

Basics of electronic structure calculations for magnetic systems: Tight binding, LDA, DMFT

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-- Digest --

A trial will be made to squeeze some of the basic ideas of 80 years electronic structure theory, applied to magnetic systems, into a two-hours lecture. I will subdivide the subject into six units. Unit one to five will be devoted to ground state properties and their description in an effective single-particle quantum theory, while unit six will sketch recent developments in the field of excited states modelling. A list with suggested literature is appended, where the topics are treated in much more detail than in this brief overview.

Unit 1: Hohenberg-Kohn-Sham theory, basics of the local density approximation (LDA)

This unit will not deal with magnetism at all, but it 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 mentioned.

Unit 2: 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. The celebrated hydrogen-molecule will be used 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 the hypothetical simple cubic hydrogen crystal. Atomic levels are replaced in extended systems by the density of states (DOS). An important technical point is the appropriate choice of atomic-like orbitals in a multi-band tight-binding calculation. Sorry, there is no magnetism in this unit either.

Unit 3: Exchange, the root of condensed matter magnetism

Given the magnetic properties of elementary particles, quantum mechanical exchange is what provides their cooperation to form local atomic or molecular moments, or even long-range magnetic order. 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 discuss 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 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 two 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, $\text{La}(\text{Fe},\text{Si})_{13}$, 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, $\text{La}(\text{Fe},\text{Si})_{13}$ is a favored candidate for magneto-caloric applications.

Unit 5: Magnetic response without exchange, a detour to diamagnetism

Diamagnetism is fundamentally different from the exchange-dominated effects (in particular, para- and ferromagnetism and their flavors) discussed before. I will introduce the basic idea of diamagnetic response and discuss the size of the closed-shell effects in comparison with open-shell magnetic response. This detour has been suggested by Olivier Fruchart at the Nanomagnets 2009 workshop (Aussois).

Unit 6: Dynamical mean field theory

DFT is a ground state theory that can be used, e.g., to calculate lattice geometries, magnetic states, charge- and spin-densities. Sometimes the Kohn-Sham levels, though in general not suited for this task, compare well with excited states measured by photoemission or related techniques. The appropriate approach for excited states, however, is a Greens functions technique. A recent, very successful implementation of such a technique is dynamical mean field theory (DMFT). I will sketch the basic ideas of DMFT and its field of application to magnetic phenomena.

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 and 2)

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

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 2)

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, 4, and 5; 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 3, 4, and 5; 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.

(Units 3 and 5; general interest)

Michael D. Kuz'min and Manuel Richter, *Mechanism of the strong magnetic refrigerant performance of $\text{LaFe}_{13-x}\text{Si}_x$* , *Phys. Rev. B* **76**, 092401 (2007).

(Unit 4)

A. Georges, G. Kotliar, W. Krauth, and M.J. Rozenberg, *Dynamical mean-field theory of strongly correlated fermion systems and the limit of infinite dimensions*, *Rev. Mod. Phys.* **68**, 13-125 (1996).

(Unit 6)