Publishable Summary for 20IND04 ATMOC

Traceable metrology of soft X-ray to IR optical constants and nanofilms for advanced manufacturing

Overview

The optics and semiconductor industries have been using innovative materials and complex nanostructures whose optical properties are difficult to measure and often not accurately known. This project will develop advanced mathematical methods to traceably characterise these materials for wavelength ranges, from soft X-ray to IR. This will be achieved by creating a database of optical constants with associated uncertainties for bulk materials and ultra-thin film systems and industrially relevant datasets. This database will provide the opportunity to relevant industries to run simulations and eventually develop new materials with tailored properties.

Need

Innovations based on novel materials and devices in photonics and semiconductor manufacturing play a key role in addressing the grand challenges ranging from the European digitalisation strategy to the European Green Deal. For the development of novel technologies and high-precision manufacturing techniques in semiconductor and optics industries and in nanotechnologies, precise knowledge of the optical properties of these materials is vital, providing the foundation for novel nanoelectronic devices, high-quality sensors or effective photovoltaic elements.

Next generation materials with tailored and optimised properties in combination with progressing miniaturisation in named nanotechnologies need accurate characterisation of optical properties. Shrinking structure dimensions and the demands on increased functionalities are challenging. Established photonic measurement methods in the soft X-ray to IR wavelength range like scatterometry, Mueller ellipsometry and reflectometry, need to further be developed. Without traceability, rigorous uncertainty estimations, harmonised modelling and data analysis approaches, future technological developments will be difficult to maintain.

Industry and research require reliable optical constant databases (objective 4) to assist photonic industry developments of innovative and tailored next generation materials. For this purpose, publicly available databases have been established for optical materials across different wavelength ranges. However, the underlying datasets are of poor quality, relying on calculated and estimated values without reliable uncertainty estimations.

Three components are required to build a database of high quality that meets the needs of the industry:

i) The test and reference samples to be characterised are of relevance for industry
ii) The measurements must be reliable and of high quality with an uncertainty budget
iii) For the indirect measurements, algorithms must be developed that reliably determine the uncertainties

For optical metrology, this mission represents a major challenge that cannot be met by one NMI alone but requires collaborative efforts of several NMIs and research institutes. For wide wavelength range, from soft X-rays to IR, physical properties of samples vary considerably. As a result, the models for analysing measurement data, uncertainty estimations and the sensitivities of the instruments are very different and therefore need to be compared, combined and further developed.

Objectives

The overall objective of the project is to develop traceable measurement techniques for optical constants of thin-film systems and nanostructures and to use these techniques to support the introduction of an improved optical properties database for industrial users. The specific objectives are:

1. To identify, select and produce test samples including ultra-thin layer systems, complex nanostructures (e.g. PillarHalls) and novel materials (high k-materials and other materials, e.g. 2D nanosheets). In addition, to provide metrologically characterised optical response data for use in the existing databases and models that are used by industry.
2. To develop reflectometry, Mueller ellipsometry and scatterometry as reliable and traceable thin-film metrology techniques in the soft X-ray to IR spectral range, for determining the layer thickness, optical properties (such as the refractive index, absorption coefficient, reflectivity) and dielectric tensors of the test samples developed in objective 1. This should include the development of reference materials that are suitable for industrial use.
3. To develop and apply advanced mathematical models for virtual and real measurements in order to determine the optical response of the test samples developed in objective 1 and their dependence on complex nanostructures. The uncertainties associated with ab initio methods, interlayer roughness, crystal structures, model reduction techniques, surrogate modelling, machine learning and inverse modelling should also be determined.
4. To determine the optical constants and the corresponding measurement uncertainties of thin stratified layer systems and to estimate the geometrical parameters of these complex nanostructures in the soft X-ray to IR spectral range. In addition, to assemble a database of optical constants, dielectric tensors and estimated geometrical parameters, including both measurement values and virtual/simulated measurement data.
5. To facilitate the take up of the technology and measurement infrastructure developed in the project by the measurement supply chain (photonics industry), standards developing organisations (DIN, ISO) and end users (photovoltaics, advanced manufacturing and healthcare sectors).

Progress beyond the state of the art

Accompanied by leading stakeholders, the project has identifed, manufactured and characterised key structures (test and reference samples) to support the development of next generation layer systems and nanostructures, including novel materials such as high-k or 2D materials with high innovation potential.

The consortium carefully selected test and reference samples of ultra-thin layer systems, complex nanostructures, and novel materials. This includes ultra-thin layer systems of various thicknesses, nanostructures (line gratings, pillar gratings, pillarHalls), and fabrication procedures like PVD, PLD, Epitaxy, and Spin coating. The materials and geometries used in these samples are diverse, covering a broad spectrum of parameters.

