Centre of excellence

“Mesosystems - Theory and Applications”

Contents

Rationale for the Centre

The current research of materials and processes delves progressively into micro- and nano- (on spatial scale) and to femtolevel (in time-domain). Mesoscopic systems and phenomena lie in between the macro world and atomic dimensions. Such spatial scale is a challenge for the current theories as both, the ones developed to describe macroscopic bodies as well as the theories for isolated atoms and molecules, fail. On the other hand, the meso-range of dimensions and processes hides potential for numerous innovative approaches for sensorics and informatics, and is also pertinent when talking of living systems (covering loosely the size range between viruses and bacteria). Hence, there is a need for new theoretical approaches and closely related experimental studies in order to "bridge" the gap between the atomic and macro dimensions. The distinguishing feature of the proposed research centre is the unification of theoretical and experimental groups in solving one common task: **How do atomic and macroscopic processes and properties interconnect on mesolevel?**

The goal of the CoE is to generate new ideas and principles for applications for the times the existing technical solutions are pushed to their limits already and the nowadays applied research has exhausted its ideological basis. These applications pertain to quantum information processing and communication, nanofabrication, medical molecular diagnostics, environmental monitoring, green energy.

The research is focused on three closely interrelated topics

T1. Coherence

The very essence of mesophysics is the extension of quantum coherence from atomic to macroscopic dimensions. Phenomena like superconductivity and superfluidity (Bose-Einstein condensation in general), ferromagnetic and ferroelectric ordering, and plasmonic resonances are important examples of such effects, all to be studied by the CoE. It is also important to know how these phenomena can be „squeezed down" to lower dimensions and confined geometries (e.g., „downscaling" of optical coherence into nanodimensions via plasmonic effects).

T2. Dynamics

While linear dynamical approaches (T1) are to a certain extent applicable to systems with a fixed structure, they (i) do not account for all phenomena in such systems and (ii) they fail to describe structural changes (formation of structures) in mesosystems. As a solution, nonlinear models of energy localization and defect formation in microsolids will be developed and tested in experiments. Transport processes in mesostructured systems are important both for their formation (synthesis, e.g. in sol-gel technology) as well as for functioning (e.g., in living cells) of such structures. An important problem to be addressed here, is how from uncorrelated molecular events a directed transport arises („molecular ratchet").

T3. Structures

Conquering the mesodimension necessarily presumes mastering of the methods of fabrication and understanding of the structure formation in mesosystems. This is a prerequisite for the studies of both mesoscopic coherence (T1) and dynamics (T2). Results on nonlinear dynamics (T2) are closely related and give an essential support to this topic. The CoE addresses phenomena like defect formation, „stripe structures" and incommensurate modulation, cracking of sol-gel films, molecular recognition in living cells. A particular emphasis is put on low-dimensional (2D, 1D, "0D") systems, as far as (i) the dimension modifies the physical properties in the most drastic way and (ii) the novel methods of material preparation have made the fabrication of such systems viable.

These problems will be addressed by **theoretical modeling**, **numerical simulations** and **various experimental techniques**. The latter involves different spectroscopic (absorbtion, fluorescence, Raman), microscopic (optical, electron, SPM) and combined (micro-Raman) methods. Existing international collaborations extend the scope of available methods to synchrotron and neutron scattering.

The systems to be investigated within the aforementioned studies include metal-oxide sol-gel systems of various shapes (films, fibers, micro- and nanotubes, nanopowders) activated with metal nanoparticles and semiconductor nanodots; plasmonic metal mesostructures; carbon-based structures (CNT and graphene + adsorbates); droplets of quantum liquids ^{4}He & ^{3}He; superconductors including HTSC; frustrated quasi-2D magnetics; biological cell membranes and receptors, microtubules (cytoscelecton). Methods such as sol-gel synthesis, laser ablation, pyrolysis, ion-beam, and colloidal lithography will be used in sample preparation.

