Magnetic interactions from first-principles

Mikhail Katsnelson
Outline

1. Introduction
2. Time-dependent DFT and magnetic susceptibility
3. Exchange interactions from first principles
4. Beyond DFT: correlated systems and LDA+DMFT
5. Applications: Fe, Ni, Gd, NiO, CrO$_2$...
6. Dzyaloshinskii-Moriya interactions
7. Application: Molecular magnets
8. Orbital and spin contributions
9. Towards consequent theory of ultrafast spin dynamics
To the theoretical physicists, ferromagnetism presents a number of very interesting, unsolved and beautiful challenges. Our challenge is to understand why it exists at all.

*(Feynman Lectures on Physics)*

Make things as simple as possible but not simpler

*(A. Einstein)*
Magnetite – first known magnet

Very complicated structure, still a lot of open questions

Two types of Fe sites (tetra and octa);
Metal-insulator transition;
Charge ordering;
Role of orbital degrees of freedom;
Half-metallicity...
Types of magnetic ordering

Textbook wisdom

- Ferromagnetic
  - M≠0
  - Fe, Co, Ni...
- Antiferromagnetic
  - M=0
  - NiO....
- Ferrimagnetic
  - M≠0
  - Magnetite...

Sometimes very complicated

α-Mn

Spin spirals

- γ-Fe
- UO₂
Relation to superconductivity and other phenomena

Simplified phase diagram of Cu-O high-Tc superconductors

Layered cobaltates $Na_xCoO_2$
Types of magnetic interactions

The first term: exchange interactions (Heisenberg model)
Quantum, nonrelativistic (Coulomb interaction plus Pauli principle).
Determine the type of magnetic ordering (mostly)

The second term: magnetic anisotropy
Quantum, relativistic (due to spin-orbit interaction). At least, second-order in SOC. Determine “practical” magnetism (hard and soft magnetic materials, hysteresis loop, etc.)

The third term: Dzyaloshinskii-Moriya interactions
Quantum, relativistic (due to spin-orbit interaction). First-order in SOC but require broken inversion symmetry. Responsible for weak FM, skyrmiones etc.
**General formulation**

System of interacting electrons (many-body problem) + crystal potential

External strong time-dependent laser field (nonequilibrium problem)

Temperature effects (thermal bath, open system, basic statistical mechanics)

Collect all difficulties of modern theoretical physics
Levels of description

- Macroscopic (LLG equations + temperature balance, etc.)

- Microscopic, classical Heisenberg model

- Microscopic, quantum itinerant-electron model

- Ab initio, time-dependent density functional

Multiscale problem
Time-dependent DFT

SE for many-body wave function in configurational space is replaced by single-particle nonlinear self-consistent equation

Spinor

\[ \Psi = \begin{pmatrix} \Psi^+ \\ \Psi^- \end{pmatrix} \]

\[ i \frac{\partial \Psi}{\partial t} = \left[ H_L - \hat{\sigma} \cdot \mathbf{B}(\mathbf{r}, t) \right] \Psi \]

\[ H_L = -\nabla^2 + \sum_{R} V_{rR} + 2 \int d\mathbf{r}' \frac{n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} + V_{xc} \]

\( \mathbf{B} \) is self-consistent magnetic field
Simplifications

Adiabatic approx.: $V_{xc}$ and $B_{xc}$ are the same as in the equilibrium + local (spin) density approx.

\[
i \frac{\partial \psi}{\partial t} = H \psi
\]

\[
H = -\nabla^2 + V(\mathbf{r}) - \frac{1}{2} (\mathbf{B}_{xc}(\mathbf{r}) + \mathbf{B}_{\text{ext}}(\mathbf{r})) \sigma
\]

\[
V(\mathbf{r}) = V_{\text{ext}}(\mathbf{r}) + \int d\mathbf{r}' \frac{n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} + \frac{\partial}{\partial n} [n \varepsilon_{xc}]
\]

\[
\mathbf{B}_{xc} = -2 \frac{m}{m} \frac{\partial}{\partial m} [n \varepsilon_{xc}]
\]

\[n,m\] are charge and spin densities
**Linear response: magnetic susceptibility**


\[ B_{\text{ext}}(r) \rightarrow 0 \]

\[ \delta m^\alpha = \hat{\chi}^{\alpha\beta} \delta B^\beta_{\text{ext}} \]

\[ \delta B_{\text{tot}}^\alpha = \delta B_{\text{ext}}^\alpha + \frac{\delta B_{xc}^\alpha}{\delta m^\beta} \delta m^\beta \]

\[ (\hat{\chi} \varphi)(r) = \int dr' \chi(r, r') \varphi(r') \]

At the same time (Runge-Gross theorem, 1984) in TD-DFT

\[ \delta m^\alpha = \hat{\chi}_0^{\alpha\beta} \delta B^\beta_{\text{tot}} \]

A response of effective system of noninteracting Kohn-Sham particles (Liu & Vosko 1989 for magnetic case)
Linear response: magnetic susceptibility II

Rigorous expression:

\[ \hat{\chi}^{\alpha\beta} = \hat{\chi}_0^{\alpha\beta} + \hat{\chi}_0^{\alpha\gamma} \frac{\delta B_{xc}^\gamma}{\delta m^\delta} \hat{\chi}^{\delta\beta} \]

Adiabatic approximation plus LSDA:

\[ \frac{\delta B_{xc}^\gamma}{\delta m^\delta} = \frac{B_{xc}}{m} \left( \delta_{\gamma\delta} - \frac{m^\gamma m^\delta}{m^2} \right) + \frac{m^\gamma m^\delta}{m^2} \frac{\partial B_{xc}}{\partial m} \]

Transverse susceptibility is separated from (longitudinal spin + charge) susceptibilities.
Transverse susceptibility

\[ \chi^{+-}(\mathbf{r}, \mathbf{r}', \omega) = \chi_0^{+-}(\mathbf{r}, \mathbf{r}', \omega) + \int d\mathbf{r}'' \chi_0^{+-}(\mathbf{r}, \mathbf{r}'', \omega) I_{xc}(\mathbf{r}'') \chi^{+-}(\mathbf{r}'', \mathbf{r}', \omega) \]

\[ I_{xc} = \frac{B_{xc}}{m} \]

