ERC Synergy Grant 2019
Research proposal [Part B1][1]

Ultrafast dynamics of correlated electrons in solids
FASTCORR

Cover Page:
- Olle Eriksson, Uppsala University, Sweden
- Mikhail Katsnelson, Radboud University, Nijmegen, Netherlands
- Alexander Lichtenstein, Hamburg University and XFEL Hamburg, Germany
- 72 month project period

Experimental activities at advanced photon sources, such as pulsed lasers, high harmonic generation facilities, and X-ray free electron lasers, generate results that challenge our understanding of light-matter interaction and ultrafast dynamics at the femtosecond and sub-femtosecond timescales. These results are particularly difficult to interpret for materials with correlated electrons, where a driving pulse can produce strong non-linear effects.

In FASTCORR, we answer this challenge with the development of a theory for driven quantum many-body systems that goes well beyond existing methods. This will be accomplished by developing dynamical mean-field theory and its generalizations, e.g., the dual fermion and dual boson theory, to cover out-of-equilibrium phenomena.

We aim to create a solid theoretical foundation on which we will build practical tools that allow to interpret and predict ultrafast time-resolved phenomena of correlated electron systems. This involves (i) the development of fundamental mathematical and physical concepts, (ii) software implementation, and (iii) numerical simulations that will be compared to experiments. Synergies between the three applicants are crucial to achieving the goals of this project.

FASTCORR will result in novel high-performance software that we will distribute freely. These computational tools will enable designed and targeted calculations for driven materials where the electronic structure is determined by strong correlation effects. The developed theory will be used hand in hand with world-leading experimental works in the field of pump-probe measurements and spectroscopy, e.g., as investigated at X-ray free-electron laser laboratories.

[1] Instructions for completing Part B1 can be found in the '*Information for Applicants to the Synergy Grant 2019 Call'*.

Important challenges tackled in FASTCORR

The experimental breakthroughs with dramatically new measurement techniques, such as high-intensity femtoscale optical lasers [1] and x-ray free-electron lasers [2] put focus on the out-of-equilibrium situation. Interestingly, in these experiments the light-matter interaction is of similar strength as all the relevant interactions, causing entirely new states of matter. A class of materials that is of particular interest in this regard is the correlated electron systems, where the independent electron picture breaks down. In Fig.1 we illustrate how, in out-of-equilibrium situations, the interesting and unexpected intertwining of charge-, spin-, orbital- and lattice-degrees of freedom (upper part of figure), becomes drastically more complex for many-electron systems, where the optical response is special, due to the unique electronic structure (lower part of figure). Materials with a correlated electronic structure have attracted attention for a long time, dating back to ground-breaking works that earned P. W. Anderson, N. F. Mott and J. H. van Vleck the Nobel Prize in Physics in 1977. A new era in this field is now emerging, and is intimately connected to the recent development of new experimental facilities that study out-of-equilibrium phenomena of these materials [3]. However, the experimental breakthroughs mentioned above need to be followed by an equally demanding theoretical leap. Currently this is missing, which is detrimental for the field of out-of-equilibrium science, since an interpretation of these experiments can only be made reliably when observations are coupled to an accurate theory. Since theory is missing, when it comes to making a comparison to experimental data of out-of-equilibrium situations of correlated electron systems, **FASTCORR** will ambitiously fill this knowledge gap.

Experimentally, often using pump-probe technique, outstanding observations have been made that challenge the understanding of light-matter interaction in solids. Several interesting measurements have been reported, e.g. the excitation and relaxation processes of correlated electron systems at times scales that are relevant for the hopping processes of electrons from one lattice site to another [4], the photo-induced metal-insulator transition in Mott systems [5], the photo-induced melting and recovery of charge density waves [6], and the light-induced demagnetisation and recovery of ferromagnets [7] and antiferromagnets [8]. Another unique feature of time-resolved measurements of correlated systems is that an external perturbation may cause cooperative changes and drive the system into hidden states that are not accessible in other ways [9]. For technology, pump-probe experiments have shown that strong few-cycle optical fields can drive electric currents within one femtosecond, showing potential for high speed electronics in the petahertz domain [10,11]. Interestingly, the development of higher harmonic generation (HHG) sources opens possibilities for in house, time resolved studies. An even more exciting development for time-resolved spectroscopies is the European x-ray free electron laser (XFEL) in Hamburg, that recently has started its regular operation, generating pulses of photons in the energy range of 0.24 to 24 keV, with a brilliance that is a billion times higher than that of conventional x-ray sources (https://www.xfel.eu/).

