Comparison of different approaches to ab-initio calculations of the spin wave stiffness

How to get similar results from equivalent expressions

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Outline

- Spin stiffness D / exchange stiffness A_{ex}:
  What it is good for?
- Two ways to evaluate spin stiffness D.
- Issues with D calculations: "larger than small" spread of results.
- Checking potential risk factors.
  Drawing practical advice.
- Effects of spin-orbit coupling on the spin stiffness D.

Why to care about the stiffness?

Micromagnetics:

Continuum approximation, the angle of the magnetization changes slowly over atomic distances, spin vectors are replaced by a continuous function $\mathbf{m}(\mathbf{r})$.

$$E[\mathbf{m}] = \int_V \mathrm{d}\mathbf{r} \left\{ A_{\text{ex}} \left[\left(\frac{\partial \mathbf{m}}{\partial x} \right)^2 + \left(\frac{\partial \mathbf{m}}{\partial y} \right)^2 + \left(\frac{\partial \mathbf{m}}{\partial z} \right)^2 \right] + \dots \right\}$$

Exchange stiffness A_{ex}, spin wave stiffness D:

$$A_{\text{ex}} = \frac{D\, M_s}{2 g \mu_B}$$

M_s is saturation magnetization, g is Landé factor ($g \approx 2$ for metals).

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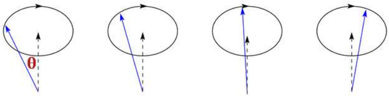
Spin stiffness D from spin spirals

Long wavelength limit of the acoustic mode of magnon dispersion:

$$\epsilon(\mathbf{q}) \;=\; D\,|\mathbf{q}|^2 \;+\; \beta\,|\mathbf{q}|^4 \;+\; \ldots$$



For cubic systems:

Dispersion relation for magnon spectra $\epsilon(\mathbf{q})$ can be obtained from the change of the total energy per unit cell due to a spin spiral state with wave vector $\mathbf{q}$, spiral cone angle θ, and magnetic moment per site M,

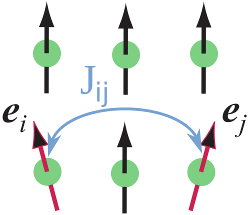
$$\epsilon(\mathbf{q}) \;=\; \lim_{\theta\to 0}\, \frac{4\mu_B}{M}\, \frac{E(\mathbf{q},\theta) \;-\; E(0,\theta)}{\sin^2\theta} \qquad .$$

[Kübler, Theory of itinerant electron magnetism (2000)]

Spin stiffness D from exchange coupling constants

By relying on the Heisenberg Hamiltonian

$$H = -\sum_{ij} J_{ij}\, \hat{\mathbf{e}}_i \cdot \hat{\mathbf{e}}_j \quad ,$$



one arrives (for isotropic systems) to

$$D = \sum_j \frac{2\mu_B}{3M} J_{0j}\, R_{0j}^2 \quad .$$

R_{0j} is the interatomic distance.

Convergence issues solved by introducing the damping parameter η:

$$D = \lim_{\eta\to 0} D(\eta) = \lim_{\eta\to 0} \sum_j \frac{2\mu_B}{3M} J_{0j}\, R_{0j}^2 \exp\left(-\eta \frac{R_{0j}}{a_0}\right) \quad .$$

a_0 is lattice parameter.

[Pajda *et al.* PRB **64**, 174402 (2001)]

Expressions are simple. So why should we care?

- Differences between theory and experiment (even for nominally simple systems).

- Differences between various calculations (even for nominally simple systems).

- It is hard to assess the impact of physical approximations in the theory if the technical evaluation of the expression itself is unreliable.

