

ERC Synergy Grant 2019
Research proposal [Part B2]¹
(not evaluated in Step 1)

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- Project duration: 72 months

Part B2: The scientific proposal (max. 15 pages, excluding the Resources section and References)

Section a. State-of-the-art and objectives

State-of-the Art

General considerations

All physical properties of materials are the result of well-known laws of quantum mechanics. In practice, this leads to 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's. This was first achieved by the invention of density functional theory (DFT) [1] and software to solve the DFT-equations, and later, for strongly correlated electron systems, by the development of dynamical mean field theory (DMFT) [2] and its further implementation into electronic structure calculations [3,4]. For some materials, an effective single electron picture is appropriate (so called weakly correlated systems), as was justified by Landau Fermi liquid theory [5]; in this case, the main many-body effects are simply renormalizations of the electron energy spectrum. In such a situation, parameter-free calculations based on DFT have shown great ability in reproducing a huge variety of properties, including equation of state, lattice dynamics, phase stability, elastic response, optical properties, magnetic moments, magnetic structures, magnetic anisotropy and magnon excitations.

When DFT breaks down due to strong correlations, such as for example in rare-earth metals, a more sophisticated approach is needed. DMFT coupled to DFT (DFT+DMFT) naturally allows for multi-configurational aspects of the electronic structure [3]. The success in practical use of this theory for describing ground-state and excited states properties of materials with correlated electrons is well established [3,6]. In fact, the local correlation effects taken into account via DMFT are crucially important for almost any group of *d*- or *f*-electron materials, including dilute magnetic semiconductors [7], elemental rare-earth metals [3], mixed-valence compounds [8], and exotic actinide-based superconductors [9,10]. It should be noted that the DFT+DMFT approach has significantly changed the way how we interpret experimental data. The central quantity in DMFT is the single-particle spectral density, which describes not only the quasiparticle bands but also quasiparticle damping and incoherent features of the electron spectrum, via the spectral density transfer phenomena. Therefore, the most natural applications of DMFT is photoemission spectroscopy (PES) and angle-resolved photoelectron spectroscopy (ARPES) where one can compare with experiment not only positions of energy bands, like in DFT calculations, but the whole energy and momentum distribution of the spectra [11]. The DFT+DMFT was also a breakthrough in our understanding of moderately correlated, itinerant-electron magnetism, as the first method which takes into account the quantum character of local magnetic moments [12,13].

The experimental breakthroughs with dramatically new measurement techniques, such as high-intensity femtoscale optical lasers [14] and X-ray free-electron lasers [15] put focus on the out-of-equilibrium situation. Interestingly, in these experiments the light-matter interaction can be of similar strength as all the relevant interactions, causing entirely new states of matter. For the correlated electron materials, there are unfortunately few theoretical tools to use to establish a theoretical understanding, especially if one has an ambition to make a direct comparison between measured and calculated properties. In **FASTCORR** we will meet this challenge.

¹ Instructions for completing Part B2 can be found in the '*Information for Applicants to the Synergy Grant 2019 Call*'.

DMFT appears to be the minimal theory which takes into account the most important properties of strongly correlated systems at equilibrium, being able to describe both incoherent (e.g. Hubbard) and coherent (quasiparticle) features of the excitation spectra [19-21]. Recently, the DMFT equations were generalized and applied for idealized models of non-equilibrium systems [22]. Generally speaking, this is an extremely complicated problem since the Green's function of a time dependent system depends on two times, and any approximation must involve a solution of cumbersome nonlinear integral equations. Equilibrium DMFT and DMFT-based approaches has been successfully applied to spectroscopy in the linear regime, where the radiation is not intense or a pump pulse is absent (examples will be given below, Figs. 1-4). However, experiments with high-intensity light sources or pump probe experiments, put entirely new demands on the

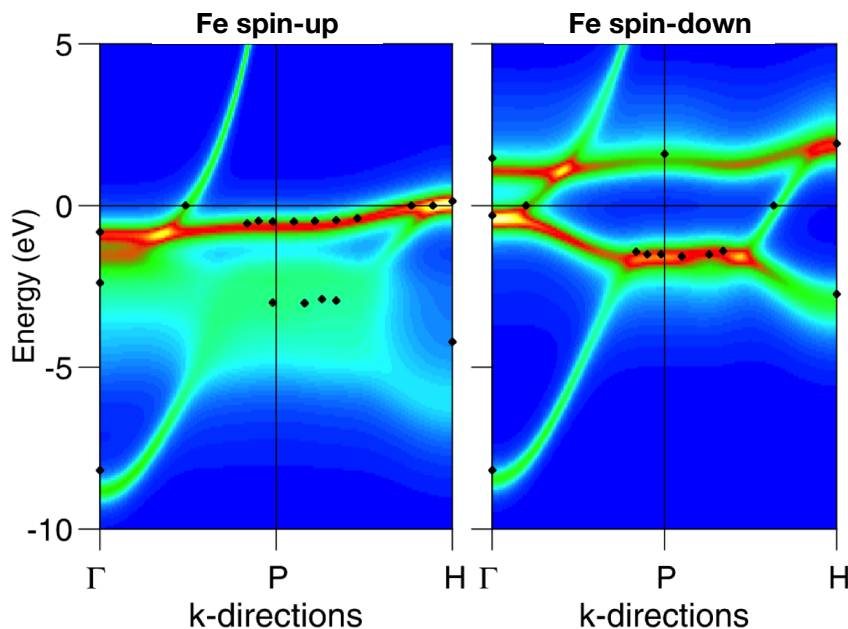


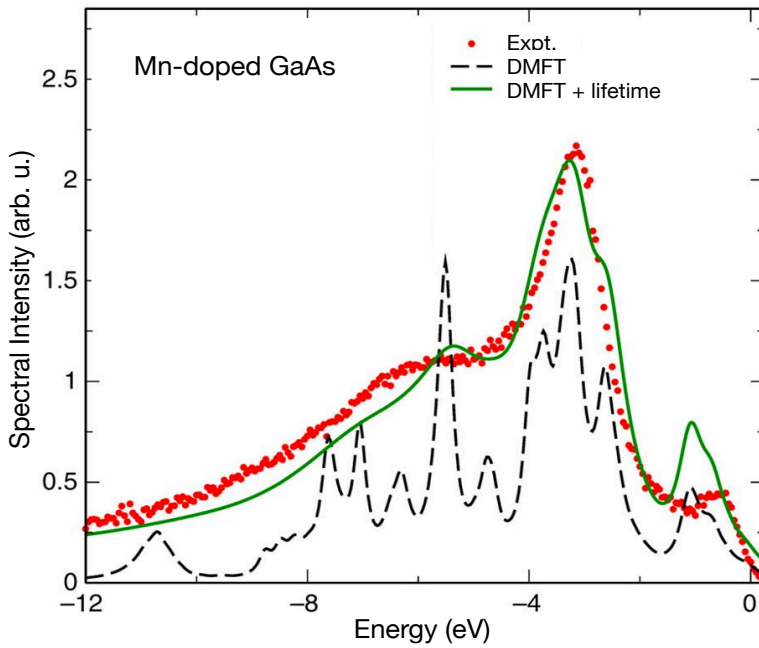
Fig.1 Spectral function of ferromagnetic bcc iron for spin-up and spin-down electrons in DFT+DMFT scheme [13] (this was the first calculation of ARPES with DFT+DMFT) compared with ARPES experiment (black points) for two k-directions in the Brillouin zone.

understanding of light-matter interaction and the interpretation of new states that emerge in these situations. Based on the experience of DMFT in the linear regime, in **FASTCORR**, we will develop a theory for correlated electron systems in the strongly *nonlinear regime*, that is, for pump-probe experiments where the first (intensive) photon beam is used to *prepare* the state under investigation and the second (weak) beam is used to *probe* the response of this strongly non-equilibrium state. As always, at the transition from the linear case to strong non-linearity, incremental development of existing tools is not sufficient, and dramatically new approaches are required.

Results for excited state properties in the linear regime near equilibrium

We describe below where theory stands with regard to being able to reproduce observations in the linear, semi-static regime of correlated electron systems. This range of temporal scales (picosecond time scales and slower) enables a separation of fast and slow variables. The results we present here serves to illustrate that DFT+DMFT is an excellent platform which **FASTCORR** will be built upon, in order to meet challenges of ultrafast phenomena (femto- and atto-second scales). The DFT+DMFT approach has for these situations been extensively applied to PES and ARPES. The most basic approach to these spectroscopies consists in the direct comparison between the measured spectra and the theoretical spectral functions. In the last two decades a variety of studies adopted this strategy, starting with the very first reports on δ -Pu [23], Ce [24] or Ni [25]. The latest generation of DFT+DMFT codes, based on all-electron full-potential implementations [26] and improved quantum impurity solvers [27], have been applied to a variety of materials, spanning various degrees of electron localization.

A significant example of a direct comparison between theoretical data, in the linear response regime, and ARPES is shown in Fig.1, as an early attempt to analyze the spin resolved spectrum for iron [13]. It is clear from this figure that theoretical data coincide with measurements, and that there is a strong broadening for spin-up electrons, which results in spectral features for the spin up states that cover an energy range of 2-5 eV binding energy, e.g. around the P-point of the Brillouin-zone. This broadening was also observed in photoemission experiments.



valence band spectrum of Mn projected states for Mn doped GaAs. Theory is shown both with (green curve) and without (dashed black line) lifetime broadening [31].