Local Stoner parameter

\[ m = \sum_{\mu\sigma} \sigma f_{\mu\sigma} |\psi_{\mu\sigma}(\mathbf{r})|^2 \]
\[ n = \sum_{\mu\sigma} f_{\mu\sigma} |\psi_{\mu\sigma}(\mathbf{r})|^2. \]

\[ \chi_0^{+-}(\mathbf{r}, \mathbf{r}', \omega) = \sum_{\mu\nu} \frac{f_{\mu\uparrow} - f_{\nu\downarrow}}{\omega - \varepsilon_{\mu\uparrow} + \varepsilon_{\nu\downarrow}} \psi_{\mu\uparrow}^*(\mathbf{r}) \psi_{\nu\downarrow}(\mathbf{r}) \psi_{\nu\downarrow}^*(\mathbf{r}') \psi_{\mu\uparrow}(\mathbf{r}') \]

Kohn-Sham states

\[ (H_0 - \frac{1}{2} \sigma B_{xc}) \psi_{\mu\sigma} = \varepsilon_{\mu\sigma} \psi_{\mu\sigma} \]
\[ H_0 = -\nabla^2 + V(\mathbf{r}) \]
Longitudinal susceptibility

\[ \chi^{zz} = \frac{1}{4} \left( K^{\uparrow\uparrow} + K^{\downarrow\downarrow} - K^{\uparrow\downarrow} - K^{\downarrow\uparrow} \right) \]

\[ K^{\uparrow\uparrow} = X^{\uparrow} + X^{\uparrow} U^{\uparrow\uparrow} K^{\uparrow\uparrow} + X^{\uparrow} U^{\uparrow\downarrow} K^{\downarrow\uparrow} \]
\[ K^{\downarrow\downarrow} = X^{\downarrow} + X^{\downarrow} U^{\downarrow\downarrow} K^{\downarrow\downarrow} + X^{\downarrow} U^{\uparrow\uparrow} K^{\uparrow\downarrow} \]
\[ K^{\uparrow\downarrow} = X^{\uparrow} U^{\uparrow\downarrow} K^{\downarrow\downarrow} + X^{\uparrow} U^{\uparrow\uparrow} K^{\uparrow\downarrow} \]
\[ K^{\downarrow\uparrow} = X^{\downarrow} U^{\downarrow\uparrow} K^{\uparrow\uparrow} + X^{\downarrow} U^{\downarrow\downarrow} K^{\downarrow\uparrow} . \]

\[ X_\sigma (r, r') = \sum_{\mu \nu} \frac{f_{\mu \sigma} - f_{\nu \sigma}}{\omega - \varepsilon_{\mu \sigma} + \varepsilon_{\nu \sigma}} \psi_{\mu \sigma}^*(r) \psi_{\nu \sigma} (r) \psi_{\mu \sigma} (r') \psi_{\nu \sigma}^* (r') \]

\[ U_{\sigma \sigma'} = \frac{\partial^2 (n \varepsilon_{xc})}{\partial n_{\sigma} \partial n_{\sigma'}} \]
\[ n_{\sigma} = \frac{1}{2} (n + \sigma m) \]
Separation of magnon poles

After rigorous manipulations

\[ \hat{\chi}^{+-} = (m + \hat{\Lambda})(\omega - I_{xc} \hat{\Lambda})^{-1} \]

\[ \Lambda(r, r', \omega) = \sum_{\mu \nu} \frac{f_{\mu \uparrow} - f_{\nu \downarrow}}{\omega - \varepsilon_{\mu \uparrow} + \varepsilon_{\nu \downarrow}} \psi_{\mu \uparrow}^*(r) \psi_{\nu \downarrow}(r) \nabla[\psi_{\mu \uparrow}(r') \nabla \psi_{\nu \downarrow}^*(r') - \psi_{\nu \downarrow}^*(r') \nabla \psi_{\mu \uparrow}(r')] \]

Magnon pole

\[ \omega(q) = \frac{4}{M} \left[ J(0) - J(q) \right] \]

\[ J(r, r', \omega) = \frac{1}{4} \sum_{\mu \nu} \frac{f_{\mu \uparrow} - f_{\nu \downarrow}}{\omega - \varepsilon_{\mu \uparrow} + \varepsilon_{\nu \downarrow}} \psi_{\mu \uparrow}^*(r) B_{xc}(r) \psi_{\nu \downarrow}(r) \psi_{\nu \downarrow}^*(r') B_{xc}(r') \psi_{\mu \uparrow}(r') \]

Im part corresponds to Stoner damping
Alternative definition of exchanges

Static susceptibility

\[ \hat{\chi}^{+-}(0) = m(\hat{\Omega}^{-1} - B_{xc}^{-1}) \]

\[ \hat{\Omega} = \hat{\Omega}(1 - B_{xc}^{-1}\hat{\Omega})^{-1} \]

The first way (poles of susceptibility) corresponds Liechtenstein, MIK & Gubanov, J. Phys. F 1984, the second way (static suscept.) Bruno, PRL 2003. The expressions for stiffness constant coincide and are rigorous within the adiabatic approximation + LSDA.
Nonlocal corrections to magnon stiffness


Exchange and correlation in spiral state of homogeneous electron gas

Angular gradient corrections

\[
E_{xc} = \int d\mathbf{r}\left\{ n \varepsilon_{xc}(n_{\uparrow}, n_{\downarrow}) + \lambda(n_{\uparrow}, n_{\downarrow}) D \right\}
\]

\[
D = (\nabla_{\alpha} e_{\beta})(\nabla_{\alpha} e_{\beta}) = (\nabla \theta)^2 + \sin^2 \theta (\nabla \varphi)^2
\]

\[
\lambda(n_{\uparrow}, n_{\downarrow}) = -\frac{e^2}{16\pi^2}\left(\frac{1}{F} - \frac{4}{3}\right)(V_{xc}p_{F\uparrow} + V_{xc}p_{F\downarrow})
\]

\[
-\frac{e^2}{96\pi^2F^2}(V_{xc} + V_{xc})(p_{F\uparrow} + p_{F\downarrow}).
\]

\[
F = (p_{F\uparrow} + p_{F\downarrow})I(n_{\uparrow}, n_{\downarrow})/2\pi^2
\]

Corrections to stiffness constant
<table>
<thead>
<tr>
<th>Material</th>
<th>Method</th>
<th>LSDA</th>
<th>Gradient Corrections</th>
<th>Experiment Range</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe</td>
<td>LSDA</td>
<td>239</td>
<td>251</td>
<td>280 - 310</td>
</tr>
<tr>
<td></td>
<td>with gradient corrections</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ni</td>
<td>LSDA</td>
<td>692</td>
<td>735</td>
<td>550-630</td>
</tr>
<tr>
<td></td>
<td>with gradient corrections</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Corrections are quite small.
Stoner damping in Fe and Ni