Spread of results: Spin wave stiffness D for Fe

Experiment:

	D (meV Å^2)
Hatherly (1964)	325
Phillis (1966)	350
Stringfellow (1968)	314
Riede (1973)	311
Pauthenet (1982)	270
Loong (1984)	307

Theory:

	D (meV Å^2)
Mryasov (1996)	214
Rosengaard (1997)	247
Brown (1999)	135
Schilfgaarde (1999)	280
Kübler (2000)	355
Pajda (2001)	250
Moran (2003)	200
Shallcross (2005)	313/322
Pan (2017)	320/466

Our results (KKR-ASA):

by fitting spirals at $\mathbf{q} \to 0$	$D = 280 \pm 5$ meV Å^2
from $J_{ij} R_{0j}^2$'s	$D = 302 \pm 5$ meV Å^2

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Spread of results: Spin wave stiffness D for Ni

Experiment:

	D (meV Å^2)
Hatherly (1964)	400
Stringfellow (1968)	470
Mook (1973)	555
Hennion (1975)	525
Maeda (1976)	390
Riede (1977)	398
Lynn (1981)	593
Pauthenet (1982)	413
Nakai (1983)	530
Mitchell (1985)	398

Theory:

	D (meV Å^2)
Mryasov (1996)	527
Rosengaard (1997)	739
Brown (1999)	480
Schilfgaarde (1999)	740
Kübler (2000)	790
Pajda (2001)	756
Shallcross (2005)	541
Pan (2017)	707

Our results (KKR-ASA):

by fitting spirals at $\mathbf{q} \to 0$	$D = 705 \pm 5$ meV Å^2
from $J_{ij} R_{0j}^2$'s	$D = 675 \pm 10$ meV Å^2

Spread of results: Permalloy (Py) $Fe_{0.2}Ni_{0.8}$

Experiment:

	D (meV Å^2)
Hatherly (1964)	400
Hennion (1975)	335
Nakai (1983)	390

Theory:

	D (meV Å^2)
Yu (2008)	515
Pan (2017)	655/620

Our results (KKR-ASA):		
	by fitting spirals at $\mathbf{q} \to 0$	$D = 503 \pm 5$ meV Å^2
	from $J_{ij} R_{0j}^2$'s	$D = 527 \pm 5$ meV Å^2

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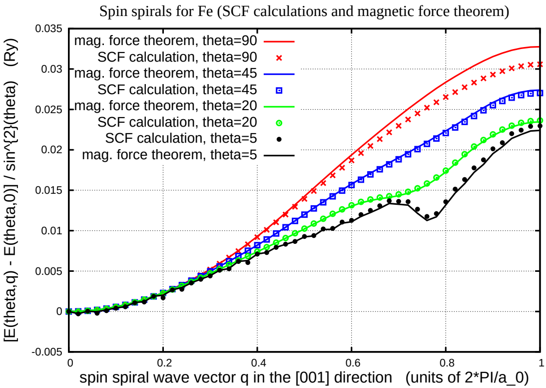
Fitting spin spiral dispersion for $\mathbf{q} \to 0$

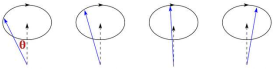
Self-consistent calculation vers. magnetic force theorem



Fe

$$\epsilon(\mathbf{q}) = \lim_{\theta \to 0} \frac{4\mu_B}{M} \frac{E(\mathbf{q},\theta) - E(0,\theta)}{\sin^2\theta}$$

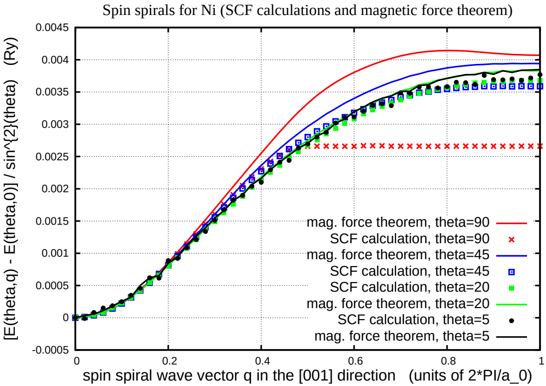
$$\epsilon(\mathbf{q}) = D\,|\mathbf{q}|^2 + \beta\,|\mathbf{q}|^4$$



Doing the limit $\theta \to 0$ is not a problem, the ratio $\Delta E(\mathbf{q},\theta)/\sin^2\theta$ depends on θ only midly.