Pre-characterization of most samples was completed using techniques like SEM, XRR, or ellipsometry measurements (Objective 1).

While reflectometry, conventional or Mueller ellipsometry and scatterometry are widely used for model-aided optical measurements, their traceability and uncertainty estimation are still a subject of current research. In this context, SI traceable measurements are very challenging due to strong parameter correlations, model approximations and setting up a fully documented uncertainty budget. Only few studies and for specific samples have been performed until now. This JRP will derive complete uncertainty budgets for all these methods and different tools for various case studies pushing forward the development of traceable measurement that involves inverse modelling. The case studies will serve as templates for different future areas of application in the industry. For that purpose several instruments used for scatterometry, Mueller ellipsometry and reflectometry measurements were characterised and calibrated (Objective 2).

Indirect measurement techniques, such as Mueller ellipsometry and scatterometry, require a solution of the inverse problem in order to determine the measurand. While for direct measurements, the determination of the measurement uncertainties is clearly covered by the GUM, for indirect measurements it is not, requiring substantial modelling. In particular, the role of stochastic parameters for the sample and the measurement system in context, with the coherence properties of light and the resulting degree of depolarisation, has hardly been investigated. Significant progress has been made in the understanding of depolarization and decoherence in Mueller ellipsometry by developing a phenomenological model (paper was submitted).

Further development and harmonisation of different inverse model approaches and uncertainty estimations will be addressed by the project.This will include determination of uncertainties, when novel modelling techniques are used for data analysis, interlayer roughness, surrogate modelling, and machine learning.

Progress has been made in the performance of optimization algorithms. The Bayesian optimization method has been extended to optimize target vectors, which is very useful for high-dimensional problems such as those encountered in the determination of optical constants. In addition, a polynomial chaos-based surrogate model for computationally intensive models (e.g., column lattices) was implemented to enable the Bayesian approach. (Objective 3).

Innovations in nanotechnologies involving new materials and architectures require reliable material parameters. Although several databases exist with these parameters, there is inconsistency between them, especially when measuring specific wavelength ranges or when displaying references without uncertainties. As a result, it is difficult to assess the quality of these data. Moreover, metadata such as environmental variables, calibration procedures, instrument parameters or model algorithms and parameters are often missing. The project will address this gap by setting up a reliable database for optical constants that will include uncertainties, detailed measurement, and modelling information. In addition, due to miniaturisation and novel materials (such as 2D or high-k) the conceptual validity of these parameters is under scrutiny since they do not address size dependencies, quantum effects or anisotropy issues (e.g. for 2D materials). The project will go beyond the state-of-the-art by developing measurement modalities and inverse modelling. (Objective 4)

Results

Selection, production and characterisation of test and reference samples (Objective 1)

The consortium has discussed and selected test and reference samples of ultra-thin layer systems, complex nanostructures and novel materials that are of interest for stakeholders (collaborating high-level stakeholders were actively involved).

Ultra-thin layer systems consisting of different layer thicknesses, material compositions and fabrication procedures (e.g. PVD, PLD, Epitaxy, Spin coating) have been selected for two case studies (case study 1: ultrythin layer systems and case study 3: layer systems produced with different manufacturing techniques). Complex nanostructures such as line and pillar gratings or pillarHall structures relevant for semiconductor industry were defined. Finally, novel materials comprising 2D nano-sheets, high-k-layer systems and thin bilayers were selected. All test and reference samples have been specified and described for the manufacturing process.

All test and reference samples for the project were manufactured and all were shipped to the partners for measurements:

- Case study 1: more than 20 samples of one-material layer systems with varying layer thickness (30nm – 3 μm) and material parameters (e.g. Si, Mo, Ru, Al, Ni, Mo, Nb, Pt, Ta, Cr, TiN, SiO_2, HfO_2, Al_2O_3) were produced and shipped to the partners.
- Case study 2: more than 20 samples (on 5 batches) with laminar grating and 5 samples (one batch) with 2D nano-pillar structures were produced and shipped to the partners. Variations includes changes in geometry (linewidth, periodicity, line height, diameter) and material parameters (e.g. Si, SiO_2).
- Case study 3: more than 8 samples of layer systems with different manufacturing techniques were produced and shipped to the partners (e.g. PVD, PECVD, ALD).
- Novel materials: more than 15 samples of novel systems were produced and shipped to the partners (e.g. coated and uncoated PillarHalls, High-k layers with varying stoichiometry, bi-layers).