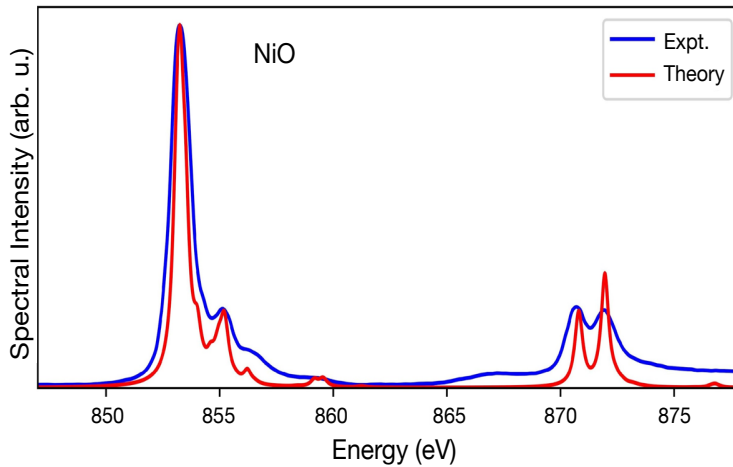


Fig.3 Comparison between the experimentally measured and the calculated L-edge X-ray absorption spectrum of NiO (for details see text)[30].

Transition metal oxides [28] or dilute magnetic semiconductors [29] are examples where the electron-electron repulsion is even more pronounced compared to the case of bcc Fe. Spectral features emerge from the theory, that are completely absent in a simplified effective one-electron approach. For correlated electron systems these features, that may be obtained from the DMFT calculations, are in agreement with observations. In Figure 2, the DFT+DMFT spectral function of Mn doped GaAs is compared to resonant photoemission data, showing good agreement. These results have also helped in clarifying the mechanism driving the ferromagnetic order in this material.

By including relevant core states into the many-body problem, it is also possible to accurately calculate X-ray absorption spectroscopy (XAS) [30] or even resonant inelastic X-ray scattering (RIXS) [31] with the DMFT method. For XAS in the linear response regime, good correspondence between theory and experiment is attainable, as illustrated in Figure 3 for L-edge spectra of NiO. The real power of these approaches lies in their accuracy and capability of reproducing observed spectroscopic features, and that this is achievable for materials where electron correlations of the valence band produce a complex multi-configurational state.

Results for semi-static magnetism for correlated systems

For longer time scales of the excitations in materials (> 0.5 ps), the division into slower and faster degrees of freedom results in the possibility of introducing an adiabatic approximation for magnetic moment orientations (and for nuclear positions). In this case it is possible to evaluate complex magnetic interactions, that often are mapped onto an effective Heisenberg model,

$$\hat{H}_{exch} = - \sum_{i \neq j} J_{ij} \mathbf{e}_i \cdot \mathbf{e}_j$$

Here \mathbf{e}_i is a unitary vector for the direction of the magnetization at the site i , J_{ij} are the exchange interactions between atomic moments of sites i and j , and the sum is over pairs of magnetic atoms. Often the effective Heisenberg model is coupled to atomistic spin-dynamics simulations [32], to address effects that are of pico-

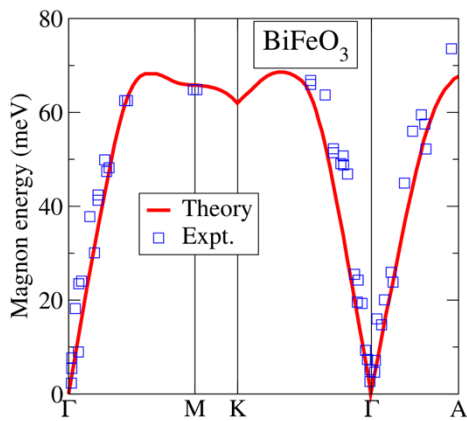


Fig.4 Calculated and measured magnon dispersions in BiFeO_3 . The results are published in Ref.[35].

second time-scales and longer (for examples see textbook of Ref. [33]). The formulation for extracting the J_{ij} from first principles theory was developed by Lichtenstein and Katsnelson, and is based on a Green's functions linear response formalism [34]. As an example of the success of this approach we show in Fig.4 the calculated magnon spectrum [35] of BiFeO_3 . Its correlated electronic structure was obtained from DFT+DMFT and the magnon dispersion was then evaluated using the Heisenberg Hamiltonian combined with atomistic spin-dynamics simulations. As one can see, theory reproduces the measured data accurately, and this is in fact a typical level of agreement one finds for correlated materials in the linear response regime. This is a very powerful approach to address finite-temperature magnetic properties, as well as magnon excitations. However, for the time-scales and experimental investigations that are relevant for **FASTCORR**, the idea of separating time-scales breaks down, and as described in this proposal, and one needs to take a different approach, both when it comes to describing spectroscopic information as well as the time-evolution of magnetism.

A few observations from pump-probe experiments

Experimentally, several outstanding observations have been made that challenge contemporary theoretical understanding. Typically, these experiments are made in pump-probe mode, that involve laser setups and higher harmonic generation, as well as photon sources from synchrotron radiation facilities and free electron lasers. 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 lattice site to lattice site [36], the photo-induced metal-insulator transition in Mott systems [37], the photo-induced melting and recovery of charge density waves [38], and the light-induced demagnetisation and recovery of ferromagnets [39] and antiferromagnets [40]. 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 [41]. For technology, pump-probes with lasers have shown that strong few-cycle optical fields can drive electrical currents within one femtosecond, showing potential for high speed electronics in the petahertz domain [42,43]. Many of the experimentalists in this field benefits from the development of higher harmonic generation (HHG) sources, that allows for in-house investigations. In fact, such experiments now reach K and L edges of 3d elements, with large photon intensity [44], which offers unique experimental possibilities. 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, with generate 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/>).

Examples of phenomena that have been observed in experiments, are the photo excitations of the VO_2 system, in which a metal-insulator transition was induced on a timescale of tenths of fs [45]. The speed of the transition depends on the details of the microstructure, doping, strain, dimensionality, as well as on the characteristics of the pump-probe. Furthermore, using time-resolved X-ray diffraction and optical reflectivity, de Jong et al. studied the time-resolved aspects of the structural-electronic Verwey transition in Fe_3O_4 . [46] They identified that the metal-insulator transition consists of two steps: an ultrafast (<300 fs) destruction of so-called trimerons, three-Fe-site lattice distortions, followed by a 1.5 ps rearrangement of these trimerons and the occurrence of a phase separation into metallic and insulating regions. As a final example of this short section on experimental backgrounds, we discuss the light-induced transition from an insulating anti-ferromagnetic ground state of strained $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$, into a hidden metallic state [47]. Using photo excitations with sub-50 fs pulses, Zhang et al. [47] were able to tune the conductivity of strain-engineered $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$ films from an insulating- to metallic-like state, as illustrated in Fig.5. The energy of the photon pulses (1.55 eV) was chosen as to resonate with the $\text{Mn}^{3+} - \text{Mn}^{4+}$ inter site transitions, and Terahertz time domain spectroscopy was used to measure the conductivity. The ultrafast, non-thermal transition to transient/metastable states, induced by the photoexcitation, were discussed to be the cooperative result of non-equilibrium dynamics of sub-picosecond timescales. The observations in Ref.47 found a light-

induced response that occurred during a fraction of a phonon period and it was speculated that the main mechanism was caused by an ultrafast modification of the exchange integral.

The examples mentioned above serve to illustrate the need for a new level of theory in this field. The existing theoretical tools, e.g. those that lie behind the results of Figs.1-4, are incapable of explaining the observations mentioned above. Neither are they apt for analysis of any of the observed extreme, out-of-equilibrium situations of the electronic structure. This knowledge-gap prohibits a successful analysis and understanding of the growing number of experimental investigations in this field, where dramatically new experimental techniques [14,15], emphasize out-of-equilibrium physics. These novel experimental techniques provide a lot of information, but theory, in its current form, can help very little with interpretation of observations or with predictions of new physical phenomena.