Fitting spin spiral dispersion for $\mathbf{q} \to 0$

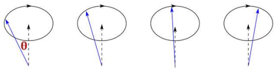
Self-consistent calculation vers. magnetic force theorem



Ni

$$\epsilon(\mathbf{q}) \;=\; \lim_{\theta \to 0} \frac{4\mu_B}{M} \, \frac{E(\mathbf{q},\theta) - E(0,\theta)}{\sin^2\theta}$$

$$\epsilon(\mathbf{q}) \;=\; D\,|\mathbf{q}|^2 \;+\; \beta\,|\mathbf{q}|^4$$



Doing the limit $\theta \to 0$ is not a problem, the ratio $\Delta E(\mathbf{q},\theta)/\sin^2\theta$ depends on θ only midly.

Stiffness D by fitting $\epsilon(\mathbf{q}) \approx D\mathbf{q}^2$

Robust with respect to

- the size of the interval within which the fit is made,
- use of magnetic force theorem,
- spiral cone angle θ,
- other technical parameters (such as density of the **k**-mesh in the Brillouin zone.)

So why should we care about convergence issues of the sum

$$\lim_{\eta\to 0} \sum_j \frac{2\mu_B}{3M} J_{0j} R_{0j}^2 \exp\left(-\eta \frac{R_{0j}}{a_0}\right) ?$$

Because it can be applied also for alloys with more atomic types which may be non-magnetic or may carry induced moments.

Such atoms should be excluded, which is difficult to be done with spin spirals.

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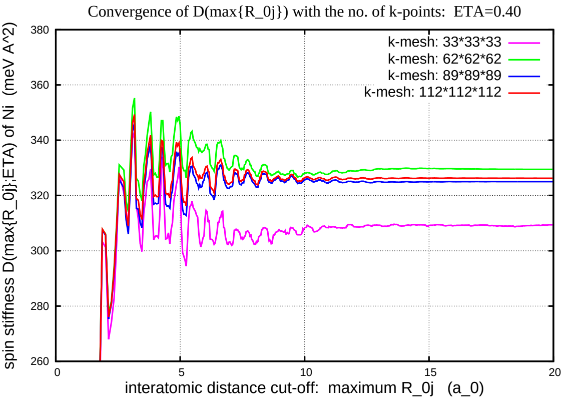
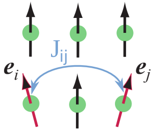
$\sum_j J_{0j} R_{0j}^2 \exp(-\eta R_{0j}/a_0)$ issues: R_{0j}, **k**-mesh

Dependence of D on the maximum distance R_{0j}



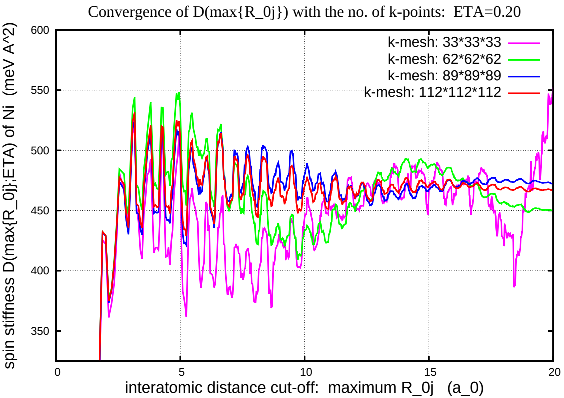
For fcc Ni,
the cluster
within $R_{0j} \approx 20a_0$
contains
about 130000 atoms.

$\eta = 0.4$



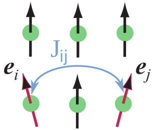
$\sum_j J_{0j} R_{0j}^2 \exp(-\eta R_{0j}/a_0)$ issues: R_{0j}, **k**-mesh

Dependence of D on the maximum distance R_{0j}



For fcc Ni,
the cluster
within $R_{0j} \approx 20a_0$
contains
about 130000 atoms.

$\eta = 0.2$



For small damping η, reliable values of D obtained only if $\sum_j$ extends to long distances, which requires very fine **k**-mesh.

