suitable for plasmonics and metamaterials. The processing addresses the important aspect of the hydrogenation to clean silver (Ag) regions, to passivate grain boundaries and stabilize chemically and in time Ag nanostructures. The developed processing does not need additional heating of the structure that can lead to uncontrolled huge enlargement of silver grain size and thus avoids disturbing effects on the original topography of silver nanostructures.

This processing can be further implemented by graphene transfer on top of the metallic stabilised nanostructure. Graphene possesses a unique combination of properties that are suitable for coating applications. Graphene layers are exceptionally transparent (~90% transmittance for 3-4-layered graphene), so graphene does not perturb the optical properties of the underlying metal. Our process can be applied to other metals, enlarging the possibilities of using metal based nanostructures in optoelectronic, plasmonic and sensing devices and paves the way for low-loss plasmonic metamaterials at high frequencies.

AFM topographies of Ag fishnet structure before and after H\textsubscript{2} plasma processing with a view of the H\textsubscript{2} plasma and corresponding SEM picture; XPS of the Ag\textsubscript{3d} core level before and after treatment demonstrate the effectiveness of the process in reducing silver oxide; optical data demonstrates the de-oxidation and stabilisation in time of the Ag fishnet structure.

List of partners involved in the specific result

- CNR-IMIP, Bari (Consiglio Nazionale delle Ricerche, Instituto di metodologie inorganiche e dei plasmi, IT)
- JENA (Institute of Applied Physics, Abbe Center of Photonics, Friedrich-Schiller-Universität Jena, DE)
- PRO (PROFACTOR GmbH, AT)
Main goals within the NIM_NIL project are the design, fabrication and comprehensive characterisation of large-scale optical metamaterials with exotic and negative refractive indices. Within the consortium, the Institute of Applied Physics at the Friedrich-Schiller-Universität in Jena/Germany is responsible for the fabrication of nanostructured master stamps as required for nanoimprint lithography. The technological challenges are to establish a process chain based on electron-beam lithography and a suitable dry etching for this purpose.

Among others, a main achievement was the establishment of large-scale metamaterial imprint stamps with smaller feature sizes of less than 50 nm. Further efforts include the accurate optical far-field characterisation of the final metamaterial samples on the basis of a combination of optical spectroscopy and a dedicated interferometric setup, and a meaningful physical interpretation of their unique electromagnetic properties.

List of partners involved in the specific result

- JENA (Institute of Applied Physics, Abbe Center of Photonics, Friedrich-Schiller-Universität Jena, DE)
Micromagnetic modeling of static and dynamic properties of devices containing magnetic components

List of partners involved in the specific result

- INNOJENA (Innovent Technology Development e.V., DE)
- UNEXE (University of Exeter, UK)
- TUM (Technische Universität München, DE)

EXPLOITABLE RESULTS

Micromagnetic simulations of the equilibrium magnetic state and dynamic magnetization processes of any ferromagnetic structure with typical sizes from ~ 50 nm to ~ 100 nm and frequencies in the MHz and GHz regions is available and can be used as an efficient and reliable tool to optimize the performance of different devices which make use of magnetic components.

In particular, existing software packages allow to calculate in advance (i.e., to predict without the actual manufacturing of the device) the following properties of ferromagnetic systems:

- Static magnetization curves and hysteresis loop, including initial dc-susceptibility, remanence and coercivity
- Eigenmodes of the magnetization oscillations, including their spatial power distribution and quality factor
- Field and frequency dependencies of the magnetic ac-susceptibility
- Reflection and transmission coefficients of magnetic structures for various types of incident waves

Potential consumers of this result are all companies and academic research groups interested in manufacturing and fundamental studies of magnetic structures and their technological applications.

Example result of micromagnetic simulations: Color maps of the real (left) and imaginary (right) parts of the magnetic ac-susceptibility $\chi(f,H)$ for a hexagonal array of nanodisks.
Our objective is to investigate the longitudinal localized surface plasmon resonances of an infinite doubly periodic array of nanorods. Integral equation approaches are exploited because (i) important underlying analytical results are represented in the form of Green's functions, (ii) the radiation condition is fulfilled implicitly and (iii) unknowns are limited to interfaces between homogeneous media. In view of the high level of detail, we find reduced-order representations of the currents on nanorods with the help of Macro Basis Functions, i.e. limited sets of field solutions on each object, in which the final results are decomposed. Compression techniques are exploited to calculate fast interactions between Macro Basis Functions. In Fig. 1, the longitudinal localized surface plasmon resonance has been investigated for an infinite, doubly-periodic array of gold nanocylinders. The case of excitation by a single electric point source is analyzed using the Array Scanning Method.

Magnitude of $z$ and $x$ components of electric field obtained with the Array Scanning Method, over 5 unit cells in the test plane, as shown on the left (spacing $a=0.098\lambda_0$, diameter $D=0.039\lambda_0$, length $L=0.184\lambda_0$, $\lambda_0=588\text{nm}$). $P_{z\text{source}}$ represents $z$-oriented electric current density of unit amplitude.
Our objective is to investigate the magnetic response of an infinite doubly periodic array of triangular nanoclusters, proposed by Dr. C. Simovski of Aalto University. Integral equation approaches are exploited because (i) important underlying analytical results are represented in the form of Green’s functions, (ii) the radiation condition is fulfilled implicitly and (iii) unknowns are limited to interfaces between homogeneous media. In view of the high level of detail, we find reduced-order representations of the currents on the magnetic nanocluster with the help of Macro Basis Functions, i.e. limited sets of field solutions on each object, in which the final results are decomposed.
The 2D material graphene exhibits unusual infrared (IR) characteristics that make it highly interesting for optical and electronic device engineering. NIM_NIL aims to incorporate graphene into optical metamaterials using nanoimprint lithography to pattern it on a large scale. An exact knowledge of graphene dielectric function (DF) required in order to design such structures is most accurately measured by spectroscopic ellipsometry, as shown by recent ellipsometric studies of graphene in the visible. Considering that graphene is a highly promising material for applications in tunable IR plasmonics, we performed the first near and mid-IR measurements of research-quality exfoliated graphene using a unique microfocus IR ellipsometer located at the BESSY synchrotron in Berlin. This was required due to the limited size of our exfoliated graphene flake, which are generally restricted to dimensions of a few hundred micrometers. Locating the tiny, almost-invisible flake was a major challenge, which we overcame by mapping the area (figure on the left). We could then measure the DF and compared it to theoretical models.

