

# I Introduction

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## 1 Emergent behavior

Understanding the laws that govern the universe is dream as old as mankind. Its motivation is the quest to reduce complexity to simplicity, succinctly expressed by Einstein [1]

*The supreme test of the physicist is to arrive at those universal laws from which the cosmos can be built up by deduction.*

This reductionist program achieved remarkable successes, starting with Newton's understanding that the gravitational pull on an apple is the same that holds the moon on its orbit around earth. Maxwell managed to describe the quite different phenomena of electricity and magnetism on the same footing, developing the theory of electromagnetism. The electromagnetic force could later be unified with the weak interaction into the electro-weak force, one of the pillars of the *standard model*. But there were also problems along the way. Theories became more and more complex as a whole zoo of elementary particles was discovered. Then, with the introduction of quarks, today's elementary particles became tomorrow's compound objects, while the new elementary particles ceased to have a meaning as independent objects. Interestingly, the considerable changes in what was believed to be the fundamental *Theory of Everything* had remarkably little influence on our understanding of the physics at lower energy scales: The quark-dynamics has hardly any relevance for understanding chemical bonding. When vastly different energy scales are involved, a clear-cut separation of the physical description into almost independent layers takes place.

The relevant elementary particles for describing matter at the energy scale of our everyday experience are the atomic nuclei, usually considered as point-charges, and, of course, the electrons, giving rise to *electronic structure*. The fundamental laws governing these are given by the innocent looking eigenvalue problem

$$H|\Psi\rangle = E|\Psi\rangle \quad (1)$$

where the Hamiltonian for a set of atomic nuclei  $\{\alpha\}$  with atomic numbers  $\{Z_\alpha\}$  and nuclear masses  $\{M_\alpha\}$  and their accompanying  $\{i\}$  electrons is given, in atomic units, by

$$H = -\frac{1}{2} \sum_i \nabla_i^2 - \sum_\alpha \frac{1}{2M_\alpha} \nabla_\alpha^2 - \sum_{\alpha,i} \frac{Z_\alpha}{|\mathbf{r}_i - \mathbf{R}_\alpha|} + \frac{1}{2} \sum_{ii'} \frac{1}{|\mathbf{r}_i - \mathbf{r}_{i'}|} + \frac{1}{2} \sum_{\alpha\alpha'} \frac{Z_\alpha Z_{\alpha'}}{|\mathbf{R}_\alpha - \mathbf{R}_{\alpha'}|}.$$

This equation, augmented by gravitational potentials and lowest-order relativistic corrections (spin) as the microscopic basis of magnetism, account for the phenomena of our everyday experience, i.e., they are a *Theory of almost Everything* [3, 4]. Therefore, already shortly after the formulation of the Schrödinger equation, Dirac [5] remarked that the theory behind atomic and condensed-matter physics, as well as chemistry is completely known

*The underlying laws necessary for the mathematical theory of a large part of physics and the whole of chemistry are thus completely known, and the difficulty is only that exact applications of these laws lead to equations which are too complicated to be soluble. It therefore becomes desirable that approximate practical*

*methods of applying quantum mechanics should be developed, which can lead to an explanation of the main features of complex atomic systems without too much computation.*

From the reductionist point of view, this closes the case on chemistry and condensed-matter physics. Solving (1) is an exercise in applied mathematics, merely a practical problem. However, because of the quantum many-body nature of the problem, finding the solution, though possible in theory, is impossible in practice. To understand why, we consider a canonical example, a single atom of iron. Having 26 electrons, its wavefunction  $\Psi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_{26})$  is a function of 78 coordinates. What does it take to store such a wave function? If we record  $\Psi$  at merely ten values for each coordinate, we would have to record  $10^{78}$  values. There is not enough matter in the visible universe for storing even such a ridiculously crude representation of the wave function of a single iron-atom. This complexity of the wave function is the essence of the *many-body problem*. Already Laplace [6] realized that indeed the grand goal of simulating the world, even if it was possible in theory, in practice can never be reached

*An intelligent being, who, at a given moment, knows all the forces that cause nature to move and the positions of the objects that it is made from, if also it is powerful enough to analyze his data, would have described in the same formula the movements of the largest bodies of the universe and those of the lightest atoms. [...] Although scientific research steadily approaches the abilities of this intelligent being, complete prediction will always remain infinitely far away.*

