Multiscale modeling of magnetic materials: Synergy of ab-initio and model methods

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Multiscale modeling

Simple 'brute force' methods

dynamics: 'let's run our program longer' methods

statics: 'let's describe several length scales problem using single scale technique'

not practical!

Clever 'brute force' methods

Wavelets, multigrids and different coarse graining methods in MD and SD

Still not practical!

Multiscale modeling

$$\dot{\mathbf{m}} = \mathbf{m}\times\mathbf{B} + \lambda\mathbf{m}\times\mathbf{B}\times\mathbf{m}$$

$$m\frac{d^2x}{dt^2} + c\frac{dx}{dt} + kx = 0$$

introduction of the 'stiffness'
c and λ
is a profound way of masking the effect of the unaccounted for degrees of freedom which a repository for the center of mass motion accosiated with the particle.

In reality this is a progressive donation of kinetic energy from this particle to the rest of the world which is responsible for the ostensible loss of energy

Even so elementary a model of magnetization dynamics reflects the same philosophy of separation of scales

A key factor is that a diversity of spatial and temporal scales produces properties which could not be achieved at different scales separately

Length scales and theory applicability

	Rare-earth magnets	3d-Pt	3d
DW width	**~ 5 nm**	**~ 10 nm**	**~ 100 nm**

LDA

model

Computing problems

We would like to have supercell smaller as possible but LDA should work

Parameters versus full ab-initio in CoPt computational time

$$t_{model} \,/\, t_{ab\text{-}initio} \sim 1/50$$

Synergistic approach: a combination of different single scale technique

bringing more than one theoretical tool with different regions of applicability under the same roof

first principle methods
micromagnetics
microstructural simulation

Practical!

But!

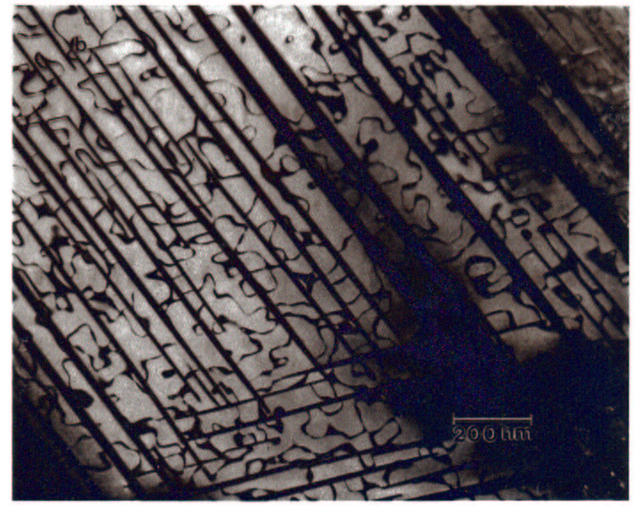
(i) it requires professional experts from each area who can handle all the tools

(ii) unfortunate feature that it is exceedingly difficult to make analytical progress with them

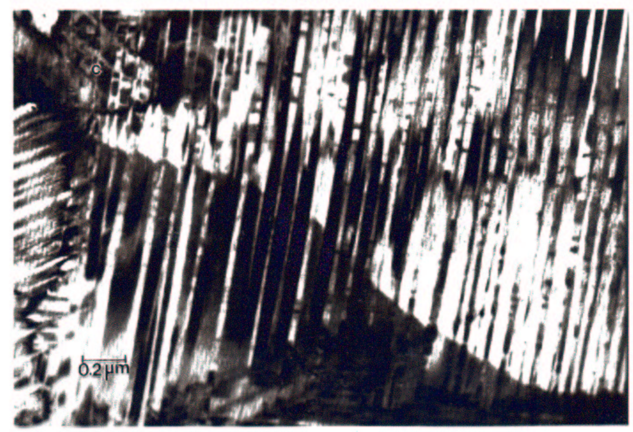
MSM should also be seen as a way of answering to our inability to rely on pure brute force approaches in modeling.

Polytwinned FePd near peak hardness

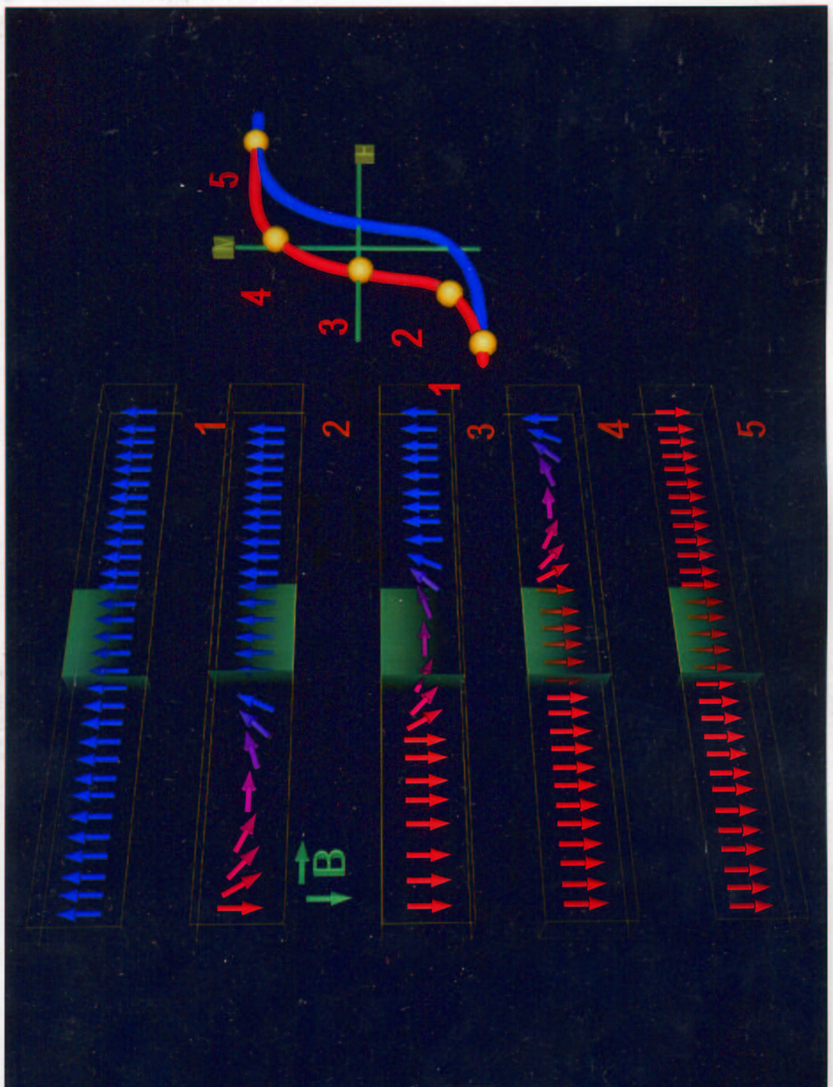
MICROSTRUCTURE (TEM)



