

# ***SpinWave* and *CefWave* User Manual**

Version 2p2 (2016)

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# 1. Basic Concepts

## 1.1 Introduction

Owing to recent developments in numerical and computer science, numerical simulations have become an important trend in physics and a useful tool to design and explain experiments. In this context, the LLB has developed a simulation program able to calculate the spin excitations spectrum and the corresponding correlation functions for a large variety of situation.

The *SpinWave* software addresses spin Hamiltonian of the form

$$\mathcal{H} = \frac{1}{2} \sum_{m,i,n,j} \vec{S}_{m,i} J_{m,i,n,j} \vec{S}_{n,j} + \sum_{m,i} B_{i,20} \mathcal{O}_{i,20} \quad (1.1)$$

where

- $\vec{S}_{m,i}$  is the spin at site  $(m,i)$  ; throughout this text,  $m$  is associated with the unit cell and  $i$  with the crystallographic site within a unit cell
- $J_{m,i,n,j}$  is the exchange tensor between spins at  $(m,i)$  and  $(n,j)$ .
- $B_{i,20} = \alpha_i/2 (3S_Z^2 - S(S+1))$ ,  $Z$  being the local quantification axis, useful to describe a single ion anisotropy.  $\alpha_i$  is a coefficient that depends on the ion (see below).

$\mathcal{H}$  is solved in the harmonic approximation using the Holstein-Primakov representation of the spin operators. It calculates various neutron scattering cross section :

$$\frac{\partial^2 \sigma_M}{\partial \omega \partial \Omega} = \sum_{m,i,n,j} \int dt e^{iQ(R_m+u_i-R_n-u_j)} \sum_{a,b} \langle S_{m,i}^a M^{ab}(Q) S_{n,j}^b(t) \rangle \quad (1.2)$$

where  $M(Q)$  is a  $3 \times 3$  matrix :

- $M(Q) = \left( \delta_{a,b} - \frac{Q_a Q_b}{Q^2} \right)$  is the usual tensor that picks out spin components perpendicular to the scattering wavevector  $\vec{Q}$
- $M(Q) = \vec{y} \vec{y}$  extracts the spin correlations along the  $y$  axis, with the standard definition,  $y$  being the axis perpendicular to  $\vec{Q}$  within the scattering plane

- $M(Q) = \vec{z}\vec{z}$  extracts the spin correlations along the  $z$  axis perpendicular to  $\vec{Q}$  and perpendicular to the scattering plane.

This is associated with the **EZX**, **EZY**, **EZZ** keywords that specify the coordinates of the vetical axis.

The code also provides other correlations functions (which are not observed by neutron scattering), allowing one to have a better understanding of the polarization of the correlations:  $M(Q) = \vec{a}\vec{a}$  extracts the spin correlations along the  $a$  axis,  $M(Q) = \vec{b}\vec{b}$  along the  $b$  axis, and  $M(Q) = \vec{c}\vec{c}$  along the  $c$  axis.

The *CefWave* software addresses Hamiltonians of the form

$$\mathcal{H} = \frac{1}{2} \sum_{m,i,n,j} \vec{\mathcal{J}}_{m,i} J_{m,i,n,j} \vec{\mathcal{J}}_{n,j} + \sum_{i,n,m} B_{i,n,m} \mathcal{O}_{i,n,m} \quad (1.3)$$

where

- $\vec{\mathcal{J}}_{m,i}$  is the magnetic moment at site  $(m,i)$ ;
- $J_{m,i,n,j}$  is the exchange tensor,
- $B_{i,n,m}$  and  $\mathcal{O}_{i,n,m}$  are the Weybourne coefficients and operators respectively,  $Z$  being the local quantification axis. This term of the Hamiltonian describes the crystal field of the magnetic moment at site  $i$ .

More precisely, we the following convention us used:

$$\sum_{n,m} B_{n,m} \mathcal{O}_{n,m} = \alpha/2B_{20}\mathcal{O}_{20} + \quad (1.4)$$

$$\beta/8B_{40}\mathcal{O}_{40} + \beta(-\sqrt{35}/2)B_{43}\mathcal{O}_{43} + \quad (1.5)$$

$$\gamma/16B_{60}\mathcal{O}_{60} + \gamma(-\sqrt{105}/8)B_{63}\mathcal{O}_{63} + \gamma(\sqrt{231}/16)B_{66}\mathcal{O}_{66} \quad (1.6)$$

Here, the spin excitations spectrum consists in dispersing collective excitons. It is calculated in the RPA approximation.