But would it really be desirable to know the full wave function of a solid, even if it was possible? On the one hand, yes, because from the wave function we could readily calculate all expectation values. Thus we would be able to make reliable predictions of the properties of any given material. But *predictive power* does not just mean that calculations agree with experiment. Predictive power also means that we know what problems are worth looking at, i.e., for what materials interesting properties are to be expected. For this, knowing the full wave function would be of little help; it would just lead to information-overload. The physics would be buried in the masses of data, and the life of the universe would be too short to analyze it, let alone to understand it. Remarkably, Wigner and Seitz recognized this very early on [7], when electronic-structure calculations were still mainly done by hand, i.e., by human computers [8]

*If one had a great calculating machine, one might apply it to the problem of solving the Schrödinger equation for each metal and obtain thereby the interesting physical quantities, such as the cohesive energy, the lattice constant, and similar parameters. It is not clear, however, that a great deal would be gained by this. Presumably the results would agree with the experimentally determined quantities and nothing vastly new would be learned from the calculation. It would be preferable instead to have a vivid picture of the behavior of the wave functions, a simple description of the essence of the factors which determine the cohesion and an understanding of the origin of variation in properties [...]*

To understand Nature, we have to identify the *mechanisms* behind the phenomena. We want to know why Nature behaves the way she does, not just, by theory, reproduce some experiment. Why are metals shiny? Why do some materials superconduct while others do not? What opens the gap in a Mott insulator? What drives the ordering of orbitals? What is the mechanism behind colossal magneto-resistance? These properties do not descent in any obvious way from the Theory of almost Everything (1). They are examples of *emergent behavior*. When a system is made of many interacting components, no matter how simple the interactions are, complex many-body phenomena arise. We meet examples of emergence everyday, maybe the most astonishing being the human brain itself.

The limitations of the reductionist approach were probably most strongly voiced by P. W. Anderson in his famous article *More is different* [9]

*The ability to reduce everything to simple fundamental laws does not imply the ability to start from those laws and reconstruct the universe. [...]*

*The behavior of large and complex aggregates of elementary particles, it turns out, is not to be understood in terms of a simple extrapolation of the properties of a few particles.*

Instead of a reduction to some specific Theory of Everything, science should rather be seen as a hierarchy of structures

*Thus, with increasing complication at each stage, we go up the hierarchy of the sciences. We expect to encounter fascinating and, I believe, very fundamental questions at each stage in fitting together less complicated pieces into the more complicated system and understanding basically new types of behavior which can result.*

The stages in this hierarchy are best defined, when the energy scales differ significantly. High energy physics deals with the interactions among elementary particles. On the next stage the elementary particles condense into bound states, the subject of nuclear physics. Going up in the hierarchy, at energy-scales of everyday-life, it is only the interaction between atoms and molecules that matters. This is the realm of the chemical bond. Each stage is well separated from the others. This means that new discoveries at a lower stage do not fundamentally change the description at the higher stage. On the one hand, this effective decoupling is what makes science possible without knowing the ultimate *Theory of Everything*. On the other hand, the same decoupling implies that describing the complexity at a higher stage in terms of the theory at the lower stage is a practical impossibility. At each level in the hierarchy fundamentally new properties emerge, which are largely independent of the details on the previous level. Moving up in the hierarchy we need to devise *effective theories* describing the physics at that level, which are derived from physical understanding rather than from an exact mathematical treatment of the underlying microscopic dynamics.