MACRODOMAIN WALL (Lorentz microscopy)

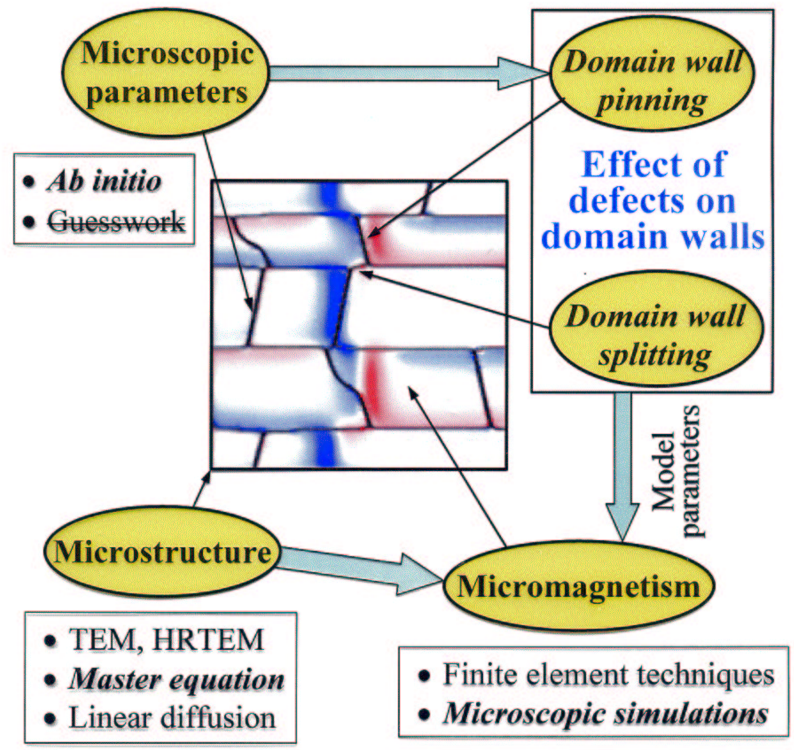


B. Zhang and W.A. Soffa, 1992



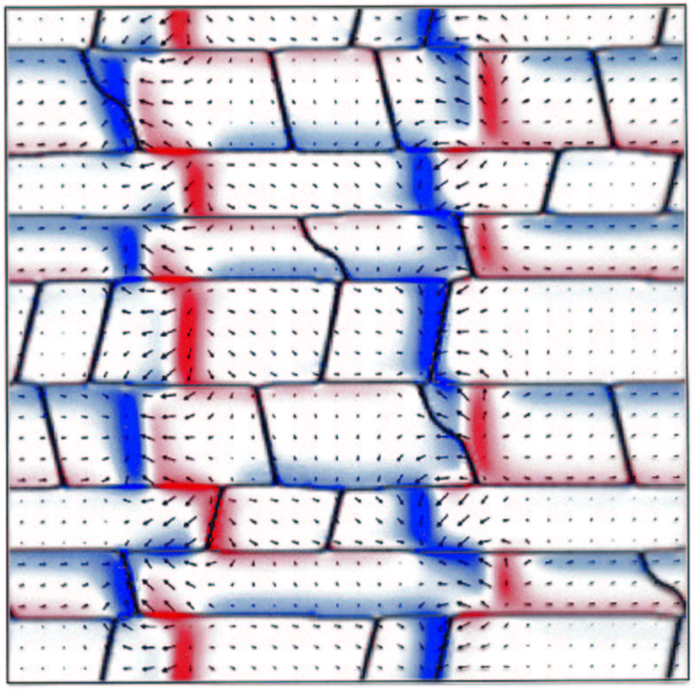
Multiscale physics of coercivity

- Defect size ~ 1 nm
- Domain wall width 5-10 nm
- Microstructural scales:
  - Thickness of twins 15-100 nm
  - Distance between defects 15-300 nm
  - Size of a twinned stack ~ 1 μm



Macrodomain walls in the CoPt model with realistic microstructure

Magnetic charges $\rho = -\operatorname{div} \mathbf{M}$, color (+/–)
Dipole fields $\mathbf{H}_{dip}(\mathbf{r})$, arrows



Simulation box: 256 x 256 x 1 *a* (70 nm edge)

Conclusions

1. Magnetization processes are inherently **multi-scale** and **material-specific**. The problem itself calls for a **synergistic** approach.
2. **Microstructure** of CoPt type magnets is dominated by twin boundaries and antiphase boundaries. Both of them play an important role in the formation of **coercivity**:

   A. **Antiphase boundaries** are strong **pinning centers** due to local suppression of the magnetocrystalline anisotropy.