Scientific novelty of proposed research

Plasmonic structures and processes

One of the heuristic sources of the proposed research is the conversion of "classical" optical phenomena into plasmonics. The CoE seeks to develop plasmonic metamaterials, "plasmonic crystals" (as analogues of photonic crystals) and study "plasmonic invisibility". Proceeding from theoretical models and simulations, plasmonic mesostructures will be fabricated, characterized and optimized. In particular, we expect to catch interaction of plasmons with biphotonic states on single-photon level. Generation, propagation and detection of coherent excitations (plasmons) in mesosystems of different dimensionality will be studied with an expected contribution to sensitive monitoring of molecular processes *in vivo* and in model biosystems. The investigation of plasmon-coupled emission will result in the development of highly efficient fluorescent materials, optical amplifiers and active optical waveguides, while other research results can be applied in the (nano)sensorics and for harvesting solar energy. It's our vision that these results will facilitate the birth of a new generation of (plasmonic) devices when the existing technologies get exhausted.

Mesoscale coherence and orderings

Mesoscale ordering governs a number of superconducting, magnetic and electronic phenomena in condensed matter, including activated 4He and 3He quantum liquid droplets, surface-activated graphene, novel multi-component superconductors and frustrated quasi-2D magnetic crystals. The CoE studies all the phenomena. In particular, whereas the ions constituting cuprate and nickelate perovskites possess magnetic moments, there appear various opportunities for manipulating volume charges and depletion layers in the heterostructures based on these crystals with the use of magnetic field. In contrast to semiconductor junctions such a control of the heterostructures is implemented without introducing additional heterogeneities. On the other hand, the novel low-dimensional magnetic systems may have important contribution to the emerging field of spintronics. Theoretical models and materials characterizations are essential in order to reach these goals.

Non-linear dynamics & stochastic processes

The nonlinear dynamics of mesoscopic systems presents a serious challenge and will be addressed here. Such processes are of utmost importance in biological systems of high water content with the dynamical network of forming-reforming hydrogen bonds: processes of molecular recognition, dynamics of cytoskeleton and cellular membranes, addressed by this project. In contrast to “bulk models”, our approaches stem essentially from local considerations. With the experimental techniques like tip-enhanced microscopy, we will be able not only to monitor but also stimulate locally such non-linear (structural) processes in order to test the developed theoretical models. Subtle interplay between stochastics and non-linearity governs also several phenomena in novel multi-gap superconductors (Josephson junctions, vortex transport), drives noise-induced phase transitions, and is manifested in the anomalies related to the phase transitions in spatially restricted samples. A general understanding of these phenomena may thus contribute to several fields and a number of applications.

Defects in mesosystems

While the effects of defects on non-coherent transport processes are widely known to be detrimental for a number of applications, the influence of defects on coherent processes on mesoscale is much less studied. Also, the influence of defects becomes more prominent in lower dimensions (nanotubes, graphene, nanolaminates), which are one of the foci of the proposed research. We approach these problems proceeding from theoretical and numerical simulations of defects in mesostructures. The novel methods of computer modeling of defects with account of both short-range and long-range interactions will be developed. The latter interactions play a crucial role in mesosystems leading to the dependence of their properties on size and shape. Our theoretical and experimental study of defects creation should allow us, for example, to increase the stability of materials in respect to radiation damage through the optimization of the size and type of the meso-structures.

Consortium – overview and synergies

Synergies from joint research

The Centre pools together **six internationally recognized research groups** (see the descriptions below) from three Estonian universities: University of Tartu (UT), Tallinn University of Technology (TUT), and Tallinn University (TU). Joint interdisciplinary (physics, biochemistry, technology) research between the groups has led to a number of publications and doctorate degrees during the past 5 years. The groups have agreed upon an ambitious long-term research agenda resulting from in-depth consultations during the preparatory phase of the Centre. The CoE will encourage the mostly bilateral research connections to be considerably diversified.

The underlying principles of CoE are fixed by the Consortium Agreement (CA, attached to the present Action Plan). The CA regulates intellectual property issues among other strategic matters. The consortium is led by Prof Vladimir Hižnjakov (G1). The Steering Committee (SC) consisting of the leaders of the groups (G1-G6) is responsible for regular benchmarking and overall coordination of the activities of the Centre. The SC is also responsible for first-hand dispute resolution should a settlement according to the stipulations of the CA cannot be reached. The SC has initiated an International Advisory Board (IAB) consisting of 5 members acting as a body evaluating the scientific excellence of the research results. Prof Giorgio Benedek*[1], Prof Ernst Sigmund*[2] and Prof Albert J Sievers*[3] are already confirmed members of IAB.