## 2 Electronic Structure

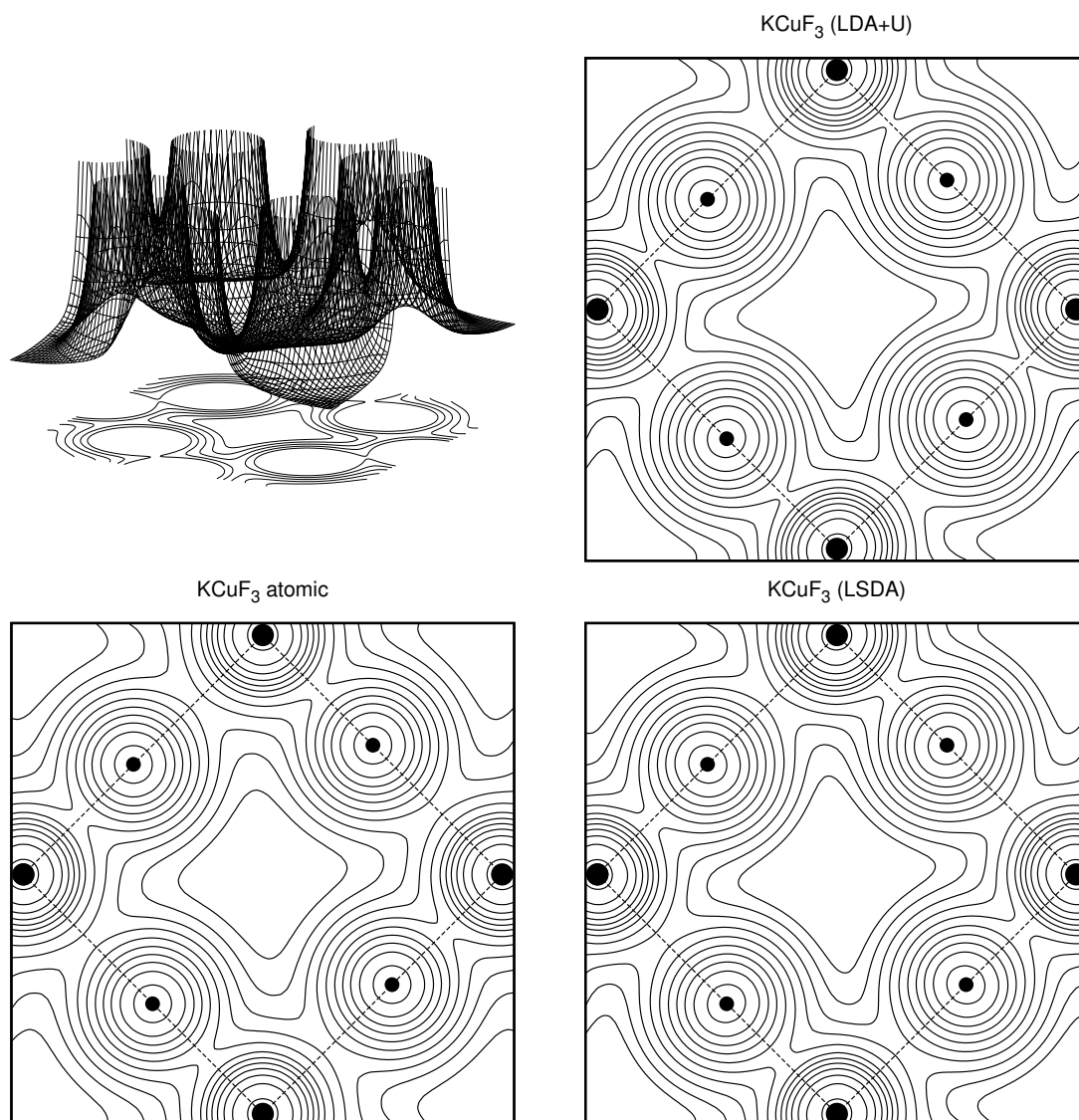
As we have seen, the fundamental problem in condensed-matter physics is solving (1). The most fascinating properties emerge from its many-body nature. This makes, except in the simplest cases, an exact solution a practical impossibility. Dirac's *approximate practical methods* for its solution therefore have to focus on specific aspects of the problem.

The most immediate many-body effect is Fermi-Dirac statistics. It gives us the *Aufbauprinzip*, letting us understand the structure of the periodic table and the basis of chemical bonding. This is well described in the Hartree-Fock approach. An even more practical approach is density-functional theory (DFT) [10]. It rephrases the many-body problem as the problem of minimizing a functional of the electron density – a tremendous simplification from a problem involving  $N$ -body wave-functions with  $3N$ -coordinates to densities  $n(\vec{r})$  with merely 3 variables. In practice, however, only approximations to the universal density-functional are known. The most successful route to constructing approximate density-functionals is via a mapping of the interacting system onto a non-interacting reference system. In this Kohn-Sham framework already very simple approaches like the local-density approximation (LDA) result in functionals that are surprisingly accurate for vast classes of materials. The basis for their success, again, is that the *Aufbauprinzip* is firmly built into the functional via the filling of the single-electron levels of the reference system.