Antropov, Harmon, Smirnov, JMMM 200, 148 (1999)
Magnetic force theorem

(Lichtenstein & MIK 1984)

Total energy in DF:

\[ E = E_{sp} - E_{dc} \]
\[ E_{sp} = \sum_{\text{occ}} \varepsilon_v \]
\[ E_{dc} = E_{\text{Hartree}} + \int d\mathbf{r} Tr \left[ \rho \frac{\delta E_{xc}}{\delta \rho} \right] - E_{xc} \]

Variation at fixed potential:

\[ \delta E = \delta^* E_{sp} + \delta_1 E_{sp} - \delta E_{dc} = \delta^* E_{sp} = \delta^* \int d\varepsilon \left[ \frac{1}{\pi} Tr \text{Im} \hat{G}(\varepsilon) \right] \]

\[ \delta^* \quad \text{at fixed potential} \]

\[ \delta_1 \quad \text{due to change of potential} \]
Magnetic force theorem II

- Torque can be written in terms of variation of the density of states
- Decomposition of the torque in pair terms gives exchange integrals (LK)
- These exchange parameters are local (near given magnetic configuration)
- Adding constrain to stabilize rotated configuration gives exchange parameters (Bruno)

Exchange parameters for $d$ metals are strongly non-Heisenbergian (depend on magnetic configuration) (Turzhevskii, Lichtenstein & MIK, Fiz. Tverd. Tela 1990)
Example: magnetism of Fe, Co, Ni

Ferromagnetism of iron is known from ancient times.
Itinerant-electron ferromagnetism at finite temperatures

T = 0

Stoner

Heisenberg

Spin-fluctuation
**Stoner criterion**

The equation for the Curie temperature is:

\[ I_{\text{eff}} N(E_F) > 1 \]

where \( I_{\text{eff}} \) is an on-site interaction parameter, and \( N(E_F) \) is the density of one-electron states.

Stoner parameter \( \approx 0.9 \text{ eV for all 3d metals; DOS is crucially important} \)

Equation for the Curie temperature:

\[ I_{\text{eff}} \int dE \left( -\frac{\partial f}{\partial E} \right) N(E) = 1 \]

If Fe would be Stoner magnet it would have \( T_C \approx 4000 \text{ K} \) (in reality \( 1043 \text{ K} \))

In reality, \( T_C \) is determined by spin fluctuations. That is, exchange parameters.
Iron: some details

Crystal field splitting

DOS for nonmagnetic bcc Fe

Stoner criterion is fulfilled due to $e_g$ states only; they should play a special role in magnetism of Fe (Irkhin, Katsnelson, Trefilov, JPCM 5, 8763 (1993))
Non-Heisenberg character of exchange interactions in Fe and Ni


Rotation of a central spin: magnetic moment is not constant, energy change is not cosine

Fig. 4.4. Magnetic moment in Bohr magnetons (the full curve) and the first derivative of energy with respect to angle of rotation in Ry (the dashed curve) according to calculations in [168]: (a) Fe, (b) Ni.

Electronic structure is angle-dependent

Fig. 4.5. Electronic density of states for an Fe atom in a metal with the magnetic moment turned through $\theta = 0$ (a), $\theta = 0.2\pi$ (b), $\theta = 0.35\pi$ (c), and $\theta = 0.5\pi$ (d).
Iron: detailed analysis

Microscopic Origin of Heisenberg and Non-Heisenberg Exchange Interactions in Ferromagnetic bcc Fe


FIG. 1. Orbitally decomposed NN exchange interaction in elemental 3d metals in the bcc structure.

\( t_{2g} \) are itinerant electrons providing (Heisenberg-like) RKKY exchange with Friedel oscillations; \( e_g \) are more correlated providing (non-Heisenberg) “double exchange” typical for narrow-band systems.
Problem with DFT: coexistence of localized and itinerant behavior

Local magnetic moments do exist above $T_C$ (Curie-Weiss law, spectroscopy, neutrons...)

d electrons are itinerant (FS, chemical bonding, transport...)

Iron, majority spin FS

4f electrons are normally pure localized but not 3d
From atomic state to itinerant

Correlated Electrons Step by Step: Itinerant-to-Localized Transition of Fe Impurities in Free-Electron Metal Hosts


Experiment: disappearance of multiplets

Calculations: increase of hybridization

Blue line: exact diagonalization for free atom
**Dynamical Mean Field Theory I**

A. Georges, G. Kotliar, W. Krauth and M. Rozenberg, Rev. Mod. Phys. ‘96

A natural generalization of the familiar MFT to the problem of electrons in a lattice

**Key idea:** take one site out of a lattice and embed it in a self-consistent bath = mapping to an effective impurity problem
Dynamical Mean Field Theory II

\[ \hat{G}(i\omega_n) = \frac{1}{\Omega} \sum_{\vec{k}} \hat{G}(\vec{k}, i\omega_n) \]

\[ \hat{G}_0^{-1}(i\omega_n) = \hat{G}^{-1}(i\omega_n) + \hat{\Sigma}(i\omega_n) \]

\[ \hat{\Sigma}_{\text{new}}(i\omega_n) = \hat{G}_0^{-1}(i\omega_n) - \hat{G}^{-1}(i\omega_n) \]

Single Impurity Solver

W. Metzner and D. Vollhardt (1987)
A. Georges and G. Kotliar (1992)
Ferromagnetism of transition metals: LDA+DMFT

Ferromagnetic Ni DMFT vs. LSDA:

- 30% band narrowing
- 50% spin-splitting reduction
- -6 eV satellite

LDA+DMFT with ME
J. Braun et al.
PRL (2006)