*[1] CV available at: http://www2.academieroyale.be/academie/documents/CVdeGiorgioBenedek623.pdf

*[2] Information is available at: www.tu-cottbus.de/btu/de/universitaet/presse/presseinformationen/archiv2006/2000/februar-2000/prof-sigmund-fuer-das-amt-des-praesidenten-gewaehlt.html

*[3] Brief CV available at: http://www.lassp.cornell.edu/~sievers/ajsvita.html

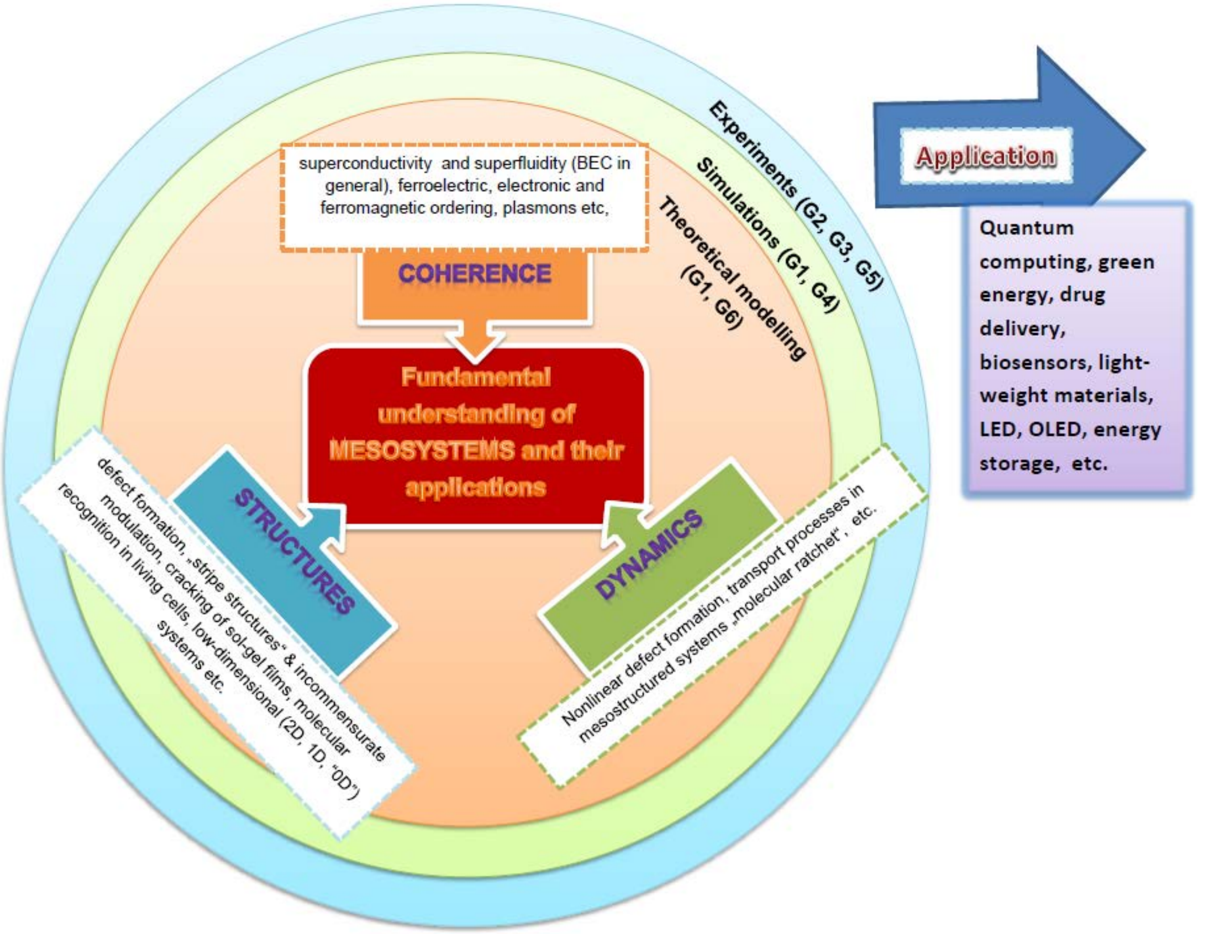

Figure 1. The joint research concept of the Centre. The CoE focuses on coherence, dynamic and structural properties of mesostrucures. The combination of different theoretical approaches, computer simulations and experiments leads to a qualitatively new level of research of mesosystems.

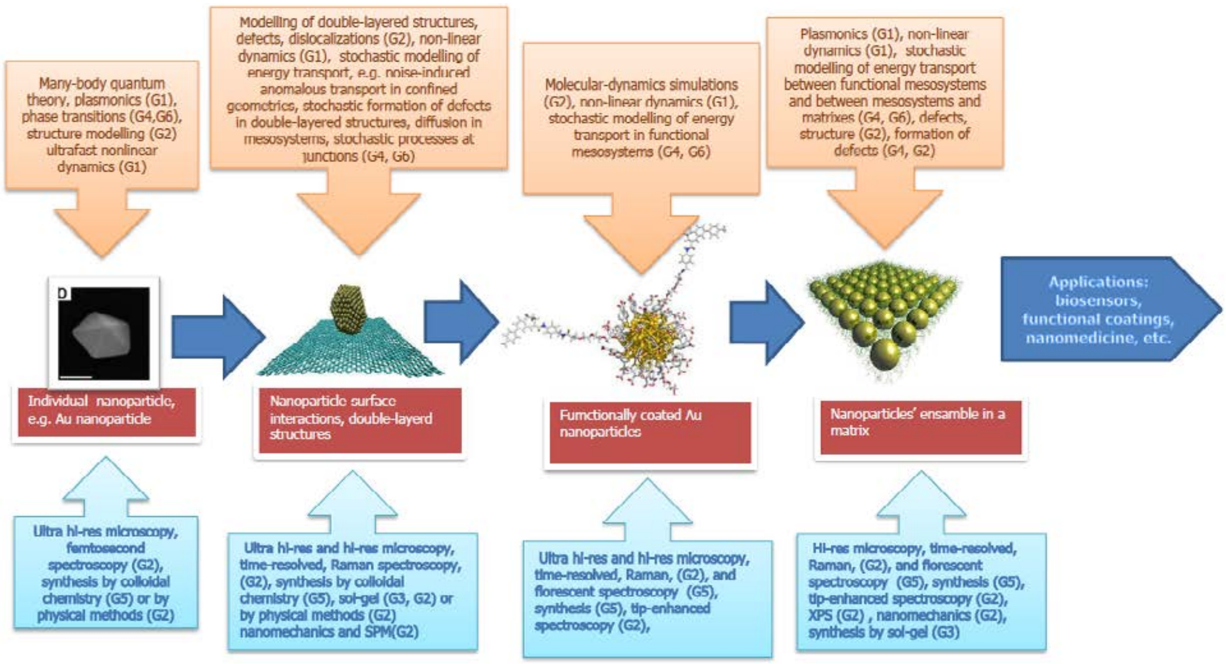

Figure 2. An example of how a synergy of various expertises provides fundamentally new knowledge about complex mesostructures leading towards hi-tech applications.

G1. Theory of condensed matter (Prof Hižnjakov)

The group is a renowned contributor to the theory of condensed matter and quantum fields. Some of the recent successes of the group include the discovery of percolative phase separation in high-temperature superconductors (in collaboration with Nobel prize winner A.Müller[1]; discovery of the effect of the zero-point motion of atoms in optical spectrum of superfluid droplet of 4He[2] [4]; general theory of vibronic transitions[3]; prediction of local modes associated with vibrational solitons[4]; theory of the magnetic incommensurability in cuprates[5], and modeling of defect formation and increasing of radiation stability of crystals[6]. G1 has a number of joint publications with experimental group G2 in the fields of glasses, optical properties of solids, and plasmonics.