## 1.2 Installation

The softwares are freely available on the LLB web page at : [http://www-llb.cea.fr/fr-en/soft-neut\\_p.php](http://www-llb.cea.fr/fr-en/soft-neut_p.php)

The .zip package contains the following files : spinwave2p2.exe, cefwave3.exe, SW-Manual.pdf, and a series of example files (.txt). The .exe file should be copied in the desired working directory.

## 1.3 Running

The program should be executed from a command shell, using the following syntax :

$$spinwave2p2 < input\ file.txt \quad (1.7)$$

where 'inputfile.txt' is the command file which is described in 2. To save the screen output in a file :

$$spinwave2p2 < input\ file.txt > listing.txt \quad (1.8)$$

An output file is generated with the name given in the command file .The listing.txt file contains detailed information about the calculation performed, and should be checked regularly.



## 2. Structure of a *SpinWave* input file

All lines starting with `#` are considered as comments. The input file contains a list of keywords, which can be modified according to the calculation to be performed. These keywords can appear in any order that suits the user. Some of them should however appear on the same line, as explained in the following.

### 2.1 Defining the magnetic cell

#### 2.1.1 Lattice parameters

**AX=, AY=, AZ=, ALFA=, BETA=, GAMMA=**

are respectively, the magnetic cell parameters  $a$ ,  $b$ ,  $c$  and cell angles  $\alpha$ ,  $\beta$ ,  $\gamma$ . Note that these parameters can be different from the crystalline ones.

**EZX=, EZY=, EEZ=** are the coordinates of the vertical axis.

#### 2.1.2 Defining moments and anisotropy

For each atom carrying a magnetic moment, one should define :

**I=, NOM=, X=, Y=, Z=, PHI=, THETA=**

that is, respectively, label, name, reduced coordinates  $x$ ,  $y$ ,  $z$ , spherical coordinates of the magnetic moment  $\phi$  (within the  $(a, b)$  plane) and  $\theta$  (perpendicular to the  $(a, b)$  plane).

The magnetic moment can also be defined with Cartesian coordinates along  $x$ ,  $y$ ,  $z$  :

**I=, NOM=, X=, Y=, Z=, SX=, SY=, SZ=**

The label keyword **I** allows one to distinguish identical spins located at non equivalent crystallographic sites.

*Example :*

`I=1, NOM=S3D2, X= 0.00, Y= 0.00, Z=0.00, PHI= 0, THETA= 90`

describes an atom 1, located on site (0,0,0), with a spin  $S=3/2$  parallel to the  $a$  axis of the cell.

It is equivalent to :

```
I=1, NOM=S3D2, X= 0.00, Y= 0.00, Z=0.00, SX= 1.000, SY= 0.00, SZ= 0.00
```

To include magnetic anisotropy, four more keywords should be added on the same line : **CX = ,CY = ,CZ = ,B20 =**.

(**CX**, **CY**, **CZ**) give the coordinates of the (local) anisotropy axis, and **B20** defines the single ion anisotropy value defined according to  $B20 \propto (3S_Z^2 - S(S+1))$ .

*Example :*

```
I= 1, NOM=S3D2, X=0.0, Y=0.0, Z=0.0, SX=0.7, SY=0.7, SZ=0.0, CZ=1, B20=0.1
```

defines an atom with  $S=3/2$  whose magnetic anisotropy is the (*ab*) plane.

### 2.1.3 Form factor

The keyword **FFORM** will multiply the result with a magnetic form factor. The value of **FFORM** is tabulated according to the **NOM** of the magnetic atoms, but the form factor parameters can also be addressed directly with the **fA=, fa=, fB=, fb=, fC=, fc=, fD=** keywords.