Lichtenstein, MIK, Kotliar, PRL (2001)
Orbital magnetic moments

For Fe\textsubscript{x}Co\textsubscript{1-x} alloys
LDA+Disordered Local Moments

The best first-principle Spin-fluctuation model with classical moments

J. Staunton and B. Gyorffy
PRL 69, 371 (1992)
**DMFT Effective Magnetic Moments: \( T > T_c \)**

<table>
<thead>
<tr>
<th>W</th>
<th>exp</th>
<th>eff</th>
<th>loc</th>
<th>DLM</th>
<th>( T_c )</th>
<th>exp</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe</td>
<td>3.13</td>
<td>3.09</td>
<td>2.8</td>
<td>1.96</td>
<td>1900</td>
<td>1043</td>
</tr>
<tr>
<td>Ni</td>
<td>1.62</td>
<td>1.5</td>
<td>1.3</td>
<td>1.21</td>
<td>700</td>
<td>631</td>
</tr>
</tbody>
</table>

\[ \chi(\bar{T}) \]

\[ \frac{M(\bar{T})}{M(0)} \]

\[ \frac{\chi(\bar{T})}{M_{\text{eff}}^2 / 3T_c} \]
Agreement is not bad (much better than LDA/GGA) but essentially worse than in nickel. Correlations in iron are not quite local.
**ARPES for 3d metals**

Effects of spin-dependent quasiparticle renormalization in Fe, Co, and Ni photoemission spectra: An experimental and theoretical study


**TABLE I.** Values of the experimental and theoretical mass enhancement factors $m^* / m_0$ for majority spin states at high symmetry points of the BBZ of Fe, Co, and Ni, respectively. The theoretical values are derived for $U(\text{Fe}) = 1.5$ eV, $U(\text{Co}) = 2.5$ eV, $U(\text{Ni}) = 2.8$ eV.

<table>
<thead>
<tr>
<th></th>
<th>Fe</th>
<th>Co</th>
<th>Ni</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Gamma$</td>
<td>1.7</td>
<td>1.2</td>
<td>1.26</td>
</tr>
<tr>
<td>$\Lambda$</td>
<td>1.1</td>
<td>1.2</td>
<td>1.29</td>
</tr>
</tbody>
</table>

Variation of $U$ does not help too much for Fe

Black – spin up, red – spin down

Upper panel – exper, lower - DMFT
Why Ni is more local than Fe?

Nickel is almost half-metallic: majority-spin FS almost coincides with the boundaries of the Brillouin band.

But the difference for minority spin is even more dramatic.

Occupations for majority (minority) electrons:
- Fe: 4.6 (2.34)
- Ni: 4.82 (4.15)
**Why Ni is more local than Fe II**

Friedel oscillations originating from FS are much weaker in nickel

As a result:

Magnons are much softer in Fe than in Ni (Curie temp. Higher but magnon frequencies lower)

The softer magnons the stronger nonlocal e-m intercation
Exchange and Functionals

Magnetic force theorem

\[ \Omega^d = \Omega^d_{sp} - \Omega^d_{dc} \]

\[ \Omega^d_{sp} = -Tr \left\{ \ln \left[ \Sigma - G_0^{-1} \right] \right\} \]

\[ \Omega^d_{dc} = Tr \Sigma G - \Phi \]

\[ G^{-1} = G_0^{-1} - \Sigma \]

\[ \Sigma = \frac{\delta \Phi}{\delta G} \]

\[ \delta \Omega = \delta^* \Omega_{sp} + \delta_1 \Omega_{sp} - \delta \Omega_{dc} \]

\[ \delta_1 \Omega_{sp} = \delta \Omega_{dc} = Tr G \delta \Sigma \]

\[ \delta \Omega = \delta^* \Omega_{sp} = -\delta^* Tr \ln \left[ \Sigma - G_0^{-1} \right] \]
**LDA+DMFT**


<table>
<thead>
<tr>
<th>LSDA</th>
<th>LDA++</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density functional</td>
<td>Baym-Kadanoff functional</td>
</tr>
<tr>
<td>Density $\rho(r)$</td>
<td>Green-Function $G(r, r', E)$</td>
</tr>
<tr>
<td>Potential $V_{xc}(r)$</td>
<td>Self-energy $\Sigma_i(E)$</td>
</tr>
<tr>
<td>$E_{tot} = E_{sp} - E_{dc}$</td>
<td>$\Omega = \Omega_{sp} - \Omega_{dc}$</td>
</tr>
<tr>
<td>$E_{sp} = \sum_{\lambda&lt;\lambda_F} \varepsilon_\lambda$</td>
<td>$\Omega_{sp} = -Tr \ln[-G^{-1}]$</td>
</tr>
<tr>
<td>$E_{dc} = E_H + \int \rho V_{xc} dr - E_{xc}$</td>
<td>$\Omega_{dc} = Tr \Sigma G - \Phi_{LW}$</td>
</tr>
<tr>
<td>Temperature:</td>
<td>Matsubara frequencies: real-T</td>
</tr>
<tr>
<td>in the Fermi function</td>
<td>for collective excitations</td>
</tr>
</tbody>
</table>
Exchange interactions from DMFT

Heisenberg exchange:

\[ H = - \sum_{ij} J_{ij} S_i S_j \]

Magnetic torque:

\[ \delta \Omega = \delta^* \Omega_{sp} = V_i \delta \varphi_i \]

\[ V_i = 2 Tr_{\omega L} [\Sigma_i^s \times G_i^s] \]

Exchange interactions:

\[ J_{ij} = - Tr_{\omega L} (\Sigma_i^s G_{ij}^\uparrow \Sigma_j^s G_{ji}^\downarrow) \]

Spin wave spectrum:

\[ \omega_q = \frac{4}{M} \sum_j J_{0j} \left( 1 - \cos q R_j \right) \equiv \frac{4}{M} [J(0) - J(q)] \]

Non-collinear magnetism:

**Alternative view**

First- and second-order smallness in theta angle!

\[ \delta H = \sum_{ij} \text{Tr}_{L \sigma} \left[ t_{ij} c_i^+ (U_i^+ U_j - 1) c_j \right] = \delta_1 H + \delta_2 H \]

\[ \delta_1 H = \sin^2 \frac{\theta}{2} \sum_k \text{Tr}_{L \sigma} \left[ \left( t (k+q) - t (k) \right) c_k^+ c_k \right] \]

\[ \delta_2 H = \frac{1}{2} \sin \theta \sum_{ij} \text{Tr}_L \left[ t_{ij} c_{i \downarrow}^+ c_{j \uparrow} \right] \times \left( \exp (iq \mathbf{R}_i) - \exp (iq \mathbf{R}_j) \right) . \]

Total energy corrections by diagram technique *neglecting vertex corrections* → our exchanges

\[ \omega_q = D_{\alpha \beta} q_\alpha q_\beta , \quad q \to 0 \]

\[ D_{\alpha \beta} = - \frac{2}{M} \text{Tr}_{\omega L} \sum_k \left( \sum_s \frac{\partial G^\uparrow (k)}{\partial k_\alpha} \sum_s \frac{\partial G^\downarrow (k)}{\partial k_\beta} \right) \]

Exact within DMFT (local self-energy!)
Applications

Nontrivial: electronic structure is very different!