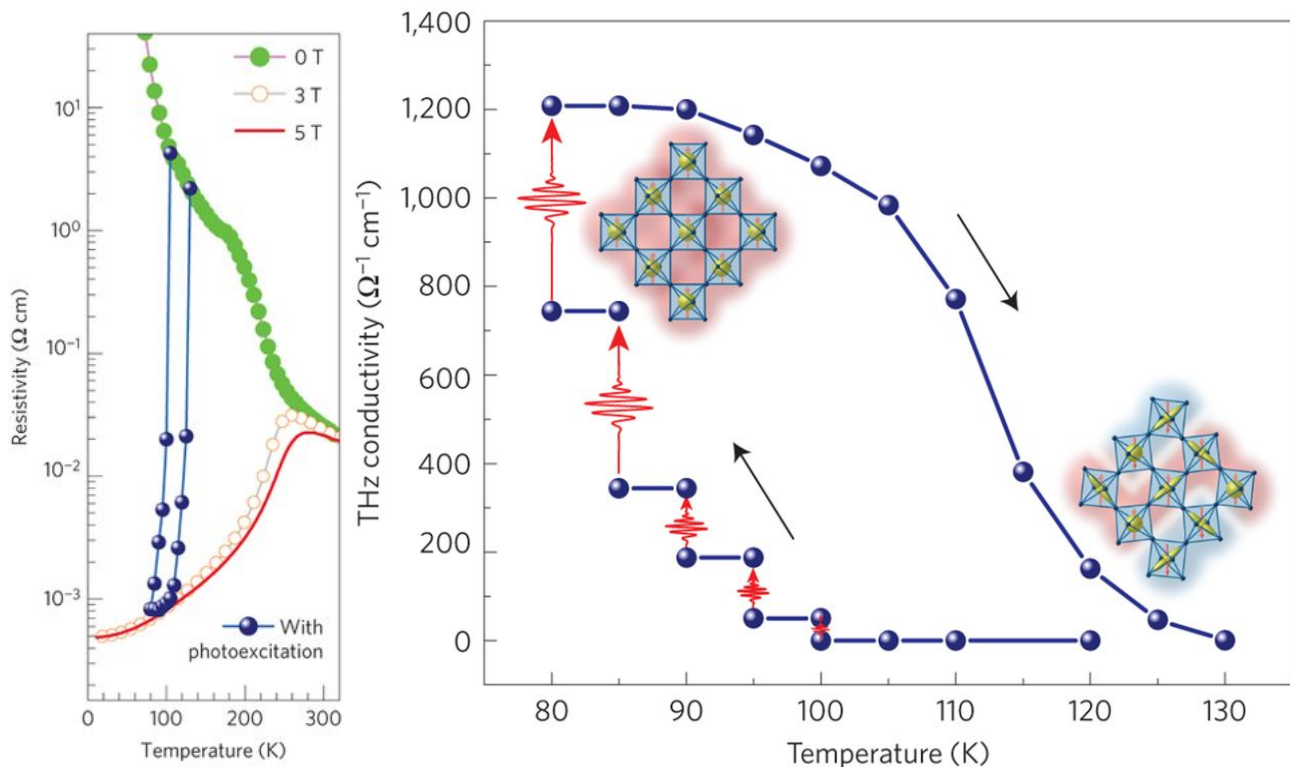


Fig.5 Photo-induced insulator-to-metal transition in a strain-engineered $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$ thin film. Left-hand side: Resistivity-temperature curves for various applied magnetic fields. Photoexcitation under zero field can initiate the insulator-to-metal transition and reach the 'hidden' metallic state that would only be accessible under finite magnetic field in equilibrium. Right-hand side: Photoexcitation also enables access to new states that are not thermally accessible. Figure redrawn after Ref.47.

Objectives

The ambition is that **FASTCORR** will result in a general theory of driven quantum systems beyond the linear response regime, specifically for correlated electron materials. This will allow for a level of understanding that goes well beyond existing theories, e.g. based on linear response theory of the many-body problem or from time-dependent (TD) 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 software 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. We also foresee predictions of new phenomena, that result from the developed methods that will be combined with high-performance materials specific calculations, that simulate pump-probe experiments. Properties that will be under focus here are spectroscopic features, magnetism, optical and transport data.

With **FASTCORR** we aim to take leading position in the theory of pump-probe investigations of correlated electron materials, and to establish collaborations with world-leading experimentalists and other theorists in this field. Examples of laboratories active in these investigations are XFEL Hamburg, synchrotron radiation facilities with capability to generate X-rays, such as MAX IV, DESY, PSI and BESY, as well as high intensity laser facilities (e.g. the HHG source of the HELIOS laboratory in Uppsala). In fact, we have years of experience with such collaborations [48-50]. Relevance for experimental work is in the field of XAS, XMCD (X-ray magnetic circular dichroism), magneto-optical Kerr effect (MOKE) and resonant inelastic X-ray spectroscopy (RIXS). Special emphasis will be put on pump-probe experiments that focus on excitations of valence states followed by spectroscopic detection, either by excitations of deeper lying core states (XAS, XMCD) or from valence states (MOKE). These measurements give information about the time evolution of magnetism and/or of the conductivity. A successful outcome of **FASTCORR** will enable direct interpretation and a deeper understanding of the phenomena of correlated electron systems investigated at these laboratories. Filling the knowledge gaps in this field is expected to open up new technologies, new functional materials and new scientific vistas.

This objective of **FASTCORR** is very ambitious and a combination of different approaches, both analytical and computational, is necessary for success. Therefore, synergetic aspects and intense collaborations within the network proposed here, are very important. The three project teams are complementary, while at the same time having overlapping scientific interests and methodology. This is outlined also in the B1 part of **FASTCORR**.

Section b. Methodology

The basic formalism for quantum many-body theory in out-of-equilibrium condition is provided by Schwinger-Baym-Kadanoff-Keldysh Green's functions [16-18]. The Keldysh diagram technique in conjunction with non-equilibrium DMFT is probably one of the most convenient ways to calculate these functions. Two specific cases, which are easier than the generic case, are typically considered: *i)* the case of a Hamiltonian that is periodic in time, which corresponds, in the limit of non-interacting electrons, to Floquet theory, and *ii)* the case of a quenched system (an instant change of the Hamiltonian, where after the quench the Hamiltonian remains time-independent). Both these problems are physically relevant for condensed matter systems: the Floquet case corresponds, in particular, to physical systems under a strong, periodic laser field, while the quench case is relevant to describe experiments where short, intense pulses used in pump-probe measurements. Although some preliminary work has already been done [22], there are still many complications to address before developing methods that can be applied to material-dependent problems. Interaction parameters entering, e.g. in a Hubbard model or in DMFT, are themselves time-dependent, since they are determined by screening effects [51]. Therefore, for fast processes such as the ones of focus for **FASTCORR**, theory must be extended to allow for a continuous feedback effect, while keeping computational effort within reasonable limits. Both for the Floquet case and the quench will we follow the time evolution of the electronic structure, magnetism and conductivity. We will also develop a theory to extract spectroscopic data (XAS, XMCD, XPS) during the dynamic processes, similar to the data of the semi-static situation reported in Figs.1-4.

Our work will commence within the framework of simplified models, e.g. the multi-orbital Hubbard model, where fundamental aspects will be worked out in detail. In parallel with these efforts, we will start the development of software capable of describing pump-probe experiments in a materials specific, laboratory realistic way. We describe below in detail how we will carry out the project and the four work packages forming the backbone of **FASTCORR**: Fundament aspects (WP1), Practical implementations and efficient algorithms (WP2), Materials simulations and connection to experiments (WP3) and Project coordination (WP4). The main responsible for WP1 is Prof. Katsnelson, while WP2 will be directed by Prof. Lichtenstein. Finally, for WP3 and WP4 Prof. Eriksson is the leading scientist. The description below outlines the scientific steps that needs to be taken in **FASTCORR**, as well as details on decision making and how resources will be distributed for all subprojects.

Fundamental aspects (WP1, main responsible Katsnelson)

(a) *Development of non-equilibrium DMFT beyond the current level*

Development of non-equilibrium DMFT will be carried out that goes beyond the current level. The latter is characterized by the use of a very limited number of solvers, each of them with serious shortcomings [22], e.g. the non-crossing approximation is applicable only near the atomic limit and quantum Monte Carlo at real time domain suffers from sign/phase problem. We are going to develop a solver for the case of a quenched system, using the newly suggested method of tight-binding propagation, able to incorporate multiple orbitals and a general description of the electronic interaction [52]. This method is based on the expansion of the time evolution operator in Chebyshev polynomials. Here the time evolution of the wave function $|\Psi\rangle$ follows the expression:

$$|\psi(t)\rangle = \hat{U}(t,0)|\psi(0)\rangle = \left[J_0(t)\hat{T}_0(\hat{H}) + 2\sum_{m=1}^{\infty} J_m(t)\hat{T}_m(\hat{H}) \right] |\psi(0)\rangle \quad (1)$$

where $|\Psi(0)\rangle$ is the initial state, \hat{H} is the Hamiltonian, t is the time, and J_m are Bessel functions. For the modified Chebyshev polynomials, T_m , one can use a very efficient recursion relation:

$$\hat{T}_{m+1}(\hat{H})|\psi\rangle = -2i\hat{H}\hat{T}_m(\hat{H})|\psi\rangle + \hat{T}_{m-1}(\hat{H})|\psi\rangle \quad (2)$$

with the initial conditions: $\hat{T}_0(\hat{H})|\Psi\rangle = I|\Psi\rangle$, $\hat{T}_1(\hat{H})|\Psi\rangle = -i\hat{H}|\Psi\rangle$.

The expression in Eqn. 1 becomes exact in the limit $m \rightarrow \infty$. This method has proven to be highly efficient in numerical applications for single- and many-body quantum systems with the dimensionality of the Hamiltonian up to 10^{10} assuming that the corresponding matrix is sparse (which is usually the case in real physical problems), with a wide range of applications from foundations of statistical mechanics, to astrochemical calculations.

In the case of a time-dependent Hamiltonian with fermionic and bosonic degrees of freedom, which is a crucial part of DMFT extended to the ultrafast regime, or from the dual boson approach, **FASTCORR** will use a closely related iterative diagonalization scheme within the time dependent Lanczos method in the Krylov-space approach. This method was developed recently in Hamburg [53]. To minimize numerical effort, we adopt the many-body space partitioning from the continuous-time quantum Monte Carlo (CT-QMC) scheme [54], which transforms the initial Hamiltonian in a block-diagonal form $\hat{H} = \hat{H}_1 \oplus \hat{H}_2 \oplus \dots \oplus \hat{H}_s$. The evolution operator for short time propagations in the simplest rectangle approximation reads

$$\hat{U}(t_n + \Delta t, t_n) = T_t \exp \left[-\frac{i}{\hbar} \int_{t_n}^{t_n + \Delta t} \hat{H}(t) dt \right] \approx \exp \left[-\frac{i}{\hbar} \Delta t \hat{H}(t_n + \frac{\Delta t}{2}) \right] \quad (3)$$

The later operator exponent is efficiently evaluated by Lanczos scheme using the k -th order Krylov space for initial quantum state $|\Psi_0\rangle$: $K(|\Psi_0\rangle) = \text{span}\{|\Psi_0\rangle, \hat{H}|\Psi_0\rangle, \dots, \hat{H}^{k-1}|\Psi_0\rangle\}$.