Ellipsometry is also a valuable tool for in-situ real time monitoring the growth and processing of graphene during CVD fabrication. At the CNR we used ellipsometry in the visible as an optical non-destructive method for controlling and optimizing the catalyst cleaning and annealing and, consequently, the graphene deposition and properties. The kinetic ellipsometry monitoring also highlighted the mechanism and kinetics of CVD graphene formation.
The fabrication of metallic nanostructures has attracted an outstanding interest in recent years because of their potential in extraordinary optical devices such as photonic metamaterials and the exploration of novel effects in low-symmetry nanostructures. However, from the point of view of nanofabrication, most metallic nanoparticles were fabricated as single functional layers only, limiting the control of the structural variation in the third spatial dimension. The design guidelines for more elaborate, three-dimensional nanostructures will benefit enormously from solutions to fabricate 3D metallic nanostructures.

We lifted this issue by the further miniaturization of truly three-dimensional metallic nanoparticles and a dedicated top-down nanofabrication technology [1]. Furthermore, we established a unique interferometric setup which allows for the direct measurement of the complex Jones matrix in the visible and near-infrared spectral domain, applicable not only to optical metamaterials, but rather to a very general class of dispersive media [2]. The performance of the method was applied at genuine 3D nanostructures revealing their outstanding dispersive characteristics. With respect to exploitation platforms, we could show how to reveal an optical activity larger than in any natural or artificial material [3] and a previously unknown optical effect, namely the asymmetric transmission of polarized light [4].

Interferometric setup to measure the complex transmission response of complex metamaterials.

Visualization of a chiral metamaterial composed of 3D nanostructures and its optical activity.

ELLIPSOMETRY AS CHARACTERISATION METHOD IN MASS PRODUCTION OF OPTICAL STRUCTURES/NIMs

List of partners involved in the specific result
- SEN (Sentech Instruments GmbH, DE)
- ISAS Berlin (Leibniz-Institut für Analytische Wissenschaften, DE)
- JKU/ZONA (Johannes Kepler Universität, Zentrum für Oberflächen- und Nanoanalytik, AT)
- IF (Institute of Physics Belgrade, University of Belgrade, RS)

EXPLOITABLE RESULTS

Ellipsometry is the method of choice for the experimental determination of the permittivity of bulk and thin film materials. It has significant advantages over normal incidence reflection and transmission. Ellipsometry measures obliquely incident reflection and transmission polarization ratios which significantly reduces errors in calibration. Oblique incidence also lifts the polarization degeneracy and expands the number of measurable parameters to four complex values for both reflection and transmission. Thus, for NIMs four real values are available by which the complex permittivity $\varepsilon$ and permeability $\mu$ may be retrieved.

We measured the spectroscopic ellipsometric parameters at multiple incident angles for fishnet NIMs fabricated using nanoimprint lithography. The magnetic resonances associated with the negative refractive regions are identified and the data is used as input for the extraction of effective parameters using the Berreman 4x4 matrix method. Our results showed that there is significant spatial dispersion in the structures and that the material may not be described by effective tensors.
We show a possibility to exploit the thin film made of one-dimensional (1D) or two-dimensional (2D) magnonic crystals (MC) (also in the form of antidots lattices) to fabricate metamaterials with tailored electromagnetic properties in broad range of frequencies, from few GHz up to hundreds of GHz. It can be used to create the metamaterial with negative refraction index or the metamaterial with required absorption of electromagnetic fields (see Figure (a)). The innovation consists also in the possibility to a precise estimation of a magnetic susceptibility for different geometries of 1D, 2D and three-dimensional lattices of magnetic nanoelements (see Figure (b) and (c)).

Figure: (a) The permeability function of the stack of a thin layers of slabs of 1D MC (the structure is shown in the inset) composed of alternating Co and permalloy stripes of 25 nm width, 5 nm thickness in the external magnetic field of $\mu_0 H_0 = 0.2$ T. The filling fraction of the magnonic crystal in the dielectric matrix is 25%. (b) Calculated magnetic susceptibility as a function of the frequency for a 1D array of interacting stripes of width 260 nm and periodicity 500 nm at $\mu_0 H_0 = 50$ mT. (c) $S_{21}$ scattering parameter, a quantity proportional to the magnetic susceptibility, measured with Vector Network Analyzer-FMR technique. In panels (b) and (c) the black (red) line denotes the real (imaginary) part.

List of partners involved in the specific result

- AMU (Uniwersytet im. Adama Mickiewicza w Poznaniu, PL)
- UNEXE (University of Exeter, UK)
- UNIFE (Università di Ferrara, IT)
- INNOJENA (Innovent Technology Development e.V., DE)
- TUM (Technische Universität München, DE)
Magnonic metamaterials with tailored band gap and effectively continuous magnonic (spin wave) properties

List of partners involved in the specific result
- UNIFE (Università di Ferrara, IT)
- AMU (Uniwersytet im. Adama Mickiewicza w Poznaniu, PL)
- UNEXE (University of Exeter, UK)
- CNISM, Perugia (Consorzio Nazionale Interuniversitario per le Scienze Fisiche, Unità di Perugia, IT)
- TUM (Technische Universität München, DE)

EXPLOITABLE RESULTS

Band gaps are studied in two-dimensional magnonic crystals consisting of holes embedded into a ferromagnetic medium, using the dynamical matrix method. A comparison with experimental dispersions obtained by means of Brillouin light scattering is performed. The occurrence of band gaps at the Brillouin zone boundaries can be interpreted as due to the Bragg diffraction for propagating spin waves, because of the presence of the artificial periodicity of the internal field. However, the relevant scattering potential for Bragg reflection is not provided by the holes themselves, but by the concomitant internal field inhomogeneity between holes. Magnonic band structure of thin films of magnonic crystals with a small lattice constant is determined mainly by exchange interactions. For small filling fractions, the magnonic gap width is only weakly dependent on the shape of inclusions, showing the effective magnonic properties of this sets of parameters.