For Fe (and Ni) quite small difference between DFT and DMFT

Error cancellation?!
Applications II

Does not follow a naive formula
Difference between Mott and charge transfer insulator

NiO: not too big difference between DMFT and LDA + U

Gd: also, DFT works quite good
Applications III

Half-metallic FM DMFT shows non-quasiparticle states in the gap
MIK et al, RMP 80, 315 (2008)
Applications IV

Important consequences from DMFT contributions to exchange

Without magnetic polarization of oxygen FM state is unstable within DMFT (but not in simpler approaches)

Direct exchange also plays an important role

FIG. 9. (Color online) Results of calculations of the spin-wave dispersion with the DMFT parameters obtained for the isolated $t_{2g}$ band (solid line) and after taking into account the additional FM contribution $\Delta J_2 = 17.81$ meV, arising from magnetic polarization of the oxygen band and direct exchange interactions in the $t_{2g}$ band (dotted line). Notations of the high-symmetry points of the BZ are taken from [55].
Dzialoshinskii-Moriya interactions

MIK, Kvashnin, Mazurenko & Lichtenstein, PRB 82, 100403 (2010)

LDA+U

$\hat{H} = \hat{H}_t + \hat{H}_u$

$= \sum_{12} c_1^+ t_{12} c_2 + \frac{1}{2} \sum_{1234} c_1^+ c_2^+ U_{1234} c_3 c_4$

DM interactions (weak FM, etc.)

$H_{D M} = \sum_{i j} \tilde{D}_{i j} [\vec{e}_i \times \vec{e}_j ]$

Small rotations

$\hat{R}_i = e^{i\delta \phi_i} \hat{J}$

$\hat{J} = \hat{L} + \hat{S}$
Dzialoshinskii-Moriya interactions II

Starting from collinear configuration

\[ \delta \hat{H}_t = \sum_{ij} c_i^+ (\delta \hat{R}_i^+ \hat{t}_{ij} + \hat{t}_{ij} \delta \hat{R}_j) c_j \]

\[ = -i \sum_{ij} c_i^+ (\delta \bar{\varphi}_i \hat{\bar{J}} \hat{t}_{ij} - \hat{t}_{ij} \hat{\bar{J}} \delta \bar{\varphi}_j) c_j \]

\[ = -\frac{i}{2} \sum_{ij} c_i^+ (\delta \bar{\varphi}_i - \delta \bar{\varphi}_j) (\hat{\bar{J}} \hat{t}_{ij} + \hat{t}_{ij} \hat{\bar{J}}) c_j \]

\[ \tilde{D}_{ij} = -\frac{i}{2} Tr_{m,\sigma} \langle c_i^+ [\hat{\bar{J}}, \hat{t}_{ij}] + c_j \rangle = -\frac{i}{2} Tr_{m,\sigma} N_{ji} [\hat{\bar{J}}, \hat{t}_{ij}] + \]

\[ N_{ji} = \langle c_i^+ c_j \rangle = -\frac{1}{\pi} \int_{-\infty}^{E_f} \text{Im} G_{ji}(E) dE \]
Applications to La$_2$CuO$_4$

Canting angle 0.005
Exper. 0.003

TABLE II: Different contributions to Dzyaloshinskii-Moriya vector (in meV).

<table>
<thead>
<tr>
<th></th>
<th>$\vec{R}_{1j}$</th>
<th>$\vec{D}_{1j}^{spin}$</th>
<th>$\vec{D}_{1j}^{orb}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>(1,2)</td>
<td>(-0.005; -0.006; 0.0)</td>
<td>(-0.07; -0.03; 0.0)</td>
<td></td>
</tr>
<tr>
<td>(1,3)</td>
<td>(-0.005; 0.006; 0.0)</td>
<td>(-0.07; 0.03; 0.0)</td>
<td></td>
</tr>
<tr>
<td>(1,4)</td>
<td>(-0.005; -0.006; 0.0)</td>
<td>(-0.07; -0.03; 0.0)</td>
<td></td>
</tr>
<tr>
<td>(1,5)</td>
<td>(-0.005; 0.006; 0.0)</td>
<td>(-0.07; 0.03; 0.0)</td>
<td></td>
</tr>
</tbody>
</table>
FeBO₃

A novel exper. technique to measure DM vector and not only canting angle (resonant X-ray scattering)

Measuring the Dzyaloshinskii–Moriya interaction in a weak ferromagnet

V. E. Dmitrienko¹, E. N. Ovchinnikova², S. P. Collins³*, G. Nisbet³, G. Beutier⁴, Y. O. Kvashnin⁵, V. V. Mazurenko⁶, A. I. Lichtenstein⁷ and M. I. Katsnelson⁵,⁸

TABLE I. Calculated values of isotropic exchange interactions between magnetic moments in FeBO₃ (in meV). The number in parentheses denotes the coordination sphere.

<table>
<thead>
<tr>
<th></th>
<th>Fe(1)</th>
<th>Fe(2)</th>
<th>Fe(3)</th>
<th>Fe(4)</th>
<th>Fe(5)</th>
<th>Fe(6)</th>
<th>Fe(7)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>10.28</td>
<td>0.21</td>
<td>0</td>
<td>0.54</td>
<td>-0.08</td>
<td>0</td>
<td>0.02</td>
</tr>
</tbody>
</table>

TABLE III. Parameters of Dzyaloshinskii–Moriya interaction (in meV) calculated by using Eq. (6).