Expected results of future research: development of methods of simulations of ultrafast nonlinear dynamics of mesosystems (together with G6), development of new models of mesosystems (together with G2 and G4), development of theory of nonlinear quantum optics and nonlinear plasmon-photonics on discrete photon level (together with G2 and G3), prediction and study of new superconducting and magnetic mesosystems (together with G6), and new materials stable against radiational damage (together with G3).

G2. Physics of mesostructures (Prof Kikas)

The group has excellent track-record in material and optical research with experimental methods. It has been active in developing advanced spectroscopic methods like spectral hole-burning and single-molecule optical spectroscopy and in their application to large variety of systems and processes ranging from complex biological structures to diamond-based materials. In recent years the group has put a lot of attention on techniques utilizing dopant molecules and ions in solids as nanoemitters/markers or as extremely selective and sensitive probes of local conditions. Novel techniques such as single-molecule imaging (SMI)[7] [8], confocal Raman- and luminescence microscopy, photon correlation spectroscopy have been implemented[9]. In recent years these methods have been complemented by AFM and intensive numerical simulations[10] [11]. Pulsed laser deposition, sol-gel technology and focused ion beam lithography enable the fabrication of a large variety of meso- and nanosystems[12].

The group provides an essential support to G1 (experimental tests of theoretical models including plasmonic effects), G3 (characterization of synthesized structures), G4&G6 (experimental tests of theoretical models: stochastic diffusion by SMI) and G5 (experiments on plasmonic amplification of marker fluorescence in biological systems).

G3. Synthesis of functional structures (Prof Krunks)

G3 from Tallinn University of Technology is active in the field of chemistry and deposition of metal oxide and of binary and ternary metal sulfide thin films by wet chemical methods such as sol-gel spray and chemical spray pyrolysis (CSP). G3 main strength is the chemical analysis of formation, structure and thermal behaviour of intermediate complexes as precursors for the films. The parameters of chemical processes are used to model the material via the solutions, deposition and/or calcination temperatures[13]. G3 has obtained notable success on the preparation of metal oxide and metal sulfide thin films for electronics[14], sensorics[15] and photovoltaics[16]. G3 invented the technology for manufacturing ZnO layers comprising nanorods/nanowires by CSP[17] and solar cells based on those[18]. G3 has joint publications with G2 on sensor materials.

[4] Highlights, Europhysics News, 40/5, 9, 2009.

G3 will develop functional mesomaterials by wet chemical methods for light harvesting, plasmon-photonics (together with G1 and G2) and biosensorics (G5), and new materials stable against radiational damage (G1).

G4. Stochastic processes (Prof Mankin)

The main goal of our activity is to develop theoretical basis for various manifestations of stochastic processes and their applications in inter-disciplinary spheres. Possible applications range from superconductors to intercellular protein transport to methods of particle separation in nanotechnology, thus enabling to establish a fruitful collaboration with all other research groups in the CoE.

The group has significant experience in performing inter-disciplinary research in stochastic processes[19] [20] [21] [22]. The main research topics are: anomalous transport; structure formation and phase transitions; influence of plasma on materials; noise-generated phenomena in superconductors; cell/nanoparticle sorting in molecular biology. We will elaborate corresponding theoretical models that will be tested in collaborative experiments. One of the strengths of the group is the knowledge on capillary electromigration methods[23] [24] which together with molecular biology knowledge (G5) and development of new materials (G3) helps to improve the system-level understanding of the functioning of living cells. Specially designed microfluidic devices would be developed to apply stochastic effects on cell/nanoparticle sorting (G6).