Experimental Brillouin light scattering data (circles) and calculated bands (lines). The red curves represent localized Damon-Eshbach-like mode (DEloc). The dashed vertical lines mark the borders between adjacent Brillouin zones. Inset: profiles of DEloc and DEint modes at the border of the first Brillouin zone. The couple of modes is separated by a frequency band gap of 0.6 GHz.

Width of the first magnonic band gap for a triangular lattice of square (red), hexagonal (green) and circular (blue) Fe inclusions versus the filling fraction. Magnonic crystal has the form of a thin film (5 nm) with Fe inclusions in Ni matrix. The insets below the plot illustrate changes in the structure as the filling fraction increases.
Design of 2D and 3D metamaterials with optical response in the visible regime

List of partners involved in the specific result
- FORTH (Foundation for Research and Technology Hellas, GR)
- IF (Institute of Physics Belgrade, University of Belgrade, RS)
- ISAS Berlin (Leibniz-Institut für Analytische Wissenschaften, DE)
- JKU/ZONA (Johannes Kepler Universität, Zentrum für Oberflächen- und Nanoanalytik, AT)

EXPLOITABLE RESULTS

The realization of low loss negative index metamaterials in the visible regime is highly desired for various practical applications [1]. Fishnets, a category of perforated metal-dielectric-metal structures, are found very promising to obtain negative index in the optical regime. Therefore, we need to find the best option as metallic elements to mostly reduce the loss of the system. In the meantime, nano-imprinting lithography technique makes possible multilayered fishnet configuration, i.e., a real three dimensional (3D) metamaterial. Considering the characterisation of negative index metamaterials, the demonstration of negative refraction by building a wedge configuration is the most intuitive way. However, no such simulation or measurement result has been reported in the visible regime until now.

We developed a model to describe the dissipative loss in resonant metamaterials. The model leads to an identification of what conducting materials are useful for metamaterials, and silver is found as the best conducting material at optical wavelength [2, 3]. Through the retrieval procedure for multilayered systems, we may get the effective electromagnetic parameters of the designed 3D metamaterials, so that we are able to optimize the fishnet structure for a low-loss negative index metamaterial in the visible regime. We improve the wedge system by applying a relatively narrow incident beam compared to the studied system, and it renders us an unambiguous demonstration of negative refraction for our designed negative index metamaterials in the visible regime.

(a) Retrieved real part of refractive index for a 15-layer structure of our designed optical metamaterial.

(b) Wedge demonstration of negative refraction for our designed metamaterial at wavelength 590 nm.

Within the MAGNONICS project AMU is responsible for the development of theoretical models, the calculation of the magnonic band structures in 2D and 3D magnonic crystals (MCs), and the modeling of the effective continuous properties of magnonic metamaterials. The main objective is to adapt the plane wave method to the calculation of the spin-wave spectra of the MCs fabricated using various techniques by other groups in this project. The electromagnetic metamaterial properties (i.e. the magnetic susceptibility and permeability) of the magnonic structures (see the Figure) in the GHz-THz frequency range are calculated, too.

Proposition of electromagnetic metamaterial based on an antidot lattice magnonic crystal, and the respective spin-wave resonance (SWR) spectra with two pronounced peaks in the high-frequency range. Around these frequencies the negative permeability appear.

Tasks of CNISM:
Within the MAGNONICS project, CNISM is responsible for dynamical characterisation in the GHZ frequency range of the 1D and 2D Magnonic Crystals by means of conventional and micro-focused Brillouin Light Scattering technique. The main CNISM activity was the measurements of the spin wave band structure in continuous and discrete Magnonic Crystals in order to achieve a quantitative description of fundamental physical phenomena induced by the artificial periodicity, such as the existence of allowed and forbidden frequency bands, and the appearance of acoustic and optical spin waves due to the presence of a complex base for the Magnonic Crystal. We measured for the first time the band diagram for a two-dimensional MC constituted by a square array of coupled NiFe disks.
The general task of the Innovent theory group in the frames of the MAGNONICS project is numerical micromagnetic modelling of the spin wave dynamics in 2D and 3D magnonic crystals fabricated and measured in the project. In particular,

- static simulations are performed, in order to find the ground state of the system magnetization (to be used in dynamic simulations)
- dynamic simulations of 2D and 3D periodic structures are carried out, with particular attention to the effects of the spatial structure on the properties of magnonic crystals

- in order to optimize the performance of magnonic logic devices, dynamic simulations of their characteristics are also performed.
- conversion utilities and post-processing tools for comparison of the simulations output with analytical theories and experimental results are developed.

Eigenmodes of the magnetization oscillations for a hexagonal array of nanodisks at various external fields

A reprogrammable magnonic crystal formed by a periodic array of ferromagnetic nanowires exhibiting alternating magnetization directions (green and red arrows). Using a small in-plane field $H$ a spin wave (blue) is stopped.
In addition to coordination of the MAGNONICS consortium, UNEXE is involved in the design, fabrication and characterisation of magnonic metamaterials as well as devices containing such metamaterials as their functional elements. Along with the more conventional processes, UNEXE develops self-assembled etched nanosphere lithography (ENSL) for fabrication of large area magnonic metamaterials. The characterisation of the fabricated meta-materials and devices is performed using time-resolved scanning Kerr microscopy (TRSKM) in the GHz domain, broadband spectroscopy and time resolved optical pumped scanning optical microscopy (TROPSOM) in the THz domain, and Magnetic Transmission X-ray Microscopy (MTXM at Advanced Light Source in Lawrence Berkeley National Lab) at statics. The TRSKM is also used for testing magnonic devices. The design activities are based on numerical simulations of all micromagnetic aspects of the magnonic technology.
The University of Bristol group focuses on nanofabrication for metamaterial applications by novel wet-chemical processes. Compositionally modulated magnetic alloy films with repeat distances down to less than one nanometre are produced by precision electrodeposition. We are preparing a range of monodisperse magnetic nanoparticles including pure and cobalt-doped magnetite in protein and virus templates. The nanoparticles can be assembled into large-scale three-dimensional periodic arrays by crystallizing the carrier proteins, and patterned by either top-down or bottom-up techniques.
Within the METACHEM project, CRPP is involved in three tasks.
(i) Fabrication of 3-dimensional assemblies of plasmonic resonators by Langmuir-Blodgett technique. The assembled objects are core-shell nanospheres with metallic core and silica shell synthesized at CRPP (gold core) and ICMCB (silver shell).
(ii) Fabrication of nanostructured metal-dielectric composites made from self-assembled diblock copolymer templates. Optical characterisation by spectrophotometry and spectroscopic ellipsometry of materials assembled in (i) and (ii) are performed at CRPP.
(iii) Theoretical studies of compensation of losses by active gain media.

