Publishable Summary for 17FUN01 BeCOMe
Light-matter interplay for optical metrology beyond the classical spatial resolution limits

Overview

The EU wants to hold a dominant global position in nanotechnology, micro-nanoelectronics, photonics and advanced materials: four of the six Key Enabling Technologies (KETs) identified by the European Commission. However, such dominant position strongly depends on the availability of proper tools that can serve the metrology needs of those KETs in terms of speed, non-invasiveness, reliability and integrability. This project addresses these needs by exploring novel metrology paradigms exploiting light-matter interplay, the topological information encoded in optical fields and the most recent accomplishments in the areas of quantum optics and inverse problems to achieve disruptive advances in optical metrology.

Need

The EU has formally identified six KETs, which have been given the highest priority within the EU strategic research agenda. KETs have a *"…significant impact on how Europeans will live and work, and on how European industries and economies will grow to provide sustainable employment for its citizens*". KETs represent the *technology building blocks* for advanced products and their manufacture, and they form the backbone of the European competitiveness on the global market. Research efforts meant to strengthen the position of Europe in these KET areas are of uppermost importance, since "*once the manufacturing base is lost, it never comes back*".

Metrology runs side by side with such scientific and technologic progress. Manipulating matter at the nanoscale, in a scientifically reliable and predictable way, urges the metrology community to provide the appropriate metrology solutions. Such novel or improved metrology solutions then, in turn, stimulate again the next technological and scientific developments.

In this context, optical measurement methods play an instrumental role as the development in four out of the six KETs (Nanotechnology, Micro-nanoelectronics, Photonics and Advanced materials) is strongly underpinned by optics-based measurement methods. The importance of holding a dominant scientific and technologic position in these sectors is clear if one considers that altogether they are worth more than € 800 billion on the global market and Photonics and Micro-nanoelectronics already employs more than 400,000 people in Europe.

Despite the many advantages of optical systems (speed, non-invasiveness, high-precision, moderate investments involved, integrability) the operational spatial resolution attainable in classical optical metrology is still essentially limited by the wavelength used for the optical probe. Therefore, novel and robust metrology solutions are needed that maintain all the recognised benefits of optical methods while substantially overcoming the current limitations.

Objectives

The overall goal of this project is to set the basis for the realisation of the next generation of optical metrology systems, with unprecedented performances in terms of spatial resolution, traceability, reliability, robustness. The specific objectives of the project are:

1. **To develop stable and reliable methods to achieve deep sub-wavelength spatial resolution by exploiting higher-order (beyond Born regime) probe-target interactions**. To design metamaterials-based structures that can enhance such interaction and bring it to detectable levels for a large class of targets, not only strong scatterers. The goal is to reach, for a well-defined class of samples (e.g. diffraction gratings on silicon substrates, isolated nanoparticles on both opaque and

transparent substrates), a traceable spatial resolution at the λ/10 level and sub-nanometre uncertainty, with λ being the wavelength of the light probe

2. **To exploit invariant topological structures in electromagnetic fields,** in their polarisation, amplitude and phase distributions**, and map how such topological information transforms after interacting with matter**, especially in the case of nanostructured materials endowed with specific geometric symmetries (e.g. diffraction gratings, spiral geometries, bio-inspired circularly-symmetric objects). The ultimate goal is to implement spectroscopy-like measurement concepts, leading to robust and high-precision dimensional and physical measurement results
3. **To realise and demonstrate near-field techniques to measure deep sub-wavelength gratings down to the regime << λ/10** which allow accurate and traceable optical procedures to characterise nanostructured optical components and to measure effective optical material parameters. In addition to link such near-fields methods to far-field optical methods of specific applied interest
4. **To apply sub-shot noise quantum technologies to optical systems, addressing both low and high Numerical Aperture (NA) systems**. To realise input fields with spatially-entangled optical channels and to map their coupling with the geometry of nano-targets. The potential of quantum metrology in optical systems will be explored through spatial modes entanglement and its integration into existing optical systems. The aim is to find a natural link with the exploitation of topologic information in classical fields, as discussed hereinabove
5. **To facilitate the take up of the technology developed in the project by the end users.**

Progress beyond the state of the art

Rayleigh's criterion for assessing the resolution limit of optical systems, no matter how complex and which wavelength λ they employ, states that the smallest detail that can be resolved in an object is of the order of 0.6 λ/NA, where NA is the numerical aperture of the system. For systems operating in air, NA is a positive number smaller than 1, which results in a resolution of about half of the wavelength. While different criteria can be adopted to quantify such limit, they all currently agree on the fact that the wavelength is the limiting factor.

