Research Project

Correlations and coherence in quantum materials and structures (CCQM) - unique properties on macro and nano scales

1. A brief summary

The principal aim of this 3-year project is to develop a novel *quantitative* (or *semiquantitative* in complex systems, *see below*) unified and mutually consistent approach and to apply it to a wide class of quantum materials and structures. The systems range from those close to the metal-semiconductor (of Mott type) to the strongly correlated systems, where spectacular phenomena of new type such as unconventional and/or high-temperature superconductivity or spin-dependent heavy masses of electrons or quantum critical phenomena, are all taking place. These phenomena bear their origin in microscopic correlations on atomic (nano) scale, so therefore our method will be tested on all scales from nano to macro. The question of an adequate description of those systems presents itself as a fundamental problem for the present-day condensed matter and materials science, and thus the approach consists of applying a novel quantum and thermodynamic description to new materials and structures, some of which (e.g. high temperature superconductors or correlated-metal thermoelectrics) have very good prospects for applications. The starting point of the project is formulation by our group in the last 12 months or so of a new statistically consistent approach [1] to the correlated fermions in the whole range of interactions (with respect to single-particle energy) and its subsequent application to high-temperature superconductivity [2, 3], to an unconventional superconductivity in heavy fermion systems [4, 5, 6], and to quantum critical behavior of the wave function [7]. Within the project framework we plan to apply our original approach to concrete materials, as well as to extend it to the fermions/bosons in optical lattices, and to the model quantum liquid - condensed helium 3, in both normal state close to the quantum solidification and to its superfluid phases. In such manner, we would like to demonstrate a degree of universality of our approach in describing normal, superconducting, and quantum critical properties. As an additional result, the project principal investigator proposes including a monograph "*Correlated Fermion Systems*" to be finalized for a renowned international publisher.

2. Motivation for the project: correlated quantum matter

Strongly correlated electronic systems encompass the systems ranging from magnetic insulators/semiconductors through correlated metals undergoing insulator - metal (Mott) transition [8] to the strongly correlated systems incorporating both high temperature superconductors [9] and metals with very heavy electrons (*heavy fermion compounds* [10]). They contain the systems for which the XX-century paradigms - the concept of electron gas, Landau-Fermi liquid, and the Bardeen-Cooper-Schrieffer theory of superconductivity, are not entirely adequate to describe their physico-chemical and materials properties. Instead, the concepts of nonstandard quasiparticles, real-space pairing induced by electronic correlations, non-Fermi (non-Landau) Fermi-liquid or quantum critical behavior, all have been observed and await a coherent description within the emerging theory of materials and systems with strong electronic correlations. It is fair to say that formulation of the theory of strongly correlated systems is one of the most fundamental problems not only in condensed matter/materials physics, but also in other disciplines such as quantum chromodynamics [11], astrophysics [12], and atomic physics in optical/laser lattices [13]. This is because in all those disciplines we deal with situations, in which the interaction among particles is comparable or even much larger than the characteristic, Fermi or single-particle energy of constituent particles or whole atoms [14]. In effect, we cannot start from an ordinary addition of single particles to form a condensed-matter or even nanoscopic systems, as sizable if not predominant interparticle correlations appearing already on a nanoscale determine the system quantum and thermodynamic behavior. Such correlated state composes a true *correlated quantum matter*. In such situation, the

Hartree-Fock, or band theory, or perturbational approaches regarded as standard methods become insufficient, if not simply inapplicable. On the other hand, modeling those systems on a nanoscale with the help of advanced computer techniques such as Quantum Monte Carlo or exact diagonalization methods, allow minimally to test specific approximate methods that are applicable to quantum condensed macro systems or the systems on different scale (of various dimension), description of which is our main goal. Parenthetically, formulated by us earlier rigorous mathematical treatment [15] serves as a proper description of intrinsic nanosystems [7], but has a rather limited application to larger systems unless not properly reformulated.

