Higher Order exchange interactions: 2D materials and metamagnets

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VAMPIRE Workshop 2023

Magnetic Hamiltonian

$$\mathbf{h}_i = -\frac{1}{\mu_i \mu_0}\frac{\partial \mathcal{H}}{\partial \mathbf{S}_i} + \xi_i$$

Effective field + thermal bath

$$\mathscr{H} = -\frac{1}{2}\sum_{i,j}\mathbf{S}_i^{\alpha}\mathscr{J}_{ij}^{\alpha\beta}\mathbf{S}_j^{\beta} - \frac{1}{2}\sum_{i,j}K_{ij}(\mathbf{S}_i\cdot\mathbf{S}_j)^2 - \sum_i D_i(\mathbf{S}_i\cdot\mathbf{e})^2 - \sum_i \mu_i \mathbf{S}_i\cdot\mathbf{B}_{\mathbf{dp}}$$

Billinear exchange

Biquadratic exchange

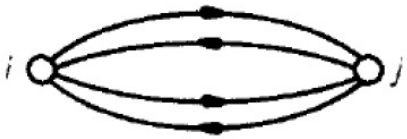
Uniaxial anisotropy

Dipolar interaction

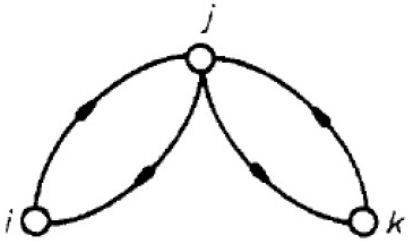
$$\mathscr{H} = -\frac{1}{2}\sum_{i,j}J_{ij}(\mathbf{S}_i\cdot\mathbf{S}_j) - \frac{1}{3}\sum_{i,j,k,l}D_{ijkl}(\mathbf{S}_i\cdot\mathbf{S}_j)(\mathbf{S}_k\cdot\mathbf{S}_l) - k_u\sum_i(\mathbf{S}_i\cdot\mathbf{e})^2$$

Four-spin exchange

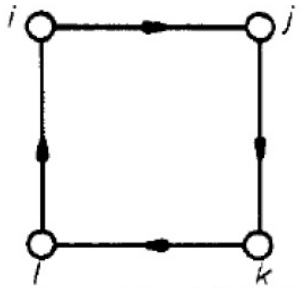
Higher order exchange



$K_{ij}\,(S_i \cdot S_j)^2$



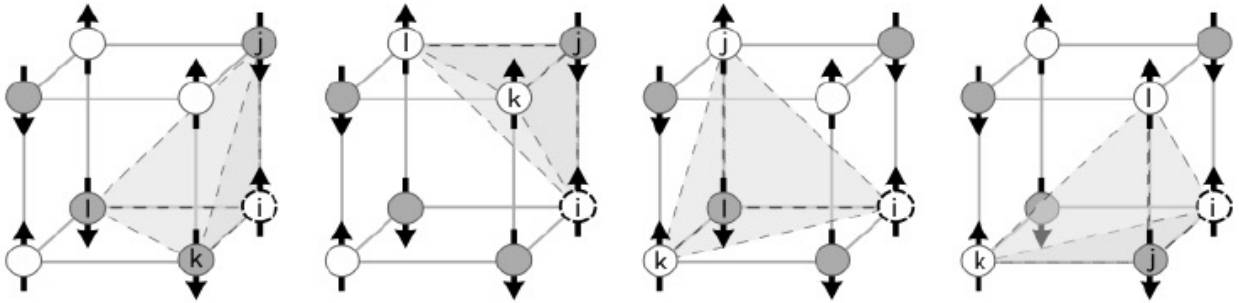
$L_{ijk}\,(S_i \cdot S_j)(S_j \cdot S_k)$



$K_{ijkl}\,(S_i \cdot S_j)(S_k \cdot S_l)$

Adler, Oitma, 1978

! They scale with the magnetic moment to the 4th-order !



Each site **i** there will be included in **32** nearest neighbours quartets.