This is a severe limitation for optical methods, especially in view of the growing impact of nanostructured devices and materials in current and expected technological developments in our society. For specific applications, smart alternatives have been found e.g. for the imaging of biological samples, they can be stained by fluorophores, and hence time-resolved fluorescent methods (such as Stimulated Emission Depletion (STED), Stochastic Optical Reconstruction Microscopy (STORM), Photo Activated Localisation Microscopy (PALM) etc.) have flourished in the last years, with proven resolutions at few tens of nanometres level [6-8]. Unfortunately, such techniques are of no use for inorganic materials in the semiconductor sector, where no natural fluorescent response exists, nor is a contamination with external markers allowed. Additionally, the quantification of the measurements results is complex and currently rarely addressed.

One way to approach this issue is by progressively moving to smaller wavelengths and/or is to make use of all the prior information available on the target. Going towards smaller wavelengths does not completely solve this problem because shorter wavelengths have short (around 100 nm) penetration depth. Furthermore, for some applications no prior information on the target is available.

This project is going beyond the state of the art by developing novel methods to improve the spatial resolution limit of far-field optical systems well beyond the classical resolution limit. Specifically, it has begun investigating ways to exploit higher-order interactions between the probe and sample of interest. This will imply a full implementation of multiple scattering models into imaging systems to account for strong spatial light-matter interaction regimes. For weak scatterers, where such interaction can be too feeble to be detected, amplification of sub-wavelength information, such as evanescent wave amplification, are being realised through the design of specially engineered nanodevices and structures. The targeted resolution, for systems working in far field illumination-far-field detection mode, will be one tenth of the wavelength, with targeted sub-nanometre uncertainty. The project is also investigating the existence of invariant modal information in vectorial optical fields and how that information transforms after interacting with matter. The goal is to identify robust information channels that can be used to establish a direct mapping between the light probe and scattered field and to setup the basis for the new field of spatial spectroscopy. The project also aims to link the performance of methods developed with the project to near-field techniques in use at NMIs, in order to validate the novel solutions investigated in the project. Furthermore, it is developing sub-shot noise quantum technologies, quantum-based metrology schemes and novel spatial entanglement-based measurement concepts to be implemented in existing metrology systems, such as optical scatterometers, atomic force microscopes (AFM) and optical ptychographic systems.

Results

To develop stable and reliable methods to achieve deep sub-wavelength spatial resolution by exploiting higher-order (beyond Born regime) probe-target interactions

This objective aims to find ways to solve direct strong scattering problems using perturbative methods. Perturbative methods are semi-analytical and can provide more insights on the physical mechanism behind a measurement result than a brute force numerical approach. It is however a known fact that for strong scatterers (strong scattering regime) perturbative methods lead to highly divergent solutions, something which hampers their practical usefulness. Providing a solution to this problem holds high theoretical and practical impact because direct scattering problems are instrumental to addressing and solving inverse scattering problems, which are the main target of the project. Partners from BeCOMe have been able to solve the problem of the divergence for wildly divergent Born series and proved the method for 1-D and 2-D scattering problems. This important result from BeCOMe has been recently published on a scientific journal (Phys Rev. Research **2**,013308, 2020), Partners from the consortium working on this topic have now moved to the application of this theory to the creation of novel inverse methods where more insights on the mechanism behind superresolution can be gained. Inverse electromagnetic problems have also been approached from a different viewpoint, namely by using Bayesian inversion methods. The potential of Bayesian inversion has been proven by benchmarking its performances with respect to other commonly used methods.

Another output has been the design and the fabrication of metamaterials-based superlenses, which are meant to boost the spatial resolution of existing optical systems, such as optical scatterometers, in the visible range of the electromagnetic spectrum. Some of the devices designed have been fabricated and tested on direct bright field imaging. The impact of the different metamaterials-based superlenses on the quality and resolution of the images is clearly visible from the first tests performed and currently further investigations are on-going to assess the actual performances of these resolution enhancers. More on this topic will be reported in future updates on the project outputs.