*Example :*

```
FFORM
```

```
fA=-0.3094, fa=0.0274, fB=0.3680, fb=17.0355, fC=0.6559, fc=6.5236, fD=0.2856
```

### 2.1.4 Predefined magnetic moments

The keyword **NOM** refers to a list of predefined magnetic moments :

Ion	$J$	$g_J$	$\alpha$
ND	9/2	8/11	$-7/(9 \times 121)$
ER	15/2	6/5	$4/(9 \times 25 \times 7)$
YB	7/2	8/7	$2/63$
HO	8	5/4	$-1/(2 \times 9 \times 25)$
TB	6	3/2	$-1/99$
PR	4	4/5	$-4 \times 13/(9 \times 25 \times 11)$
DY	15/2	4/3	$-2/(9 \times 5 \times 7)$
CE	5/2	6/7	1
S1	1	2	1
S2	2	2	1
S3	3	2	1
S4	4	2	1
S5	5	2	1
S6	6	2	1
S7	7	2	1
S8	8	2	1
S9	9	2	1
S10	10	2	1
SD2	1/2	2	1
S3D2	3/2	2	1
S5D2	5/2	2	1
MN3	2	2	1
MN4	3/2	2	1
FE3	5/2	2	1

## 2.2 Defining couplings

### 2.2.1 Generalized Heisenberg coupling

Each coupling path should be defined on a single line, with the following keywords :

**I1=, I2=, J1=, D1=**

that is, respectively, atom 1 label, atom 2 label, coupling value and maximum coupling distance.

*Example :*

I1=1, I2=1, J1=-3.14, D1=3.5

couples atom 1 and atom 1 with a ferromagnetic exchange of -3.14 meV, if the distance between the two atoms is less than 3.5 Å, and greater than 0.

To introduce a second-neighbour coupling between two atoms, similar keywords are used, **J2=** and **D2=**.

*Example :*

I1=1, I2=1, J1=-3.14, D1=3.5, J2=0.8, D2=7.0

couples first neighbour atoms with a ferromagnetic exchange of -3.14 meV, and couples second neighbours (less than 7 Åpart) with an antiferromagnetic exchange of 0.8 meV.

The same procedure applies for **J3** and **J4** with distances **D3** and **D4**, if relevant.

It is also possible to address directly the coefficients of the exchange tensor with the following keywords:

$$J = \begin{pmatrix} J1XX & J1XY & J1XZ \\ J1YX & J1YY & J1YZ \\ J1ZX & J1ZY & J1ZZ \end{pmatrix} \quad (2.1)$$

If **I2 = ALL**, the coupling is defined for all possible ions within the unit cell.

### 2.2.2 Bond directed couplings

Using the **MODELE=ANISOBV** keyword, it is possible to alter the definition of the exchange tensor. For each couple  $(i, j)$ , a local frame  $(\vec{u}, \vec{v}, \vec{w})$  is defined with  $\vec{u} = R_{m,i} - R_{n,j}$ ,  $\vec{v} = R_{n,j} + R_{m,i}$  and  $\vec{w} = \vec{u} \times \vec{v}$ . We then have :

$$J_{i,j} = J1\vec{v}\vec{v} + J2\vec{w}\vec{w} + J3\vec{u}\vec{u} + J4\sqrt{2} \begin{pmatrix} 0 & w_z & -w_y \\ -w_z & 0 & w_x \\ w_y & -w_x & 0 \end{pmatrix} \quad (2.2)$$

This is usefull to describe strongly anisotropic couplings, for instance along the bond of the structure as in the Kitaev model.

### 2.2.3 Dzyaloshinskii-Moriya coupling

The Dzyaloshinskii Moryia interaction between spins  $i$  and  $j$  belonging to the magnetic cells  $m$  and  $n$  is of the form :

$$\mathcal{H} = g_{\text{DM}} \vec{D}_{i,m,j,n} \cdot (\vec{S}_{i,m} \times \vec{S}_{j,n}) \quad (2.3)$$

The user should provide  $g_{\text{DM}}$  (**GDM**), and for each couple  $(i, j)$  (**I1, I2**) within the cell, the vector  $R_n - R_m$  (**UX, UY, UZ**) and the corresponding  $\vec{D}_{i,m,j,n}$  (**(DMX, DMY, DMZ)**).

*Example GDM = 0.01*

I1= 1, I2= 2, UX= 0., UY= 0., UZ= 0, DMX=0, DMY=0, DMZ=1

I1= 2, I2= 3, UX= 0., UY= 0., UZ= 0, DMX=0, DMY=0, DMZ=1

I1= 3, I2= 1, UX= 0., UY= 0., UZ= 0, DMX=0, DMY=0, DMZ=1

I1= 4, I2= 5, UX= 0., UY= 0., UZ= 0, DMX=0, DMY=0, DMZ=1

*Warning :* (**DMX,DMY,DMZ**) is not normalized, and is defined in Cartesian coordinates.