J. Barker, R. Chantrell, 2015

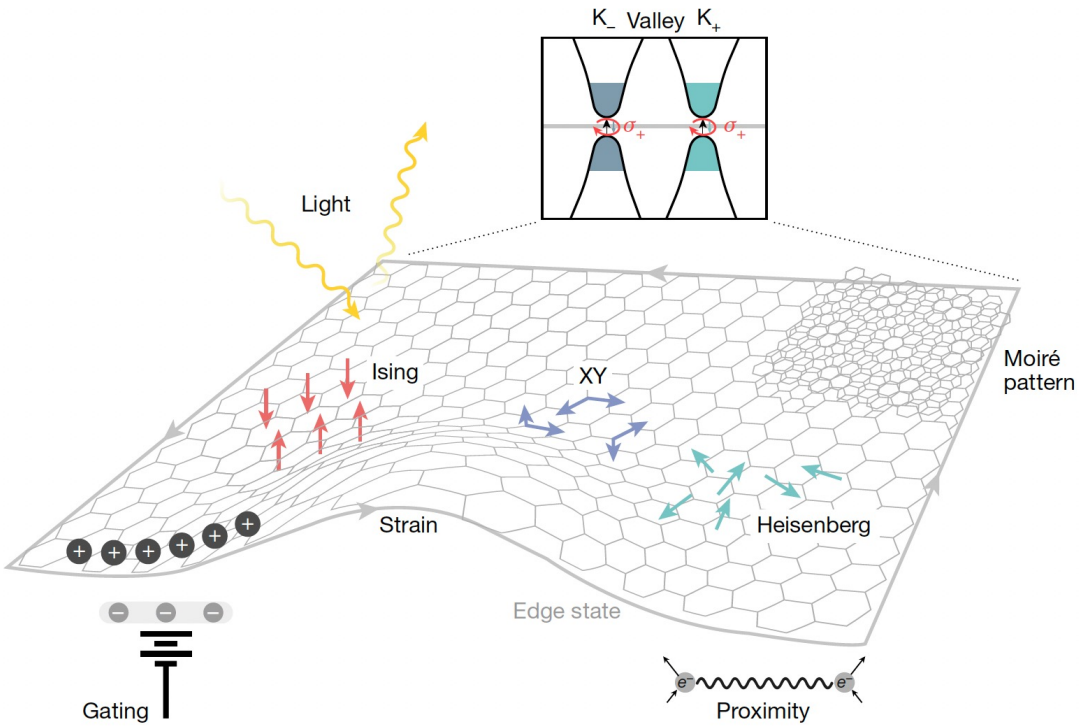
2D magnetic materials (2017)

LETTER

doi:10.1038/nature22060

Discovery of intrinsic ferromagnetism in two-dimensional van der Waals crystals

Cheng Gong[1]*, Lin Li[2]*, Zhenglu Li[3,4]*, Huiwen Ji[5], Alex Stern[2], Yang Xia[1], Ting Cao[3,4], Wei Bao[1], Chenzhe Wang[1], Yuan Wang[1,4], Z. Q. Qiu[3], R. J. Cava[5], Steven G. Louie[3,4], Jing Xia[2] & Xiang Zhang[1,4]



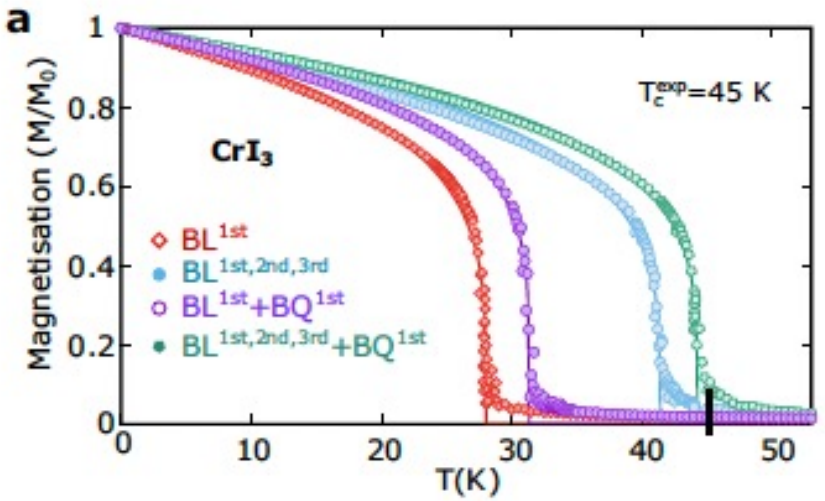
Burch, K. S., Mandrus, D., & Park, J. G. (2018)
Nature, *563*(7729), 47–52.