G5. Bioorganic chemistry (Prof Rinken)

The bioorganic chemistry research group at the Institute of Chemistry is dedicated to the studies on the mechanisms of the functioning of heptahelix receptors and their regulatory proteins, the key players in chemical signal transduction in nervous system. During the last years, classical molecular pharmacological methods with radioligands were implicated to study molecular recognition mechanisms[25] [26] as well as their connection with different physiological behaviors[27] [28]. At the same time we have started to implement new fluorescence based methods [29] together with new virus-based targets. In parallel, we have worked out new synthetic strategies of substituted hydrazines[30] which are valuable precursors for generation of new drugs and materials. Fluorescence applications created several new dimensions of molecular recognition studies, allowing monitoring of reactions in time (new parameters & models with G4 and G6) and in combination with surface plasmonic methods. This, in collaboration with G1 (theory) and G2 (optics), increases determination sensitivities, giving a new approach for understanding molecular recognition processes, and so is base for generation of new biosensors for determination of different substances.

G6. Critical phenomena, orderings and phase transitions (Prof Örd)

The group is active mostly in the field of multi-band superconductivity. Some of the successes include the modelling of the properties of MgB_2[31], doping-created interband coupling mechanism for high-T_C cuprates[32], and the study superconducting fluctuations[33]. We are also active in such directions as nonlinear quantum optic effects[34] and stochastic effects in phase transitions[35].

Further research together with the other groups includes the study of superconducting, magnetic and electronic mesoscale orderings (G1, G2); stochastic phenomena in multi-gap superconductors (Josephson junctions, vortex transport) (G4); phase transitions and nonlinear dynamics in spatially restricted systems (G1, G2, G4); the development of new models of mesosystems (G1, G2, G4, G5).

Synergies from common infrastructure

The infrastructural resources of a single group are not adequate for investigating such complex objects as mesostructures. , Hence, all available infrastructures for materials research should be considered. In recent years there has been a tremendous effort to modernize the research infrastructure in Estonia. This has widened our capabilities and better integrated our research with the European Research Area. Materials science and technology has been one of the priority areas of the modernization. The high-end set-ups (e.g. FIB-SEM system), realization of the roadmap programme (e.g. high-resolution TEM through NAMUR programme[5]), and new possibilities for international collaboration and improved access to the large-scale European facilities (e.g. Estonian beam-line at Max-Lab, ESS) form a solid and comprehensive basis of experimental investigations of materials including mesosystems.