### 2.2.4 KSEA coupling

The KSEA interaction between spins  $i$  and  $j$  belonging the magnetic cells  $m$  and  $n$  is of the form :

$$\mathcal{H} = g_{\text{KSEA}} \vec{S}_{i,m} \begin{pmatrix} 0 & D_{i,m,j,n}^x D_{i,m,j,n}^y & D_{i,m,j,n}^x D_{i,m,j,n}^z \\ D_{i,m,j,n}^x D_{i,m,j,n}^y & 0 & D_{i,m,j,n}^y D_{i,m,j,n}^z \\ D_{i,m,j,n}^x D_{i,m,j,n}^z & D_{i,m,j,n}^y D_{i,m,j,n}^z & 0 \end{pmatrix} \vec{S}_{j,n} \quad (2.4)$$

As above, the user should provide  $g_{\text{KSEA}}$  (**GKSEA**), and for each couple  $(i, j)$  (**I1**, **I2**) the vector  $R_n - R_m$  (**UX**, **UY**, **UZ**) and the corresponding  $\vec{D}_{i,m,j,n}$  (**DMX**, **DMY**, **DMZ**).

*Warning* : (**DMX**, **DMY**, **DMZ**) is not normalized and is defined in Cartesian coordinates.

## 2.3 Mean Field option

The Mean Field **MF** allows to determine the magnetic configuration at temperature  $T = \text{TEMP}$ , in the mean field approximation, given the exchange couplings and the definition of the unit cell. The Hamiltonian is a sum of contributions for each site  $i$  of the cell :

$$\mathcal{H}_{i,\text{MF}} = \vec{S}_i \left( \sum_j J_{i,j} \langle \vec{S}_j \rangle + \sum_i B_{20} \mathcal{O}_{i,20} \right) \quad (2.5)$$

It is diagonalized in the space spanned by the kets  $|n\rangle$ , with  $n = -S, \dots, S$  :

$$\langle \vec{S}_i \rangle = \text{Tr} \left( e^{-\mathcal{H}_{i,\text{MF}}/k_B T} \vec{S}_i \right) \quad (2.6)$$

The convergence is obtained by an iterative process controlled by the **NITER** keyword. Note that in that case, the **THETA**, **PHI**, **SX**, **SY**, **SZ** parameters are considered as initial values of the iterations. They are replaced, after **NITER** iterations by their actual values. The convergence can be checked in the output file in two different ways: by checking that the magnetic moments are constant, or by checking that the total energy has converged. Note that when the anisotropy is large, the convergence can be difficult; in that case, the initial guess is extremely important to "help" the code getting out of local minima.

## 2.4 Calculation controls

The diagonalization of the spin Hamiltonian is performed in the spin wave approximation. Diagonalization of the spin wave matrix can be tricky, and may require the use of regularization parameters **REG1**, **REG2**, which respectively, add a small value to the real and imaginary part of the matrix of interest. A third regularization parameter **REG3** is used in case of coupled magnon-phonon calculations.

### 2.4.1 Defining the energy grid of the spinwave calculation

**WMAX=**

This keyword defines the upper value of the energy range in which the calculation is performed.

**NW=**

defines the number of energy steps.

### 2.4.2 Standard "single crystal" Q-cut in reciprocal space

Defining the direction of the spinwave calculation in the reciprocal space is obtained with the following commands :

**Q0X=, Q0Y=, Q0Z=**

Starting point in the reciprocal space

**DQX=, DQY=, DQZ=**

Step size along *x*, *y* and/or *z*

**NP=**

Number of steps

*Example :*

Q0X=0.0, Q0Y=0.0, Q0Z=0.0

DQX=0.00, DQY=0.00, DQZ=0.005

NP=400

WMAX=25, NW=150

The calculation will be performed along the *c* axis, with 400 Q-steps between points (0 0 0) and (0 0 2), 150 E-steps between 0 and 25 meV.