   B. Macrodomain walls **split** in segments at **twin boundaries**. These magnetostatically coupled segments may **adapt** to local pinning potential, increasing pinning efficiency.

   C. Coercivity emerges as a **combined effect** of domain wall pinning and splitting.
3. Macrodomain walls were simulated for a **realistic microstructure** obtained using the **master equation approach**.

K.D. Belashchenko, V.P. Antropov, cond-mat/0110526.

Three main parameters

Microscopical theory can produce for the next level methods following:

Exchange parameters J

Spin wave stiffness D (A)

Magnetic anisotropy K

Spin wave stiffness

$$\omega_q \approx Dq^2$$

all famous magnets are anisotropic

Co-Pt, Sm-Co, Nd-B-Fe

in reality D has a matrix structure

$$D_\perp , \qquad D_\parallel$$

??

Magnetic anisotropy

$$K = E(100) - E(001)$$

Definition problem in the case of defects
Range of perturbation

High level of accuracy!

Conclusions

Exchange interactions in DFT

$$J_{\gamma\delta} = \delta^2 \mathrm{E} / \delta \mathrm{m}_\gamma(\mathbf{r}) \delta \mathrm{m}_\delta(\mathbf{r}')$$

rigid spin $J_{ij} = \partial^2 \mathrm{E} / \partial \mathbf{m}_i \partial \mathbf{m}_j$

local approximation $\mathbf{B(r)} = I\, \mathbf{m(r)}$

rigid spin+ local definition

$$J_{ij} = I_i \partial^2 \mathrm{E} / \partial \mathbf{B}_i \partial \mathbf{B}_j I_j$$

models:
Morya (1974), S. Liu (1978), Korenman (1979)

$$J_{ij} = I_i \chi_{ij} I_j$$

DFT implementation: Lichtenstein (1984)

$$J_{ij} = \int \Delta_i T_{ij}^{\uparrow} T_{ji}^{\downarrow} \Delta_j d\varepsilon$$

approximations:

(i) local approximation is true only for exchange correlation field;
(ii) spin is rigid, no gradients of **m(r)**

$$\delta E / \delta \mathbf{m}(\mathbf{R}_i + \mathbf{r}) \approx \partial E / \partial \mathbf{m}_i$$

Physical perturbation

$$m_\alpha(\mathbf{r}) = \chi_{\alpha\beta} B_\beta(\mathbf{r}'),$$

$$\chi_{\alpha\beta} = \delta m_\alpha / \delta B_\beta = \chi_{\alpha\gamma} \delta^2 E / \delta m_\gamma(\mathbf{r}) \delta m_\delta(\mathbf{r}') \chi_{\delta\beta} = \chi_{\alpha\gamma} J_{\gamma\delta} \chi_{\delta\beta}$$

$$\int dr'' J(\mathbf{r},\mathbf{r}'',\omega)\chi(\mathbf{r}'',\mathbf{r}',\omega) = \delta(\mathbf{r}-\mathbf{r}')$$

enhanced exchange coupling

$$J(\mathbf{r},\mathbf{r}',\omega) = J_0(\mathbf{r},\mathbf{r}',\omega) - I_{exc}(\mathbf{r},\mathbf{r}',\omega)$$

static version of the exact exchange

$$J(\mathbf{q}) = \chi(\mathbf{q})^{-1}$$

localized moment definition

$$J(\mathbf{q}) = I\chi(\mathbf{q})I$$

itinerant description

$$\omega_{\mathbf{q}} = m\left(\chi^{-1}(\mathbf{q}) - \chi^{-1}(0)\right)$$

localized description

$$\omega^0_{\mathbf{q}} = m\left(J(0) - J(\mathbf{q})\right) = mI\left(\chi(0) - \chi(\mathbf{q})\right) I$$

$$\omega_{\mathbf{q}} = \omega^0_{\mathbf{q}}\left(1 - \chi_0 \omega^0_{\mathbf{q}}\right)^{-1}$$

smallness parameter in q-space and r-space

$$\left(\chi(\mathbf{q}) - \chi(0)\right)/\chi(0) << 1$$

$\chi_{ij}/\chi_{ii} << 1$ or $\boldsymbol{\omega} / \mathbf{mI} << 1$

I~0.9 eV

fcc FM **Ni**:	**ω** ~ 0-0.4 eV	0.4-0.5
bcc FM **Fe**:	**ω** ~ 0-0.3 eV	0.3-0.4
hcp FM **Gd**:	**ω** ~ 0-0.01 eV	0.01-0.02

$$J(\mathbf{q}) = \chi(\mathbf{q})^{-1}$$

vs

$$J(\mathbf{q}) = I\chi(\mathbf{q})I$$

Long wave length
small q – small errors

$$\omega_q \approx Dq^2$$

D **is correct!**

Short wave length
large q -- large errors

nearest atoms exchange is bad

high temperatures, non-collinear structures

one –site approximation: DMFT, LDA, LDA+U, Gutzwiller …

(1) in all previous calculations the interatomic coupling parameters were at least underestimated when the long wave approximation has been used (localized model). NN J can be wrong.

(2) Spin wave stiffness is OK and as a rule has a matrix structure (anisotropy of D can be as low as 30%).

(3) magnetic anisotropy is very sensitive quantity. Very high accuracy is required.