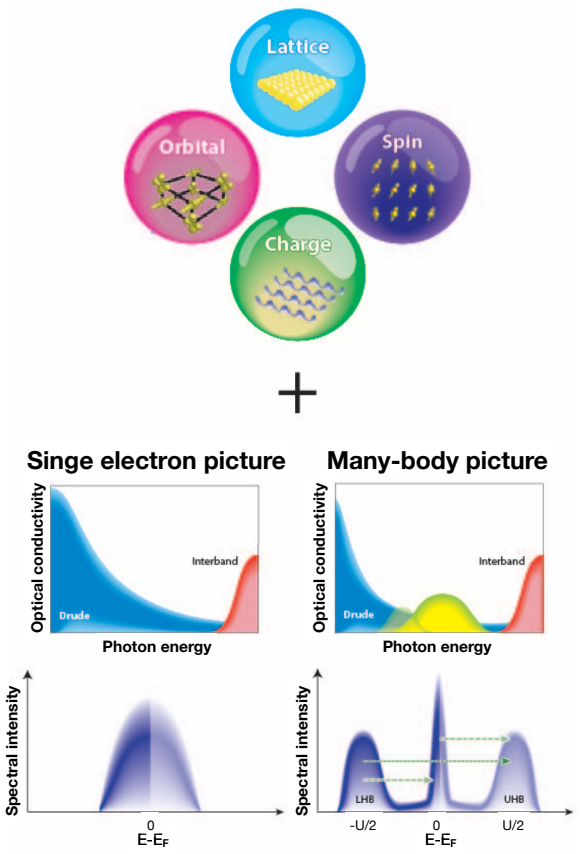

Fig.1 The coupling between lattice, charge, spin, and orbital degrees of freedom (upper figure) in pump-probe experiments is complicated by new features in the optical response of correlated electron systems (lower figure).

Although the foundations for developing a theory for ultrafast phenomena and non-equilibrium spectroscopy of correlated electron systems must have the Dynamical Mean Field Theory (DMFT) equations as a starting point [12], there exists no practical working tool that can be used to analyze strongly driven materials, and e.g. to interpret any of the experiments mentioned above. For materials without significant electron correlations, time-dependent density functional theory (TD-DFT) can be applied in out-of-equilibrium situations, but a large class of materials have electronic configurations that can't be characterized by a single-electron wave-function, causing TD-DFT to break down. Moreover, in certain

cases the DMFT is not sufficient, since nonlocal correlations and interactions play a crucial role. The lack of understanding of correlated electron materials in out-of-equilibrium conditions, as is the situation in the emerging field of ultra**FAST** dynamics of strongly **CORR**elated materials, motivates **FASTCORR**.