Within the METACHEM project, ICMCB is involved in two main tasks:
(i) Synthesis and structural characterisation of raspberry-like nanoresonators composed of silver spherical or triangular nanocrystals located around a dielectric central nanoparticle of larger size. Production in sufficient quantities.
(ii) Preparation of large quantities of patchy colloidal particles with a precise number of patches for specific decoration with Ag triangular particles.

Within the METACHEM project, CNRS-Paris is involved in two tasks.
(i) Layered deposition of fishnet structures of plasmonic resonators and dielectric materials by dip-coating (bottom-up synthesis).
(ii) Latex templating to produce a range of pore sizes (50-350 nm) to study the tunability of the network and to confirm theoretical calculations.
The objective of the work of Fraunhofer ISC is the support of Metachem partners to identify at an early stage needs of industrial customer which have to be considered when results will be exploited. This support is performed by different approaches: (1) Fraunhofer ISC assists by providing special designed proprietary nanomaterials, such as hybrid polymers, in order to increase the possibilities to process metamaterials and to manufacture demonstrators. (2) Fraunhofer ISC provides some top-down technologies in order to create guiding patterns for (self-assembling) metamaterials. (3) Fraunhofer ISC increases the awareness with respect to industrial requirements such as representative, chemical, mechanical, thermal requirements.

The role of UVIGO on METACHEM is linked to the fabrication of plasmonic nanocluster by means of colloidal chemistry approaches. In particular the effort is focused on the development of novel synthetic strategies for the production of magneto electric nanoclusters (MENC) and magnetic nanocluster (MNC). The MENC consist on a central metallic (Au or Ag) surrounded by smaller (Au or Ag) satellites, while MNC are similar but with a dielectric (SiO2, polystyrene) central particle.

Typical transmission electron microscopy image of clusters made with two different sizes of Au nanoparticles, a central 60 nm surrounded by 15 nm nanoparticles.
The role of AALTO in METACHEM is to offer novel design solutions for magnetic and magnetoelectric nanoclusters, to develop analytical and numerical models of them and to obtain theoretical evidences that ensembles of these nanoclusters possess the target optical properties as follows:

- Resonant isotropic magnetic response in the visible and interband ranges
- Isotropic negative permeability in these ranges
- Isotropic near-zero permeability and/or permittivity
- Isotropic negative refraction index

We show using HFSS and CST Studio simulations the negative refraction in an isotropic lattice of core-shell plasmonic nanoparticles operating in the interband range. The design parameters were obtained theoretically.

The main task of CNR-IPCF in METACHEM are to (i) develop nanosized building blocks (nanoparticles, nanocrystals) with fine control of size, shape and composition for fabrication of novel metamaterials by using colloidal material chemistry tools, (ii) properly engineer and functionalize the synthesized nano-objects in order to conveniently exploit them for the fabrication of metamaterial with a engineered electrical and magnetic response (iii) assemble the suitably engineered nanomaterials by using spontaneous assembly, exploiting the specific surface chemistry of the prepared building blocks to achieve 2/3 D organization into large scale architectures (iv) synthesize suitably tailored active component to use to enhance optical gain (v) implement chemical and physical characterisation of the fabricated structures.

Schematic representation of the drop-casting and solvent evaporation procedure for NC self-assembly. (a) TEM images of 3.9 nm±0.5 nm PbS nanocrystals (NCs), (b) of 1.9 nm±0.5 nm and 4.1 nm±0.5 nm PbS NCs, (c) of 1.9 nm±0.5 nm and 5.4 nm±0.5 nm PbS NCs in toluene solutions, with the corresponding close up on the geometry (scale bar = 20 nm), the FFT and the sketch of the assembled geometry. (d) TEM image of Au 15 nm ±1 nm nanoparticles.
CNR-LICRYL, IT
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CNR- LICRYL overall aim is to tackle and solve the fundamental problem of optical losses in nano-engineered plasmonic structures. We proposed and explored a multi-scale complementary approach to introduce optically active components right at the heart of the engineered meta-materials (dyes, quantum dots, semiconductor nanocrystals). Therefore, the main objective is to demonstrate the validity of loss compensation in metamaterials both gain functionalized or gain assisted in order to enable their numerous potential applications. In particular, the main tasks are:

- Study of loss compensation feasibility in the framework of the meta-structures engineered within the consortium as function of dye or nanocrystals concentrations;
- Definition of the main geometrical parameters and physical properties from single element to the assembled structures.

Dedicated microfluidic tools based on evaporation are developed to obtain densely packed NPs with the advantage of providing control of the crystal nucleation stage at the nanolitre scale and to permit directed growth in confined geometries (shaping-up of materials).

3D structure made of densely packed functional nanoparticles (here Ag@SiO2).
LOUVAIN is mainly involved in the fast numerical simulation of complex metamaterials at optical frequencies. An integral-equation approach is chosen, since it offers the possibility of limiting unknown fields (equivalent currents) on the interfaces between piecewise homogeneous media and does not require absorbing boundary conditions. In view of the high level of detail, we find reduced-order representations for the fields on the objects. This is done here with the help of Macro-Basis Functions, i.e., limited sets of field solutions on each object, in which the final results is decomposed. Fast and accurate solutions (benchmarked by comparison with commercial softwares) are obtained for arrays of plasmonic nanorods (see figure), for arrays of resonant particles (spherical or triangular) arranged around a spherical cell and other templated or self-arranged metamaterials.