Microscopical methods

Density functional

$$(H_L - \sigma \mathbf{B})\Psi(\mathbf{r}) = E\Psi(\mathbf{r}); \quad H_L = -\nabla_r^2 + V_{rR} + 2\int d\mathbf{r}' \frac{n(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} + V_{xc}$$

Hubbard model

$$H = w\sum n_{i\sigma} + t\sum\left[a^+ a + c.c.\right] + U\sum n_\uparrow n_\downarrow$$

Theoretical and experimental characterics of band structure for 3d metals: specific heat and bandwidth (mJ/mol K^2 and eV)

3d	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu
γ theor	5.25	1.4	4.31	1.67	6.49	2.63	2.08	4.13	0.7
γ exp	10.7	3.35	9.26	1.4	9.20	4.98	4.73	7.02	0.69
W theor	5.13	6.08	6.77	6.56	5.6	4.82	4.32	3.78	2.8
W exp	6.2	6.6	6.8	6.5	8.5	8.5	6.9	5.4	2.6

Realistic models of correlated systems

Density functional theory (LDA)

Self-consistent, fast

Many body perturbation theory

Green function approach

No self-consistency, slow, limited diagrams

LDA+U, dynamic mean field

Model parameters (U,J), exact solution of impurity problem, slow

'Self-consistent Green function approach for calculations of electronic structure in transition metals'

N.E.Zein, V.P.Antropov, cond-mat/0202483

one site approximation
effective contour integration
GW and beyond GW

Self-consistent, fast.

For transition metals our approach can be considered as a practical ab-initio alternative to modern DFT methods with a much wider range of applicability.

Ab-initio correlated electron theory

Slater (1950-1960)
All results similar to modern LDA

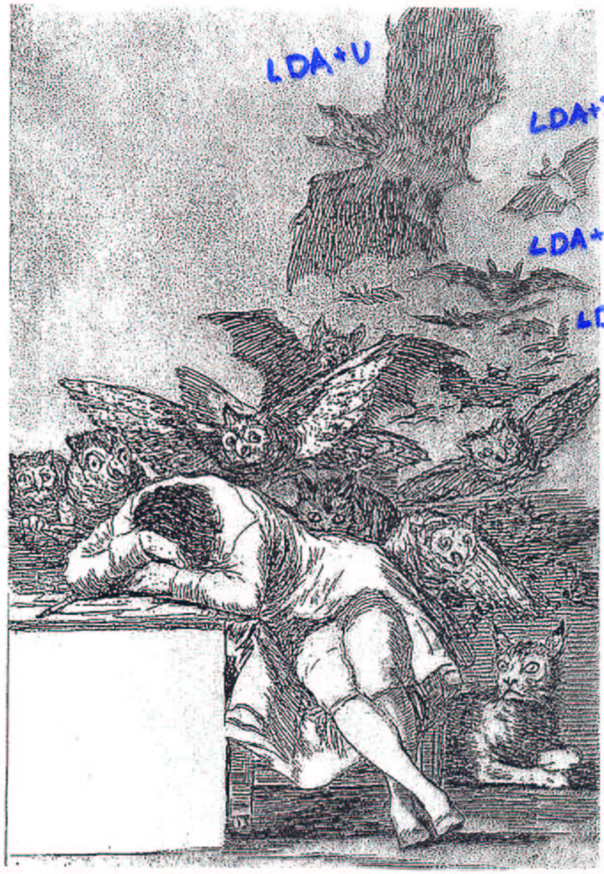
GW (Hedin 1965)

Nothing after

Model techniques

1991 – LDA+U
('manual' technique, parametrized)

The Sleep of Reason Produces Monsters.



Many body Green function technique

(HF → GW → FLEX →.....)

+ one site approximation for the correlations (metals, not for semiconductors).

Faster :

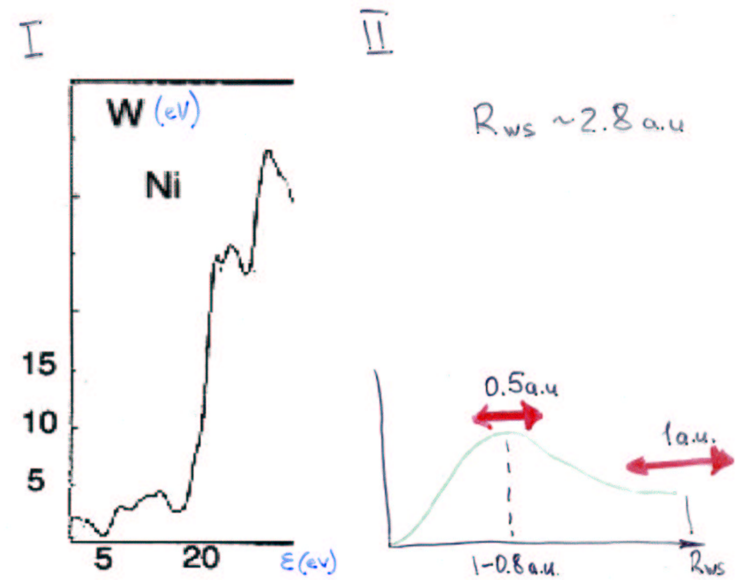
→ self-consistency (independent on initial wave functions)

→ more diagrams (can be improved)

Only screened exchange effects (only LDA+U effects).

Advantage:

effective interaction is energy dependent and resolved on intraatomic scale (non-locality/ l-dependence)



$$\lambda = \chi^{-1}, \chi^2 = 4\pi e^2 \Pi_0(\rho(r))$$

$$\Pi_0(\rho(r)) = p_F / \pi^2 = (3/\pi\rho)^{1/3} / \pi$$

U must be resolved on atomic scale and be energy dependent

Many body Green-function approach

one-site approximation

$$\Sigma(\mathbf{r},\mathbf{r}',\varepsilon,\mathbf{k}) \approx \Sigma(\mathbf{r},\mathbf{r}',\varepsilon)$$

self consistency

$$G^{-1}(\mathbf{k},\varepsilon) = \left\langle \dot{\phi}\, h_{\mathbf{k}} + \phi \middle| \varepsilon - \hat{T} - V_H - \Sigma(r,r',\varepsilon) \middle| \phi + h_{\mathbf{k}}\dot{\phi} \right\rangle$$