To exploit invariant topological structures in electromagnetic fields and map how such topological information transforms after interacting with matter

The consortium has been working since the beginning of the project on schemes to probe specific spatial features of objects making use of use of a spatial modes decompositin of a light field. The goal is to use this scheme as a general procedure to be applied to a wide class of objects of interest. The approah chosen uses results from quantum Fisher information theory to evaluate the maximum sensitivity one can obtain in a given experiment. The approach uses a formalism developed within quantum estimation theory and it is mainly aimed at understanding what the fundamental limitations are on the amount of information that can be extracted from an optical wavefield. The work done in BeCOMe highlights the impact that the evaluation of the quantum Fisher information of particular measurement schemes might have on the limitations and possibilities of optical metrology. A consequence of this approach is the identification of an ideal measurement setup and procedure that consists of projecting the wavefield reflected or transmitted by the object in the spatial modes selected previously. The experimental demonstration that makes use of these ideas is curently being implemented.

In regards to the mapping of the spatial degrees of freedom carried by an optical field, the research is progressing according to two different paths (i) theoretical/numerical studies are currently being performed to estimate the effect of sub-wavelength structures (such as diffraction gratings) on the spatial channels used to describe the field. This is addressed by the partners involved using complementary approaches. For instance, HNMs decomposition, decomposition in spatial modes containing orbital angular momentum and spatial modes with radial or azimuthal polarization, use of quantum Fisher information to estimate resolution limits. (ii) several groups of the consortium are now implementing experimental setups based on different imaging methods to tailor and detect the amplitude and phase of the wavefields and measure how they transform after interaction with some objects of reference.

<u>To realise and demonstrate near-field techniques to measure deep sub-wavelength gratings down to the regime << λ/10</u>

First calculations of the near field of a grating SIL (Solid Immersion Lens) or photonic nanojet lens system have been performed as well as a program has been realised to calculate the photonic nanojet field after a ball lens using Mie theory. The numerical simulations are currently extended to circularly polarized light. Additionally, a numerical method for rather substantial control of the position of a photonic nanojet has been developed and successfully tested. The method currently works in the 2D setting and has recently successfully generalised also to 3D. A publication has been on this part of the work is under preparation.

Significant progress has been achieved in setting up a novel spectroscopic SNOM tool. New SNOM probes have been fabricated by focused ion beam and have been characterised and successfully tested in measurements. A spectroscopic set-up has been realised and tested. First test measurements have shown unexpected discrepancies between measured and expected intensity distribution in the reflected spectra. Rigorous FDTD simulations are currently performed to clarify the observed discrepancies.

Investigations on Structured Illumination Microscopy are currently being performed, including NSIM, PSIM Design and FEM simulations of metallic plasmonic lens structures. The original design of the plasmonic lens showed severe manufacturability issues and an alternative design, an inverted lens design, has been discussed and developed. The optimisation of the inverted design has been performed and successfully finished using a particle swarm algorithm. The resulting inverted design was discussed and investigated, and the results showed a good compromise between performance and manufacturability. A corresponding lens has been designed (the results of this research are now available at Meas. Sci. Technol. **31** (2020), 1-10), Investigations on the optical performances of this lens are ongoing.

Further to this, an enhanced sensitivity in scatterometry by analysing structure-induced resonances was performed, experimentally demonstrated. The result of this work is now published (Opt.Express **28** (2020), 23122.).

A systematic numerical study based on rigorous RCWA modelling has been performed to investigate the parameter dependencies of localised surface plasmon resonances (LSPR) in silicon gratings observed in the DUV (l = 266.3 nm). The results demonstrate not only a significantly enhanced sensitivity of structure width and hight of < 0,2 nm and < 0,5 nm, respectively, for goniometric DUV scatterometry, but also a measurement capability with a sensitivity of about 1 nm for pitch measurements for sub-diffraction periods between 50 nm and 250 nm, which are not accessible without exploiting the LSPR.

<u>*To apply sub-shot noise quantum technologies to optical systems, addressing both low and high NA systems*</u>

Regarding the low-noise imaging schemes, a new protocol has been demonstrated experimentally for improving resolution-sensitivity trade off in sub-shot noise imaging (SSNI) of weak transmitting masks, leading to the publication (**Appl. Phys. Lett. 116, 214001 (2020)**). This will help get better results also in SSNI of phase object trough small intensity pattern perturbation detection (that we call shadow imaging).

The consortium has identified a scheme that is suitable for phase retrieval of an unknown object, using spatially multimode entangled light sources (Noise Reduction Factor (NRF) < 0.3) that are produced by BeCOMe partners. Fruitful collaborations among partners from different countries have started to combine expertise on SSNI and classical phase retrieval.