A more elaborate Q-cut, taking into account an integration around a given Q-point, can be defined with the following commands:

**COUP1D**

**Q0X=, Q0Y=, Q0Z=**

Define the starting point in the reciprocal space

**DQ1X=, DQ1Y=, DQ1Z=**

**DQ2X=, DQ2Y=, DQ2Z=**

**DQ3X=, DQ3Y=, DQ3Z=**

Define the step size along **DQ1**, **DQ2** and **DQ3**

**NP1=, NP2=, NP3=** Define the number of steps in each direction.

An integration is performed along **DQ1**, **DQ2** and **DQ3** to obtain a single Q-point data.

### 2.4.3 Standard "single crystal" energy-maps

Defining a constant energy cut is possible with :

**COUPE**

**EN0=**

Energy of the cut

**Q0X=, Q0Y=, Q0Z=**

Starting point in the reciprocal space

**DQ1X=, DQ1Y=, DQ1Z=**

**DQ2X=, DQ2Y=, DQ2Z=**

Step size along **DQ1** and **DQ2**

**NP=**

Number of steps

**2.4.4 Powder average**

It is possible to calculate the powder average of the cross section with the **TOF** keyword. **NP** Q values between **QMIN** and **QMAX** are considered. For each Q-point, a shell of radius  $|\vec{Q}|$  is sampled based on a Fibonacci algorithm. The quality of the sampling is controlled by **NFIBO**. **NFIBO=14** is usually a good compromise between convergence and time.

**2.4.5 Resolution**

The keyword **SIG=** spreads over the energy grid the intensity  $I$  corresponding to a given spin wave mode  $E$  according to a constant "resolution" like factor:

$$I e^{-(\omega-E)^2/\text{SIG}^2} \quad (2.7)$$



## 3. Advanced options

### 3.1 Defining twins

It is possible to take into account the contribution of crystalline twins defined with the following keywords : **TWIN**, **ATW=**, **TWX=**, **TWY=**, **TWZ=**

The twin direction is obtained by rotation of **ATW** degrees around the vector (in real space coordinates) (**TWX**,**TWY**,**TWZ**). Several **TWIN** lines must be used to calculate the contributions of different twins.

*Example :*

**TWIN**, **ATW=60**, **TWX=1**, **TWY=0**, **TWZ=0**

The twinned direction is obtained by rotation of 60° around the vector (100).

### 3.2 Defining an incommensurate structure

Defining an incommensurate magnetic structure is possible with the **INCOM** keyword. The keywords **AX=**, **AY=**, **AZ=**, **ALFA=**, **BETA=**, **GAMMA=** now correspond to the crystalline cell parameters  $a$ ,  $b$ ,  $c$  and cell angles  $\alpha$ ,  $\beta$ ,  $\gamma$ .

**KROPX=**, **KPROPY=**, **KPROPZ=** are used to define the coordinates of the magnetic propagation vector. The rotation plane of the magnetic moment should also be defined, for each atom of the crystal cell, using **UPX=**, **UPY=**, **UPZ=** (which defines the vector perpendicular to the rotation plane).

*Example :*

**INCOM**

**KPROX=0**, **KPROPY=0**, **KPROPZ=0.329**

**I1=1**, **UPX=0**, **UPY=0**, **UPZ=1**

This defines an incommensurate structure with propagation vector  $\mathbf{k} = (0, 0, 0.329)$ , in which the spins are confined to the  $ab$  plane.



## 4. Troubleshooting

This chapter provides a list of common error messages

### 4.1 Negative Eigenvalues (VP<0!)

The magnetic structure that has been entered by the user or calculated via the **MF** option does not correspond to an energy minimum. This probably means that the magnetic exchange values were not chosen appropriately, and should be modified. Another possibility is that the magnetic exchange paths were not entered properly : in this case, the user should refer to the screen output listing to check that each atom has the appropriate number of neighbors. It can be that some couplings are missing or equal to zero, for instance if the distance between two coupled atoms is too short with respect to the crystal structure.

### 4.2 Too many iterations

This happens during a diagonalization process, either in the **MF** option or in the normal process of calculating the eigenvalues. Check the input file, and especially ensure that no coupling is accidentally set to zero.





## 5. Examples

All of the SpinWave files (.txt) corresponding to the examples described below can be found in the download package (<http>).

