Electronic structure calculations for magnetic systems

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I will present basic ideas of density functional theory (DFT) in four units with a focus on magnetic systems. Some of the ideas presented before in the lectures on basic concepts (Coey: Magnetism of single atoms) and on magnetism in matter (Blundell) will be re-considered from the viewpoint of DFT and supplemented with several application examples. In a related tutorial, I will demonstrate that such calculations can be even performed by a fearless novice .

Unit 1: Hohenberg-Kohn-Sham theory and Local Density Approximation (LDA)

In this unit, I will introduce one of the most powerful models applied in recent condensed matter physics. As a starting point I will explain why we are not able to solve the Schrödinger many-electron equation for systems larger than a few atoms and why it makes no sense to aim on this. As a consequence, we have to resort to **model theories** as opposed to quantum chemical *ab initio* calculations. One possible model, taking into account details of the system chemistry and geometry, is the Local Density Approximation (LDA). This is a **parameter free** but not a first-principles (a synonym for *ab initio*) approach. The LDA is based on the formally exact Hohenberg-Kohn theory and on the interacting electron gas model that can be solved numerically with an accuracy sufficient for all practical purposes. The related Kohn-Sham equations will be explained, their strength and limitations will be discussed.

Unit 2: Exchange, the root of condensed matter magnetism

Given the magnetic properties of electrons, quantum mechanical exchange is what provides their cooperation to form local atomic or molecular moments, or even long-range magnetic order in crystals. Elementary magnetic moments arrange themselves to lower the mutual Coulomb interaction of the electrons, being subject to symmetry restrictions of the fermionic wave function. I will dicuss the particular cases of the two-electron wave function and of the homogeneous polarized electron gas. The Local Spin Density Approximation (LSDA) will be introduced and the LSDA Stoner parameter will be defined. Calculated spin polarization energies for atomic shells will be compared with related spectroscopic data. The spin splitting of the Kohn-Sham states will be evaluated.

Unit 3: Tight-binding approach, chemical binding in a nutshell

As a corollary of the virial theorem, the reduction of potential energy will be identified as the driving force behind chemical binding. I will use the hydrogen-molecule to explain the basic ideas of the linear combination of atomic orbitals (LCAO, or tight-binding) approach, as well as the notation of bonding and antibonding states. Bloch's theorem will be introduced, and the tight-binding formalism will be applied to a crystal. In this way, discrete levels of finite systems are replaced by the density of states (DOS) in extended systems. An important technical point is the appropriate choice of atomic-like orbitals in a multi-band tight-binding calculation.

Unit 4: Tight-binding meets exchange, real systems and applications

In order to summarize and to apply the knowledge gained in the previous units, the electronic structure and the magnetic ground state of appropriate model systems will be discussed. First, I will analyze the spin-polarized molecular levels of the iron dimer. Second, the band structure and spin-polarized DOS of bulk bcc iron will be presented and explained. A third system, $La(Fe,Si)_{13}$, will be used to extend the scope to systems of emerging practical relevance. The peculiar electronic structure of this compound yields an extraordinarily flat dependence of the total energy on the

magnetic moment and a related strong susceptibility to external influence (magnetic field, temperature, pressure). As a consequence, $La(Fe,Si)_{13}$ is a favored candidate for magneto-caloric applications. A final remark will be spent on topological insulators, recently disclosed non-magnetic systems with surface spin currents protected by the topology of the bulk band structure.

Suggested reading:

Helmut Eschrig, *The Fundamentals of Density Functional Theory*, Teubner-Texte zur Physik, Band 32, B.G. Teubner Verlagsgesellschaft, Stuttgart 1996, ISBN 3-8154-3030-5.
(Units 1, 2 and 3)

Manuel Richter, *Band structure theory of magnetism in 3d-4f compounds*, J. Phys. D: Applied Physics **31**, 1017-1048 (1998).
(Units 1, 2, and 4)

Manuel Richter, *Density Functional Theory applied to 4f and 5f Elements and Metallic Compounds*, Handbook of Magnetic Materials (Ed. K.H.J. Buschow), Vol. 13, Elsevier, Amsterdam 2001, pp. 87-228, ISBN 0-444-50666-7.
(Units 1, 2, and 4)

Claude Cohen-Tannoudji, Bernard Diu, and Franck Laloë, *Quantum Mechanics*, Vol. II, Hermann, Paris 1977, ISBN 0-471-16435-6.
(Units 2 and 3)

John Singleton, *Band Theory and Electronic Properties of Solids*, Oxford Master Series in Condensed Matter Physics, Oxford University Press, Oxford 2006, ISBN 0-19-850644-9.
(Unit 3)

Manuel Richter, Klaus Koepernik, and Helmut Eschrig, *Full-Potential Local-Orbital Approach to the Electronic Structure of Solids and Molecules*, in: Condensed Matter Physics in the Prime of the 21st Century, Ed. J. Jedrzejewski, World Scientific, Singapore 2008, pp. 271-291, ISBN 981-270-944-8.
(Unit 3)

Jürgen Kübler, *Theory of Itinerant Electron Magnetism*, International Series of Monographs on Physics, Vol. 106, Oxford Science Publications, Clarendon Press, Oxford 2000, ISBN 0-19-850028-9.
(Units 2, 3, and 4; general interest)

Stephen Blundell, *Magnetism in Condensed Matter*, Oxford Master Series in Condensed Matter Physics, Oxford University Press, Oxford 2006, ISBN 0-19-850591-4.
(Units 2 and 4; general interest)

Daniel C. Mattis, *The Theory of Magnetism Made Simple*, An Introduction to Physical Concepts and to Some Useful Mathematical Methods, World Scientific, Singapore 2006, ISBN 981-238-671-8.
(Unit 2; general interest)

Michael D. Kuz'min and Manuel Richter, *Mechanism of the strong magnetic refrigerant performance of* $LaFe_{13-x}Si_x$, Phys. Rev. B **76**, 092401 (2007).
(Unit 4)

C. Pauly *et al.*, Nature Physics 2015.
(Unit 4)