The samples were pre-characterized by SEM, XRR or ellipsometry measurements and the results are summarized in a report. Objective 1 has been achieved.

Development of traceable reflectometry, Mueller ellipsometry and scatterometry (Objective 2)

For optical measurement modalities, i.e. reflectometry, scatterometry, Mueller ellipsometry, characterisation methods are reported. The methods include different instruments ranging from Soft X-ray beamline, VUV beamline, EUV scatterometer, coherent Fourier scatterometer to Mueller-ellipsometer.

In particular, the following instruments were characterised: soft-X ray reflectometer (synchrotron, BESSY), soft-X ray scatterometer (synchrotron, BESSY), IR reflectometer (synchrotron, BESSY), EUV reflectometer (MLS), VUV ellipsometer (MLS), DUV scatterometer, Mueller ellipsometer (DUV- NIR), Spectroscopic Mueller ellipsometer, Empyrean X-ray reflectometer, Coherent Fourier scatterometer, Stand-alone EUV spectrometer, DUV spectrometer. The characterisation results will be summarized in a review article.

To develop reflectometry, Mueller ellipsometry and scatterometry as reliable and traceable thin-film metrology techniques in the soft X-ray to IR spectral range, for determining the layer thickness, optical properties, uncertainty budgets are necessary to establish for each measurement methodology. Uncertainty budgets for each measurement modality was set up and measurements are performed. The first comparative measurements were successfully carried out.

Advanced inverse modelling and virtual measurements (Objective 3)

Existing forward and inverse models, optimisation methods and approaches to determine measurement uncertainties were explored and summarised using results from the survey. Depending on the measurement modality (scatterometry, reflectometry, Mueller ellipsometry), the following forward models are identified:

- Transfer matrix method for layered systems
- Finite Element Method (FEM) and rigorous-coupled wave analysis (RCWA) for nanostructures.

For the (statistical) inverse problem the following approaches have been selected (sorted by simple non-accurate uncertainties to complex reliable uncertainties):

- (weighted) Least squares optimisation, uncertainties are determined by the covariance matrix
- maximum likelihood estimation, uncertainties are determined by the Fisher information matrix method
- maximum posterior estimation, uncertainties are determined by the Fisher information matrix method
- Bayesian inference, uncertainties are given by the posterior distribution.

Identified models (RCWA, Transfermatrix, FEM) were implemented for reference and test samples (for case studies 1-3 and for novel materials). Novel advanced methods (like Polynomial Chaos, Bayesian target optimization) for Bayesian inference were applied and tested. A method has been developed that enables the adaptive determination of model errors in Bayesian inversion.

A novel phenomenological model for treating depolarisation and decoherence in Mueller ellipsometry was developed and a procedure for experimental validation was proposed.

The effect of surface roughness onto reconstruction results was investigated and results are published in a scientific paper.

A first version of the open software tool PyThia, which enables Bayesian inference for computationally expensive models (e.g., FEM, RWCA forward models) including the polynomial chaos-based method, has been released. The open source software tool PyThia can be downloaded[1]. A scientific article on the software tool has been published.

Determination of optical response functions and assembling a database (Objective 4)

A measurement plan that allocates test and reference samples has been selected and produced in objective 1 and measurement and modelling activities were carried out. The measurement plan includes test and reference samples, production times, measurement times, simulation times and shipping addresses. This measurement plan was complemented by a tracking of shipped samples and regularly updated.

For the database of optical constants, a draft on data formats and metadata was compiled and first datasets are shared among the partners. A first dataset on EUV optical constants was published at Zenodo. A study was started to investigate the effect of prior knowledge of optical constants on the reconstruction results of nanostructures.

Impact

[1] https://gitlab1.ptb.de/pythia/pythia

The project partners have provided presentation and contributions to standardisation bodies such as VDI/VDE-GMA, DIN NA 062-01-61 AA, ISO/TC 107 JWG 4, IMEKO TC10 and METSTA SR229. Several stakeholders were contacted and a stakeholder committee with 10 members was established.

The project partners have given more than 45 presentations at various national and international conferences among them the 9th int. Conference on Spectroscopic Ellipsometry, TBO Nanophotonics, MATHMET2022, Internat. Conf. of Quantum, Nonlinear and Nanophotonics' 2022, EOSAM, QUNOM 23, IMEKO TC10, Congress of ICO and AES2022.

Individual partners of the consortium offered training on measurement methodologies to other partners. The open-source software tool PyThia was launched and can be downloaded[2] and a training course was provided.

Impact on industrial and other user committees

The project will engage with many participants, key stakeholders, and leading experts in order to develop a database with tailored optical materials and improved semiconductor manufacturing processes. Furthermore, extensive datasets from the planned three case studies will be provided.