State-of-the-art

In principle, all physical properties of materials are the result of well-known laws of quantum mechanics. In practice, this is a problem of extraordinary difficulty, primarily due to the decisive role of electron-electron interactions and therefore the inherent many-body character of the problem. The theoretical description of the quantum nature of the electronic structure, and the materials properties that are the direct consequence of it, has developed tremendously since the 60'ies. This was first achieved by the invention of density functional theory (DFT) [13] and softwares that solve the DFT-equations, and later, for strongly correlated electron systems, by the development of DMFT [14] and its further implementation into electronic structure calculations [15,16]. For some materials an effective single electron picture is appropriate (so called weakly correlated systems), as was justified by Landau Fermi liquid theory [17]; in this case, the main many-body effects are basically renormalizations of the electron energy spectrum. In such a situation, parameter-free calculations based on DFT have shown great ability in reproducing a great variety of properties, including equation of state, lattice dynamics, phase stability, elastic response, optical properties, magnetic moments, magnetic structures, magnetic anisotropy and magnon excitations. In fact, the observed tunneling magnetoresistance [18] in Fe/MgO/Fe sandwich structures, where a few layers of insulating MgO are inserted between thin films of Fe, represents an important sensor technology that followed theoretical predictions based on DFT calculations of the electronic structure [19]. The materials science community now puts firm trust in these simulations, to such extent that major efforts are made in building large data-bases of electronic structures for which one may develop data-mining algorithms in the search for functional materials [20]. When DFT is not enough due to strong correlations, such as for rare-earth metals, DMFT, that naturally allow for multi-configurational aspects of the electronic structure, has been proven very powerful [21]. Unfortunately, for out-of-equilibrium properties the situation is different, since the theoretical understanding of driven, correlated electron systems in general is lacking.

Ground-breaking approach and synergy

The main aim of **FASTCORR** is to provide a broad theoretical framework to address the exciting experimental development in the field of out-of-equilibrium phenomena at atomic time scales, in condensed matter systems with correlated electrons. Since this field to a large degree is unexplored theoretically, it is necessary to start on a fundamental level and even to provide a proper theoretical language. **FASTCORR** will meet this challenge by the development of mathematical models and numerical methods. Coupled to efficient implementations this will allow for a full materials specific description. This must necessarily involve groups with complementary skills, that can establish fruitful collaboration and synergies. All partners of **FASTCORR** are needed in order to achieve the desired goals. The complementarity in expertise among the present constellation of applicants, range from fundamental theory, via numerical implementation and high performance computing to interaction with experimentalists. The breadth in expertise, combined with demonstrated ability amongst the applicants to collaborate with each other, provides synergy effects that will ensure far reaching goals.

The tasks of the project involve work on model Hamiltonians, where idealized situations of more simplistic consideration allow for a deepened understanding of essential mechanisms. We will also pursue a development that ultimately has all essential ingredients that allow for a full materials specific description, e.g. of the experiments of Refs. [4-11]. Among the more significant tasks, we will develop DMFT and its generalizations to more advanced many-body approaches (e.g. the so called dual fermions and dual boson approach, see below), in order to handle out-of-equilibrium situations. The aim of these efforts is to provide a realistic description of the electron dynamics of materials on time scales ranging from femto- to atto seconds. In addition to electron dynamics we will focus on phenomena that accompany electronic motion, such as magnetism and transport properties**. FASTCORR** also strives to provide an accurate theoretical description of x-ray (XAS and RIXS) and photo-electron spectroscopy for these systems.

Research objectives and Work Packages

FASTCORR will be organized in four WP's, that will be carried out in parallel, in a collaborative effort by all three nodes of the proposal. WP1 has special focus on development of fundamental, theoretical concepts and is as such the most mathematically intense part of the project. In WP2 we focus on finding efficient numerical solutions to the theories developed in WP1, both for model Hamiltonians as well as for

computationally very demanding theories that allow for materials specific calculations and a direct comparison to experiment. In WP3, high-performance computations will be employed using the methods developed in WP2. The obtained theoretical results will be compared to experimental data, both for spectroscopic and optical data, conductivity as well as magnetic properties. WP4 involves project planning and decision making. The research directions in the different WP's are:

Workpackage 1: Basic development and model Hamiltonians
We will develop non-equilibrium DMFT on a model level, involving simplified Hamiltonians (e.g. one-band Hubbard model). This will create prerequisites for more advanced approaches discussed in WP2. In this sense, this development is auxiliary, but very important. Also, results from these efforts can be relatively straightforwardly generalized to realistic multiband cases, although significant challenges have to be met [22].