Important progress has been made on the realisation of new sources of squeezed light and their application. More specifically, quantum enhanced stimulated Raman imaging results has been now published (**Optica 7 (5), 470-475 (2020)**)**.** The consortium is currently building a new OPO system for generating squeezing in high-order modes.

Impact

In October 2019 (21-23 October), within the framework of the Face2Phase conference BeCOMe organised a workshop focused on Phase retrieval and its applications to metrology. Four lectures were provided by recognised experts in the field. It was an extremely succefull event, with more than thirty participants who received an attendance diploma. More information is available here.

On 29 May 2019, a special section dedicated to this project and entitled " *BeCOMe - Current and future trends in Quantum Optics-based measurements methods*" was organised within the workshop "*From Foundations of*

Quantum Mechanics to Quantum Information and Quantum Metrology & Sensing" (Quantum2019), held from 26 May to 1 June 2019 at the University of Torino (Italy), with more than 250 attendees.

In addition, in January 2019, two press releases addressed to the general public were issued by the University of Torino, on the "UNITONews" (link) and "Frida" portals (link), respectively. These press releases reported the first achievements of the project on a new class of single photon emitters, with potential applications to high-resolution imaging.

In 2019 members of the consortium organised specific conferences dedicated to Optical Metrology and Nanometrology, the SPIE conference Modelling Aspects in Optical Metrology (June 2019, Munich, Germany, partly combined with CLEO/EQEC) and the Nanoscale (October 2019, PTB Braunschweig, Germany), which used to disseminate the project's results via several presentations.

The consortium has organised a school of physics on optical metrology within the framework of the European Optical Society (EOS) Annual Meeting (EOSAM2020, 07-11 September 2020). Although originally planned in Porto, Portugal, due to the COVID-19 outbreak that event has been turned into a Virtual event. The focus of the school, open to professional, students and young scientists, was on innovation and on how fundamental research on metrology can underpin the scientific and industrial role of Europe in optical metrology. More information can be found here. The School was very successul and attracted attendees from all other the world.

Impact on industrial and other user communities

This project performs innovative research that impacts such high-intensity technologies such as software, biotechnology and computer electronics. Additionally, the project targets optical measurement instruments and methods which are relevant for these applications, such as optical scatterometers and high-contrast phase sensitive systems. The adoption of the project's methods in instruments relevant to the end-users, will also support their uptake in such technology areas.

Engagement with industry is considered very important by the consortium, irrespective of the fact that the project deals with fundamental metrology. Members from the consortium have strong links with leading companies in the semiconductors and inspection sectors. At the last two progress meetings representative from high-tech industry has joined the meeting in order to provide feedback to the consortium. During the first 27 month of the project, the consortium of BeCOMe has engaged with representatives from key high-tech companies operating on metrology for the semiconductor and nanodimensional metrology sector. This interaction is leading to new forms of partnerships and works as spin-off of new cooperation.

Impact on the metrology and scientific communities

This project will provide new solutions for optics-based dimensional metrology by demonstrating the possibility of integrating modern field theories and material science into classical measurement schemes, in order to progress beyond the current-state-of-the-art. New CMCs, regarding higher spatial resolutions and determination of materials properties with smaller uncertainties can be foreseen as future outputs of the project, as well as inputs to the EURAMET Technical Committee Length (TC-L). More specifically, metrology systems, such as optical scatterometers, bright-field microscopes and similar contact-less metrology tools, will be able to extend their spatial resolution by integrating methods, and devices, developed within the project. This will range from the implementation of new super-resolution models to classical experimental data to the integration of metamaterials-based superlenses (resolution enhancer devices), into existing systems. In this way, end-users from the metrology and the scientific community will learn how to boost the performances of tools already at their disposal without resorting to major investment and technology shifts. One distinctive feature of most of optics-based metrology systems is their reliance on physical models for the extraction of desired information on a measurement target from acquired data. This often means rigorously solving Maxwell equations, using subtle inversion models and regularisation methods. Spatial spectroscopy, as addressed by objectives 3 and 4 of the project, would directly map the measured data to the measurand in a more straightforward way. This will facilitate the adoption of optical measurements techniques by the scientific communities by lowering the threshold represented by the development of complex data post-processing and analysis. Finally, the integration of quantum-enhanced methods, such as spatial modes entanglements and sub-shot noise shadow imaging, into optical systems of industrial and metrological interest will further promote the integration of quantum technologies and classical optical systems and will represent an excellent showcase for long-term uptakes, especially in the field of non-invasive optical metrology, an area of research currently challenged by the metrology targets set by the needs of industry. To support this, the consortium has welcomed a new collaborator fom University of Purdue (USA) who will contribute to the integration of quantum theories into optical metrology.

