Annals of Physics 360 (2015) 61–97

Contents lists available at ScienceDirect

Annals of Physics

journal homepage: www.elsevier.com/locate/aop

# Magnetic interactions in strongly correlated systems: Spin and orbital contributions

A. Secchi [a,*], A.I. Lichtenstein [b], M.I. Katsnelson [a]

[a] Radboud University, Institute for Molecules and Materials, 6525 AJ Nijmegen, The Netherlands
[b] Universitat Hamburg, Institut für Theoretische Physik, Jungiusstraße 9, D-20355 Hamburg, Germany

ARTICLE INFO

*Article history:*
Received 19 December 2014
Accepted 4 May 2015
Available online 11 May 2015

*Keywords:*
Strongly correlated systems
Anisotropic exchange interaction
Dzyaloshinskii–Moriya interaction
Green's functions
Orbital magnetism

ABSTRACT

We present a technique to map an electronic model with local interactions (a generalized multi-orbital Hubbard model) onto an effective model of interacting *classical* spins, by requiring that the thermodynamic potentials associated to spin rotations in the two systems are equivalent up to second order in the rotation angles, when the electronic system is in a symmetry-broken phase. This allows to determine the parameters of relativistic and non-relativistic magnetic interactions in the effective spin model in terms of equilibrium Green's functions of the electronic model. The Hamiltonian of the electronic system includes, in addition to the non-relativistic part, relativistic single-particle terms such as the Zeeman coupling to an external magnetic field, spin–orbit coupling, and arbitrary magnetic anisotropies; the orbital degrees of freedom of the electrons are explicitly taken into account. We determine the complete relativistic exchange tensors, accounting for anisotropic exchange, Dzyaloshinskii–Moriya interactions, as well as additional non-diagonal symmetric terms (which may include dipole–dipole interaction). The expressions of all these magnetic interactions are determined in a unified framework, including previously disregarded features such as the vertices of two-particle Green's functions and non-local self-energies. We do not assume any smallness in spin–orbit coupling, so our treatment is in this sense exact. Finally, we show how to distinguish and address separately the spin, orbital and spin–orbital contributions to mag-

* Corresponding author.
*E-mail addresses:* a.secchi@science.ru.nl, andrea.secchi@gmail.com (A. Secchi).

http://dx.doi.org/10.1016/j.aop.2015.05.002

netism, providing expressions that can be computed within a tight-binding Dynamical Mean Field Theory.


## 1. Introduction

Establishing a rigorous connection between magnetic and electronic descriptions of condensed matter systems is a challenging problem [1], whose formal statement can be formulated as follows: Given a physical system described by means of a completely known electronic Hamiltonian, what is the *spin* Hamiltonian (supposing that it exists) that most closely reproduces the spectral and dynamical features of the system?

The answer to this important question is, of course, far from being straightforward. It is well known, e.g., that the spectrum of the lowest energy band of the single-orbital Hubbard model at half filling with nearest-neighbour hopping $T$ and strong on-site Coulomb repulsion $U$ can be effectively represented in terms of the antiferromagnetic quantum Heisenberg Hamiltonian, as follows from perturbation theory in small $|T|/U$. Dynamics of the electronic system, however, may involve hopping transitions via intermediate higher bands, which are not captured in the Heisenberg Hamiltonian alone, as well as real hopping processes become relevant at other electronic fillings (as a first correction, one should consider the $T$-$\mathcal{J}$ model [2]). A non-Heisenberg character of magnetic interactions in itinerant systems was explicitly demonstrated, e.g., for the narrow-band Hubbard model on the Bethe lattice beyond half filling [3]. The problem gets much more complicated if one attempts to map more realistic electronic systems to magnetic models: for example, the natural extension of the single-orbital Hubbard model is the *multi*-orbital Hubbard model [4–8], which includes more than just one orbital per site, being a more appropriate description of relevant systems such as $d$ and $f$ materials. Moreover, when both spin and orbital degrees of freedom of the electrons are taken into account, their interplay gives rise to relativistic interactions such as spin–orbit coupling and anisotropies [9].

When no smallness in some characteristic energy parameters of the system can be assumed (such as $|T| \ll U$ in the Hubbard model), the parameters describing the magnetic interactions in an electronic system can be *defined* by imposing the equivalence between the response to spin rotations of a quantity characterizing the system and the analogous response computed for a reference *classical* spin model [1]. In the case of symmetry-broken phases, the quantity which is generally considered is the thermodynamic potential [10–12] computed for an *out-of-equilibrium* state or statistical superposition, that is, either a pure state which is *not* an eigenstate of the electronic Hamiltonian, or a statistical superposition of eigenstates whose weights do *not* depend only on their energies (which would be the case for the Boltzmann distribution, with weights $W_n(\beta) = \mathrm{e}^{-\beta E_n}/Z$, where $E_n$ is the eigenenergy of state $n$, $\beta$ is the inverse temperature and $Z$ is the partition function). The idea of using a symmetry-broken state is similar in spirit to the Higgs mechanism: we need first to solve the non-perturbative many-body problem and find the local moments (massive Higgs fields) and then use the information contained in the single-particle Green's functions and the vertex functions to find perturbatively the soft modes related with exchange interactions. It has been shown that the expressions for the exchange parameters obtained by applying this approach in the non-relativistic case, within the framework of time-dependent density functional theory in the adiabatic approximation, provide an accurate expression for the spin-wave stiffness [13], while the computation of static properties requires the introduction of constraining magnetic fields to equilibrate the non-equilibrium spin configuration [14,15]. However, the corresponding corrections to the exchange parameters [15] are small in the adiabatic approximation, that is, when typical magnon energies are small in comparison with the Stoner splitting [13]. This justifies our approach. In the non-relativistic case, we have recently extended the treatment of Ref. [10] to systems driven explicitly out of equilibrium by time-dependent external electric fields, by considering the potential arising from the non-equilibrium Kadanoff–Baym partition function [16] (in Refs. [10–12,16] the electronic system was modelled by means of the multi-orbital Hubbard model).