Then two-time correlations function, for example, the greater Green's function can be represented in the compact form

$$G_{ij}^>(t, t') = -\frac{i}{\hbar} \text{Tr} \left[\rho_0 \hat{c}_i(t) \hat{c}_j^+(t') \right] \approx -\frac{i}{\hbar} \sum_{mss's''} w_m \langle \psi_m | \hat{U}_{ss}(0, t) c_{iss'} \hat{U}_{s's'}(t, t') c_{js's''}^+ \hat{U}_{s''s''}(t', 0) | \psi_m \rangle \quad (4)$$

In this expression, the index s is a symmetrized block index, naturally defined in the method [53]. For the time-dependent DMFT scheme we need to evaluate exactly such fermionic and bosonic two-point correlation functions, while for the simplest form of the dual boson scheme (see sub-project b) one needs in addition, a three-point correlation function. The effective interactions in the simplest

form of the dual boson approach are related with three-point impurity correlation functions:

$G_{ijk}^{(3)}(t, t', t'') = -\frac{i}{\hbar} \text{Tr}[\rho_0 \hat{c}_i(t) \hat{c}_j^\dagger(t') \hat{n}_k(t'')]$ where $\hat{n}_k(t) = \hat{c}_k^\dagger(t) \hat{c}_k(t)$. **FASTCORR** will develop an efficient scheme to calculate vertex function on the Keldysh contour.

This sub-project will require 1 postdoc and 1 PhD student.

(b) Dual fermions and dual bosons

The dual fermion and dual boson theory was suggested and developed by the Nijmegen and Hamburg groups [76,77] to describe collective phenomena in strongly correlated systems beyond the local correlation assumptions of DMFT, or with nonlocal interactions (e.g., Coulomb or dipole-dipole). Basically, these methods introduce a smart, essentially nonlinear, change of variables in the path integral representation for the Green's functions, in such a way that the DMFT (or extended DMFT – E-DMFT) corresponds to the bare particles (zeroth order approximation), and the so-called GW+DMFT scheme is included as a simplified case [76].

This means that all important atomic physics such as the formation of the Hubbard bands is taken into account from the very beginning, and we can build a perturbation expansion in the *nonlocal* part of correlations only (for a review, see [78]).

In order to develop an efficient diagrammatic scheme for the non-equilibrium partition function $Z = \int e^{iS} D[c^*, c]$ on the Keldysh contour, we will start from a generalized Hubbard model subjected to a strong instantaneous electromagnetic pulse, described by the action:

$$SS = \int \int dt dt' \left\{ \sum_{\vec{q}\sigma} c_{\vec{k}}^*(t') \left[i\delta(t-t') \partial_t - \varepsilon(\vec{k} - \vec{A}(t-t')) \right] c_{\vec{k}}(t') - \frac{1}{2} \sum_{\vec{q}\sigma\sigma'} n_{\vec{q}\sigma}^*(t) V_{\vec{q}}^{\sigma\sigma'}(t, t') n_{\vec{q}\sigma'}(t') \right\}, \quad (5)$$

with a non-local interaction $V_{\vec{q}}$ which depends on the spin projection σ , and on time due to the screening from high-energy conducting electrons [79]. Using an optimal time-dependent EDMFT impurity problem as a reference system we can obtain exact local correlation functions. The renormalized Keldysh perturbation series, giving diagrammatic contribution for the non-local self-energy

$$\tilde{\Sigma}^\sigma(t, t') = i \sum_{\vec{q}\sigma'} \int dt_1 dt_1' dt_2 dt_2' \gamma(t, t_1, t_2) \tilde{G}_{\vec{k}-\vec{q}}^{\sigma'}(t_1, t_1') \tilde{W}_{\vec{q}}^{\sigma\sigma'}(t_2, t_2') \gamma^*(t_2', t_1', t') = \text{Diagram} \quad (6)$$

where $\tilde{G}_{\vec{k}-\vec{q}}^{\sigma'}(t_1, t_1')$ is the non-local part of the DMFT lattice Green's function and $\tilde{W}_{\vec{q}}^{\sigma\sigma'}(t_2, t_2')$ is the screened Coulomb interactions $\tilde{W}_{\vec{q}}^{-1}(t, t') = V_{\vec{q}}^{-1}(t, t') - \Pi_{\vec{q}}(t, t')$, γ is the three-time correlation function obtained from $G^{(3)}$. The non-equilibrium non-local polarization function can be calculated within the dual boson theory $\tilde{\Pi}_{\vec{q}}(t, t') = \text{Diagram}$. The non-trivial combination of numerically exact solutions of a small time-dependent impurity problem, with renormalized analytical perturbation theory with vertex corrections on the Keldysh contour, bring qualitatively new tools in the complex field of correlated many-body systems that are found in out-of-equilibrium conditions.

In equilibrium, this approach has demonstrated its great power. Some highlights of the results are the theory of flat band formation near a Van Hove singularity in the two-dimensional Hubbard model [80], the prediction of spectral weight transfer in plasmonic spectra of strongly correlated systems [81], a generalization of the theory of exchange interactions in strongly correlated systems [82] and the theory of charge ordering and correlation-induced Lifshitz transitions for dipolar ultra-cold gases [83]. With **FASTCORR** we aim to generalize this method for the Keldysh diagram technique which will allow to build the theory of plasmon and magnetic properties of strongly correlated systems out of equilibrium. Initially the case of an instantaneous light-matter interaction will be considered (the quench), but the ultimate goal of this sub-project is to consider any time-scale for which an optical field can interact with a solid. This is a clearly a demanding sub-project. The initial state is clear, since it is not difficult to write a formal transformation to the dual variables for the Keldysh contour.

However, serious efforts need to be made to check whether the same approximations (e.g., ladder approximation with the four-leg diagrams only) which lead us to the success for the case of plasmonic excitations near the equilibrium [81] will work equally well for the out-of-equilibrium situation. The case of magnetic degrees of freedom is especially challenging since in general it requires also a consideration of six-leg vertices [82]. The development of novel computational methods and algorithms to deal with time-dependent self-consistency equations is a vital step towards realization of this theory. Once successful, we can address materials specific questions with a theoretical tool dramatically more powerful than any existing approach of quantum many-body theory out-of-equilibrium.

This sub-project will require 1 postdoc and 1 PhD student.

(c) *Time-periodic driven systems*

In the limit of a time-periodic photon field, we will use Floquet theory, extended for strongly correlated systems. In the limit of a sufficiently high frequency ω of the external laser field, an efficient asymptotic method has been developed [69] and has been successfully applied to several problems [70-72]. This method is based on a regular procedure representing the effective *static* Hamiltonian as a series in the inverse frequency ω^{-1} , with a suitable recurrent expression of the n -th expansion term via the previous ones, by solution of simple ordinary differential equations. It was demonstrated that the use of a high-frequency mono-chromatic light can change the sign of exchange interactions transforming an anti-ferromagnet to a ferromagnet and, more generally speaking, create quantum Hamiltonians with exotic terms or unusual ratio of parameters (for example, spin Hamiltonians with exchange interactions much weaker than Dzyaloshinskii-Moriya interactions). We note that this is by no means a hypothetical case, real materials have in static situations been identified that belong to this class [73-75].

The methods in Refs. [70-72] can for practical reasons only be applied to few-band Model Hamiltonians. The ultimate step of **FASTCORR** is a generalization of the Floquet approach to a broader class of magnetic materials, involving multi-orbital situations. This requires several steps of improvements in efficiency, both on a formal level when finding time-saving approximations, but also on the implementation side. Once successful, this will enable applications of the theory to ultrafast dynamics of essentially any correlated materials (transition metal oxides, actinides and lanthanides), and in addition to spectroscopic data, we will focus on conductivity and magnetism.

This sub-project will require 1 postdoc.

(d) *Exchange interactions from TD-DMFT*

Some time ago, theory of exchange interactions of strongly correlated systems [13] was generalized to the non-equilibrium case [84]. The method was developed by the Hamburg and Nijmegen groups, and started with the (multi-band) Hubbard model, for which local moments and classical fields were introduced, that describe the rotations of the magnetization direction. The fermionic degrees of freedom were integrated out, and the resulting effective action was compared to that of a non-stationary classical spin model. Several interesting features were found that relate to the lack of time-reversal symmetry, e.g., a new term in the effective spin action which was named *twist exchange*. This work is just in its infancy and **FASTCORR** will extend these initial efforts to realistic materials specific situations. Such work requires a lot of additional developments, both analytical and computational. In particular, it is necessary to (1) perform realistic calculations based on the time-dependent DMFT (using the methods from the project (a) and (c)). Initially this will be made within the Hubbard model, but multi-orbital, materials specific calculations are the ultimate goal. (2) To calculate the twist exchange within the Hubbard model and to study its effects on spin dynamics. (3) To describe experimental results on magnetization dynamics on the ultra-fast time-scale, one of the main focus points of **FASTCORR**, it is necessary to go beyond the semi-classical Langevin equation. We will in this sub-project study *colored* noise terms that originate from the “quantum-quantum” part of the effective action, and will compare these results to those of the semiclassical Langevin equation, that describes spin dynamics with random thermal torques [85]. We will study the effects of colored noise on spin dynamics, especially for weakly correlated/itinerant-electron

systems. (4) It will also be necessary to generalize the consideration of orbital exchange for the equilibrium case [86] to the time-dependent situation. This project is ambitious and risky, with several technically challenging aspects that require attention from both analytical and computational point of view. The results will however be important: a successful outcome will generate a microscopic theory for the laser-induced ultra-fast spin dynamics in weakly correlated/itinerant-electron ferromagnets.