<table>
<thead>
<tr>
<th>Bond m − n</th>
<th>( R_{mn} )</th>
<th>( D_{mn} ) (meV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0-1</td>
<td>(1.0 ; 0.0 ; -0.904)</td>
<td>(-0.25; 0.0; -0.24)</td>
</tr>
<tr>
<td>0-2</td>
<td>(-0.5 ; -( \sqrt{3}/2 ); -0.904)</td>
<td>(0.12 ; 0.22 ; -0.24)</td>
</tr>
<tr>
<td>0-3</td>
<td>(-0.5 ; ( \sqrt{3}/2 ); -0.904)</td>
<td>(0.12 ; -0.22 ; -0.24)</td>
</tr>
<tr>
<td>0-4</td>
<td>(-0.5 ; 0.0 ; 0.904)</td>
<td>(-0.25; 0.0; -0.24)</td>
</tr>
<tr>
<td>0-5</td>
<td>(0.5 ; -( \sqrt{3}/2 ); 0.904)</td>
<td>(0.12 ; -0.22 ; -0.24)</td>
</tr>
<tr>
<td>0-6</td>
<td>(0.5 ; ( \sqrt{3}/2 ); 0.904)</td>
<td>(0.12 ; 0.22 ; -0.24)</td>
</tr>
</tbody>
</table>

Agrees well with exper.
Molecular magnets

Example: $V_{15}$

AFM ground state $S = 1/2$

$V_{15}(K_6[V_{15}As_6O_{42}(H_2O)] \cdot 8H_2O)$
**LDA+U calculations**

PHYSICAL REVIEW B 70, 054417 (2004)

Electronic structure and exchange interactions in $V_{15}$ magnetic molecules: LDA+U results

D. W. Boukhvalov, V. V. Dobrovitski, M. I. Katsnelson, A. I. Lichtenstein, B. N. Harmon, and P. Kögerler

---

TABLE II. The exchange parameters (in Kelvin), electronic gap, and the magnetic moments of $V$ ions for different magnetic structures of $V_{15}$. The calculations have been made for $U=4$ eV, $J=0.8$ eV.

<table>
<thead>
<tr>
<th>parameter</th>
<th>AFM1</th>
<th>AFM2</th>
<th>FM</th>
</tr>
</thead>
<tbody>
<tr>
<td>$J$</td>
<td>−910</td>
<td>−905</td>
<td>−942</td>
</tr>
<tr>
<td>$J'$</td>
<td>−45</td>
<td>−46</td>
<td>−53</td>
</tr>
<tr>
<td>$J''$</td>
<td>−136</td>
<td>−139</td>
<td>−156</td>
</tr>
<tr>
<td>$J_1$</td>
<td>−219</td>
<td>−247</td>
<td>−255</td>
</tr>
<tr>
<td>$J_2$</td>
<td>−134</td>
<td>−128</td>
<td>−132</td>
</tr>
<tr>
<td>$J_3$</td>
<td>−5</td>
<td>−6</td>
<td></td>
</tr>
<tr>
<td>$J_4$</td>
<td>−13</td>
<td>−12</td>
<td>−15</td>
</tr>
<tr>
<td>$J_5$</td>
<td>−3</td>
<td>−3</td>
<td>−3</td>
</tr>
<tr>
<td>$J_6$</td>
<td>−3</td>
<td>−3</td>
<td>−3</td>
</tr>
<tr>
<td>gap</td>
<td>1.08</td>
<td>1.02</td>
<td>1.16</td>
</tr>
<tr>
<td>$\mu_{V1}$</td>
<td>−0.94</td>
<td>−0.93</td>
<td>−0.99</td>
</tr>
<tr>
<td>$\mu_{V2}$</td>
<td>+0.91</td>
<td>+0.92</td>
<td>−0.97</td>
</tr>
<tr>
<td>$\mu_{V3}$</td>
<td>−1.00</td>
<td>+0.97</td>
<td>−1.00</td>
</tr>
</tbody>
</table>
**LDA+U calculations II**

Exact diagonalization for Heisenberg model

- Diagram showing energy of the state vs total spin of the state.
- Graphs showing magnetic moment vs temperature for different values of $U$.

- Graph (a) has data points for experiment and constant $U$ values ranging from 4.2 to 5.4 eV.
- Graph (b) has similar data with a focus on different temperatures from 0 to 300 K.

Charts illustrate the relationship between the energy of a state and its total spin, as well as the magnetic moment as a function of temperature for various $U$ values.
**Motivation**

The prototype molecular magnet

Dimension of Hilbert space:

\[(2^2+1)^8(2\times3/2+1)^4=10^8\]

A real challenge!
Inelastic neutron scattering data: cannot be explained without strong DM interactions (MIK, Dobrovistki & Harmon, PRB 1999)

Eight-spin model: \( S = \frac{1}{2} \) dimers from \( S=2 \) and \( S=3/2 \)

Dimensionality of Hilbert space decreases to \( 10^4 \)

Cannot be justified quantitatively!

**Full LDA+U calculations plus Lanczos ED**

\[
\hat{H} = \sum_{ij} J_{ij} \hat{S}_i \hat{S}_j + \sum_{i \mu \nu} \hat{S}_i^\mu A_{i}^{\mu \nu} \hat{S}_i^\nu + \sum_{ij} \vec{D}_{ij} [\hat{S}_i \times \hat{S}_j]
\]

**TABLE I.** Intramolecular isotropic exchange interaction parameters (in meV) calculated by using the LDA + U approach. Positive sign corresponds to the antiferromagnetic coupling.

<table>
<thead>
<tr>
<th>Bond ((i, j))</th>
<th>1–6</th>
<th>1–11</th>
<th>1–9</th>
<th>6–9</th>
<th>7–9</th>
<th>1–4</th>
<th>1–3</th>
</tr>
</thead>
<tbody>
<tr>
<td>( J_{ij} ) (this work)</td>
<td>4.6</td>
<td>1.0</td>
<td>1.7</td>
<td>-0.45</td>
<td>-0.37</td>
<td>-1.55</td>
<td>-0.5</td>
</tr>
<tr>
<td>( J_{ij} ) (Ref. [4])</td>
<td>4.8</td>
<td>1.37</td>
<td>1.37</td>
<td>-0.5</td>
<td>-0.5</td>
<td>-1.6</td>
<td>-0.7</td>
</tr>
<tr>
<td>( J_{ij} ) (Ref. [26])</td>
<td>7.4</td>
<td>1.72</td>
<td>1.72</td>
<td>-1.98</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
### Mn\textsubscript{12}: full calculations III

Plus anisotropy tensors...

#### TABLE II. Intramolecular anisotropic exchange interaction parameters calculated by using the LDA + U approach. \( R_{ij} \) is a radius vector connecting \( i \)th and \( j \)th atoms (in units of \( a = 17.31 \) Å).