LETTER

doi:10.1038/nature22391

Layer-dependent ferromagnetism in a van der Waals crystal down to the monolayer limit

Bevin Huang[1]*, Genevieve Clark[2]*, Efrén Navarro-Moratalla[3]*, Dahlia R. Klein[3], Ran Cheng[4], Kyle L. Seyler[1], Ding Zhong[1], Emma Schmidgall[1], Michael A. McGuire[5], David H. Cobden[1], Wang Yao[6], Di Xiao[4], Pablo Jarillo-Herrero[3] & Xiaodong Xu[1,2]



Kartsev A. et al.
npj Computational Materials (2020) 6:150

$CrCl_3$ unit cell file

```
# Unit Cell Size
 7.079000  12.26120 25.000
# Unit Cell Vectors
1.0000000000000000   0.0000000000000000    0.0000000000000000
0.0000000000000000   1.0000000000000000    0.0000000000000000
0.0000000000000000   0.0000000000000000    1.0000000000000000
# Atoms num, id cx cy cz mat lc hc
4 1
0   0.0000000000000000  0.0000000000000000  0.0000000000000000 0 0 0
1   0.5000000000000000  0.5000000000000000  0.0000000000000000 0 0 0
2   0.0000000000000000  0.3333333333333333  0.0000000000000000 0 0 0
3   0.5000000000000000  0.8333333333333333  0.0000000000000000 0 0 0
# Interactions n exctype, id i j dx dy dz Jij
48 normalised-tensorial
0  0 3  -1  -1  0  4.614268507200001 0 0 0 4.614268507200001 0 0 0 4.686366452625
1  0 3   0  -1  0  4.614268507200001 0 0 0 4.614268507200001 0 0 0 4.686366452625
2  0 2   0   0  0  4.614268507200001 0 0 0 4.614268507200001 0 0 0 4.686366452625
3  1 2   0   0  0  4.614268507200001 0 0 0 4.614268507200001 0 0 0 4.686366452625
4  1 2   1   0  0  4.614268507200001 0 0 0 4.614268507200001 0 0 0 4.686366452625
5  1 3   0   0  0  4.614268507200001 0 0 0 4.614268507200001 0 0 0 4.686366452625
```

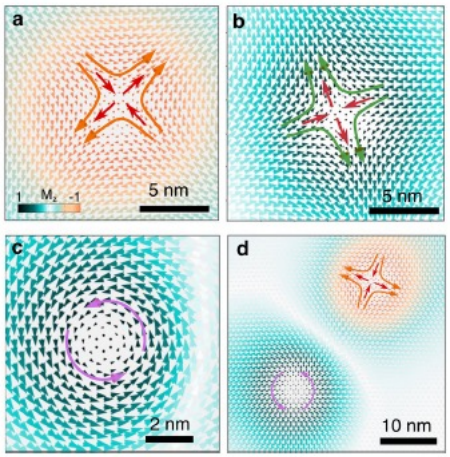
.....

```
12 isotropic
0  0 3  -1  -1  0  1.7844241492687502e-22
1  0 3   0  -1  0  1.7844241492687502e-22
2  0 2   0   0  0  1.7844241492687502e-22
3  1 2   0   0  0  1.7844241492687502e-22
4  1 2   1   0  0  1.7844241492687502e-22
5  1 3   0   0  0  1.7844241492687502e-22
6  2 1   0   0  0  1.7844241492687502e-22
7  2 1  -1   0  0  1.7844241492687502e-22
8  2 0   0   0  0  1.7844241492687502e-22
9  3 1   0   0  0  1.7844241492687502e-22
10 3 0   1   1  0  1.7844241492687502e-22
11 3 0   0   1  0  1.7844241492687502e-22
```

Biquadratic exchange

2D magnetic $CrCl_3$

Strungaru, M., Augustin, M. & Santos, E.J.G. Ultrafast laser-driven topological spin textures on a 2D magnet. *npj Comput Mater* **8**, 169 (2022).