- The first case study will deal with onset of quantum effects and anisotropies for decreasing layer thicknesses.
- The second case study will examine the influence of the measurement accuracy of optical constants on the reconstruction results of nanostructures.
- The third case study will explore the dependence of optical constants for ultra-thin single layers and layer systems.

The database for optical constants and the provision of datasets from the case studies will enable end users to access reliable optical constants with their uncertainties, which are necessary for the development of modern functional optical materials.

The guideline on traceable reflectometry, Mueller ellipsometry and scatterometry, as reliable and traceable thin-film metrology technique, that would be developed in the project, would also help the end-users to improve their standard measurement techniques even further, and even pave the way for innovative optical coatings, such as in lithography masks.

Impact on the metrology and scientific committees

Measurement uncertainty, reliability and traceability are core issues in metrology. Indirect methods which require advanced modelling to solve an inverse problem are not yet standardised, which is a prominent issue for the determination of optical constants. The project will meet this challenge by establishing reliable uncertainties and traceability for reflectometry, Mueller ellipsometry and scatterometry. For this purpose, a complete uncertainty budget for the measurement methods will be set up and specific error models will be developed to deal with errors in modelling. Modern statistical methods such as Bayesian optimisation will be utilised thus extending the usability of the results beyond optical metrology.

Impact on relevant standards

Partners will contribute to national and international standards and guidelines throughout the project, especially for nanometrology and optical materials characterisation. This will include dissemination of the projects' results to standard committees to propagate the results and make them available to the user community. Special attention will be given to the standardisation of spectral ellipsometry as a generally applicable tool, an effort which is already ongoing. The project anticipates a high impact of the mathematical tools and advanced uncertainty evaluation methods through the international committee IMEKO TC 21.

Longer-term economic, social and environmental impacts

Optical technology has an increasing impact to all areas of our lives and plays a key role for future markets such as the Internet of Things (IOT), cyber-physical systems, advanced manufacturing and health care. For photonics, the global market has reached a volume of more than €600 billion. The European photonics industry includes 5000 companies with more than 300.000 highly skilled jobs with an annual turnover of €60 billion. A database of optical materials, as projected in the project, is therefore at the heart of new developments in the field. New and enhanced materials with designed optical properties will enable the full development of photonics technologies across many sectors and will play an essential role in the European digital transformation.

Longer-term, results from the project will have a social impact in two ways. Firstly, the European social community will be influenced by the ongoing digitalisation and will benefit from the fact that essential parts of this transformation will be developed within Europe. Since hardware development needs high precision manufacturing, further development of metrology in this field will be inevitable. Secondly, the build-up of innovative technologies and know-how in Europe strengthens its future position worldwide. In that respect, laying out foundations for upcoming industrial processes by consolidating its pioneering role in optical technology will positively impact Europe's job market in the future.

Through the field of photovoltaics, the optics industry is directly involved in the task of decarbonisation, which is a high priority worldwide and in Europe. Development of novel solar cell technology is currently driven by new materials, surface as well as interface design, which indirectly or directly all rely on optical metrology. Apart from power generation, the development of novel materials and technologies based on tailored materials' properties will improve sustainability, efficiency and reduce energy consumption. Battery technology and water splitting are further applications with highly specific needs to surface structuring and the implementation of novel material systems. In the effort to reduce the consumption of resources, the design of products and machines is moving away from the bulk phase to the surface. The capability of surface design and characterisation will therefore be critically necessary for almost all industries within the European Economic Area in future to reach environmental goals.

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This list is also available here: https://www.euramet.org/repository/research-publications-repository-link/

Project start date and duration: 01 July 2021	36 months	
Coordinator: Sebastian Heidenreich, PTB Tel: +49 (0)30 3481 7726 E-mail: sebastian.heidenreich@ptb.de Project website address: https://www.atmoc.ptb.de/home/		
Internal Funded Partners: 1. PTB, Germany 2. Aalto, Finland 3. BAM, Germany 4. DFM, Denmark 5. TUBITAK, Turkey 6. VSL, Netherlands 7. VTT, Finland	External Funded Partners: 8. CEA, France 9. CNRS, France 10. FSU Jena, Germany 11. FVB, Germany 12. IMEC, Belgium 13. JCM, Germany 14. JKU, Austria 15. TU Delft, Netherlands 16. TUB, Germany 17. UTwente, Netherlands	Unfunded Partners: 18. RWTH, Germany 19. CZ Smith, Germany
Linked Third Parties: 20. EP, France (linked to CNRS)		