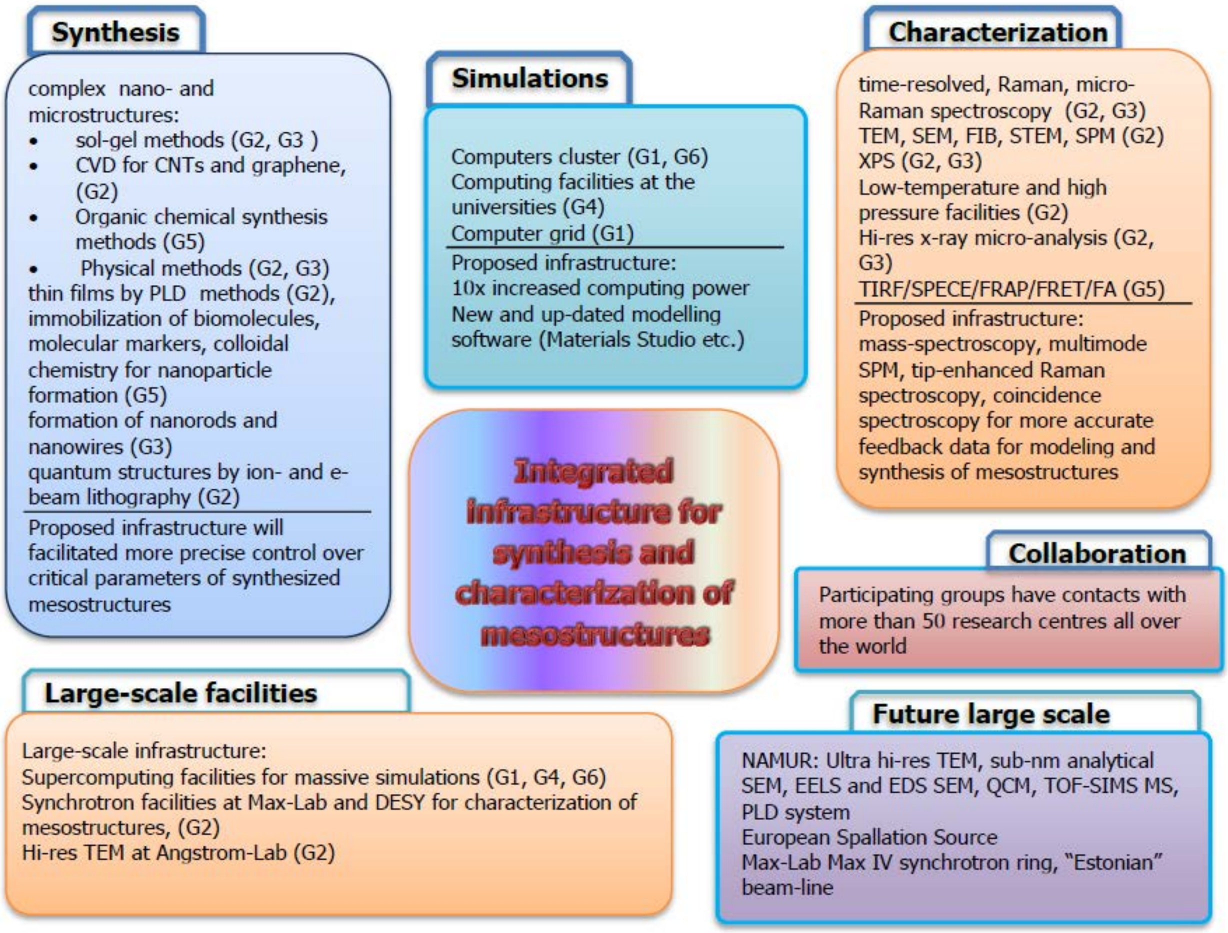

Figure 3. The combined research infrastructure of the participating groups as well as other collaboration schemes results in an experimental research and computer simulation capabilities never been possible before.

However, facilities for computer modeling have received relatively less attention creating sometimes a gap between the capabilities of theoretical predictions and experimental measurements. A significant fraction of the proposed investments will consists of new soft- and hardware that will facilitate more balanced development of materials science infrastructure. New capabilities will also facilitate large-scale simulations utilizing cluster, cloud, and super-computing currently not possible. Proposed investments into research infrastructure serve four major purposes:

- Advanced simulation of materials characteristics (e.g. software Materials Studio)

[5] https://www.etis.ee/Portaal/infrastruktuur.aspx ; funding for NAMUR is 4.94 MEUR.

- Development of synthesis methods for fabrication of larger variety of mesostructures with precisely controlled parameters (e.g. controlled environment capabilities)
- Characterization tools for better and deeper understanding of formation and dynamics of mesostructures (e.g. measurements kits for AFM, mass-spectrometer)
- Set-ups for specific measurements for feedback to theoretical modeling (e.g. tip-enhanced Raman spectroscopy).

Impact on education and internationalization

The CoE is fully integrated into the Estonian and international R&D and higher education structure. A special note should be given to the high level of international collaboration, direct links to applied research through unique collaboration with two Competence Centres[36], ENCC[37] and CCRMB[38], and integration into higher education system via common courses and other measures.