Material & input file

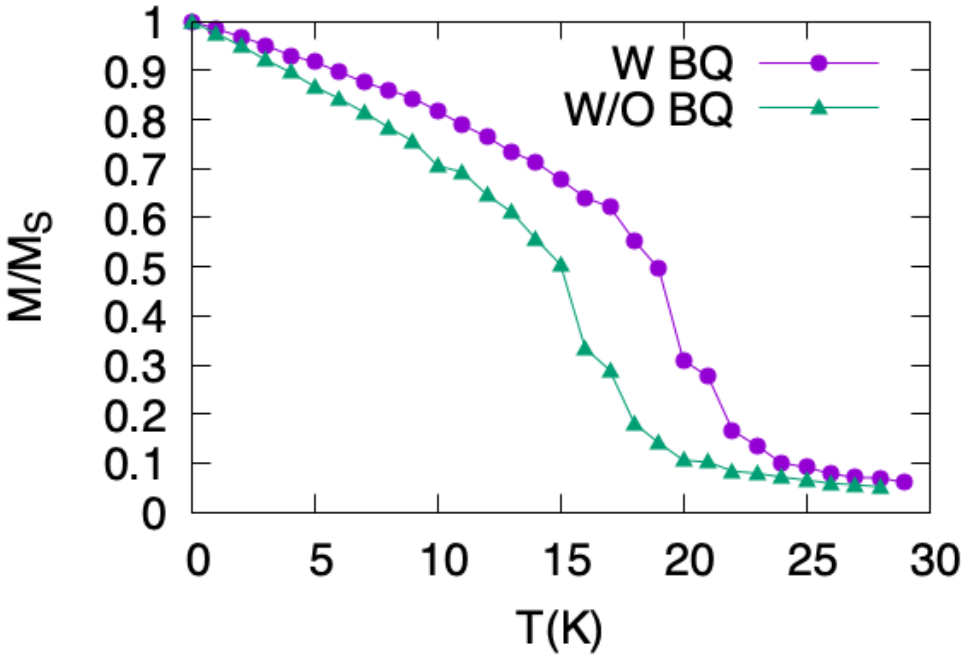
Material file

```
#-------------------------------------------------
# Number of Materials
#-------------------------------------------------
material:num-materials=1
#-------------------------------------------------
# Material 1
#-------------------------------------------------
material[1]:material-name=Cr
material[1]:damping-constant=0.1
material[1]:atomic-spin-moment=3.35500 !muB
material[1]:material-element=Cr
material[1]:exchange-matrix[1]=1.0e-22
material[1]:uniaxial-anisotropy-direction=0,0,1
material[1]:uniaxial-anisotropy-constant=3.6048972712500005e-24
material[1]:initial-spin-direction=1,0,0
```

Enabling dipole in the input file

```
dipole:solver = tensor
dipole:field-update-rate = 100
cells:macro-cell-size  =2 !nm
```

Quick M(T)




```
#---------------------------------------
dimensions:system-size-x = 20.0 !nm
dimensions:system-size-y = 20.0 !nm
dimensions:system-size-z = 0.5 !nm
create:periodic-boundaries-x
create:periodic-boundaries-y


sim:equilibration-time-steps = 10000
sim:loop-time-steps = 10000
sim:time-steps-increment = 1
```

FeRh metamagnet

O. N. Mryasov, Phase Transitions 78, 197 (2005): showed that the non-linear behaviour of Rh leads to some higher order exchange, suggesting that the first-order phase transition could be driven by the competition between bilinear and higher order exchange terms.