With model Hamiltonians we will develop theory that is capable of studies of instantaneous light-matter interaction (so called local quenches) in strongly correlated systems. An alternative to the diagram technique approach is based on the use of holography (anti de-Sitter/Conformal field theory correspondence, AdS/CFT) borrowed from string theory. This has successfully been applied to numerous problems of condensed matter physics, which turn out to be too difficult for more conventional quantum many-body tools [23]. This method has previously been applied to global [24] and local [25,26] quenches. The aim of this work package is to provide analytical results for a deeper physical understanding of the computational results obtained within the other work packages.

We will in WP1 address a Floquet problem for strongly correlated systems, that are irradiated by an intense time-periodic photon field during long times. In the limit of sufficiently high frequency of the external laser field, an efficient asymptotic method has been developed [27] and successfully applied to specific problems [28-30]. The Floquet problem will in **FASTCORR** be generalized to a multiband Hubbard model and realistic Hamiltonians with orbital degrees of freedom. We will also combine the Floquet theory with the local quench approach, in order to have a full theory of a typical pump-probe measurement. The Floquet theory will be used to evaluate the electronic structure during and after the drive pulse, while the quench will be used to mimic the probe step of an experiment. Applications of this technique will focus on correlated systems with charge ordering, typically transition metal oxides, involving also the superconducting cuprates.

Workpackage 2: Practical implementations and efficient algorithms
Dual fermion and dual boson theory was suggested and developed by the Nijmegen and Hamburg groups [31,32], and this theory allows to consider collective phenomena in strongly correlated systems. In equilibrium, this approach has delivered several outstanding results [33-35]. We will use it as a starting point to develop a Keldysh diagram technique, which will provide a description of plasmon and magnetic excitations in driven, strongly correlated systems. Initially, the case of an instantaneous light-matter interaction will be considered (quench), but the ultimate goal is to treat a general time scale for which an optical field can interact with a solid.

Theory of interatomic magnetic exchange interactions of strongly correlated systems [36] has been generalized to the non-equilibrium case [30]. However, the dynamics described by the effective action of Ref.30 has not been investigated at all. To develop a full theory of magnetization dynamics requires significant analytical work, implementation in softwares and numerical simulations for direct comparison with experiments.

Numerical methods of electronic structure theory as well as many-body physics, e.g. continuous time quantum Monte Carlo simulations [37], exact diagonalization [38], fluctuating spin-polarized T-matrix [39,40], and Hubbard-I approximation [16,21] will be generalized for the time-domain of DMFT, and their efficiency and accuracy will be evaluated. We will here consider also the time-dependence of the interaction parameters of the electron Hamiltonian, that are visible in experiments that approach the ultrafast regime. This challenging development is vital when aiming at materials-specific calculations and connection to experimental results.

Workpackage 3: Calculations and comparison to experiments
With the methodology developed under WP1 and WP2 we will address the challenges provided by the experimental community active in pump-probe laser field physics, and spectroscopy at x-ray facilities. The methodology developed will enable a theoretical description of excitations in the optical regime as well as the soft and hard x-ray regimes. Furthermore, in WP3 we will calculate theoretical spectroscopic data of driven materials, that will directly be compared to experimental results of XAS, RIXS and x-ray magnetic circular

dichroism (XMCD). We have the ambition to be able to consider pump-probe experiments, with ultra-short photon pulses as well as longer time scales for which an optical field can interact with a solid. This puts us in a favorable position to address questions connected to e.g. photo-induced metal-insulator transition in Mott systems, where the primary focus will be on complex oxides. We will also be in position to study the light-induced demagnetization and remagnetization of magnetic materials [7,8]. Examples of systems we will investigate in this regard are elemental 3d metals, half-metallic Heusler alloys [40] and magnetic transition metal oxides, e.g. colossal magnetoresistance manganites as well as high-temperature superconducting cuprates. We will in WP3 also use data-mining algorithms to find materials that are suitable for the model investigations in WP1 and WP2 that rely on single or few band approximations, i.e. we will data-mine for materials in which correlations are only relevant in a subset of all orbitals.