This sub-project will require 1 postdoc and 1 PhD student.

(e) ***Use of AdS/CFT approach for local quench effect in strongly correlated systems***

In the regime of strong correlations, a breakdown of Fermi-liquid behavior may occur [55], resulting in the destruction of long-lived quasiparticles [56]. The conventional perturbative diagrammatic expansion, which is usually defined in the basis of freely propagating particles, becomes barely applicable in this case, posing a question of finding a more suitable approach. A powerful non-perturbative alternative to the diagram technique has emerged in the context of string theory - the holographic duality [57-59] (also known as the anti de-Sitter/Conformal field theory correspondence, AdS/CFT). Originally, it appeared as a peculiar exact duality between quantum theory of gravity of a certain type and a supersymmetric Yang-Mills theory. However, over the last decade it has evolved into a universal and flexible method that has been successfully applied to numerous problems of condensed matter physics that are too difficult for conventional quantum many-body tools [60]. The fundamental idea behind this method is that a quantum theory at large coupling constant can be reformulated in terms of a dual theory of gravity of a higher dimension, which can be made classical in a certain limit. The original quantum theory is said to be defined on the *boundary* of the dual gravitational *bulk*. All the observables of interest can then be computed by manipulating with *classical* fields in a dual gravitational background. This leads to a drastic simplification of the original problem, as it reduces to solving partial differential equations with well-known methods.

The computational and conceptual power of this approach is especially pronounced when one deals with a time-dependent setting, as the language of classical gravity turns out to be very natural to address dynamical problems at finite temperature. Settings that involve sudden quenches of different types have been extensively studied within the framework of the AdS/CFT correspondence. Holographic homogeneous global quenches were used for studying the evolution of entanglement entropy in perturbed strongly correlated systems [61], time-resolved fermionic response of non-Fermi liquids [62], and effects of chemical potential [63] and angular momentum [64] on the temporal patterns of equilibration of two-point correlation functions in deformed conformal field theory. Recently, a realistic model of laser pump-probe has been suggested and applied to demonstrate instant thermalization in densely entangled quantum matter [65]. At the same time, holographic models of inhomogeneous (and, in particular, local) quenches, which are relevant for understanding the Mahan-Nozieres-de Dominicis (MND)-like phenomena in correlated quantum materials, were considered only in an oversimplified way. Essentially, the initial studies have been limited so far to the case of conformal field theory at zero chemical potential, and only thermalization of entanglement entropy and two-point correlation functions of primary operators was analyzed. In CFT of an arbitrary dimension, point-like instant energy injection has been considered (which in the holographic language dualizes to an infalling neutral massive particle in asymptotical AdS spacetime [66]), and its effects on the patterns of entanglement and complexity of quantum states [67] were studied.

In a simpler case of 1+1-dimensional CFT, an analytically tractable model of an arbitrarily shaped inhomogeneous thermal quench has been formulated. Keeping in mind possible applications to X-ray spectroscopy of real materials, one needs to proceed further on this way by generalizing the initial, simplified models to include perturbation of chemical potential/charge density and crystal lattice structure, and computing time evolution of spectral properties (including time-resolved ARPES). We plan to achieve this goal in several steps. First, we will modify the model of Ref. [68] by taking a charged massive particle on the dual side in a semi-probe limit: we will account for its back-reaction on the electromagnetic field in the bulk, but will neglect its effect on the spacetime metric. Effectively it will open a way to study charge dynamics in the quenched theory while ignoring the related energy transfer. This can be done straightforwardly, since solving Maxwell equations in a

fixed static background is relatively simple. Two-point correlation functions at this stage will be computed geometrically in the so called geodesic approximation, which allows to obtain a rough qualitative estimate on the Green's functions. The next step would be to include the dynamics of the metric and solve the full self-consistent system of Einstein-Hilbert-Maxwell equations, thus accounting for energy transport. Once we formulate the complete holographic model and develop numerical machinery for solving it, we will study fermionic response in full detail, and in this way constructing a holographic analogue of the MND theory for correlated quantum materials. This will result in a deeper understanding of the interaction between light and correlated electrons, aiding the theoretical understanding in subprojects a)-d).

This sub-project will require 1 PhD student.

Practical implementations and efficient algorithms (WP2, main responsible Lichtenstein)

All formal theories discussed above will have to be implemented in software and computer codes that allow for materials specific calculations, under realistic conditions, i.e. similar to a laboratory situation. The numerical methods will then be used to make connection to experimental observations. We have a plethora of software that are available for us, and long-term experience with developing and using these methods. WP2 is primarily focused on making the results from the analytic efforts under WP1, useable in efficient and accurate numerical models. As an example, we note that in WP1 a) this encompasses efficient ways to identify parameters that are relevant to describe the electronic structure, with starting point from equilibrium DMFT calculations, and to pass these parameters on to the effective Hamiltonian used in the time-dependent calculations. This is also an important task of WP1 b) and c). In both cases it also becomes relevant to try to establish a self-consistency loop where information from Eqn.4 is passed back to the k-resolved lattice Green's function for realistic multi-orbital scheme. With software available to us (e.g. [87]), that enable calculations of the electronic structure of solids, both for uncorrelated and correlated electron systems, we are in a good position to implement the ideas that result in parts a)-e), described above. This also involved numerical methods of many-body physics, e.g. continuous time quantum Monte Carlo simulations, exact diagonalization, fluctuating spin-polarized T-matrix, and Hubbard-I approximation [88]. These form natural building blocks which will be used as starting points for our implementations of the theories that evolve from projects a)-e).

One great numerical challenge tackled in WP2 is related to the efficient implementations of non-equilibrium dynamics within DFT+DMFT, as described in WP1. The situation is reminiscent of the first attempts of equilibrium DMFT, starting from a simple orbitals diagonal representation of three-band doped Mott insulator [89] and iterative perturbation theory for the scalar Coulomb interaction [90]. Then moving in the direction of general orbital-matrix, the down-folded Hamiltonian scheme with the Racah-tensor form of the local electron interactions, the vertex and correct multi-orbital fluctuating spin-polarized T-matrix approximation or the atomic-like Hubbard-I approximation, was used to solve the DMFT problem [91]. The new generation of the numerically exact impurity solvers of DMFT is related with developments of the CT-QMC upgrade, and the DFT+DMFT scheme is known to have predictive power of correlated electronic structures [76]. Corresponding interface exists for main electronic structure codes, like Wien2k, VASP and RSPt, with CT-QMC quantum solvers as implemented in the ALPS (Zurich) or TRIQS (Paris)[92] software.

Unfortunately, on the Keldysh contour for non-equilibrium DMFT scheme the CT-QMC quantum solver, both in the mode of an interaction expansion (CT-INT) or a hybridization expansion (CT-HYB), has unacceptable dynamical sign problem, which discards the original CT-QMC scheme for time-dependent calculations [22]. Recently, possible solutions were proposed for CT-HYB scheme related with inchworm QMC [93] and Keldysh-diagrammatic QMC for CT-INT [93]. The latter scheme can be easily generalized to the multi-orbital case for the non-equilibrium DFT+DMFT. The new generation of time-dependent CT-QMC inchworm solvers are extremely time-consuming and still in a very early stage of developments. With our expertise in continuous-time Monte Carlo it may be possible to increase the efficiency of this method for single-band case by analytical integrating out one spin component of the fermionic propagator. For this case, we will also implement an alternative method based on the exact expansion of the evolution operator in terms of Chebyshev polynomials [52], or Krylov space for time-dependent Lanczos scheme with efficient many-body symmetrization for Hamiltonian sectors [53] (as described in WP1 a)). Like in the equilibrium case, for

weakly correlated materials with time-dependence, a natural choice is the perturbative multi-orbital fluctuating T-matrix scheme.

Technically, the much more complicated problem, which can have severe consequences in a pump-probe case, is related to the realistic description of non-local correlation effects. For the equilibrium situation, we have already developed an efficient expansion around the numerically exact DMFT solution using a dual fermion (for local interactions) [76] or dual boson (for non-local interactions) [77] method, which uses a renormalized diagrammatic expansion in terms of screened impurity two-particle vertices [77]. Starting from the non-equilibrium generalization of the dual fermion scheme [76], as outlined in WP1, we will implement the multi-orbital time-dependent dual boson method. The later approach will be important for realistic problem related with time-dependent screening and change of the effective Coulomb interactions during pump-probe experiments.

Below we describe which personnel will work under the different sub-projects of WP2:

In order to make the theoretical efforts of WP1 a) we will develop an efficient iterative diagonalization scheme for three-point correlation functions (Chebyshev and Krylov), as well as investigating the numerical stability of the generalized multi-orbital inchworm and diagrammatic QMC schemes.

Two postdocs of the Hamburg node will be employed for this task.

In order to make the theoretical efforts of WP1 b) tractable, numerically we will develop a general purpose time-dependent dual fermion and dual boson code compatible with time-dependent GW scheme for non-local interactions.

In order to implement the theoretical efforts of WP1 c) into efficient software, one postdoc and one student of the Uppsala node will be employed (the student will be jointly supervised by the Nijmegen group under a double degree agreement).

One postdoc and one student of the Hamburg node will be employed for this task.

To implement the efforts of WP1 d) into numerically efficient algorithms we will connect the formalisms in a full-potential, all-electron method, for multi-orbital calculations. This involves the twist exchange as well as the time-resolved, dynamical orbital-orbital exchange.