$$G^{-1}(\mathbf{k},\varepsilon_l)\phi_l = 0 \quad G^{-1}(\mathbf{k},\varepsilon_l)\dot{\phi}_l = -\phi_l + \frac{\partial\Sigma}{\partial\varepsilon}\phi_l$$

$$G(\mathbf{r},\mathbf{r}',\omega) = \sum_{\mathbf{k}} \left(G_0(\mathbf{r},\mathbf{r}',\omega,\mathbf{k}) - \Sigma(\mathbf{r},\mathbf{r}',\omega)\right)^{-1}$$

$$\Sigma = \int GW$$

$$\mathrm{W} = \mathrm{V_c} / (1 + \mathrm{V_c}\Pi)$$

$$\Pi(\mathbf{r},\mathbf{r}',\omega) = \int \mathrm{G}(\varepsilon)\mathrm{G}(\varepsilon+\omega)\mathrm{d}\omega / 2\pi\mathrm{i}$$

Luttinger-Ward Functional

$$\Omega = -Tr\left\{\ln\left[\Sigma - G_0^{-1}\right]\right\} - Tr\Sigma G - \Phi$$

$$E = E_{sp} - E_{dc} - E_{corr}$$

(GW-approximation)

$$\Phi = -\left(\int \frac{d\omega}{2\pi i} \ln det(1 + \Pi V) - \Pi V\right)$$

self-energy $\Sigma = \delta\Phi/\delta G$

$$\Sigma(k) = -\int G(k-p)\Pi(p)\frac{d\omega}{2\pi i}$$

under the Fermi level

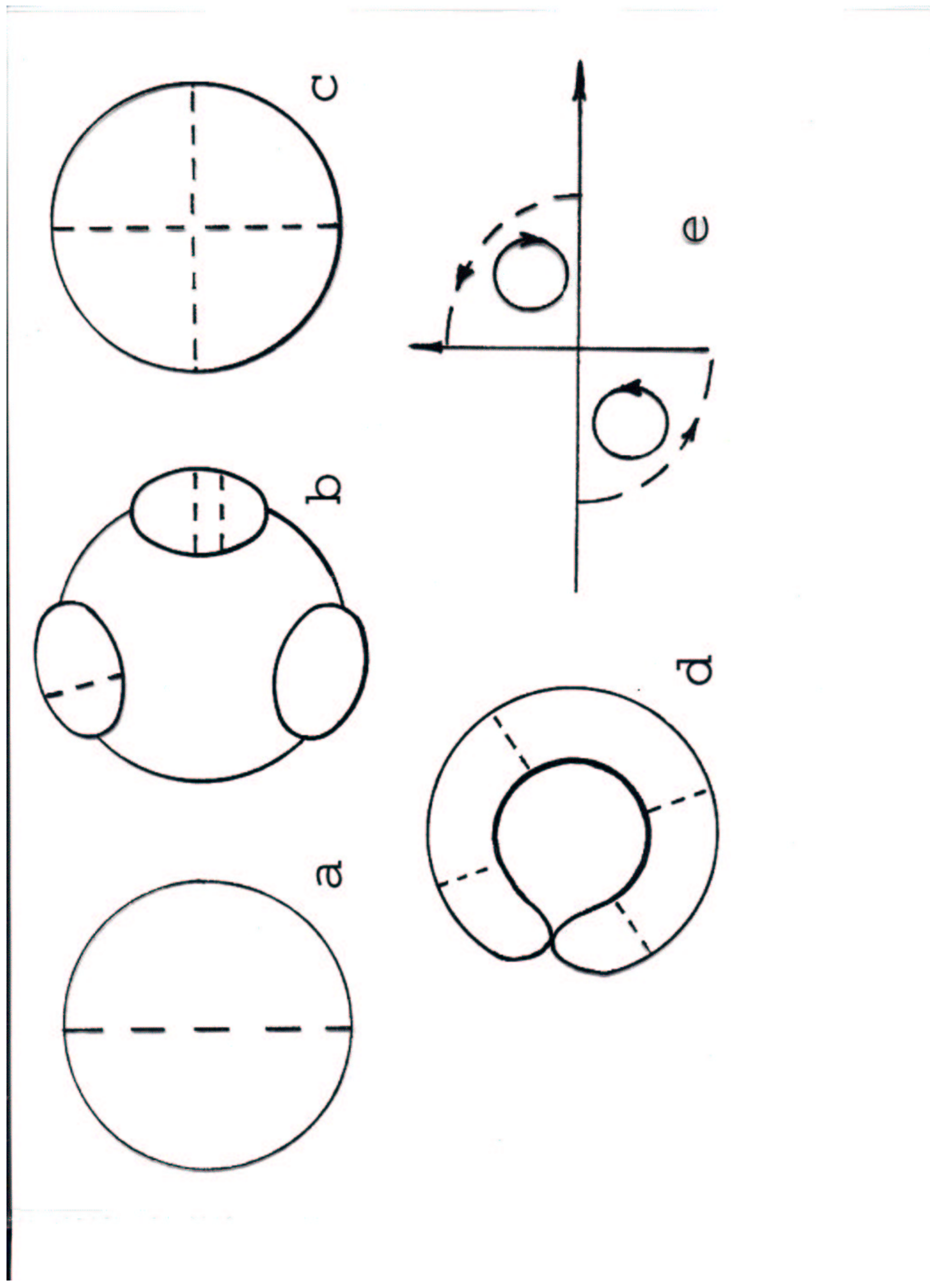
$$\Sigma^p_{ca} = \int_\varepsilon^0 g(\omega')\Pi(\varepsilon-\omega')d\omega' vD \quad \varepsilon < 0$$