To provide motivation for this project unique features is thus relatively simple. First, the research on strongly correlated electronic and atomic systems represents a forefront of condensed matter physics and materials science, and is very important in other disciplines. This concerns above all a lack of complete theoretical description and requires an advanced computer modeling of these complex materials. The exemplary unexplained so far phenomena are: (i) the nature of high temperature superconductivity, (ii) the mechanism of heavy-fermion superconductivity and its coexistence with antiferromagnetism, (iii) the quantum criticality and new quantum-liquid states, and (iv) modeling of correlated and condensed states in optical lattices.

In response to these challenging problems, the following topics are to be tackled: (i) the formulation of our statistically consistent approach (SGA) to correlated systems with inclusion of both mean-field and quantum-fluctuations aspects of the concrete materials description, (ii) the detailed studies of real-space pairing based on high-temperature and heavy-fermion systems, together with comparison with experimental results, (iii) formulation and elaboration of a new method of incorporating correlations into the electronic structure calculations of nano and macro systems, and (iv) extension of our SGA method and computer simulations to fermions and bosons in optical lattices.

3. Research methodology: problems and methods of approach

The unique contribution of our team to the whole discipline of strongly correlated systems relies on four specific subjects. We characterize first our research methodology and its consequences for the future work. Rather than elaborating on our specific methodology, we illustrate its strength with just obtained results in model situations.

3. 1. Statistically-consistent approach: high-Tc superconductors

Our main effort at the moment concentrates on the formulated very recently [1] new **statistically consistent approach** (SGA) to the theory of correlated fermions. It relies on an essential extension of the Gutzwiller and related approaches [1] to achieve a mutual consistency of quantum and statistical (thermodynamic) aspects of the quantitative description of those materials which cannot be described by standard methods. In Figs. 1 and 2 we exhibit exemplary just obtained results concerning selected properties of high temperature superconductors in the so-called overdoped regime [3]. These are the first such successful theoretical results for the condensed macro state obtained with the help of our new (SGA) method, which is also a subject of the upcoming Ph. D. Thesis [16] . It seems to be truly amazing, that the so-called *t-J* model introduced by Spałek *et al* [17] long time ago (at the very beginning of the team leader carrier) and elaborated within the new method [1] can describe with one free parameter such a complex situation. The principal nontrivial feature of the method is to introduce not only renormalization of the microscopic parameters by the correlations, but also to introduce a set of self consistent effective fields in that situation, all in such a manner that the complete physical solution can be carried out in an explicit manner.

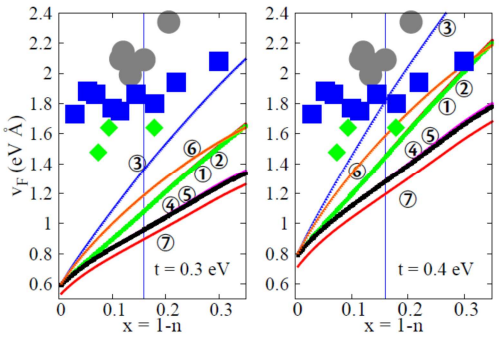

Fig. 1. Doping dependence of the Fermi velocity in the so-called nodal (0, 0) → (π, π) direction for high-T_c superconducting cuprates. The curves are labeled according to different level of theoretical analysis (for details see [3]). The hopping parameter *t* specified.

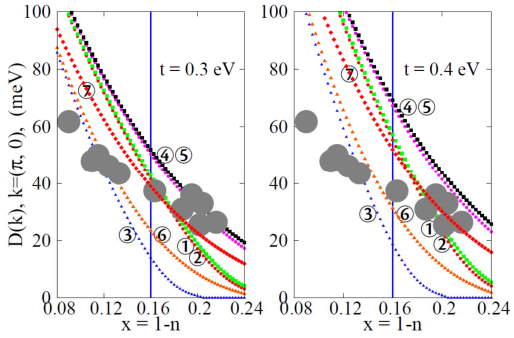

Fig. 2. Doping dependence of the superconducting gap $D_{\mathbf{k}}$ at **k** = (0, π) and for the high-T_c superconductors; experimental points - solid circles (for details see [3]). The vertical line in the Figures marks the optimal doping.