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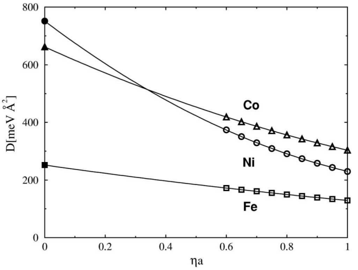
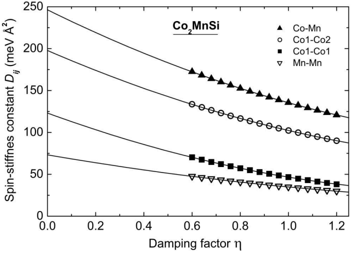
Extrapolation of $D(\eta)$ to zero damping

Sum $\sum_j J_{0j} R_{0j}^2 \exp(-\eta R_{0j}/a_0)$ has to be extrapolated to η=0.



Pajda *et al.* PRB **64**, 174402 (2001)



Thoene *et al.* J. Phys.D: **42**, 084013 (2009)

Commonly, extrapolation by means of a quadratic fit has been employed.

However, the extrapolation is not "unique".

Extrapolation of $D(\eta)$ to zero damping

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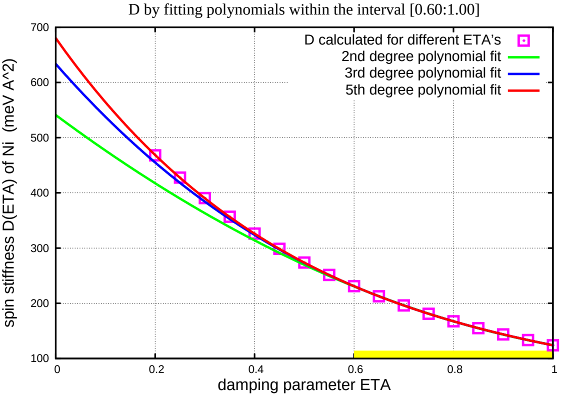
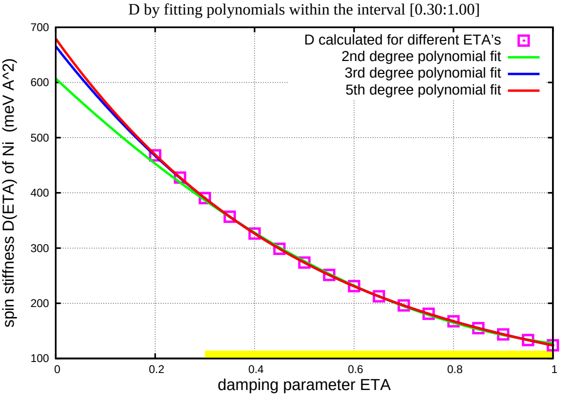


Extrapolation done by fitting a polynomial in the interval $\eta \in [\ 0.6\ : 1\ ]$.

Extrapolation by means of a quadratic fit undershoots.

Extrapolation of $D(\eta)$ to zero damping

Sum $\sum_j J_{0j} R_{0j}^2 \exp(-\eta R_{0j}/a_0)$ has to be extrapolated to $\eta{=}0$.



Extrapolation done by fitting a polynomial in the interval $\eta \in [\ 0.3\ : 1\ ]$.

Extrapolation by means of a quadratic fit undershoots.

Sensitivity to technical parameters of the extrapolation

Stiffness D for fcc Ni obtained from $\sum_j J_{0j} R_{0j}^2 \exp(-\eta R_{0j}/a_0)$ for different ways of doing the extrapolation to $\eta = 0$:

fit interval η	D (meV Å^2)		
	2nd order polynomial	3rd order polynomial	5th order polynomial
0.2–1.0	633	684	706
0.3–1.0	608	673	700
0.4–1.0	584	660	704
0.5–1.0	560	646	709
0.6–1.0	537	631	705
0.7–1.0	516	616	662

Stiffness D by extrapolating $\sum_j J_{0j} R_{0j}^2 \exp(-\eta R_{0j}/a_0)$

Extrapolation of the sum $\sum_j J_{0j} R_{0j}^2 \exp(-\eta R_{0j}/a_0)$ to $\eta = 0$ is fragile.

It is necessary to go to small η's, which requires

- large cut-off of the interatomic distance R_{0j}, and consequently
- very dense **k**-mesh.