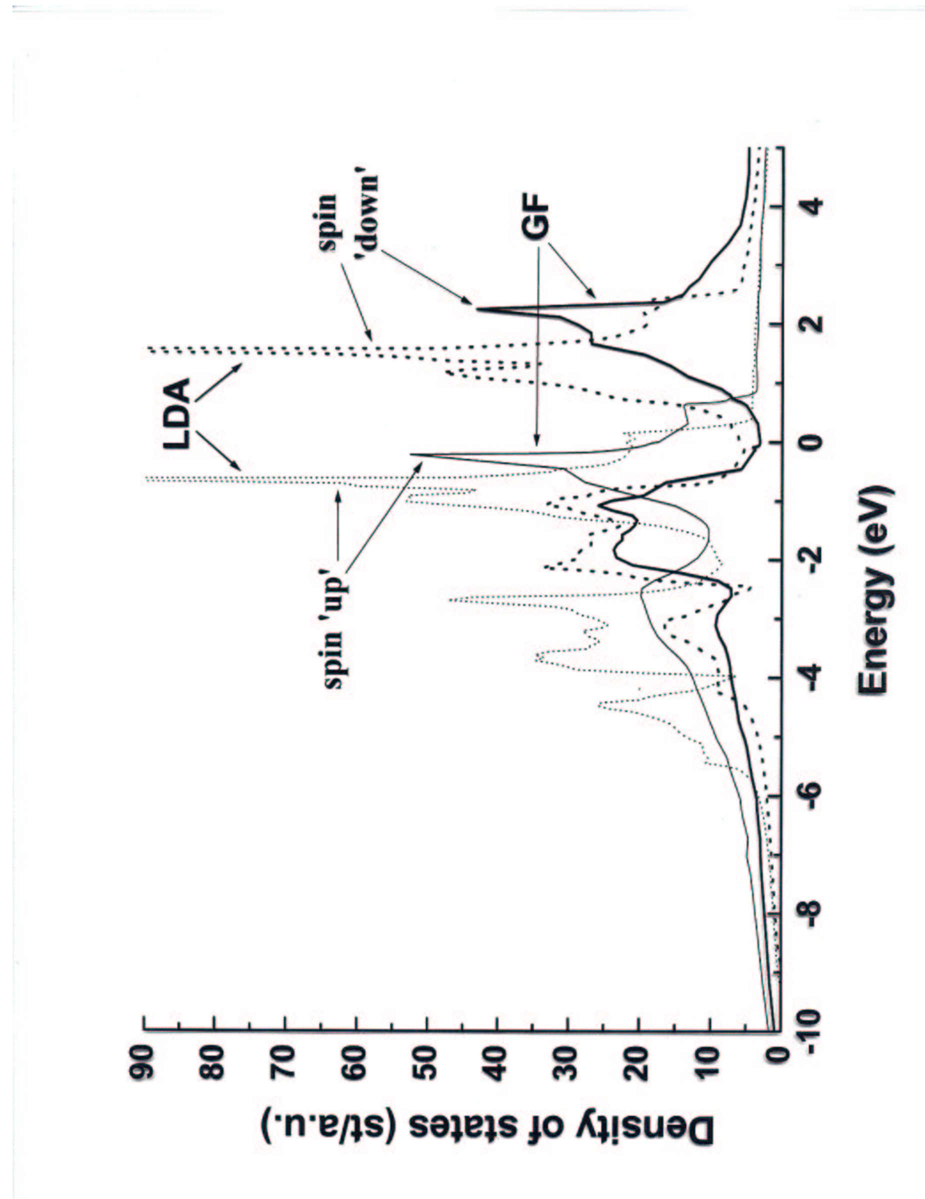
and the term, corresponding to the integration over the imaginary axe

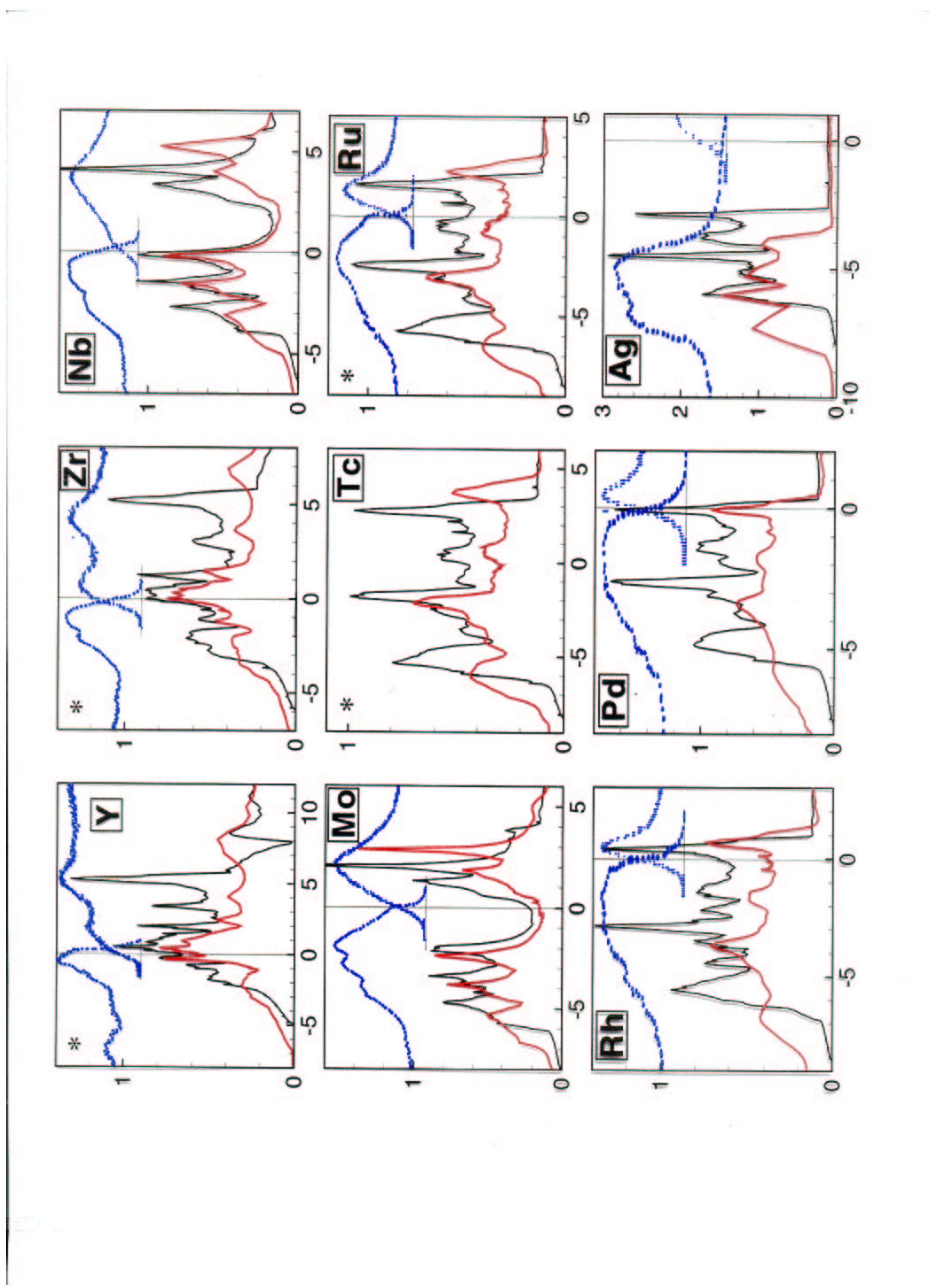
$$\Sigma^i_{ca} = \int_{-\infty}^{\infty} d\omega' \int_0^{\infty} g(\omega')\frac{2(\varepsilon-\omega')}{(\varepsilon-\omega')^2+\omega^2}\Pi(i\omega)d\omega'\frac{d\omega}{2\pi i}vD$$