3. 2. Heavy-fermion systems: new superconducting-state description

Associated with this research is our second recent result within the SGA (and related) methods: description of an **unconventional superconductivity in the strongly correlated systems with heavy masses** [4-6]. Namely, one of the current questions is to describe the long-sought Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) superconducting phase discovered experimentally [18], most probably mixed with antiferromagnetic ordering [19]. In Figs. 3 and 4 we illustrate why introduced by us some time ago an original concept of the spin-dependent effective mass [20] is instrumental in stabilizing this unconventional superconducting state. There are other additional important topics making this rather exotic state stable specifically for the correlated systems (as we explained [4, 5]) on the expense of the canonical Bardeen-Cooper-Schrieffer (BCS) superconducting state; this and related important issues is the subject of Ph. D. Thesis in progress [21]. One has to mention, that contrary to the recent analysis [22, 23], our methodology ascribes the specific role to strong electronic correlations. The research requires a thorough material-specific analysis.

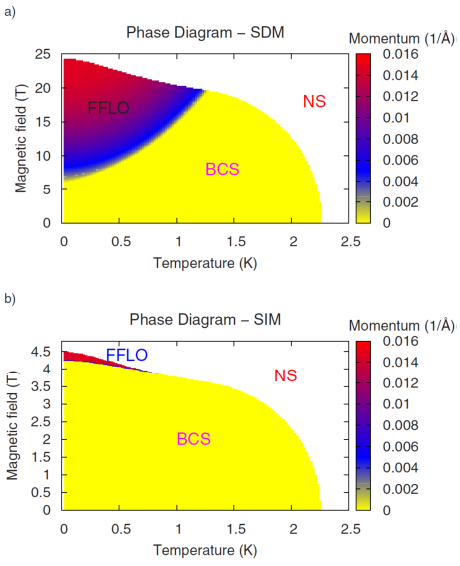

Fig. 3. Phase diagram on applied magnetic field - temperature plane for 3-dimensional correlated fluid incorporating unconventional (FFLO) superconducting phase for the system with the spin-dependent heavy masses (a) and without (b). Note that the upper critical field for the a) case (cf. [4]).

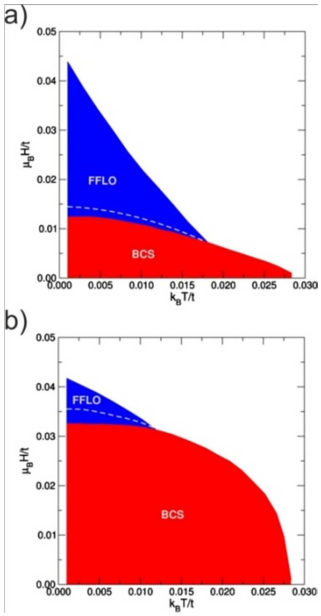

Fig. 4. Same as in Fig. 3 for two-dimensional correlated liquid. The dashed line marks the end line of the BCS existence in each situation. The axes are in units of nearest-neighbor hopping *t* (cf. [6]). Note a decisive extension of the FFLO stability for correlated systems (case a)).

3. 3. Quantum criticality at the border of localization: novel critical points

A separate, long-term interest of our group is in determining **quantum criticality at the border of Mott localization**. In this critical regime standard band-theoretical methods, such as LDA, fail. Near this border, but still on the metallic side one observes either a *nonstandard Landau-Fermi liquid*, as we proposed some time ago [24] for selected systems or a new type of quantum liquid, the so-called non-Fermi (non-Landau) liquid [10]. In this respect, we have proposed working closely with experimental groups [25, 26] a *new type of quantum critical point* separating different condensed quantum states: the (non-)Fermi liquid from a Mott or Kondo-semiconductor (insulator). In Figs. 5 and 6 we show an exemplary rationalization of experimental results achieved by us for the Verwey transition in magnetite Fe_3O_4 [25] and in doped Kondo insulator $CeRh_{1-x}Sn_xSb$ [26], respectively. A quantitative elaboration of these results requires an extensive effort from both the modeling and the experimental sides (*see below*), as the results are pioneering, but precursory.