### 5.1 Ferromagnetic and antiferromagnetic chains

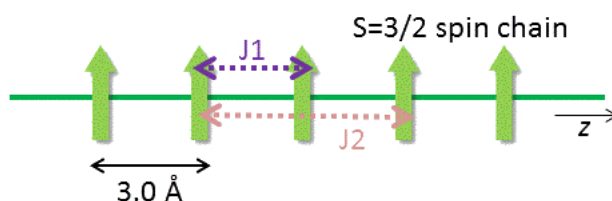


Figure 5.1: Model of a  $J_1$ - $J_2$  chain

Figure 5.1 shows the so-called  $J_1$ - $J_2$  chain, here in the case of  $S = 3/2$  spins. There is a first-neighbour coupling  $J_1$ , and a second-neighbour coupling  $J_2$  along  $z$ . The distance between magnetic atoms are  $3\text{\AA}$ . The arbitrary crystal cell is orthorhombic with  $a=b=15\text{\AA}$ ,  $c=3\text{\AA}$ ,  $\alpha=\beta=\gamma=90^\circ$ .

First we need to define the magnetic cell, which is the same as the crystal cell in this very case, the chain being ferromagnetic :

```
#Fictitious chain of Cr atoms
AX = 15.00
AY = 15.00
AZ = 3.00
ALFA= 90.0
BETA= 90.0
GAMA= 90.0
```

#

Here the cell parameters  $a$  and  $b$  are chosen deliberately very large, to emphasize the chain-like topology (parallel to  $c$ ) of the magnetic lattice.

There is only one magnetic atom per unit cell, labelled 1, with a spin parallel to  $a$  :

#Position of the Cr spin ( $S=3/2$ )

I= 1, NOM=S3D2, X= 0.00 ,Y= 0.00 ,Z=0.00, PHI= 0, THETA= 90

#

Anisotropy parameters should be added at the end of this line :

#Position of the Cr spin ( $S=3/2$ ), axial anisotropy (along  $a$ )

I= 1,NOM=S3D2, X= 0.00 ,Y= 0.00 ,Z=0.00, PHI= 0, THETA= 90, CX=1, B20=-0.5

#

To describe the magnetic interaction  $J_1$  coupling first-neighbours separated by  $3\text{\AA}$ :

#First-neighbour ferromagnetic coupling

I1= 1,I2= 1, J1=-3.14, D1= 3.5

#

To make sure that two atoms are coupled together along  $c$ , the parameter **D1** is larger than the actual interatomic distance ( $3.5\text{\AA}$ ).

In case there is a second-neighbour coupling  $J_2$ , since there is only one atom per unit cell, one has to write :

I1= 1,I2= 1,J1=-3.14,D1= 3.5, J2=0.7, D2= 7.0

#

In this example  $J_2$  is antiferromagnetic and will couple atoms separated by more than  $3.5\text{\AA}$  and less than  $7\text{\AA}$ . Note that writing this :

I1= 1,I2= 1,J1=-3.14,D1= 3.5

I1= 1,I2= 1,J1=0.7,D1= 7.0

#

would not work, as it will couple the magnetic atoms separated by less than  $7\text{\AA}$  with  $J_1=0.7\text{ meV}$ , erasing the first command line.

The last part of the input file deals with the calculation itself :

#Scan definition

SIG=0.3

#

Q0X=0.0,Q0Y=0.0,Q0Z=0.0

DQX=0.00,DQY=0.00,DQZ=0.005

NP=400

WMAX=25,NW=150

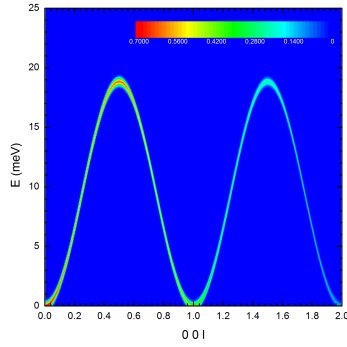
#

FICH=res-ferrochain.txt

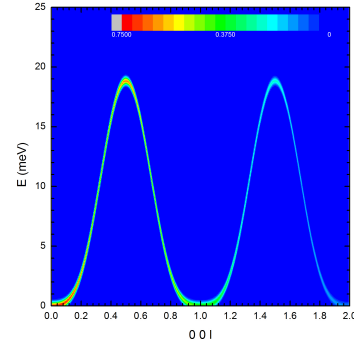
#

The spinwave dispersion is calculated along  $00l$ , between 0 and 25 meV. The results are in the file res-ferrochain.txt.