Workpackage 4: Project coordination and decision making
The efforts of WP4 connect to practical aspects of carrying out the project, dealing with decision making and resource allocation. Risk analysis will be made in this work package, as well as decisions for how resources can be reallocated to handle unexpected obstacles in WP1, WP2 and WP3. Here longer visits will be planned between the nodes of the proposal, as well as time and topics of workshops and schools organized under the umbrella of **FASTCORR**.

Scientific impact of FASTCORR
FASTCORR will result in a general theory of driven quantum systems, specifically for correlated electron materials. This will allow for a level of understanding that goes well beyond existing theories, e.g. based on static approaches to the many-body problem or from time-dependent one-electron theory, as provided by TD-DFT. In addition to a deepened insight and new paradigms in understanding driven correlated systems, we expect the project to result in new high performance softwares that will be distributed freely and will enable researchers across the world to perform designed and targeted calculations of driven correlated systems, of their choice.

The beneficiaries of the success of **FASTCORR** are both theorists in laboratories across the world that can use the developed softwares of **FASTCORR**, as well as world-leading experimentalists that work in the field of pump-probe experiments and spectroscopy. Examples of these are laboratories such as XFEL Hamburg and synchrotron radiation facilities that provide X-rays, as well as high intensity laser facilities. Relevance for experimental work is in the field of XAS and resonant inelastic x-ray spectroscopy (RIXS) as well as pump-probe experiments with lasers light, that excites valence states. With **FASTCORR** it will for the first time be possible to interpret and understand the phenomena of correlated electron systems investigated at these laboratories, and the understanding is expected to open up new technologies, new functional materials and new scientific vistas.

The geographic placement of the nodes of this project with respect to the European XFEL could not be more strategically located. The knowledge gain in this project will be important also for more applied aspects of solid state physics/materials science. Examples of such research endeavors are light harvesting applications that rely on dye-sensitized materials, information technology that rely on magnetic media, and materials for use in electronics devices that operate in the petahertz regime.

Feasibility and risk strategy
The aim of **FASTCORR** is very ambitious and involves a combination of different approaches, both analytical and computational, which is necessary for success. Therefore, synergetic aspects and intense collaborations within the network proposed are crucial. The three project teams are complementary, while at the same time having overlapping scientific interests and methodology (see Fig.2). The applicants of FASTCORR also have extensive experience in leading research teams and being involved in larger collaborations and consortia.

The feasibility of **FASTCORR** is ensured by that fact that the PIs have already shown an aptitude to collaborate. Examples of past interactions that demonstrate an ability of providing ground-breaking theory can be found in Refs. [16, 31-33, 38-57]. Furthermore, the softwares are available to us (e.g. in Refs.[16,21,31,32,38-40, 49,51, 58]), that form a stepping-stone for the planned work of **FASTCORR**. Computational facilities, infrastructure and local expertise within each node provide an excellent background for mitigating the potential risks. Nevertheless, financial and personnel resources will be reallocated to solve potential technical difficulties that might emerge at different stages of **FASTCORR.** The ambition is that the three scientific work packages will seamlessly become connected, and thus enable a full success of

FASTCORR. However, even if only part of the projects under WP1, WP2 and WP3 are connected in this way, **FASTCORR** will still be successful in answering many unknowns. For example, WP1 will focus on idealized model Hamiltonians with only few active orbitals. Even if a generalization of this work to a full multi-orbital computational software is not reached, many unresolved questions can be answered from a stand-alone model Hamiltonian investigation.

Infrastructure and Research Environment Uppsala University
The Division of Material Theory (**DMT**) UU (http://www.physics.uu.se/mattheo) hosts over 90 scientists. The researchers of the **DMT** publish annually over 130 papers in peer reviewed journals, on topics connected to electronic structures, spectroscopy and correlated electron systems. These papers annually generate between 4000 and 5000 citations (ISI web of knowledge). Sweden has outstanding computer resources for high-performance computing, where the PI (O.E.) of **FASTCORR** is the largest allocated user.