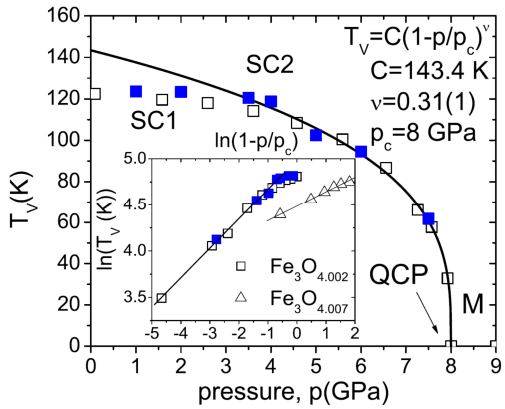

Fig. 5. Pressure dependence of the Verwey transition temperature ending at a quantum critical point (QCP) proposed by us [25]. Inset illustrates an extreme sensitivity of QCP to minute deviations from the perfect stoichiometry (cf. [25]).

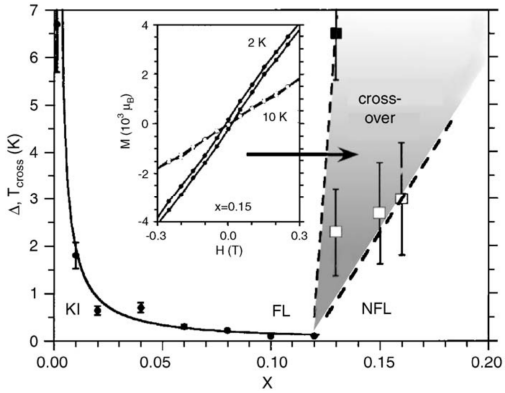

Fig. 6. Concentration *x* dependence of transition from Kondo insulator (KI) to a non-Landau fermionic liquid (NFL) (see [26]), with a QCP not marked explicitly then. Inset: magnetization curve showing an onset of a weak ferromagnetic phase in the NFL state.

3. 4. Quantum criticality in electronic structure: renormalized band theory

One of the fundamental topics in these systems is to unify the description of correlated metals and Mott insulators into a single scheme. Associated with this project on the *quantum criticality* is our recent work [7, 15] on **extending *ab-initio* approach to correlated fermion systems** by including the optimization of the single-particle wave function in the correlated state (wave function of each individual electron relaxes in the strongly interacting environment). This is in our view, a very promising method, as it can improve (in the sense of not counting twice the interaction) on the current methods (LDA+U, LDA+DMFT, and LDA+Gutzwiller [27]) of calculating the renormalized by interactions single-particle (band) structure of correlated materials (*see below*). In Figs. 7 and 8 we illustrate the observed theoretically interesting universal type of behavior obtained within our exact EDABI [15] method and its approximate extension [7] to 3-dimensional systems. This last method extends our earlier treatment of nanoscopic systems and a concerted effort will be undertaken to make it applicable to correlated *3d* systems. Also, its incorporation into the SGA scheme is regarded as crucial part of the project (see below).

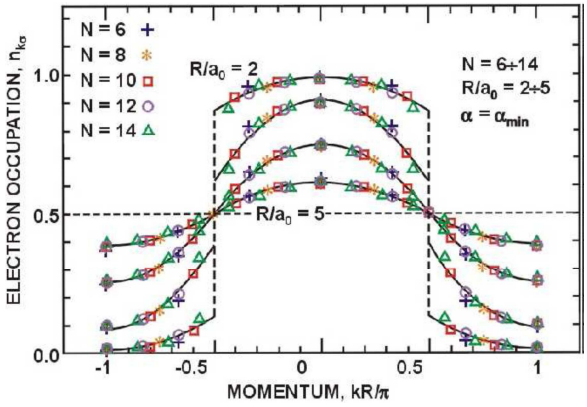

Fig. 7. Statistical distribution $n_{k\sigma}$ for electrons for a linear nanochain of N = 6 - 14 monovalent atoms, with the interatomic distance R in units of Bohr radius a_0. An evolution from nanometal (small R) to a nanosemiconductor (nanoinsulator) is observed with the increasing R.