Using brute force seems to be unavoidable.

Including the relativistic effects

Exchange interaction is anisotropic $\Rightarrow$ modified Heisenberg Hamiltonian:

$$H = -\sum_{ij} \hat{\mathbf{e}}_i \, \underline{J}_{ij} \, \hat{\mathbf{e}}_j \quad ,$$

The exchange tensor $J_{ij}^{\alpha\alpha}$ is anisotropic.

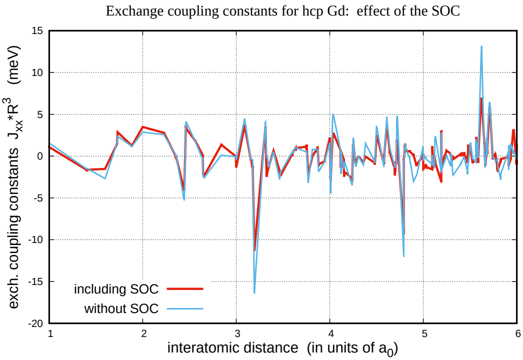
Assuming the reference direction of magnetization $\hat{\mathbf{m}} || \hat{\mathbf{z}}$:

$$D_{\nu\nu} = \frac{1}{M} \sum_{j} [J_{0j}^{xx} + J_{0j}^{yy}] \, R_{0j,\nu}^2$$

For cubic systems:
$J_{0j}^{xx} = J_{0j}^{yy} = J_{0j}^{zz}$ as well as $D_{xx} = D_{yy} = D_{zz}$, and therefore, the relativistic results shoud be similar to those obtained using the non-relativistic expression.

Effect of SOC on exchange coupling constants

Case study: Effect of spin-orbit coupling (SOC) on J_{0j}^{xx} of hcp Gd.



Values of J_{0j}^{xx} with SOC and without SOC are close to each other, the same applies to spin stiffness.

Conclusion

- Evaluating spin wave stiffness D by fitting the spin spiral dispersion relation and by extrapolating the sum $\sum_j J_{0j} R_{0j}^2 \exp(-\eta R_{0j}/a_0)$ to zero damping indeed leads to similar results *(provided that all convergence issues have been harnessed).*

- Do not use the $\sum_j J_{0j} R_{0j}^2 \exp(-\eta R_{0j}/a_0)$ sum for evaluating D unless really necessary.
  - If pressed to use it, extend the R_{0j} radii as far as you can. Deal with "ridiculosly large" clusters of $\sim$100000 atoms.

- Spin-orbit coupling modifies the values of D only slightly.

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Robustness of the fitting procedure

Stiffness D for Fe and Ni by fitting magnon dispersion relation around $\mathbf{q} \to 0$ in different $\mathbf{q}$-intervals:

$\mathbf{q}$-range ($2\pi/a_0$)	D (Fe) (meV Å^2)	standard error	D (Ni) (meV Å^2)	standard error
0–0.10	258	13 %	706	4 %
0–0.15	271	5 %	694	2 %
0–0.20	292	2 %	682	1 %
0–0.30	302	0.7 %	673	0.6 %

Self-consistent calculations for spiral cone angle θ=20°.

Spin stiffness D obtained by fitting $\epsilon(\mathbf{q}) = D\,|\mathbf{q}|^2 + \beta\,|\mathbf{q}|^4$ is robust against the range in which it is accomplished.

Magnetic force theorem, spiral cone angle θ

Stiffness D of Fe (in units meV Å^2).

θ (°)	via SCF	via mag. force th.
5	291	309
10	305	308
15	303	306
20	302	304
45	292	296
90	282	286

Stiffness D of Ni (in units meV Å^2).

θ (°)	via SCF	via mag. force th.
5	675	683
10	688	679
15	675	679
20	673	682
45	675	683
90	696	702

Fitting by $D\,|\mathbf{q}|^2 + \beta\,|\mathbf{q}|^4$ done in the range $q \in [0 : 0.3]$.

Spin stiffness D is robust with respect to whether it is evaluated from self-consistent calculations and by relying on the magnetic force theorem and with respect to the choice of the spiral cone angles θ.