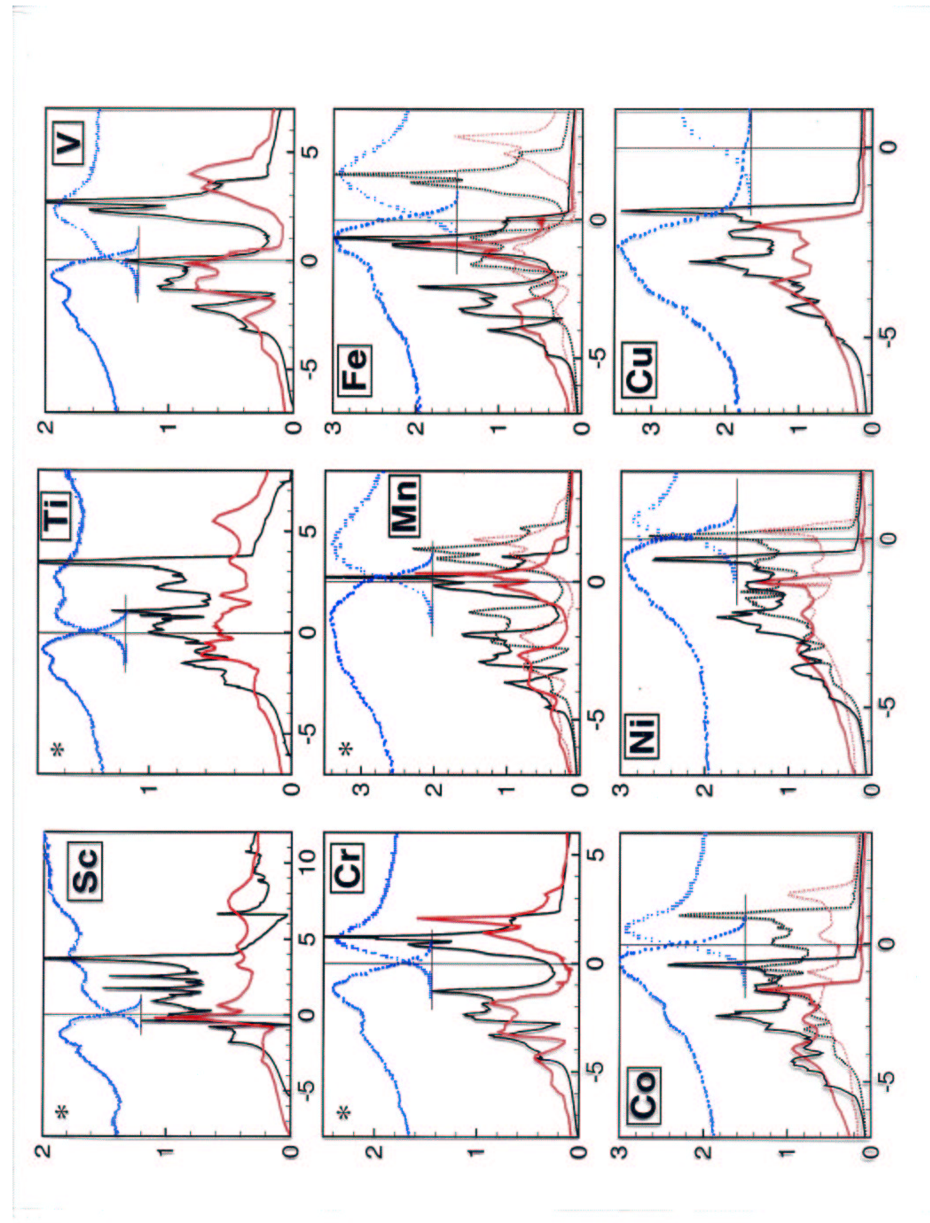
Then for the non-local potential we obtain