Magnitude of z and x components of electric field obtained with the Array Scanning Method, over 5 unit cells in the test plane, as shown on the left (spacing a=0.098\(\lambda_o\), diameter D=0.039\(\lambda_o\), length \(L=0.184\lambda_o\), \(\lambda_o=588\)nm). \(P_{\text{source}}\) represents z-oriented electric current density of unit amplitude.

The role of the University of Manchester in METACHEM is to study optical properties of fabricated nanomaterials, extract optical constants, check and study extraordinary electromagnetic behavior of plasmonic nanostructures, suggest new geometries of the plasmonic metamaterials and evaluate their properties using electron beam lithography. UNIMAN also participates in theoretical analysis of negative index metamaterials and evaluation of possible applications. In addition, the University of Manchester studies various coupling of localized plasmons and spatial dispersion in detail.
University of Siena (UniSI) is involved in the theoretical and numerical modeling of the interaction between electromagnetic waves and self-assembled metamaterials, to provide physical insight and quantitative tools for predicting peculiar optical responses. Specifically, UniSI has been developing a code based on Mie theory and single dipole approximation (but in the future will be extended to higher spherical harmonics) for the description of aggregates of spherical nanoparticles (e.g., coated spheres, clusters) both when isolated (diluted limit) and when packed in layers or crystal (2D/3D periodic arrays). The response of the particles is calculated in terms of measurable parameters like cross-sections (extinction, absorption, scattering), from which effective parameters of the metamaterial can be retrieved. The algorithm for the extraction of material parameters and their dependence on short-range-order/long-range-disorder is also subject of theoretical investigation.
At the FSU Jena we are contributing to the Nanogold project by providing theoretical understanding and by describing numerically how light interacts with self-assembled bottom-up nano-structures that constitute the target of the project. With this work we provide genuine contributions to the field of theoretical nanooptics but also strongly support the work of our experimental partners by providing ideas on potential structures to be fabricated and by supporting the characterisation of fabricated structures by devoted simulations.

The University of Hull research group is focused on the design and synthesis of organic-inorganic hybrid materials for subsequent investigation into their meta-material properties. This has involved particular focus on the development of techniques for gold nanoparticle formation, varying the size of the nanoparticles from 1.5 – 6.3 nm, as well as the design and synthesis of existing and novel liquid crystalline organic materials for the fabrication of the self-assembly hybrid meta-materials.

Our groundbreaking solution to form periodic structured bulk electromagnetic meta-metamaterials is interdisciplinary and combines inorganic chemistry, organic macromolecular synthesis, physics of electromagnetic resonances and liquid crystal technology. Resonant entities (metallic nanoparticles) are organized via self-organization on the molecular and supermolecular scale in chains or in 2D and 3D assemblies. Systematic modular variation of the chemical entities gives access to libraries of materials which will be used to arrive at systems with the desired properties.
Within the NANOGOLD project, UNICAL has contributed by designing and realizing active plasmonic systems in polymer-liquid crystal composite structures doped with gold nanoparticles (AuNPs). The most relevant result is the possibility of tuning the plasmonic resonance frequency of AuNPs by exploiting liquid crystals as reconfigurable media. The plasmonic peak position can be shifted by both applying an external electric field to the developed systems or by changing their temperature conditions. In figure, an example is reported of the morphology and functionalities of a channelled polymer structure infiltrated with AuNPs doped Cholesteric LCs (CLCs).

Within the NANOGOLD project, UNIGE is preparing plasmonic nanoparticles and develops methods for their self-assembly in two and three dimensions. We used a layer-by-layer technique based on polyelectrolytes to fabricate multilayer arrays of gold nanoparticles. The technique allows one to control on a nanometer scale inter- and intra-array distances between nanoparticles and thus the optical properties of the system. The use of curved surfaces (silica beads) resulted in the organization of gold nanoparticles in a core-shell system (see figure). The optical response of these structures with the strongly shifted plasmon resonance indicates that the leading term stems from a magnetic dipole contribution.
The UPAT team provides theoretical support and technical knowledge in molecular simulations and in electromagnetic simulations. The main target of the UPAT team is to simulate the self-assembly processes by which metallic nanoparticles (NPs) decorated with liquid-crystalline molecules organize themselves into finite clusters (aggregates) and, at the second level, how the clusters can organize themselves to macroscopic lattices or glasses so as to realize a bottom-up metamaterial exhibiting artificial magnetism. Having determined the structure of the metamaterials by molecular simulations of the self-assembly process, the electromagnetic (optical or IR) response is probed numerically by electromagnetic techniques such as the layer-multiple scattering method and discrete dipole approximation.

The Polymers, Liquid Crystals and Supramolecular Structures group has many years experience in studying the structures and physical properties of soft matter, particularly LCs and supramolecular materials, and in diffraction and complementary techniques, such as AFM, TEM, SEM and optical microscopies (polarized, confocal, fluorescence). The development of advanced instrumentation and analytical methods for x-ray and neutron scattering is complemented by the development in near atomic resolution atomic force microscopy elsewhere at Sheffield. Purpose-built in-house equipment is used, as well as synchrotron radiation sources where high source brilliance and resolution are required. Grazing-incidence scattering, particularly powerful in the study of thin film nanostructures is used extensively.

To fabricate metamaterials through self-assembly of nanoparticles, one must first understand the various structures such nanoparticle systems are able to form. Our knowledge of such structures and their self-assembly principles, could then enable us to design nanoparticle systems that will demonstrate the desired optical, or electrical properties. Scattering techniques, combined with advanced microscopies, are irreplaceable structure characterisation methods. They are versatile, non-destructive and require very small sample sizes.