The impact on metrology is also related to the effort towards the definition of standards for measurement of new quantities related to the quantum phenomena as, for example, a first informal comparison of the g(2) measurement in the visible range [Metrologia 56, 015016 (2019)]. This is related, as a by-product, to the activity on quantum-based superresolution imaging, which has received a “Research Highlight” in the Journal “Nature Physics“, (https://www.nature.com/articles/s41567-019-0432-9).

The dissemination of the first outputs of the project has been also done via the “QUILT Autumn School 2018 - *Quantum-Enhanced Imaging and Spectroscopy*” organised by the Fraunhofer Gesellschaft (IOF/IPM) in Bad Honnef, Germany. The School was aimed at providing participants with first-hand information on current trends in quantum imaging and to discuss fundamental and applied aspects in the area of research. The audience consisted of about 70 attendees, mainly PhD students but also experts and scientists working on different field of classical and quantum physics. Lectures and speakers were renowned experts working on quantum imaging.

Impact on relevant standards

The research planned in the project is fundamental by nature, hence early impact on standards is not foreseen. Project results will however be disseminated within EURAMET TC-Length (TC-L), the BIPM Consultative Committee for Length (CCL) Working Group on Dimensional Nanometrology (CCL-WG-N) and Versailles Project on Advanced Materials and Standards (VAMAS) Technical Work Area (TWA) 42 on Raman and microscopy.The work done within BeCOMe on the measurements artefacts, for assessment of the spatial resolution of optical systems, was also presented to ISO/TC 213 “Dimensional and geometrical product specifications and verification” at a working group meeting in October 2019.

Longer-term economic, social and environmental impacts

According to the European commission the expected impact of advances on nanotechnology are, amongst others, “*supporting European competitiveness through accelerated market uptake of nano-enabled products, improving in existing manufacturing processes and industrial productivity, contribution to improved technical knowledge, promoting safe-by-design approaches and contributing towards the framework of EU nanosafety and regulatory strategies (including standardisation)*“. This is particularly true for Europe's photonics industry, which is strong (e.g. in laser-based manufacturing, medical photonics, sensing, lighting, high-end fabrication of optical components) and has the possibility to exploit new emerging market opportunities, particularly by exploiting the novel opportunities provided by functional nano-optical materials.

This project will encourage collaborative scientific excellence in materials science, classical and quantum optics by stimulating the interaction of specialists in optical systems and metrology, experts in materials fabrication and metamaterial engineering. The design of fit-to-purpose resolution enhancers (such as metamaterials-based superlenses, addressed in objective 1 of the project), requires knowing the main features of optical and imaging systems, the limitations of material production and having available innovative design concepts and models for such spatial resolution enhancers that can work on a broad range of wavelengths.

The scientific communities working on classical and quantum optics have had little interaction with each other in the past, which is desirable and often counterproductive. Scientists from classical optics have always worked in close contact with industry and addressed issues as they emerge from the applications. Quantum optics communities, especially in the area of dimensional metrology, have focused on key aspects of quantum physics, such as entanglement. The uptake of such fundamental research is however encountering some resistance from an end-user viewpoint, due to the objective complexity of its implementation and, sometimes, the arguable added value offered. This project represents a unique environment where both communities can unite their forces and ideas to advance the area of optical metrology as whole. This will, for the first time realise the quantum version of systems, such as optical scatterometers, with the application of genuine quantum effects, such as weak amplification and Hong-Ou-Mandel interferometry, directly into dimensional metrology.

List of publications

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Internal Funded Partners: 1 VSL, Netherlands 2 CMI, Czech Republic 3 DFM, Denmark 4 INRIM, Italy 5 NPL, United Kingdom 6 PTB, Germany	External Funded Partners: 7 Aalto, Finland 8 FSU Jena, Germany 9 ICFO, Spain 10 SwanU, United Kingdom 11 TU Delft, Netherlands 12 TUBS, Germany 13 UNITO, Italy 14 ZIB, Germany	Unfunded Partners: 15 DTU, Denmark
RMG1: DFM, Denmark (Employing organisation); VSL, Netherlands (Guestworking organisation)		