Because of the single-electron nature of the Kohn-Sham reference system, practical density-functional calculations work well for materials with an electronic structure that can be understood in terms of the energy levels of individual electrons. In these cases density-functional calculations not only provide accurate results, but also contribute to our understanding, e.g., of bonding in materials in terms of molecular orbitals, or Bloch waves and Wannier functions in solids. The approach has proven so successful that its main creator, Walter Kohn, was recognized with the 1998 Nobel Prize in Chemistry.

The successes of the density-functional approach make it easy to forget that it was far from clear whether working density-functional approximations could ever be found. The electron density is a remarkably featureless quantity. As figure 1 exemplifies, it is almost impossible to distinguish the charge densities of a solid from the superposition of its constituent atoms or even to tell a metal from an insulator. This fact is the basis for the unexpected usefulness of the non self-consistent Harris functional [11]. It also lets us appreciate that finding good approximate density functionals is a highly nontrivial task. While the simple local-density approximation works surprisingly well for many materials, it has problems describing magnetism. In this case, expressing the functional in terms of spin-densities greatly simplifies the task of writing down a good functional. While all necessary information is included, in principle, in the density alone, only treating the spin-densities as separate variables gives a simple way, the local spin-density approximation [12], of ensuring that states with large spin tend to have lower energy.

This situation is typical for the art of devising approximate practical methods. We need to identify which details of the problem should be included. If we manage to identify the relevant details, finding a good approximation is significantly simplified. If we fail to identify them,



**Fig. 1:** Charge density for a cut through  $\text{KCuF}_3$ . The charge density contours (on a logarithmic scale) for the insulating solid calculated with LDA+U hardly differs from the charge density of the metal obtained in LSDA. Both hardly differ from the superposition of atomic charge densities. Large black circles: position of Cu atoms, small black circles: F atoms.

finding a good approximation is virtually impossible.

By the nature of the Kohn-Sham approach, density-functional calculations are largely confined to materials, for which the picture of individual electrons is adequate, and *Fermi-liquid theory*, which models weakly interacting quasi-particles, applies. There is, however, a remarkable variety of *strongly correlated* materials for which this standard model of electronic structure theory breaks down. The hallmark of these materials is that some of their electrons are neither perfectly localized, nor fully itinerant. These electrons can no longer be considered individually. The resulting behavior presents some of the deepest intellectual challenges in physics. At the same time interest in strongly correlated materials is fueled by the astounding possibilities for technological applications. Prominent examples are the transition-metal oxides, e.g., the high-temperature superconductors, and molecular crystals [13].