One senior scientist, serving as a link between WP2 and WP3, will be employed by Uppsala University (40%), spending significant time in Hamburg.

Materials simulations and connection to experiments (WP3, main responsible Eriksson)

With the methodology developed under a)-e) we will address contemporary challenges provided by the community of experimentalist active in pump-probe laser field physics, and spectroscopy at XFEL sources. The methodology developed in this project will enable the theoretical description of excitations both in the optical regime and the X-ray regime. We will consider situations that follow experimental situations as closely as possible, for instance by inducing excitations of the valence band states using intense light pulses, treated with the methods described in WP1 and WP2. With these methods, we will be able to follow the optical and magnetic response, with ambition to address questions connected to e.g. photo-induced metal-insulator transition in Mott systems. This includes the experiments of $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$, shown in Fig. 5.

Other experimental works that have drawn our attention are the reports by Först *et al.* [94], where it was shown that optical excitations of the LaAlO_3 substrate via mid-range infrared pulses could excite a phonon band. This, in turn, induces the ultrafast formation of a 2D electron gas at the interface with NdNiO_3 . The propagation of electrons into the NdNiO_3 substrate was shown to initiate a metal-to-insulator (MIT) transition. Using our theoretical tools, we will identify the microscopic origin of the formation of the 2D electron at the interface, the timescale of the transition process and its dependence on the intensity and

duration of the pulse. We shall also look into other systems that are close to MIT and are therefore highly susceptible to lattice perturbations, e.g. manganites and nickelates.

It is also of interest to study how the coupling between the lattice and the electronic and/or magnetic degrees of freedom affects the speed of the light-induced MIT. There has been a number of experimental studies on e.g. $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$ [47], Fe_3O_4 [46] and VO_2 [45]. The physical understanding gained from these investigations will hopefully provide us fundamental insights into the mechanisms and switching times of oxide electronics. Furthermore, we will investigate the magnetization dynamics following photoexcitation in correlated oxides, such as ErFeO_3 (experiments by Nova et al. [95]), Sr_2IrO_4 (experiments by Dean et al [96]) and CoO (experiments by Zheng et al. [97]). Finally, it is worth noting that the ability of optical excitations to quench the ferromagnetism of metallic systems was discovered over 20 years ago, but still lacks a convincing and comprehensive microscopic explanation. In order to analyze this phenomenon, we will initially investigate systems like Ni, Ho, and EuTe, due to the experimental efforts that have been made for these materials.

Some of the methods developed under WP1 and WP2 will only be applicable for systems where a limited number of active (correlated) orbitals can be treated. There are several known examples of materials for which this is a perfectly realistic description, e.g. for manganites, in the perovskite structure, it is often the e_g orbitals that are treated in the correlated scheme, while the t_{2g} states are considered localized and not relevant for a multi-configurational treatment. There are other materials that have only one active orbital, that become especially suitable for effective many-body models. Among them one should mention impurities of IV-group elements (Sn and Pb) at the surfaces of semiconductors Si(111) [73,74] and SiC(0001) [75]. In all these materials, a narrow, strongly correlated impurity band is formed within the semiconducting gap and a simple single-band description is possible. Some preliminary results have been reported and are quite promising, where in particular, a possibility to create a nano-skyrmion mosaic for ultra-dense magnetic storage of information has been suggested [74]. We will explore these systems initially with the methods outlined under WP1 and WP2, but as detailed below, we will actively try to also find other materials, in form of compounds, surfaces and interfaces, that have correlation effects primarily attached to a subset of the number of possible orbitals. We will, as described below, use data-mining algorithms [98] in these efforts, since they have been proven to reliably predict materials with desired functionality or character.

In data-bases of electronic structures, all necessary information of the angular momentum projected character of the electronic structure is provided for known, large groups of materials (tens of thousands). In fact, we were very early in these endeavors, and the data in <http://gurka.fysik.uu.se/ESP/> (below we will use the name Uppsala Electronic Structure Database - UESD), generated by the Uppsala group, represents ab-initio calculations of the electronic structure of approximately half of the materials listed in the International Crystallographic Structural Database (ICSD). ICSD reports structures of approximately 160.000 compounds and when combined with electronic structure information, offers a fantastic source of identifying materials with desired functionality or with a specific electronic structure. In the past we have used UESD to identify materials with specific band gap [98], new two-dimensional (2D) systems [99], and even made predictions of possible new oxide superconductors [100]. The predictions of new 2D systems have been verified experimentally with several examples [101,102].

We will in WP3 design search criteria than go through all the electronic structures listed in UESP, to find materials with electronic structures similar to those of correlated perovskites (materials with two correlated orbitals) and also to find materials with correlations in one single band, similar to the systems mentioned above for impurities at the surfaces of semiconductors. We note that transition metal oxides form in a wide range of crystal structures; Pyrochlore, Ruddelsen-Popper, and Garnets, to mention a few, which provides ample material space where one might find systems that have only a few bands that need a correlated DMFT treatment. The search we aim to do will naturally involve all oxides, but in fact materials with any chemical composition may be found relevant in this search. The optimal search criteria will be explored and form part of this investigation, but a first attempt will be to find few-band features, primarily of d - or f -character, in a gap or pseudo-gap of the electronic structure. For materials identified in such a screening, more advanced calculations of the electronic structures will be performed, first using DMFT to explore multi-configurational aspects of the electronic structure, and to gauge how important correlation effects of the compound is. We note that current DMFT implementations available to us are expedient and allow for investigations of materials with general crystal symmetry and large unit cells. As an example, we mention that in Ref.103 the

hybridization function (a critical parameter of DMFT) of supercells involving close to 100 atoms was reported. For the materials that seem most interesting and promising, we will finally use the methods of TD-DMFT, as developed under WP1 and WP2. In this way, we hope to identify new correlated materials, exhibiting novel response to a high intensity photon-field. The connection to experimental partners that may confirm or refute the theoretical findings and predictions is important, and we will actively seek collaborations among existing and new experimental colleagues.

One senior scientist (80%) employed at Uppsala will work on data-mining efforts and numerical calculations, as well as interaction to international experimental colleagues.

The ultimate goal of **FASTCORR** is to perform investigation for material with any type of multi-orbital correlations. These investigations will most likely come at the end of the time-period of **FASTCORR**, when the theoretical machinery has developed into a computationally efficient tool. Once the multi-orbital methods are available, we will put special emphasis on complex oxides and the photo-induced metal-insulator transition in Mott system. The methods of WP1 and WP2 will also allow us to study the light-induced demagnetization and remagnetization of magnetic materials [32,33], and to study how the influence of electron correlations may change the time-evolution of the magnetism. Examples of systems we will investigate here are fcc Ni, permalloy, Co₂MnGe Heuslers, lanthanides and complex oxides.

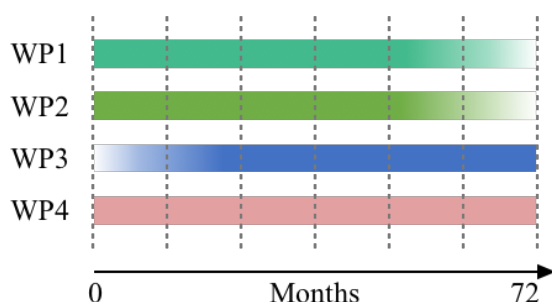
Throughout WP3 we will be open to collaborations with experimentalists in the field, and we will be eager to help analyzing new experimental results that will most certainly appear at laboratories over the world, during the project period.

One postdoc and one senior scientist (40%) will be engaged in this work, once the theoretical machinery developed under WP1 and WP2 is working.

Project coordination (WP4, main responsible Eriksson)

The efforts of WP4 are connected to practical aspects of carrying out the project, planning for longer visits among the personnel for the different nodes (Uppsala, Nijmegen and Hamburg), and for decision making for the how to manage the project. Risk analysis will be made continuously in this work package, combined with decisions for how to focus efforts on specific, technical problems. This is needed, in order to handle unexpected obstacles in WP1, WP2 and WP3. In WP4, the need for longer collaborative visits between the nodes of the proposal will be analyzed and decided. In addition, two planned topical workshops (after 36 months and after 72 months) will be organized under the umbrella of **FASTCORR**, and WP4 will deal with decisions on venue, time and topics of these events. Since the applicants of **FASTCORR** have already demonstrated the ability to interact scientifically in larger collaborative efforts, e.g. in EU funded consortia, WP4 is foreseen to run smoothly, without major risks.

The efforts spent during the project-time in the different work packages is schematized in the figure below. The activities of each work package are presented such that a darker color indicates a higher effort spend in a work package. As shown, WP1 and WP2 will require a more intense effort in the earliest part of the project, while WP3 will require an increasing effort only after an initial stage will have been completed. The reason is that some of the efforts in WP3 will rely on developments made in WP1 and WP2. Nevertheless, this should not be taken as an indication of two consecutive phases, since the feedback effects between the theoretical developments and the materials modeling in connection to experimental work will be fundamental across the full length of the project. WP4 will have full intensity during the whole project period.



For the personnel employed in FASTCORR we will in WP4 take efforts to ensure shared supervision of postdocs and PhD students. For the latter category we will explore the so called double-degree program, that enables two universities to each graduate one common student. This has already been explored by the PIs (e.g. Katsnelson and Eriksson) and is a good opportunity for two research units to commonly foster the next generation scientists, and

where all partners can reap the benefits of this effort. As a final remark of this section we note that among the applicants behind this project, Katsnelson is mostly responsible for WP1, Lichtenstein has main focus on WP2, while Eriksson will primarily be responsible for WP3 and WP4. One must bear in mind that this is just a rough division of responsibilities in the project, as everything will be done in intense collaboration.