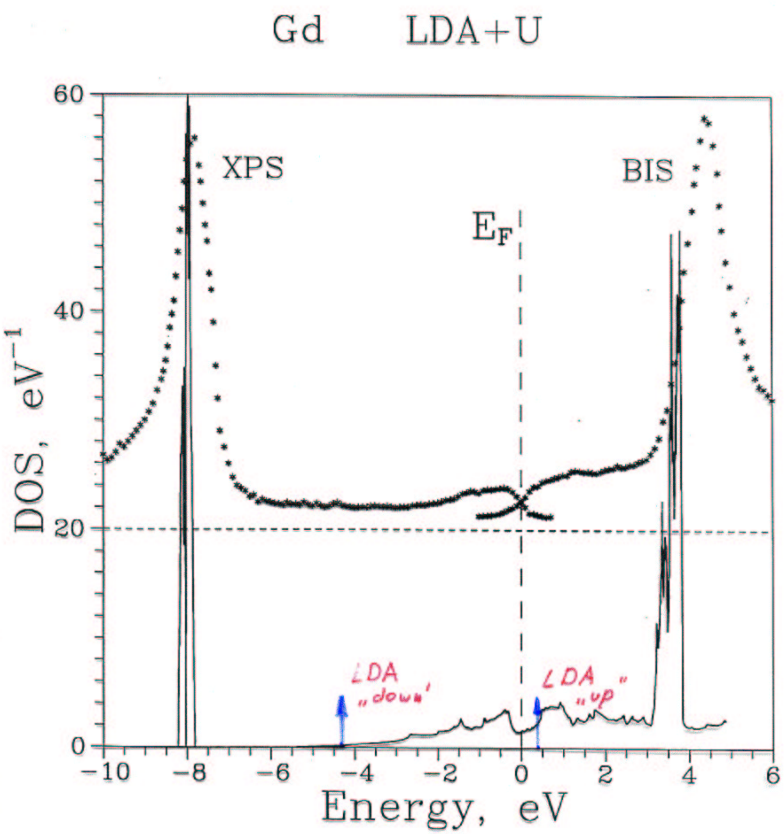
$$V(\mathbf{r},\mathbf{r}') = \Sigma_x + V_c(\mathbf{r})\delta(\mathbf{r}-\mathbf{r}') + \Sigma^p_{ca}(\varepsilon_p) + \Sigma^i_{ca}(\varepsilon_p,\varepsilon_c) + \Sigma^{iloc}_{ca}(0,\varepsilon_c)$$


a
b
c
d
e

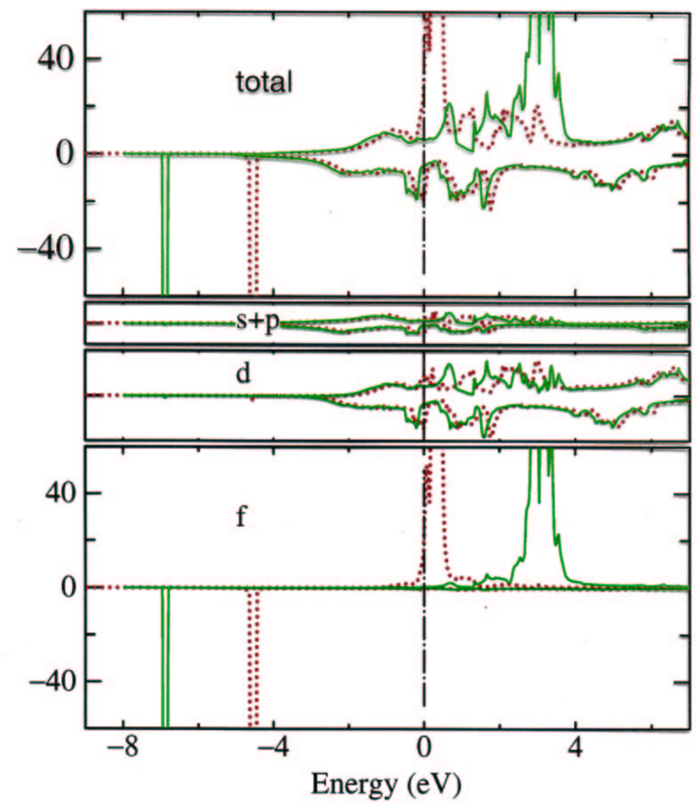

LDA
GF
spin 'down'
spin 'up'
Energy (eV)
Density of states (st/a.u.)


Y
Zr
Nb
Mo
Tc
Ru
Rh
Pd
Ag


Sc
Ti
V
Cr
Mn
Fe
Co
Ni
Cu



Gd LDA+U
XPS
BIS
E_F
LDA „down"
LDA „up"
DOS, eV^-1
Energy, eV



total
s+p
d
f
Energy (eV)

Electronic structure results

	GF	LDA	exp	GF	LDA	exp
M	2.09	2.15	2.08	0.65	0.62	0.59
$N(\varepsilon_F)$ st/a.u.	28.0	30.0	57.0	44.0	48.0	81.0
$Z_d=1-\partial\Sigma/\partial\varepsilon$	0.7			0.6		

The small value of Z is due to the polarization contribution.

Conclusions

1. A new self-consistent version of the GF approach, which uses the quasiparticle wave functions as a basis set.

2. Self-consistent GW approach produces the leading contribution to the electronic structure and magnetic properties of TM, whereas the addition of fluctuating exchange diagrams overall slightly corrects this result improving comparison with experiment.

3. While the self-consistent renormalization factors Z for s and p electrons in Ni and Fe are close to the estimations obtained from the HEG, Z_d is much smaller (0.6-0.7).

4. The values of bandwidth and density of states at the Fermi level are in reasonable agreement with experiment.

The proposed technique can be considered as a practical *ab-initio* alternative to modern DFT methods (LDA,LDA+U) with much wider range of applicability.