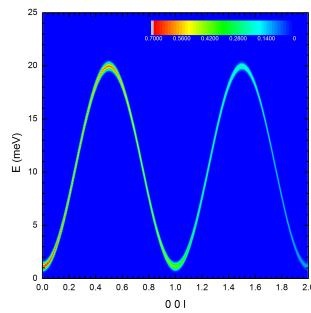
Three cases are illustrated below, always keeping  $J_1$  ferromagnetic (that is,  $J_1 < 0$ ), but varying the values of  $J_2$ , and/or the magnetic anisotropy. These three cases correspond to the SpinWave files SW-ferrochain.txt, SW-ferrochain2.txt and SW-ferrochain4.txt, respectively.



**Figure 5.2:** Ferromagnetic chain with  $J_1 = -3.14$  meV,  $J_2 = 0$ , no anisotropy



**Figure 5.3:** Ferromagnetic chain with  $J_1 = -3.14$  meV,  $J_2 = 0.7$  meV, no anisotropy



**Figure 5.4:** Ferromagnetic chain with  $J_1 = -3.14$  meV,  $J_2 = 0$ , axial anisotropy

Comparison between Figure 5.2 and Figure 5.4 shows for instance that the axial anisotropy leads to the appearance of a small gap on the Goldstone modes.

In the case of an antiferromagnetic chain, the magnetic cell is now doubled along  $c$  as spins are antiparallel :

#Fictious chain of Cr atoms

```

AX = 15.00
AY = 15.00
AZ = 6.00
ALFA= 90.0
BETA= 90.0
GAMA= 90.0
#

```

There are two atoms in the magnetic cell, with antiparallel spins :

```

#Position of the Cr spin (S=3/2)
I= 1, NOM=S3D2, X= 0.00 ,Y= 0.00 ,Z=0.00, PHI= 0, THETA= 90
I= 2, NOM=S3D2, X= 0.00 ,Y= 0.00 ,Z=0.50, PHI= 180, THETA= 90
#

```

The description of the magnetic exchange couplings becomes, for first and second-neighbours exchanges :

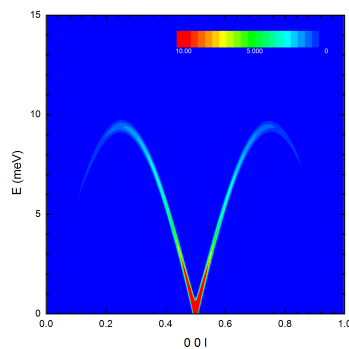
```

#First-neighbour antiferromagnetic coupling
I1= 1,I2= 2,J1=3.14,D1= 3.5
#
# Second-neighbour ferromagnetic coupling
I1= 1,I2= 1,J1=-0.8,D1= 7.5
I1= 2,I2= 2,J1=-0.8,D1= 7.5
#

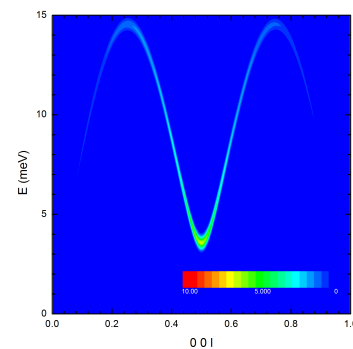
```

Note that  $J_1$  is now antiferromagnetic ( $J_1 > 0$ ). The remaining part of the text file is identical to the ferromagnetic chain example.

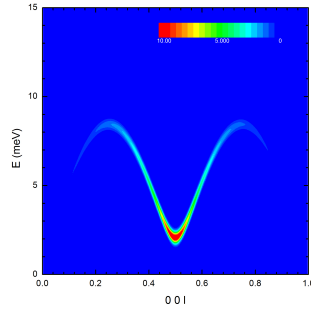
The three illustrated cases correspond to  $J_2 = 0$  (Figure 5.5),  $J_2 = -0.8$  meV and a planar anisotropy confined to the plane  $xz$  (Figure 5.6) and  $J_2 = 0.2$  meV with an axial anisotropy along  $x$  (Figure 5.7)(see also : SW-antiferrochain.txt, SW-antiferrochain3.txt, SW-antiferrochain4.txt).