<table>
<thead>
<tr>
<th>Bond ((i,j))</th>
<th>( \vec{R}_{ij} )</th>
<th>( \vec{D}_{ij} ) (meV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2–7</td>
<td>((0.03; -0.16; 0.0))</td>
<td>((-0.008; -0.013; -0.002))</td>
</tr>
<tr>
<td>4–8</td>
<td>((-0.03; 0.16; 0.0))</td>
<td>((0.008; -0.013; -0.002))</td>
</tr>
<tr>
<td>1–6</td>
<td>((0.16; 0.03; 0.0))</td>
<td>((-0.013; 0.008; -0.002))</td>
</tr>
<tr>
<td>3–5</td>
<td>((-0.16; -0.03; 0.0))</td>
<td>((0.013; -0.008; -0.002))</td>
</tr>
<tr>
<td>1–11</td>
<td>((0.06; 0.18; 0.07))</td>
<td>((-0.020; -0.03; -0.055))</td>
</tr>
<tr>
<td>3–10</td>
<td>((-0.06; -0.18; 0.07))</td>
<td>((0.020; -0.03; -0.055))</td>
</tr>
<tr>
<td>2–9</td>
<td>((0.18; -0.06; -0.07))</td>
<td>((-0.03; -0.020; -0.055))</td>
</tr>
<tr>
<td>4–12</td>
<td>((-0.18; 0.06; -0.07))</td>
<td>((0.03; 0.020; -0.055))</td>
</tr>
<tr>
<td>1–9</td>
<td>((0.11; -0.16; 0.04))</td>
<td>((0.020; 0.014; 0.03))</td>
</tr>
<tr>
<td>3–12</td>
<td>((-0.11; 0.16; 0.04))</td>
<td>((-0.020; -0.014; 0.03))</td>
</tr>
<tr>
<td>2–10</td>
<td>((-0.16; -0.11; -0.04))</td>
<td>((-0.014; 0.020; 0.03))</td>
</tr>
<tr>
<td>4–11</td>
<td>((0.16; 0.11; -0.04))</td>
<td>((0.014; -0.020; 0.03))</td>
</tr>
<tr>
<td>6–9</td>
<td>((-0.04; -0.18; 0.04))</td>
<td>((-0.006; -0.004; -0.012))</td>
</tr>
<tr>
<td>5–12</td>
<td>((0.04; 0.18; 0.04))</td>
<td>((0.006; 0.004; -0.012))</td>
</tr>
<tr>
<td>7–10</td>
<td>((-0.18; 0.04; -0.04))</td>
<td>((0.004; -0.006; -0.012))</td>
</tr>
<tr>
<td>8–11</td>
<td>((0.18; -0.04; -0.04))</td>
<td>((-0.004; 0.006; -0.012))</td>
</tr>
<tr>
<td>7–9</td>
<td>((0.15; 0.1; -0.07))</td>
<td>((0.020; -0.004; 0.012))</td>
</tr>
<tr>
<td>8–12</td>
<td>((-0.15; -0.1; -0.07))</td>
<td>((-0.020; 0.004; 0.012))</td>
</tr>
<tr>
<td>6–11</td>
<td>((-0.1; 0.15; 0.07))</td>
<td>((-0.004; -0.020; 0.012))</td>
</tr>
<tr>
<td>5–10</td>
<td>((0.1; -0.15; 0.07))</td>
<td>((0.004; 0.020; 0.012))</td>
</tr>
<tr>
<td>4–1</td>
<td>((-0.10; 0.06; 0.11))</td>
<td>((-0.014; 0.005; -0.013))</td>
</tr>
<tr>
<td>1–2</td>
<td>((-0.06; -0.10; 0.11))</td>
<td>((-0.005; -0.014; -0.013))</td>
</tr>
<tr>
<td>3–4</td>
<td>((0.07; 0.1; 0.11))</td>
<td>((0.005; 0.014; -0.013))</td>
</tr>
<tr>
<td>2–3</td>
<td>((-0.10; 0.07; -0.11))</td>
<td>((0.014; -0.005; -0.013))</td>
</tr>
<tr>
<td>1–3</td>
<td>((-0.16; -0.03; 0.0))</td>
<td>((-0.006; 0.030; 0))</td>
</tr>
<tr>
<td>2–4</td>
<td>((-0.04; 0.17; 0.0))</td>
<td>((-0.030; -0.006; 0))</td>
</tr>
</tbody>
</table>

**FIG. 2.** (Color online) Schematic comparison of the theoretical spectrum obtained by diagonalizing Eq. (1) and INS spectrum taken from Ref. [12] (Figs. 6 and 8 therein). The arrows denote the intra- and interband transitions that correspond to the excitations observed in the INS experiment.

No fitting parameters at all – not so bad!
Mn$_{12}$: full calculations IV

Also, thermodynamic quantities can be calculated
Spin and orbital contributions

Magnetic interactions in strongly correlated systems: Spin and orbital contributions

A. Secchi\textsuperscript{a,*}, A.I. Lichtenstein\textsuperscript{b}, M.I. Katsnelson\textsuperscript{a}


Spin and orbital exchange interactions from Dynamical Mean Field Theory

A. Secchi\textsuperscript{a,*}, A.I. Lichtenstein\textsuperscript{b}, M.I. Katsnelson\textsuperscript{a}

Journal of Magnetism and Magnetic Materials 400 (2016) 112–116

Rotation operator involves both spin and orbital rotations

\[ \hat{H}_T = \sum_i \sum_{i'} \hat{\psi}_{i1}^\dagger \cdot e^{-i \delta \phi_{i1} \cdot \mathbf{s}_{i1}} \cdot T_{i2}^{i1} \cdot e^{i \delta \phi_{i2} \cdot \mathbf{s}_{i2}} \cdot \hat{\psi}_{i2} \]

No smallness of SOC is assumed

Calculate the change of energy at small rotations, map to the classical spin orbital Hamiltonian

\[ H[\mathbf{e}_i] = \sum_i \mathbf{e}_i \cdot \mathbf{B}_i + \frac{1}{2} \sum_{i,i'} \sum_{\alpha,\alpha'} \mathbf{e}_{i\alpha} \mathbf{e}_{i'\alpha'} \mathcal{H}_{ii'}^{\alpha\alpha'} \]

\[ \mathcal{H}_{ij}^{\alpha\beta} = \delta^{\alpha\beta} \mathcal{H}_{ij}^{\alpha} + \sum_{\gamma} (\epsilon^{\alpha\beta\gamma} \mathcal{D}_{ij}^{\gamma} + |\epsilon^{\alpha\beta\gamma}| \mathcal{C}_{ij}^{\gamma}) \]
Spin and orbital contributions II

\[ \mathcal{J}_{00'} \equiv \mathcal{J}^{\text{spin-spin}}_{00'} + \mathcal{J}^{\text{orb-orb}}_{00'} + \mathcal{J}^{\text{spin-orb}}_{00'} \]