Synergies and strategic aspects

FASTCORR will meet scientific challenges by engaging in the development of mathematical and physical models combined with numerical methods that are coupled to efficient implementations and high performance calculations. 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 (illustrated also in Fig.2 of B1). 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 geographic placement of the nodes of this project with respect to the European XFEL provides a strategic advantage in terms of vicinity and contact to the experimental activities that **FASTCORR** are expected to be relevant for. **FASTCORR** will have an advantage in establishing contacts to first-rate experimental work concerning measurements of strongly correlated electron systems that are investigated with high intensity light sources, in the ultra-fast regime.

Feasibility

The applicants of **FASTCORR** have extensive experience in leading big research teams and to collaborate in larger research efforts and consortia. They have the technical skill to lead the scientific endeavors of **FASTCORR**, and have in addition established ability to collaborate with each other, over larger periods of time. This established collaboration involves efforts that both concern solving scientifically demanding problems as well as in education and supervision of younger scientists. The applicants of **FASTCORR** have in fact shared PhD supervision on numerous accounts, e.g. for the doctoral degrees of A. Grechnev (2005), O. Wessely (2006), D. Yudin (2015), L. Peters (2015), and I. Locht (2017). The last student in this list is an example of a double-degree examination between Uppsala University and Nijmegen University. Examples of past collaboration that is relevant for the proposal are:

- Development of the theory of exchange interactions in solids and crystals (Lichtenstein + Katsnelson) [22-25].
- Calculation of the exchange interactions for real materials (Eriksson + Lichtenstein + Katsnelson) [104-106].
- Development of spin dynamics (Eriksson + Katsnelson) [32,33].
- Development of DFT+DMFT (Lichtenstein + Katsnelson) [12].
- Implementation of DFT+DMFT for real materials (Eriksson + Lichtenstein + Katsnelson) [12,107].
- Development of dual fermion and dual boson theory (Lichtenstein + Katsnelson) [76,77].
- Dynamical core screening effects in X-ray spectra (Eriksson + Katsnelson) [108,109].

This creates a solid background for the success of **FASTCORR**. We note that the here proposed research plan is by no means an extension of an ongoing project, but represents an entirely novel effort.

Section c. Resources (including project costs)

(Note: Describe the resources needed according to the indications in the *Information for Applicants to the Synergy Grant 2019 call, section 2.3 The research proposal*).

Cost category		Corresponding PI	2 nd PI	3 rd PI	Total in euro	
PI name		Olle Eriksson	Mikhail Katsnelson	Alexander Lichtenstein		
Host Institution		Uppsala University	Radboud University	Hamburg University		
Direct Costs ²	Personnel	PI	322 605	567 382	330 000	1 219 987
		Senior Staff	652 632			652 632
		Postdocs	715 790	611 270	1 132 000	2 459 060
		Students	284 211	893 010	111 000	1 288 221
		Other				
	<i>i. Total Direct costs for Personnel (in euro)</i>		1 975 236	2 071 662	1 773 000	5 819 898
	Travel		100 000	87 000	48 000	235 000
	Equipment			50 000		50 000
	Other goods and services	Consumables	20 000	50 000	12 000	82 000
		Publications	30 000	30 000	30 000	90 000
		Other (conferences and WSs, final audits)	40 000	40 000	40 000	120 000
	<i>ii. Total Other Direct Costs (in euro)</i>		190 000	257 000	130 000	577 000
	A – Total Direct Costs (i + ii) (in euro)		2 165 236	2 328 662	1 903 000	6 396 898
	B – Indirect Costs (overheads) 25% of Direct Costs (in euro)		541 309	582 166	475 750	1 599 225
C1 – Subcontracting Costs (no overheads) (in euro)						
C2 – Other Direct Costs with no overheads (in euro)						
Total Estimated Eligible Costs (A + B + C) (in euro)		2 706 545	2 910 828	2 378 750	7 996 123	
Total Requested Grant (in euro)		2 706 545	2 910 828	2 378 750	7 996 123	

The total amount of requested EU contribution for the **FASTCORR** project is **7 996 123 Euro**, of which the majority is to cover salaries for team members. PI Eriksson will devote 30% of his time to the project, and the PIs Katsnelson and Lichtenstein 50%, respectively, and the salaries will accordingly cover these contributions. Prof. Lichtenstein will become Emeritus during the project duration, and, therefore, his

Emeritus salary will be decreased in comparison to current salary level, which is reflected by the personnel cost given in the budget table. Under the supervision of the PIs, 19 team members will contribute to the project (table below). This table summarizes the contribution of each institution and their related work packages. Three Senior Researchers (SRs) will be working in Eriksson's team, and there will be PostDocs and PhDs in the PI teams. The Senior Researchers will contribute 80% (1 SR) and 40% (2 SRs) to the project, and all PostDocs and PhDs will devote full time (100%) to the project.

We have calculated 50 000 Euro for a local computer cluster to be placed in Nijmegen, Netherlands, to be administered by Prof. Katsnelson. The work supervised by Prof. Katsnelson involves significant algebraic operations, using software such as Mathematica. The cluster will be used for these endeavours.

For consumables, we have calculated a total amount of 82 000 Euro to cover laptops and software licenses for the local computer cluster, of which the majority will be spent in the Nijmegen due to the cost of software licenses needed for their work.

For travelling, we have calculated a total cost of 235 000 Euro for the six years in this project. Dissemination at conferences is needed, and travel costs to conferences are included herein. Moreover, with intense collaboration between the three nodes, travel among personnel is vital. Meetings will be predominantly held in Hamburg due to the central location and proximity to experimental facilities such as XFEL and DESY. We plan to hold a kick-off meeting in Hamburg including PIs and the three senior researchers.

In the post "Other goods and services", we have included costs to cover conferences, workshops and final audit. Open access publication provides a significant cost, and we foresee a large amount of publications emanating from the project. We plan to arrange bi-annual workshops for development and application of the software used, in line with what has been previously done with the software packages developed by the Uppsala team. We have included costs for open access fees, publication fees and costs for dissemination and outreach. Finally, there will be three final audits, one per host institution.

The majority of the computational effort will be performed in Uppsala University, where Prof. Eriksson has extensive allocations with the Swedish National Infrastructure for Computing.

Institution/PI	Personnel	Contributions (in WPs)
Uppsala University/Eriksson	PI Eriksson	WPs 2, 3, 4
	3 SR (80%, 40%, 40%)	WPs 2, 3
	2 PostDocs (2*100%)	WPs 2, 3
	1 PhD (100%)	WPs 2, 3
Radboud University/Katsnelson	PI Katsnelson	WPs 1, 2, 4
	3 PostDocs (3*100%)	WPs 1, 2
	4 PhDs (4*100)	WPs 1, 2
University of Hamburg/Lichtenstein	PI Lichtenstein	WPs 1, 2, 3, 4
	3 PostDocs (3*100%)	WPs 1, 2, 3
	1 PhDs (1*100%)	WPs 1, 2

Table summarizing the contribution of each institution and their related work packages.

References

(references that include the applicants in blue)