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Article | Open Access | Published: 30 May 2022

Ultrafast kinetics of the antiferromagnetic-ferromagnetic phase transition in FeRh

G. Li ✉, R. Medapalli, J. H. Mentink, R. V. Mikhaylovskiy, T. G. H. Blank, S. K. K. Patel, A. K. Zvezdin, Th. Rasing, E. E. Fullerton & A. V. Kimel

Nature Communications **13**, Article number: 2998 (2022) | Cite this article

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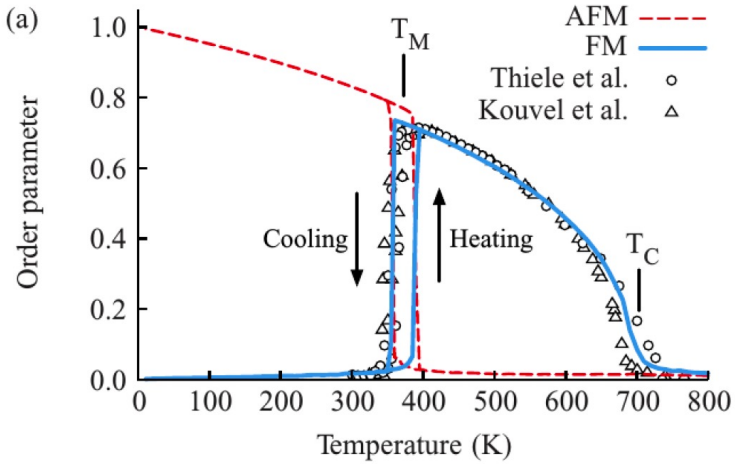
Article | Open Access | Published: 29 June 2023

Spin current driven by ultrafast magnetization of FeRh

Kyuhwe Kang, Hiroki Omura, Daniel Yesudas, OukJae Lee, Kyung-Jin Lee, Hyun-Woo Lee, Tomoyasu Taniyama & Gyung-Min Choi ✉

Nature Communications **14**, Article number: 3619 (2023) | Cite this article

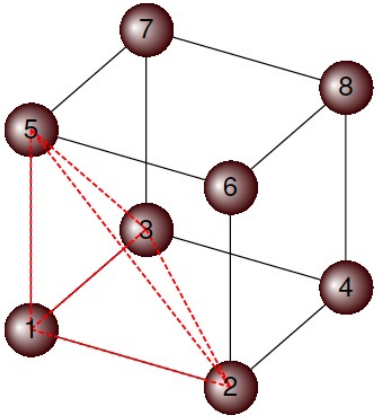
1680 Accesses | **1** Altmetric | Metrics

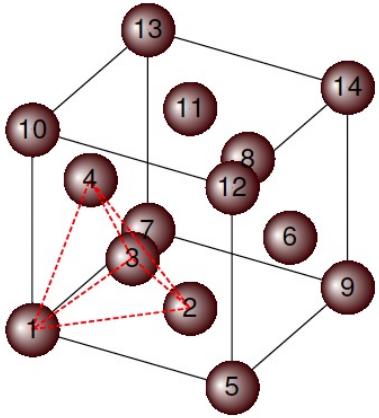


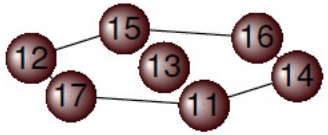
J.Barker and R.Chantrell, **Higher-order exchange interactions leading to metamagnetism in FeRh**, PHYSICAL REVIEW B 92, 094402 (2015)

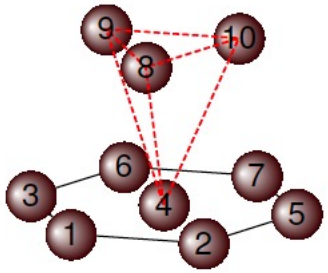
Massey, J. R., Matsumoto, K., Strungaru, M., et al. *Physical Review Materials*, *4*(2), 024403.