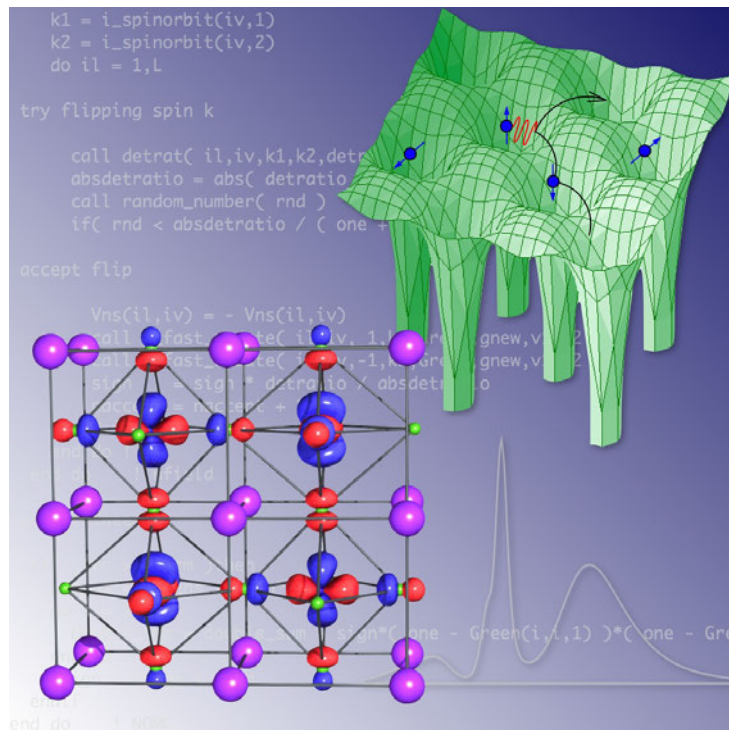
### 3 The LDA+DMFT approach

When dealing with strongly-correlated electrons we have to confront the many-body problem. For a long time the only available strategy was to come up with minimal models, tailored to describe a specific phenomenon. Even these simple many-body problems (Kondo model, Anderson model, Hubbard model, periodic Anderson model, ...), were still very hard to solve. Including the complexity of specific materials was out of the question. Most effort went into developing methods to solve these model problems. For single-impurity models, like the Kondo or the Anderson model, exact solutions were found, but lattice problems (Hubbard model, periodic Anderson model) remained unsolved, with the exception of special limits, typically one-dimensional systems. The formidable difficulty of finding approximate non-perturbative techniques to solve the Hubbard model and similar Hamiltonians lead to a decoupling of many-body physics from chemistry and electronic-structure. On the other hand, the hope of calculating model parameters from first-principles lead to largely independent developments, such as the LDA+ $U$  method, in which correlation effects are fitted into DFT calculations even if only at the crude Hartree-Fock level.

In recent years, significant progress was driven by the development of dynamical mean-field theory (DMFT). The key insight was that in the limit of infinite dimensions the self energy becomes local [14, 15]. That suggested that, in finite but not too low dimension, the infinite lattice could be mapped approximately onto an impurity problem, which has to be solved self-consistently [15]. From the many-body point of view, the techniques developed for impurity models could now be used for the Hubbard model. From the materials point of view, the LDA+ $U$  experience suggested that it would be possible to solve many-body models with parameters calculated *ab-initio*. These two lines of research culminated in the development to the LDA+DMFT method, today state-of-the-art approach for strongly-correlated systems.

While enormously reducing the cost of the simulation, non-perturbative calculations are still limited to quite simple model Hamiltonians [16]. It is therefore crucial to construct models that are as small as possible, while still capturing the essential chemistry of the real material. As we already saw in the development of approximate density-functionals, the art of modeling is to make the model as simple as possible, but not simpler. Ideally a model is simple enough that a simulation is well feasible, but detailed enough to be material-specific.

The LDA+DMFT approach works in two steps. In the first step, *ab-initio* calculations, based on density-functional theory, are used to obtain the one-electron part of the Hamiltonian. Next, the high-energy states are integrated out, so that only the low-energy partially filled ( $d$  or  $f$ ) bands are retained, and a basis of first-principles Wannier functions is constructed. These Wannier functions, by construction, carry the information on the lattice and the chemistry; furthermore, they are localized, so that the correlation part of the Coulomb repulsion is very short range in this basis. In the second step, the material-specific few-bands many-body Hamiltonians, constructed in terms of these Wannier functions, are solved in the dynamical mean-field approximation. This two steps approach has been used very successfully, e.g., to understand the role of subtle crystal-field splittings for the metal-insulator transition in  $3d^n$  transition-metal oxides [17].



**Fig. 2:** Logo showing the essential ingredients of the LDA+DMFT approach: Electrons in localized orbitals are strongly correlated as their Coulomb interaction cannot be described by a simple static mean field. The background shows a section of a quantum Monte Carlo based impurity solver which is used to calculate, e.g., spectral functions.

Still, even low-energy few-bands models can be solved only thanks to high performance computers. The task of solving the full many-body problem in a realistic setting will remain the main challenge in condensed matter for years to come. Bridging the high and low energy electronic degrees of freedom is not only one of the deepest problem in contemporary physics but should also provide a wealth of exciting materials for novel technologies.

## 4 Overview of the school

The historical development and the current state-of-the-art of the LDA+DMFT approach is reflected in the organization of the school and the present collection of lecture notes.