Figure: (a): 3D orthorhombic metamaterial made of air cavities in silica containing clusters of gold nanoparticles. Each cluster consists of 100 nonoverlapping gold nanoparticles of radius $r=8.8\text{nm}$ in a nearly close-packed arrangement, with cluster radius $R=42.67\text{nm}$. Each cluster is placed at a center of a cavity of radius $44.8\text{nm}$. The metamaterial is viewed as a succession of $(001)$ planes (square lattices) of clusters of gold NPs, parallel to the xy-plane. The lattice constant of each square lattice is $a_x=a_y=85.22\text{nm}$ whilst the lattice constant in the z-direction is $a_z=87.86\text{nm}$. (b): Transmittance $T$, reflectance $R$, and absorbance $A$ spectra for light incident normally on a slab of the metamaterial of (a) consisting of 8 unit planes.
Within the NANOGOLD project, the role of the Virtual Institute "Metamorphose" is providing consultancy on characterisation and potential applications of metamaterials developed by the project. Also, organization of the constant dialog and discussion via setup of dedicated and regular workshops, meetings and courses.

The contribution of the Optics and Photonics Technology Laboratory in EPFL to the project concerns the bottom-up organization of metallic nanoparticles (NP) into thin films or spherical assemblies and the structural (AFM, SEM, TEM) and optical characterisation (UV-Vis, POM, microspectrometry, spectroscopic ellipsometry) of those assemblies. Commercial silver NP and gold NPs functionalized with liquid crystal ligands (synthesized by our partners in Hull) were organized into different arrangement, from multilayers to spherical clusters. The optical response of NP-polymer multilayers could be tuned by tailoring the interplay between the NP plasmon mode and the Bragg mode of the multilayer. The spectral response of dense spherical NP clusters was shown to be associated with the excitation of a magnetic dipole resonance, an essential ingredient in the realization of negative index metamaterials.
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Plasma processing for cleaning, passivation and stabilisation of metal structures CNR developed a H₂ remote plasma processes for dry cleaning and annealing of metal substrates and NIM structures. The H₂ plasma processing of nickel and copper was needed to achieve good substrates for the growth of high-quality graphene, while H₂ remote plasma processing of silver based NIMs was used to reduce silver oxides and stabilize in time silver NIMs.

- Preparation of large area graphene using CVD methods also plasma assisted CNR developed a modified process for the chemical vapor deposition (CVD) growth of graphene by CH₄-H₂ on nickel and copper substrates. The graphene, both single layer and multilayer, was transferred on various substrates including SiO₂/Si, glass, quartz, sapphire, plastics.

CNR-IMIP also achieved the formation of epitaxial graphene directly on the Si-face and C-face of 4H- and 6H-SiC.

- Topographic characterisation and in-situ ellipsometry One of the peculiarities of our graphene synthesis was the use of in-situ real time spectroscopic ellipsometry, which is sensitive to monolayer formation, to investigate graphene CVD growth kinetics and achieve control of graphene homogeneity and thickness.

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Photonic, Phononic and Metamaterials Group
The Institute of Electronic Structure and Lasers (IESL) of the Foundation for Research and Technology Hellas (FORTH) is an internationally recognized centre of excellence in lasers and applications, microelectronics and devices, polymer science, and theoretical and computational physics.

FORTH-IESL is involved in the NIM_NIL project through its Photonic, Phononic and Metamaterials (PPM) group. The PPM group is among the world’s pioneering groups in the study of photonic crystals, phononic crystals and electromagnetic metamaterials, researching those materials since the birth of the associated fields. The group has played a critical role in many of the most important achievements in these fields, including the first photonic bandgap material and the first negative index metamaterials in the infrared and optical frequency bands.

Within the NIM_NIL project, the task of FORTH is the theoretical and numerical investigation and analysis of three-dimensional optical metamaterials, and the design of optimised optical metamaterial structures. Various conducting materials—including graphene, metals and transparent conducting oxides—were tested as components of metamaterials. Several 3D metamaterial structures that may exhibit negative index of refraction were analyzed and tested.
(i) preparing mechanically exfoliated graphene samples which have been used in ellipsometric measurements and to obtain high-quality structured graphene samples at PRO;
(ii) Raman and infrared spectroscopy, atomic-force microscopy and VIS ellipsometry of single- and multi-layer graphene which helped to establish an expertise in the NIM_NIL consortium on optical properties of graphene and understand its potential in metamaterial applications;
(iii) numerical simulations of the variable-angle ellipsometric response of metamaterial structures fabricated within the NIM_NIL consortium and their detailed analysis which have helped to fully understand the complicated experimental data obtained from NIM_NIL samples;
(iv) variable-angle ellipsometric measurements of NIM_NIL samples aimed at understanding the in-plane dispersion of plasmonic resonances which are important for the wide-angle operation of metamaterials;
(v) numerical simulations of the NIM_NIL prism in order to help prepare the experimental setup and interpret the experiments which should demonstrate negative refraction in the visible.

Within the NIM_NIL project, ISAS has contributed spectroscopic ellipsometry measurements in the spectral range from visible to infrared of references and negative index materials (NIM).
We primarily developed optical interpretations and established the dielectric functions for characteristic spectral regions of selected materials. We also compared ellipsometry measurements to the standard characterisation of NIMs i.e. transmission and reflection measurements and interferometry. We determined for the first time the dielectric function of a graphene flake in the mid infrared spectral range. Variable-angle ellipsometric measurements on varying substrates allowed us to identify the characteristic modes (see figure).
Main goals of the NIM_NIL project are the design, fabrication and comprehensive characterisation of large-scale optical metamaterials with exotic and negative refractive indices.

Within the consortium, the Institute of Applied Physics at the Friedrich-Schiller-Universität in Jena, Germany is responsible for the fabrication of nanostructured master stamps as required for nanoimprint lithography. The technological challenges are to establish a process chain based on electron-beam lithography and a suitable dry etching for this purpose. Among others, a main achievement was the establishment of large-scale metamaterial imprint stamps with smallest feature sizes of less than 50 nm. Further efforts include the accurate optical far-field characterisation of the final metamaterial samples on the basis of a combination of optical spectroscopy and a dedicated interferometric setup, and a meaningful physical interpretation of their unique electromagnetic properties. With all these abilities we finally contribute to the transfer of metamaterials towards real-world applications we are interested in.