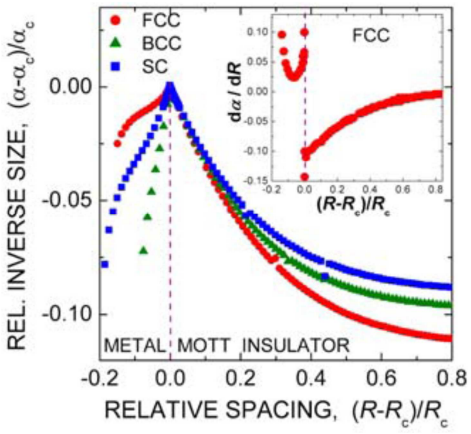

Fig. 8. Relative inverse size α of the wave-function scaling as a function of relative lattice parameter for the cubic lattices, showing a critical (singular) behavior near Mott transition point R_c. Inset: the first-derivative discontinuity showing quantum critical behavior (for details see [7]).

4. TEAM research and other activities within CCQM

The proposed research for the next 3 years is truly challenging and partly concerns continuation of ongoing work, but mainly with new undertakings, each characterized briefly below. The main aim of this research is to provide a coherent, albeit approximate, method of modeling materials on different scales.

4. 1. Strongly correlated systems: normal and novel superconducting states (1 M. Sc. student, 1 Ph. D. student, 1 post doc)

In summary, our research methodology, it relies from one side on an exact diagonalization combined with the wave-function renormalization in the correlated state ("wave-function breathing") and on a connected approximate (SGA) procedure, which allows for (semi) quantitative analysis of correlated materials and systems. In the proposed project we would like first to combine the two and/or explore further each of them to the analysis of concrete physical systems.

1) First of the problems relies on extending our SGA method by incorporating in it the so-called *quantum fluctuations* within a parametrized model such as Hubbard's or Anderson's. This extension is very important, as only then we can reach, in our view, a complete description of the thermodynamic and electrodynamic properties of the superconducting state in new materials such as $La_{2-x}Sr_xCuO_4$ or related compounds with Y and Bi. The presence of the fluctuations is one of the fundamental differences between the description of "classical" (BCS) superconductors versus novel (high-temperature, unconventional) superconductors and atomic condensates in optical lattices. The research should result in direct comparison with experiment for the cuprate superconductors.

There are other new topics related to this research. The first of them is the question how the magnetic order (antiferromagnetic in this case) coexists and/or competes with the high temperature superconductivity. This question is highly nontrivial as the high-temperature superconducting state evolves from an antiferromagnetic insulating state and in the heavy-fermion superconducting state it often coexists with the SC state. The preliminary (unpublished) results obtained within the SGA method speak in favor of an extended regime of the coexistence.

These are the tasks for 1 M. Sc. Thesis to be followed by 1 Ph. D. Thesis in years 2011-2013, in cooperation with ETH-Zürich group of Prof. Manfred Sigrist; one in preparation [16].

2) One of the new separate projects will concern **the properties of novel phases of heavy-fermion superconductors** within the same (SGA) method taking into account the microscopic model - the *so-called Anderson-lattice model*. As said above, one Ph. D. Thesis concerning the heavy quasiparticle states with spin-dependent masses is under way [21], another has just started [28]. Those Ph. D. Theses will certainly address also the question of coexistence of superconductivity and antiferromagnetism and the effect of quantum fluctuations on the properties of the unconventional superconducting states. In this manner, we can directly see the similarities and differences with high temperature superconductors. One should mention that in the latter case the real-space pairing is provided by the kinetic exchange interaction ("pairing without glue" [29]), whereas in the heavy fermion case the pairing is induced by the Kondo-type interactions [4, 6]. The obtained results will be applied to $CeCoIn_5$ and related compounds.

We expect to cooperate in this project with the ETH Zürich group.

4. 2. Correlated and coherent fermions in special systems and lattices: SGA method implementation and testing (1 M. Sc. student, 1 Ph. D. student, 1 post doc)

1) As said above, we developed a new rigorous method of approach [15] to correlated nanoscopic systems - EDABI (**E**xact **D**iagonalization **Ab I**nitio Approach). The results are highlighted in Fig. 7, where we show a rigorous evolution of statistical distribution of electrons in a quantum nano wire containing correlated electrons from Fermi gas to localized states ($n_{k\sigma}$ smeared out). We have obtained there also a coherent magnetic structure which is specific for the systems of nanoscopic size when the spin correlation range is of the system size. Therefore, one can study quantum magnetic coherence without having the symmetry spontaneously broken. The fundamental question is whether this type of structure can be observed is realistic systems. Hence, an extensive studies are to be undertaken for realistic system (Cu, Ni, Au nanowires), but the underlying structure must be modeled using supercomputing facilities (preliminary work utilizing the LDA scheme has already started).