Decomposition of exchange parameters and similar for other interactions

Hopping can be excluded using the Dyson equations

\[
(\omega - i\mu)G(i\omega) + iT\cdot G(i\omega) = 1 - \Sigma(i\omega)\cdot G(i\omega),
\]

\[
(\omega - i\mu)G(i\omega) + iG(i\omega)\cdot T = 1 - G(i\omega)\cdot \Sigma(i\omega).
\]

E.g., for DM interactions

\[
\left(\mathcal{D}_{00'}^{x}\right)^{\text{spin}} = \frac{i}{2} \text{Tr}_{m,\sigma} \text{Tr}_{\omega} \left[ s_{0x} \cdot \left( G_0^0 \cdot \Sigma_0^{0'} - \Sigma_0^{0'} \cdot G_0^0 \right) - s_{0x} \cdot \left( G_0^{0'} \cdot \Sigma_0^0 - \Sigma_0^0 \cdot G_0^{0'} \right) \right],
\]

\[
\left(\mathcal{D}_{00'}^x\right)^{\text{orb}} = \frac{i}{2} \text{Tr}_{m,\sigma} \text{Tr}_{\omega} \left[ l_{0x} \cdot \left( G_0^0 \cdot \Sigma_0^{0'} - \Sigma_0^{0'} \cdot G_0^0 \right) - l_{0x} \cdot \left( G_0^{0'} \cdot \Sigma_0^0 - \Sigma_0^0 \cdot G_0^{0'} \right) \right].
\]

and similar for exchanges. Important for actinides, RE, and 3d systems with unquenched orbital moments (e.g., CoO)
Ultrafast magnetism: Examples

- Nickel Koopmans et al, PRL 2005
- Orthoferrites, Kimel et al, Nature 2005
- Gadolinium, Melnikov et al, PRL 2003
Ultrafast magnetism: a theory

Non-equilibrium magnetic interactions in strongly correlated systems

A. Secchi\textsuperscript{a,\ast}, S. Brener\textsuperscript{b}, A.I. Lichtenstein\textsuperscript{b}, M.I. Katsnelson\textsuperscript{a}

\[ \hat{H}(t) \equiv \hat{H}_T(t) + \hat{H}_V \]

\[ \hat{H}_T(t) \equiv \sum_{i_a \lambda_a} \sum_{i_b \lambda_b} T_{i_a \lambda_a, i_b \lambda_b}(t) \sum_{\sigma} \phi^\dagger_{i_a \lambda_a \sigma} \phi_{i_b \lambda_b \sigma} \]

\[ \hat{H}_V \equiv \frac{1}{2} \sum_i \sum_{\lambda_1 \lambda_2 \lambda_3 \lambda_4} \sum_{\sigma \sigma'} V_{\lambda_1 \lambda_2 \lambda_3 \lambda_4} \phi_{i \lambda_1 \sigma} \phi^\dagger_{i \lambda_2 \sigma'} \phi^\dagger_{i \lambda_3 \sigma'} \phi_{i \lambda_4 \sigma} \]

Consider dynamics of Baym-Kadanoff-Keldysh countour

Path integral over Grassmann variables

\[ \mathcal{Z} = \int D[\phi, \phi] e^{iS[\phi, \phi]} \]
Ultrafast magnetism: a theory II

Introduce rotations

\[
\begin{align*}
\tilde{\phi}_{a\pm}(t) &= \tilde{\psi}_{a\pm}(t) \cdot R_{a\pm}^\dagger(t), & \phi_{a\pm}(t) &= R_{a\pm}(t) \cdot \psi_{a\pm}(t) \\
\tilde{\phi}_{av}(\tau) &= \tilde{\psi}_{av}(\tau) \cdot R_{av}^\dagger(\tau), & \phi_{av}(\tau) &= R_{av}(\tau) \cdot \psi_{av}(\tau)
\end{align*}
\]

Expand effective actions up to the second order in "Holstein-Primakoff" fields \( \xi, \xi^* \)

\[
R_i(z) \equiv \begin{pmatrix} \sqrt{1 - |\xi_i(z)|^2} & \xi_i^*(z) \\ -\xi_i(z) & \sqrt{1 - |\xi_i(z)|^2} \end{pmatrix}
\]

\[
\xi_i(z) \equiv -e^{i\varphi_i(z)} \sin [\theta_i(z)/2]
\]

\[
Z = \int \mathcal{D}[\tilde{\psi}, \psi] e^{iS[\tilde{\psi}, \psi]} \int \mathcal{D}[\theta, \varphi] e^{iS'[\tilde{\psi}, \psi, \xi^*(\theta, \varphi), \xi(\theta, \varphi)]}
\]

Integrate over Grassman variables neglecting vertex corrections
Ultrafast magnetism: a theory III

General expression of nonlocal in time exchange interactions in terms of Beym-Kadanoff-Keldysh Green’s functions. E.g., time-dependent stiffness constant:

\[ D_{\alpha\beta}(t) \equiv -\frac{i}{2M} \sum_{\eta} \sum_{\sigma} \int_{t_0}^{\infty} dt' \text{sign}(t' - t) \overline{\Sigma}^{S}(t) \overline{\Sigma}^{S}(t') \]

\[ \times \frac{1}{n} \sum_{k} \frac{\partial G_{k}^{\eta\sigma}(t', t)}{\partial k_{\alpha}} \frac{\partial G_{k}^{\bar{\eta}\bar{\sigma}}(t, t')}{\partial k_{\beta}}. \]

Additional terms (twist exchange) of the structure

(at equilibrium forbidden by time-reversal symmetry)

The first step is done, a lot of things to do
Beyond the talk

Finite-temperature effects

Ab initio spin dynamics for real systems

Intermediate level: TB spin dynamics

And many, many specific applications to real materials
Collaboration

Recent:

A. Lichtenstein and S. Brener (Hamburg)
A. Secchi and A. Rudenko (Nijmegen)
V. Mazurenko (Ekaterinburg)
Ya. Kvashnin and O. Eriksson (Uppsala)

and many other people involved in development of the formalism and calculations for specific materials in 1987-2013, esp. V. Antropov (Ames) and D. Boukhvalov (Seoul)

Thank you for your attention