To set the stage, Dieter Vollhardt will introduce the problem of electronic correlations and discuss the limit of infinite dimensions, which is the basis of the dynamical mean-field approach. The lecture of Peter Blöchl will then introduce the other pillar of LDA+DMFT, density-functional theory.

The second group of lectures is devoted to the art of model-building. First, Frank Lechermann will discuss some of the most important types of models. The construction of realistic models is based on Wannier functions from *ab-initio* calculations. The determination of Wannier functions is discussed in the lecture of Jan Kuneš. By construction these functions give a good representation of the kinetic part of the model Hamiltonian. In contrast, the calculation of



screened Coulomb integrals is much less developed. This challenging problem will be covered in the lecture of Ferdi Aryasetiawan. To reflect the central importance of model-building, two tutorials will cover the practical aspects of constructing Wannier functions and of calculating screened Coulomb parameters.

The dynamical mean-field approach is the central theme of the book. The foundations of this many-body method and the applications to simple models will be explained in the lecture of Marcus Kollar. To use it in a realistic contest, DMFT has to be combined with *ab-initio* techniques. The LDA+DMFT approach will be presented by Eva Pavarini.

An essential step of DMFT is to solve the auxiliary quantum-impurity problem. Three lectures are thus devoted to introducing some of the most important quantum-impurity solvers: The Lanczos method (Erik Koch), the Hirsch-Fye quantum Monte Carlo (Nils Blümer), and the continuous-time quantum Monte Carlo (Philipp Werner). Computational aspects of the quantum Monte Carlo approaches will be covered in two tutorials.

Further lectures are devoted to advanced topics. Hubert Ebert will introduce the KKR+DMFT approach, which tightly integrates DMFT in a band-structure method. Sasha Lichtenstein will discuss important extensions of DMFT to include spatial fluctuations. Finally, Karsten Held will discuss the GW+DMFT, which is based on Hedin's *GW* approach rather than DFT.

Each lecture will illustrate examples of the successes of LDA+DMFT. However, the ultimate word on a theory goes to experiments. Thus the book ends with the lecture of Hao Tjeng, maybe the most important of all. It will present challenges for theory from experiments. Facing the limits of a method is the essential step to improve a theory, and to make progress towards predictive power.

## References

- [1] A. Einstein, *Principles of Research*, in *Essays in Science* **4** (1934)
- [2] P.M.A. Dirac, *Proceedings of the Royal Society (London)* **A 123**, 714 (1929)
- [3] D. M. Ceperley, *Rev. Mod. Phys.* **71**, 438 (1999)
- [4] R.B. Laughlin, and D. Pines,  
*Proceedings of the National Academy of Sciences (USA)* **97**, 28 (2000)
- [5] P.M.A. Dirac, *Proceedings of the Royal Society (London)* **A 123**, 714 (1929)
- [6] P.S. Laplace, *Theorie Analytique des Probabilités*, Courier, Paris (1820)
- [7] E.P. Wigner and F. Seitz, *Solid State Physics* **1**, 97 (1955)
- [8] F. Hermann, *Physics Today*, June 1984, p. 56
- [9] P.W. Anderson, *Science* **177**, 393 (1972)
- [10] W. Kohn, *Rev. Mod. Phys.* **71**, 1253 (1999)
- [11] J. Harris, *Phys. Rev. B* **31**, 1770 (1985)
- [12] R.O. Jones and O. Gunnarsson, *Rev. Mod. Phys.* **61**, 689 (1989)
- [13] I.S. Osborne, *Science (Special Issue: Correlated Electron Systems)* **288**, 461 (2000)
- [14] W. Metzner and D. Vollhardt, *Phys. Rev. Lett.* **62**, 324 (1989)
- [15] A. Georges, G. Kotliar, W. Krauth, and M.J. Rozenberg, *Rev. Mod. Phys.* **68**, 13 (1996)
- [16] A. Dolfen, E. Pavarini, and E. Koch,  
*Innovatives Supercomputing in Deutschland* **4**, 16 (2006)
- [17] E. Pavarini, S. Biermann, A. Poteryaev, A.I. Lichtenstein, A. Georges, and O.K. Andersen,  
*Phys. Rev. Lett.* **92**, 176403 (2004)