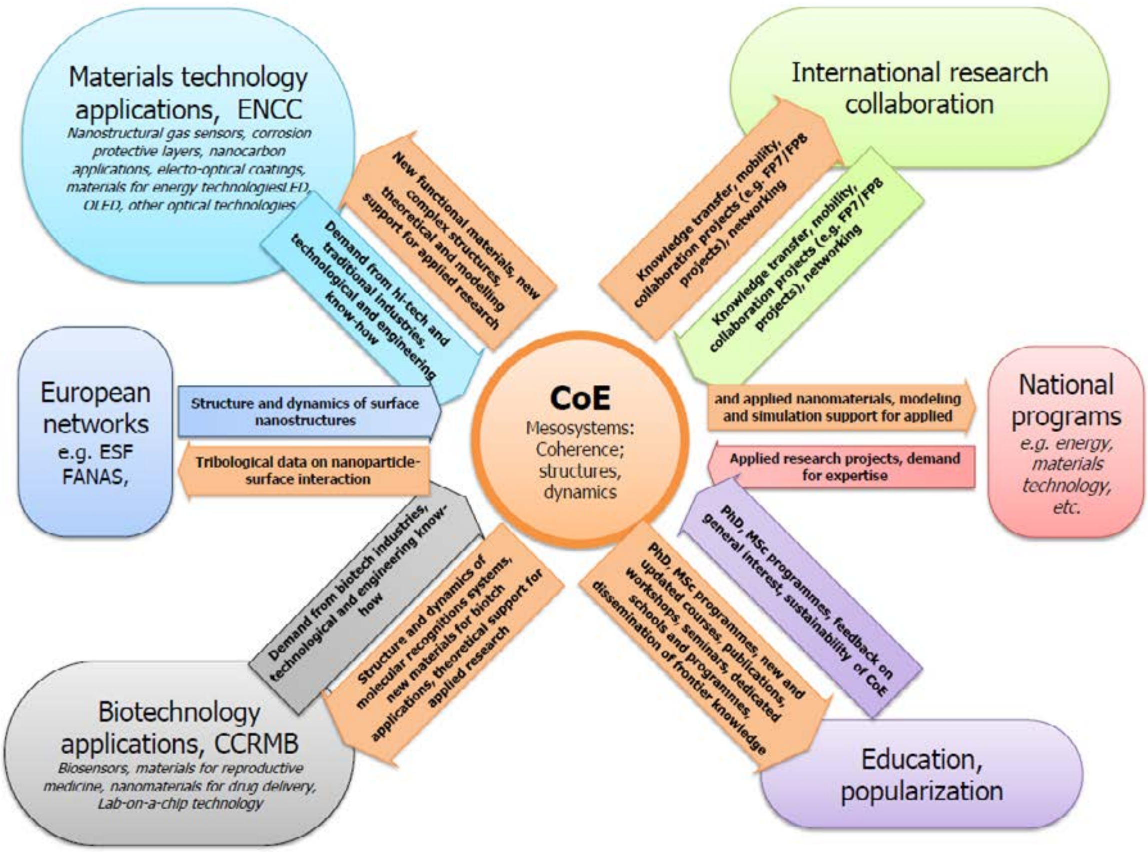

Figure 4. Partner networks of the CoE

Participating groups have traditionally been very successful in combining high level research and education. Through the CoE a particular focus will be put on increasing the level of internationalization of the teaching, one of the key priorities of the universities. E.g. 2 of 3 UT "strategic" [39] professors working at the Institute of Physics, UT, Prof M.Brik and A.Romanov involved in G2. International collaboration through CoE will significantly accelerate the internationalization process as several new cross-border courses are planned, including Quantum Liquids together with Milano-Bicocca University (Italy), and a project in the field of superconductivity together with Lublin University of Technology and with the Maria Curie Sklodowska University in Lublin.

The CoE focuses on research topics that have fundamental importance not only for academics but also for a number of industries. The new quality of the research is based on international and domestic interdisciplinary collaboration, as well as from the rapid development of research infrastructures. The sustainability of the CoE is supported by several measures ensuring steady influx of young researchers.

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Owner: Tallinn University of Technology; Authors: Malle Krunks, Ilona Oja Acik, Tatjana Dedova; Priority number: US60/671232P; Priority date: 14.04.2005. Further applications: WO2006EE00002 (13.04.2006), CA2649200 (14.10.2007), US11/911565 (15.10.2007), AU20060233503 (11.11.2007), IN4325 (13.11.2007), EP1880413 (14.11.2007), CN200680017046.8 (16.11.2007). Issued patent: CN101203948B (China, 16.06.2010).

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Owner: Tallinn University of Technology; Authors: Malle Krunks, Atanas Katerski, Tatjana Dedova, Arvo Mere, Ilona Oja Acik; Priority number US60/948508; Priority date 09.07.2007; Further applications: PCT/EE2008/000019 (WIPO) (09.07.2008), US12/668443 (11.01.2010), EP08773333.3 (01.02.2010), IN 500/KOLNP/2010 (08.02.2010), CN101861654A (22.02.2010)

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