Infrastructure and Research Environment Radboud University
Theory of Condensed Matter (**TCM**) RU (https://www.ru.nl/tcm/) hosts now about 30 scientists. The researchers of the **TCM** publish annually about 50 papers in peer reviewed journals, that annually generates about 7000 citations (ISI web of knowledge). The group has excellent facilities for high-performance computing, and uses intensively the Supercomputer Center in Amsterdam. **TCM** is a part of the Institute of Molecules and Materials of Radboud University with its outstanding experimental facilities such as the free-electron laser for infrared spectroscopy (FELIX Laboratory).

Infrastructure and Research Environment Hamburg University
The group of Theory of Magnetism and Electronic Correlations (**TMEC**) (https://theorie.physnet.uni-hamburg.de/group_magno/) is a part of the Institute of Theoretical Physics at Hamburg University (HU) and consists of full, associate and junior professors, postdocs and about 20 PhD scientists, as well as an external theory group at European XFEL with two postdocs (http://xfel-old.desy.de/research/theory/). The researchers of the **TMEC** publish annually about 20 papers in peer-reviewed journals, with more than 1500 citations (ISI web of knowledge). The Institute of Theoretical Physics has excellent computing facilities, as well as a university computer center (HUMMEL). Also the group has access to the Supercomputer Center at Berlin-Hannover.

Will this team deliver the promised ground-breaking work?
The team members of this project have contributed significantly to the understanding of complex materials, to methods of electronic structure, to many-body physics and DMFT. Among the applicants behind **FASTCORR**, Katsnelson is mostly responsible for development of fundamental theory and for interpretation and analysis of the results, Lichtenstein has main focus on the development of computational methods and theoretical implementations that allow for realistic, materials specific calculations, while Eriksson will primarily be responsible for calculations of properties of real materials and establishing connection to experimental activities. The breadth and complementarity of expertise of the applicants ensure a wide competence to address the challenges of FASTCORR, and we illustrate in Fig.2 the scientific output and competence of the team behind **FASTCORR**.

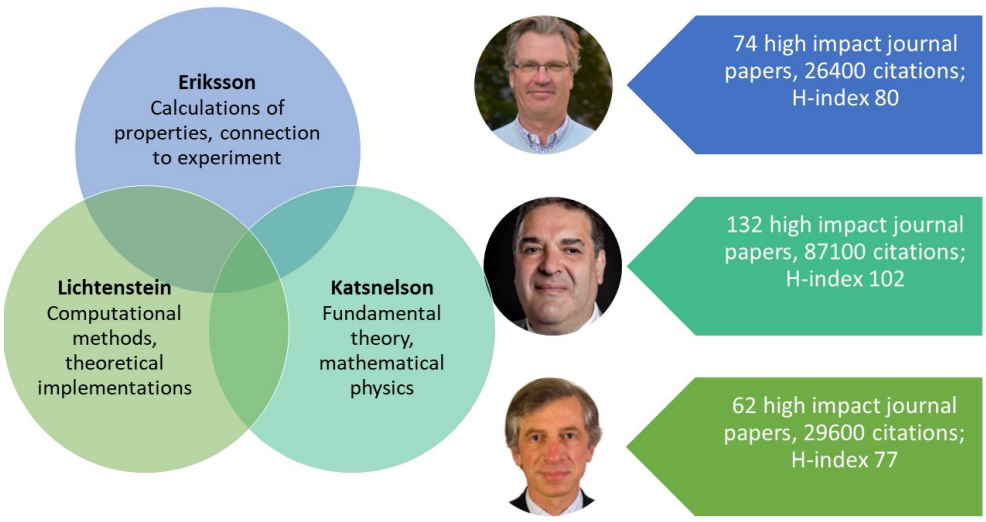

Fig.2 Expertise and bibliometric data of the applicants.

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