1) Characterisation of NIMs:
To characterize with spectroscopic ellipsometry the negative refraction and its dispersion and to measure effective parameters for permeability and permittivity. The experimental results are compared to theory and also with usual measuring techniques like reflection and transmission measurements of NIMs. Ellipsometry is a valuable tool to control the fabrication process without destroying the samples therefore it can be used for production control.

2) Ab initio modeling of structured samples
The quantitative spectroscopic studies of negative index structures in the mid infrared spectral range (3 µm – 20 µm) and UV-VIS (25 nm – 3µm) is accompanied by rigorous coupled wave analysis (RCWA). Measured results will be compared to reflection and transmission measurements. This leads to algorithms to improve the ellipsometry software.

3) Ab initio calculation of dielectric functions:
The electric susceptibility (equivalent to -dielectric function respectively permittivity) is calculated with ab initio by solving numerically the Kohn-Sham equations. At the beginning this has been done for graphene, then for noble metals and semiconductors. These results can be directly compared with measured data.
JPS designs the experimental setup for the NIM prism i.e. choice of detectors, lens system and dimensions of the NIM prism. JPS supports the whole consortium with information about the suitability of the processes for mass production from the industry driven point of view e.g. of the used materials, the used process steps.

Nano Materials and Device Laboratory in Korea University has developed a various kind of nanoimprint lithography for stacking process of Negative index Materials. For a higher stacking process, triple layer nanoimprint lithography has been developed using polyvinyl alcohol and LOL™2000 double sacrificial layer. Besides Si-contained UV curable resin(m-PDMS resin) was developed and used for higher etching selectivity. Versatile nanoimprinter with air cushion press has also been developed and used for fabricating 3D NIMs.
micro resist technology GmbH (MRT) is an SME which develops and produces specialized photoresists applicable in microelectronics and micromachining/micro-electromechanical systems (MEMS) as well as for large-area patterning and electroplating processes.

In general, MRT is responsible for the adaptation of materials in NIM_NIL for UV-NIL and lift-off processes. The materials are used as etch masks for the structuring of the different substrate candidates to produce Negative Index Materials (NIMs). MRT also provides transparent stamp materials to facilitate the low-cost fabrication of NIMs by NIL. Development of a planarization resist for the generated μm-size NIMs has also been a task of MRT, which is a key material for building up multi layer NIMs to demonstrate the successful NIM prism.

In the NIM_NIL project PROFACTOR GmbH developed processes based on nanoimprint lithography for the fabrication of metallic as well as graphene structures down to 20 nm feature sizes.

A stacking process of Negative Index Materials (NIMs) was established to achieve 3D NIMs in the visible regime. Further μm-sized optical devices like prisms were replicated into Ormoceres to be etched into 3D NIM materials. PROFACTOR GmbH offers services regarding process and material development for nanoimprint lithography as well as functional coatings and two products: an anti sticking layer BGL-GZ-83 for nanoimprint lithography stamps or photomasks as well as the HNMP-12 adhesion promoter for working stamp materials (PFPE and Ormoceres).
SENTECH Instruments GmbH, located in Berlin, develops, manufactures and sells products related to the measurement and characterisation of thin films and plasma process technology world wide.

Products for thin film metrology comprise: Film Thickness Probe, Laser ellipsometer, Spectroscopic ellipsometer and the SENDURO.

In the field of plasma process technology for the structuring and deposition of films for a variety of applications, especially in III/V, micro-optics, and nanotechnology SENTECH offers RIE plasma etcher, ICP PTSA plasma etcher, ICP PTSA cryogenic plasma etcher, ICPECVD plasma system.

Within the NIM_NIL project Sentech is involved in the development of etching processes for the fabrication of the stamps for the UV-NIL process and the fabrication of the NIM demonstrator and in development of the methods for measurement and characterisation of the structures with ellipsometry.

Mapping of Psi and Delta across the two graphene flakes with step size of 3 µm. 

SE850 spectroscopic ellipsometer with micro spots.


Under preparation, review, accepted for publication:


M. Madami, G. Carlotti, G. Gubbiotti, F. Scarponi, S. Tacchi, T. Ono, ”Magnetization ground state and spin excitations in multilayered rectangular nanodots as a function of the magnetic field“, (submitted).


Y. Au, E. Ahmad, O. Dmytriiev, M. Dvornik, T. Davison, and V. V. Kruglyak, “Resonant conversion of microwaves into spin waves”, (submitted).

M. Dvornik and V. V. Kruglyak, “Surface magnonic states in stacks of magnetic nanoelements”, (submitted).
List of the conferences from CNRS:


- "Cloaking a 3-D arbitrary shaped star-domain", Veltri A., Metamaterials 2010 - 4th International Congress on Advanced Electromagnetic Materials in Microwaves and Optics, Karlsruhe (Germany), 12-16 September 2010, Poster.


List of publications by CNRS


List of the conferences from UVIGO:


List of publications from UVIGO:

Marcos Sanles-Sobrido, Manuel Bañobre Lopez, Verónica Salgueirino,” Miguel A. Correa-Duarte,” Benito Rodriguez-Gonzalez, Jose Rivas, Luis M. Liz-Marzan, Tailoring the magnetic properties of nickel nanoshells through controlled chemical growth, Journal of Material Chemistry 2010, 20, 7360. This work has been selected for appearing at the cover of the journal.