1. P. Hohenberg and W. Kohn, Phys. Rev. **136**, B864 (1964); W. Kohn and L. J. Sham, Phys. Rev. **140**, A1133 (1965).
2. W. Metzner and D. Vollhardt, Phys. Rev. Lett. **62**, 324 (1989); A. Georges et al., Rev. Mod. Phys. **68**, 13 (1996).
3. V. I. Anisimov et al, J. Phys.: Cond. Mat. **9**, 7359 (1997); G. Kotliar et al., Rev. Mod. Phys. **78**, 865 (2006).
4. A. I. Lichtenstein and M. I. Katsnelson, Phys. Rev. B **57**, 6884 (1998).
5. L. D. Landau, Sov. Phys. JETP **3**, 920 (1957); *ibid.* **5**, 101 (1957); *ibid.* **8**, 70 (1959).
6. I. L. M. Locht et al, Phys. Rev. B **94**, 085137 (2016).
7. O.Eriksson et al. "Theory of diluted magnetic semiconductors", in *Advanced Functional Materials*, 1st Edition A Perspective from Theory and Experiment, editor O.Eriksson and B.Sanyal (Elsevier, Amsterdam, 2012).
8. P. Thunström et al. Physical Review B **79**, 165104 (2009).
9. L. V. Pourovskii, M. I. Katsnelson, and A. I. Lichtenstein, Phys. Rev. B **73**, 060506 (R) (2006).
10. A. B. Shick et al, Phys. Rev. B **87**, 020505(R) (2013).
11. M. I. Katsnelson and A. I. Lichtenstein, J. Phys.: Cond. Mat. **11**, 1037 (1999).
12. A. I. Lichtenstein, M. I. Katsnelson, and G. Kotliar, Phys. Rev. Lett. **87**, 067205 (2001).
13. M. I. Katsnelson and A.I. Lichtenstein, Phys. Rev. B **61**, 8906 (2000).
14. See e.g. A. Kirilyuk, A. V. Kimel, and T. Rasing, Rev. Mod. Phys. **82**, 2731 (2010).
15. A. Doerr, Nature Methods **15**, 33 (2018).
16. L. P. Kadanoff and G. Baym, *Quantum Statistical Mechanics* (Benjamin, 1964).
17. J. Rammer and H. Smith, Rev. Mod. Phys. **58**, 323 (1986).
18. A. Kamenev, *Field Theory of Non-Equilibrium Systems* (Cambridge Univ. Press, 2011).
19. M. I. Katsnelson et al, Rev. Mod. Phys. **80**, 315 (2008).
20. G. Sanches-Barriga et al, Phys. Rev. Lett. **103**, 267203 (2009).
21. C. Carbone et al, Phys. Rev. Lett. **104**, 117601 (2010).
22. For a review on time dependent DMFT see H.Aoki et. al Rev. Mod. Phys. **86**, 779 (2014).
23. S. Y. Savrasov, G. Kotliar, and E. Abrahams, Nature **410**, 793 (2001)
24. K. Held, A. K. McMahan, and R. T. Scalettar, Phys. Rev. Lett. **87**, 276404 (2001)
25. A. I. Lichtenstein, M. I. Katsnelson, and G. Kotliar, Phys. Rev. Lett. **87**, 067205 (2001)
26. A. Grechnev et al., Physical Review B **76**, 035107 (2007); O. Granas et al., Computational materials science **55**, 295-302 (2012); J. Minar et al., Phys. Rev. B **72**, 045125 (2005); M. Aichhorn et al., Phys. Rev. B **80**, 085101 (2009); K. Haule et al., Phys. Rev. B **81**, 195107 (2010)
27. E. Gull, A. J. Millis, A. I. Lichtenstein, et al. Rev. Mod. Phys. **83**, 349 (2011)
28. P. Thunstrom et al., Physical review letters **109**, 186401 (2012)
29. I. Di Marco et al, Nature Comm. **4**, 2645 (2013)
30. J. Lüder et al., Physical Review B **96**, 245131 (2017)
31. A. Hariki et al., Phys. Rev. Lett. **121**, 126403 (2016)
32. V. Antropov et al. Phys. Rev. B **54**, 1019 (1996).
33. O. Eriksson, A. Bergman, L. Bergqvist, and J. Hellsvik, *Atomistic Spin-Dynamics; Foundations and Applications* (Oxford Univ. Press, 2017).
35. A. I. Lichtenstein et al, J. Magn. Magn. Mater. **67**, 65 (1987).
35. S. Paul et al. Phys. Rev. B **97**, 125120 (2018).
36. S. Wall et al., Nat. Phys. **7**, 114 (2011).
37. T. Ogasawara et. al, Phys. Rev. Lett. **85**, 2204 (2000).
38. F. Schmitt et al., Science **321**, 1649 (2008).
39. E. Beaurepaire et al., 1996, Phys. Rev. Lett. **76**, 4250.
40. H. Ehrke et al., Phys. Rev. Lett. **106**, 217401 (2011).
41. H. Ichikawa et al., Nature Mater. **10**, 101 (2011).
42. A. Schiffrin et al. Nature **493**, 70, (2013).

43. M. Schultze et al. *Nature* **493**, 75, (2013).
44. D. Popmintchev et al. *Phys. Rev. Lett.* **120**, 093002 (2018).
45. M. F. Jager *et al.* *Proceedings of the National Academy of Sciences* **114**, 9558–9563 (2017).
46. S. de Jong *et al.* *Nature Materials* **12**, 882–886 (2013).
47. J. Zhang et al *Nat. Mat.* **15**, 956 (2016).
48. V. Molodtsov et al. *Phys. Rev. Lett.* **115**, 04601 (2015).
49. O. Rader, A. Varyakhalov, J. Fink et al. *Phys. Rev. Lett.* **103**, 267203 (2009).
50. P. Gambardella et al. *Phys. Rev. Lett.* **104**, 129901 (2010).
51. F. Aryasetiawan et al, *Phys. Rev. B* **70**, 195104 (2004); T. O. Wehling et al, *Phys. Rev. Lett.* **106**, 236805 (2011).
52. S. Yuan, H. De Raedt, and M. I. Katsnelson, *Phys. Rev. B* **82**, 115448 (2010).
53. M. Balzer et al, *J. Phys.: Condens. Matter* **24**, 035603 (2012).
54. P. Seth et al, *Comp. Phys. Comm.* **200**, 274 (2016).
55. C. Varma et al., *Phys. Rev. Lett.* **63**, 1996 (1990).
56. B. Meszner et al., *Phys. Rev. B* **94**, 115134 (2016).
57. J. Maldacena, *Int. J. Theor. Phys.* **38**, 4 (1999).
58. S. Gubser et al., *Phys. Lett. B* **428**, 105 (1998).
59. E. Witten, *Adv. Theor. Math. Phys.* **2**, 253 (1998).
60. J. Zaanen, Y.-W. Sun, Y. Liu, and K. Schalm, *Holographic Duality in Condensed Matter Physics* (Cambridge Univ. Press, 2015).
61. V. Balasubramanian et al, *Phys. Rev. D* **84**, 026010 (2011).
62. N. Callebaut et al., *JHEP* 10 (2014) 172.
63. E. Caceres, A. Kundu, *JHEP* 09 (2012) 55.
64. I. Aref'eva et al., *JHEP* 07 (2013) 1.
65. A. Bagrov et al., *Phys. Rev. D* **97**, 086005 (2018).
66. L. Randall, L and R. Sundrum, *Phys. Rev. Lett.* **83**, 3370 (1999).
67. D. Ageev et al, *JHEP* 08 (2018) 071.
68. M. Nozaki, T. Numasawa, and T. Takayanagi, *JHEP* 05 , 080 (2013).
69. A. P. Itin and M. I. Katsnelson, *Phys. Rev. Lett.* **115**, 075301 (2015).
70. E. A. Stepanov, C. Dutreix, and M. I. Katsnelson, *Phys. Rev. Lett.* **118**, 157201 (2017).
71. C. Dutreix and M. I. Katsnelson, *Phys. Rev. B* **95**, 024306 (2017).
72. A. Secchi et al, *prb dynamical suscept*
73. V. V. Mazurenko et al, *Phys. Rev. B* **94**, 214411 (2016).
74. E. A. Stepanov et al, *arXiv:1710.03044*
75. D. I. Badrtdinov et al. *arXiv:1804.00440*
76. A. N. Rubtsov, M. I. Katsnelson, and A. I. Lichtenstein, *Phys. Rev. B* **77**, 033101 (2008).
77. A. N. Rubtsov, M. I. Katsnelson, and A. I. Lichtenstein, *Ann. Phys. (NY)* **327**, 1320 (2012).
78. G. Rohringer et al, *Rev. Mod. Phys.* **90**, 025003 (2018).
79. T. Miyake et al, *Phys. Rev. B* **77**, 085122 (2008).
80. D. Yudin et al, *Phys. Rev. Lett.* **112**, 070403 (2014).
81. E. G. C. P. Van Loon et al, *Phys. Rev. Lett.* **113**, 246407 (2014).
82. E. A. Stepanov et al, *Phys. Rev. Lett.* **121**, 037204 (2018).
83. E. G. C. P. Van Loon, M. I. Katsnelson, and M. Lemeshko, *Phys. Rev. B* **92**, 081106 (2015); E. G. C. P. Van Loon et al, *Phys. Rev. B* **93**, 195145 (2016).
84. A. Secchi et al, *Ann. Phys. (NY)* **333**, 221 (2013).
85. C. Etz et al. *J. Physics. Cond. Matter.* **27**, 243202 (2015).
86. A. Secchi et al, *Ann. Phys. (NY)* **360**, 61 (2015).
87. J. M. Wills et al. *Full-Potential Electronic Structure Method* (Springer, Berlin, 2010).
88. E. Gull et al, *Rev. Mod. Phys.* **83**, 349 (2011).
89. I. A. Nekrasov et al, *Eur. Phys. J. B* **18**, 55 (2000).
90. M. S. Laad et al, *Phys. Rev. Lett.* **91**, 156402 (2003).
91. E. Gorelov et al, *Phys. Rev. B* **82**, 085117 (2010).
92. O. Parcollet, et al., *Comp. Phys. Comm.* **196**, 398 (2015).
93. G. Cohen et al, *Phys. Rev. Lett.* **115**, 266802 (2015); R. Porfumo et al, *Phys. Rev. B* **91**, 245154 (2015).
94. M. Först et al., *Nature Materials* **14**, 883 (2015).
95. T. F. Nova et al., *Nature Physics* **13**, 132–136 (2016).
96. M. P. M. Dean et al., *Nature Materials* **15**, 601–605 (2016).

97. Z. Zheng et al., Physical Review B **98**, 134409 (2018).
98. C. Ortiz, O.Eriksson, and M.Klintenberg, Computational Materials Science **44**, 1042 (2009).
99. S. Lebegue et al. Phys. Rev. X **3**, 031002 (2013).
100. M. Klintenberg and O.Eriksson, Computational Materials Science **67**, 282 (2013).
101. F.B. Romdhane et al., Small **11**, 1253 (2015).
102. M.W. Lin et al. J.Mat. Chem. C **4**, 315 (2016).
103. H. Herper et al., *A polaron cloud of correlated electron states in ceria* (arxiv)
104. Y. O. Kvashnin et al, Phys. Rev. B **91**, 125133 (2015).
105. S. Keshavarz et al, Phys. Rev. B **92**, 165129 (2015).
106. Y. O. Kvashnin et al, Phys. Rev. Lett. **116**, 217202 (2016).
107. A. Grechnev et al, Phys. Rev. B **76**, 035107 (2007).
108. O. Wessely, M. I. Katsnelson, and O. Eriksson, Phys. Rev. Lett. **94**, 167401 (2005).
109. O. Wessely, O. Eriksson, and M. I. Katsnelson, Phys. Rev. B **73**, 075402 (2006).