Four-spin quartets



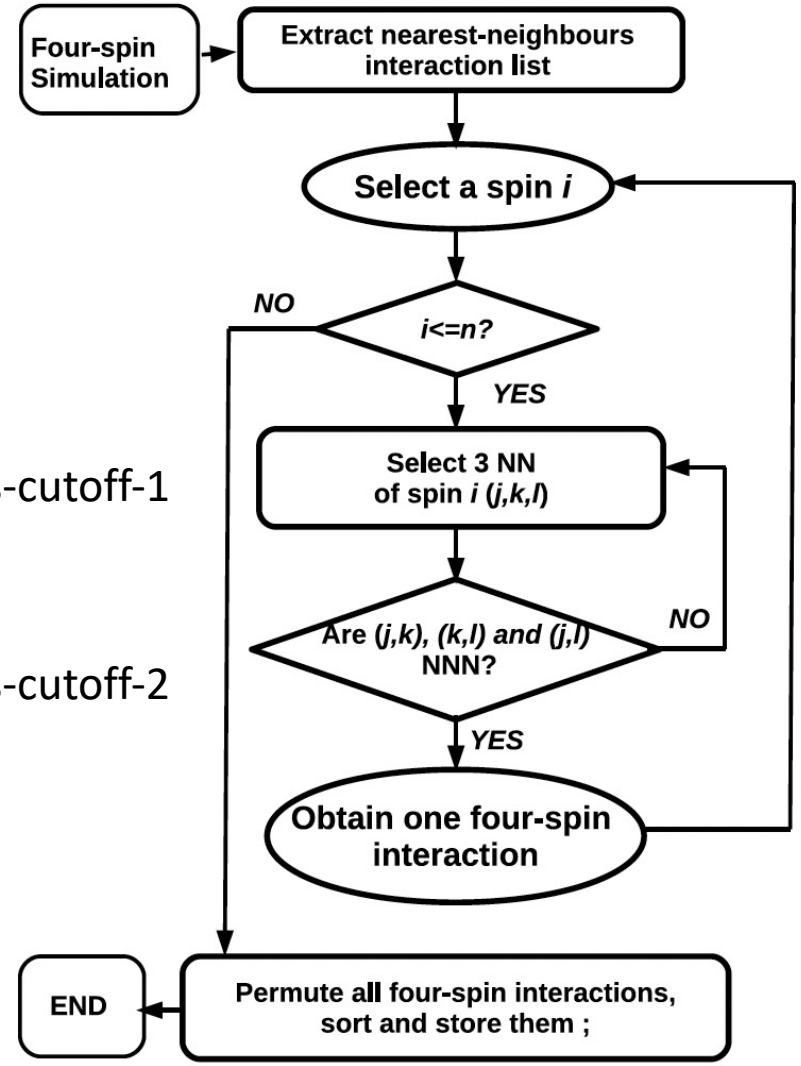






fs-cutoff-1

fs-cutoff-2



Material & input file

```
material[1]:material-name=Fe_rh
material[1]:damping-constant=1.0
material[1]:atomic-spin-moment=3.15!muB
material[1]:uniaxial-anisotropy-constant=1.404e-23
material[1]:initial-spin-direction=0,0,1
material[1]:four-spin-constant[2]=-0.23e-21

material[2]:material-name=Fe_rh
material[2]:damping-constant=1.0
material[2]:atomic-spin-moment=3.15!muB
material[2]:uniaxial-anisotropy-constant=1.404e-23
material[2]:initial-spin-direction=0,0,-1
material[2]:four-spin-constant[1]=-0.23e-21


material[1]:unit-cell-category = 1
material[2]:unit-cell-category = 2
```

```
#
material:file =ferh.mat
material:unit-cell-file = file.ucf
#---------------------------------------
# Simulation attributes:
#---------------------------------------

sim:time-steps-increment = 100
sim:total-time-steps = 100000
sim:time-step = 1 !fs
sim:maximum-temperature = 800
sim:minimum-temperature = 0
sim:cooling-time = 0.1 !ns
sim:cooling-function = linear
sim:equilibration-time-steps=0
sim:equilibration-temperature=800

exchange:four-spin-cutoff-1=1.0
exchange:four-spin-cutoff-2=1.414
```

First order phase transition

Field cooling simulation