However, the crucial step is to extend this work by implementing renormalization scheme of SGA to the original LCAO (LMTO) method with single-particle wave function variationally adjusted in SGA-correlated state.

Application of this last method to real 3d systems (magnetic oxides) is to be carried out in cooperation with our long-term coworkers Prof. Włodzimierz Wójcik and Dr. Jan Kurzyk from Technical University of Kraków. Also, a cooperation with Prof. Carmen Munoz from CSIC, Madrid (Spain) is planned on extending LDA+U method. At least one habilitation is planned as a side product when our SGA method is implemented successfully within the so-called tight-binding scheme of determining band structure.

2) Second application of our method is connected with the effect of correlations associated with doping or adsorbing atoms on the two-dimensional single sheet of graphene. In our group very interesting results have been obtained for a pure graphene [31]. It would be very interesting to consider the situation in which graphene is attached to a two-dimensional array of (magnetic) atoms producing thus a model situation for the periodic Anderson lattice, with the Falicov-Kimball type interactions [32]. The relative advantage of using our method is that it allows to treat relatively large systems and the results, obtained so-far for two-dimensional *t-J* model, provide the results comparable to those of Quantum Monte-Carlo computations, which are however limited to very small systems. Also, we would like to consider, with the help of SGA, a superconducting state for this hexagonal (honeycomb) lattice, as the analytical structure of underlying dispersion relation for electron energies is linear ("Dirac massless electrons").

This project will be executed together with Dr hab. Adam Rycerz from our group (habilitation on 23/09/2010), ***who will be an associated principal investigator*** *in the whole project. Cooperation*

with Prof. C. W. J. Beenakker from University of Leiden (NH) (a continuation of the ongoing cooperation) is also expected.

3) As a test of our SGA method we will apply it to the properties of condensed helium 3 in two dimensions, where spectacular phenomena of effective mass divergence near He solidification were experimentally observed [33]. These phenomena are also associated with observed metamagnetic behavior in very strong magnetic field [34]. Those observations square well qualitatively with our earlier theory of Mott localization [24]. It is very intriguing whether our new approach to correlated electron systems would work in a quantitative manner for this canonical Fermi liquid, solidification of which is regarded as the clearest example of Mott localization (for elementary discussion see [14]). This test case of the project is the *sine-qua-non* condition of the validity of our SGA method; also, because the system contains quantum defects (Andreev vacancies), the phenomenon of supersolidicity is observed [35] and can be addressed within our approach.

4. 3. Correlated fermions/bosons and high-spin atomic condensation in optical lattices (1 M. Sc. student, 1 Ph. D. student, 1 post doc)

Recently, atomic physics in optical/laser lattices became the test ground for older and new concepts in correlated-fermion and -boson condensed-matter physics. We plan to extend and test our SGA and related methods to those entirely new systems - correlated atomic systems in optical lattices, also in the situations with high-spin (in the simplest case - triplet) pairing [36]. The situation in these systems is unique because: (i) the single-particle potential (crucial for the system stabilization) is known unlike in electronic systems, (ii) one can create experimentally the model systems we considered extensively in the past, and (iii) no phonons are present in that case, so one can test the correlation driven mechanism of pairing (so we hope, at least). As a side effect, our team work should thus demonstrate our *method versatility*, i.e. research not limited to the solid-state or liquid-helium systems. But also, we hope, it should allow for a test of our concept of wave-function criticality. Finally, *last but not least*, we hope to address the question of the Bose-Einstein condensation vs. BCS type of state for those systems.

This project will be carried out in close cooperation with dr. hab. Maciej Maśka (Professor UŚ), as he is very experienced in that field (cf. [37]). A cooperation with Prof. J. Freericks from Georgetown University (USA) should be helpful in many respects while carrying out this project.

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