List of the conferences from CNR:
G. Strangi, A. De Luca, R. Comparelli, M. Correa Duarte, S. Ravaine, and R. Bartolino,”Optical Loss Compensation: Resonant Energy Transfer from Gain Media to Meta-Subunits” - Fourth International Congress on Advanced Electromagnetic Materials in Microwaves and Optics Karlsruhe(Germany), September 2010 (oral)

List of publications from CNR:


List of the conferences from UNIMAN:


List of publications from UNIMAN:


List of the conferences from Aalto:
„Dynamic extraction of effective material parameters of nanocomposites from reflection and transmission coefficients of a single grid”, D. K. Morits, C. R. Simovski, 43th Int. Conf. Days of Diffraction‘2010, St. Petersburg, 8-11 June 2010, oral
List of publications from Aalto:


List of the conferences from UCL:


List of the conferences from UNISI:

List of the publications from UNISI:

List of common publications
NANOGOLD PUBLICATIONS


"La nanotechnologie est-elle le premier pas vers l’invisibilité?", T.Scharf and J. Lenobel Zwahlen, Flash en ligne, 26-03-10, page 6 ; French only
Journals:

   „Oblique incidence ellipsometric characterization and the substrate dependence of visible frequency fishnet metamaterials”
   Optics Express (2012) (in press)

   „Spectroscopic ellipsometry of split ring resonators at infrared frequencies”

3. P. Tassin, T Koschny, M Kafesaki, C.M Soukoulis
   „A comparison of graphene, superconductors and metals as conductors for metamaterials and plasmonics”

4. N. H. Shen, T Koschny, M Kafesaki, C. M. Soukoulis
   „Optical metamaterials with different metals”

5. M. Losurdo, M Giangregorio, P Capezzuto, G Bruno
   „Graphene CVD growth on copper and nickel: role of hydrogen in kinetics and structure”

   „Characterization of plasmonic effects in thin films and metamaterials using spectroscopic ellipsometry”

7. M. Losurdo, M Giangregorio, P Capezzuto, G Bruno
   „Ellipsometry as a Real-Time Optical Tool for Monitoring and Understanding Graphene Growth on Metals”

   „Microfocus infrared ellipsometry characterization of air-exposed graphene flakes”

   „Single and multilayer metamaterials fabricated by nanoimprint lithography”

10. Milka Jakovljević, Borislav Vasić, Goran Isić, Radoš Gajić, Tom Oates, Karsten Hinrichs, Iris Bergmair and Kurt Hingerl
    „Oblique incidence reflectometry and spectroscopic ellipsometry of split-ring resonators in infrared”
    J. Nanophoton. 5, 051815 (Jul 01, 2011), doi:10.1117/1.3601359

    „Spectroscopic ellipsometry of few-layer graphene”
    J. Nanophoton. 5, 051809 (Jun 08, 2011); doi:10.1117/1.3598162
Conference contributions:


2. M. Kafesaki, "What are good conductors for metamaterials and Plasmonics?" – invited talk

Workshop: Novel Ideas in Optics (https://engineering.purdue.edu/~shalaev/workshop/)
3. M. Kafesaki, „What is the best conductor for metamaterials“ – talk

EIPBN 2012 (http://eipbn.org/eipbn-2012-conference-site/)

EMRS 2012 Spring Meeting (http://www.emrs-strasbourg.com/) 

META 2012 (http://metaconferences.org/oci/index.php/META/META12)

SPIE Photonics Europe 2012 (http://spie.org/x12290.xml)


Workshop at KIT 2012 (http://www.tkm.kit.edu/vortraege/workshop_woelfle_70.php)
10. C. M. Soukoulis, „Wave propagation: From electrons to photonic crystals and metamaterials“, Workshop: Electronic Correlations and Disorder in Quantum Matter; Dedicated to Peter Wölfle’s 70th Birthday – invited talk

APS March Meeting 2012 (http://wwwaps.org/meetings/march/)
11. P. Tassin, „Graphene, superconductors, and metals: What is a good conductor for metamaterials and plasmonics?“ – invited talk

EMLC2012 (www.EMLC2012.com)

IMNC2011 (http://imnc.jp)


16. C. M. Soukoulis, „Photonic Metamaterials: Challenges and Opportunities“ – oral presentation

Metamaterials2011 (http://congress2011.metamorphose-vi.or)g
17. Thomas Oates, Babak Dastmalchi, Kurt Hingerl, Iris Bergmair, Karsten Hinrichs, Michael Bergmair, Kurt Hingerl, “Spectroscopic ellipsometry of the fishnet metamaterial” – oral presentation


MNE 2011 (www.mne2011.org)


22. C. Helgert, K. Dietrich, D. Lehr, T. Kasebier, T. Pertsch, and E.-B. Kley, „A dedicated multilayer technology for the fabrication of three-dimensional metallic nanoparticles” – oral presentation

SPIE Optics+Photonics 2011 (http://spie.org/x57032.xml)


WavePro, Crete 2011 (http://cmp.physics.iastate.edu/wavepro/index.shtml)
25. P. Tassin, T. Koschny, M. Kafesaki, and C. M. Soukoulis, „What is a good conductor for metamaterials? A comparison between metals, graphene, and superconductors“ – invited talk

MediNano-3 (http://www.medinano3.ipb.ac.rs)

27. Milka Mirić, Borislav Vasić, Goran Isic, Rados Gajić, Tom Oates, Karsten Hinrichs, Iris Bergmair, Kurt Hingerl, „Analysis of the Ellipsometric Spectra of Split Ring Resonators“ – poster presentation


NNT2010 (www.nntconf.org)

MNE2010 (http://www.mne2010.org)


PECiX (http://www.pecis-ix.org)


34. I. Bergmair, Ahmad Saeed, Babak Dastmalchi, Günter Hesser, Thomas Pertsch, Holger Schmidt, Ernst-Bernhard Kley, Uwe Hübner, Raluca Penciu, Maria Kafesaki, Costas M. Soukoulis, Kurt Hingerl, Michael Mühlberger, Rainer Schöfner, “Fabrication and Characterisation of stacked NIM samples” – invited talk

Metamaterials2010 (http://congress2010.metamorphose-vi.org)


EIPBN2010 (http://eipbn.org)


ICSE 2010 (http://www.icse-v.org/web)


41. Karsten Hinrichs, Dennis Aulich, Simona Pop, Tom Oates, Michael Gensch, Arnulf Röseler, Rados Gajic, Goran Isic, Milka Miric, Raluca Penciu, Maria Kafesaki, Costas M. Soukoulis, Michael Bergmair, Kurt Hingerl, Iris Bergmair, “IR ellipsometry of split ring resonators” – poster presentation

42. Kurt Hingerl, “Photonics of two-dimensional metamaterials” – invited talk

Mautern2010 (http://www.gbpt.at)


44. Michael Bergmair, Peter Zeppenfeld, Iris Bergmair and Kurt Hingerl, “Investigation of Surface Plamon Excitations on Metallic Gratings” – poster presentation