**Figure 5.5:** Antiferromagnetic chain with  $J_1 = 3.14$  meV,  $J_2 = 0$ , no anisotropy

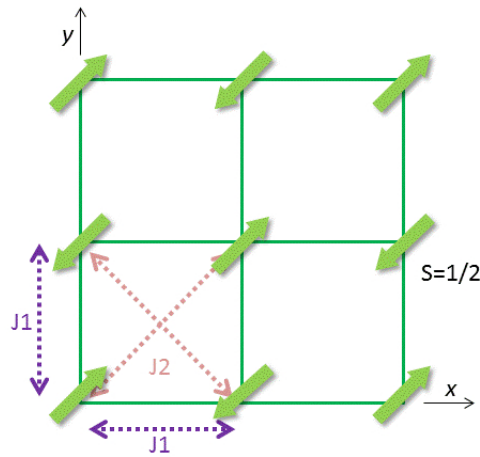


**Figure 5.6:** Antiferromagnetic chain with  $J_1 = 3.14$  meV,  $J_2 = -0.8$  meV, planar anisotropy



**Figure 5.7:** Antiferromagnetic chain with  $J_1 = 3.14$  meV,  $J_2 = 0.2$  meV, axial anisotropy

## 5.2 A square lattice : $\text{La}_2\text{CuO}_4$



**Figure 5.8:** Magnetic exchange paths in the square lattice of  $\text{La}_2\text{CuO}_4$

Figure 5.8 shows the  $J_1$ - $J_2$  square lattice of  $S = 1/2$  spins characterising  $\text{La}_2\text{CuO}_4$ . The corresponding spinwave file is (see also SWLa2CuO4.txt):

```
# La2CuO4 (magnetic exchange values from PRL86, 5377 (2001) Coldea et al.)
AX = 7.5746
AY = 7.5746
AZ = 13.2883
ALFA= 90.0
BETA= 90.0
GAMA= 90.0
#
# Spin positions (S=1/2) in the CuO2 plane
I= 1,NOM=S1D2, X= 0.00 ,Y= 0.00 ,Z=0.00 ,S=0.5 , PHI= 45,THETA= 90
I= 2,NOM=S1D2, X= 0.50 ,Y= 0.00 ,Z=0.00, S=0.5 , PHI= 225,THETA=90
I= 3,NOM=S1D2, X= 0.00 ,Y= 0.50 ,Z=0.00 ,S=0.5 , PHI= 225,THETA=90
I= 4,NOM=S1D2, X= 0.50 ,Y= 0.50 ,Z=0.00 ,S=0.5 , PHI= 45,THETA= 90
#
```

```
# First-neighbour coupling J1
I1= 1,I2= 2,J1= 11.18,D1= 3.8
I1= 1,I2= 3,J1= 11.18,D1= 3.8
I1= 4,I2= 2,J1= 11.18,D1= 3.8
I1= 4,I2= 3,J1= 11.18,D1= 3.8
#
#Second-neighbour (diagonal) J2
I1= 1,I2= 4,J1= -1.14,D1= 5.5
I1= 2,I2= 3,J1= -1.14,D1= 5.5
#
```

In Figure 5.9 is represented the dispersion relation along high symmetry directions of the 2D Brillouin zone, calculated with  $J_1 = 111.8$  meV and  $J_2 = -11.4$  meV (according to [1]).

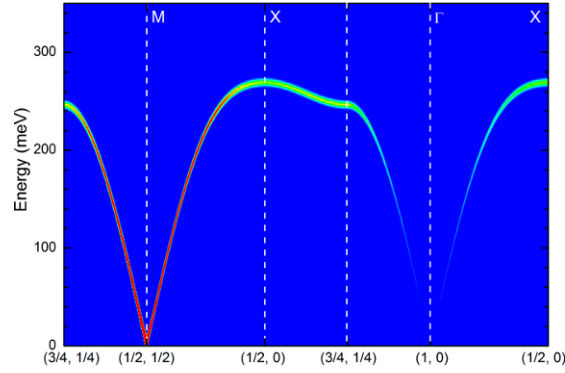


Figure 5.9: Dispersion relation along high symmetry directions of the 2D Brillouin zone in  $\text{La}_2\text{CuO}_4$

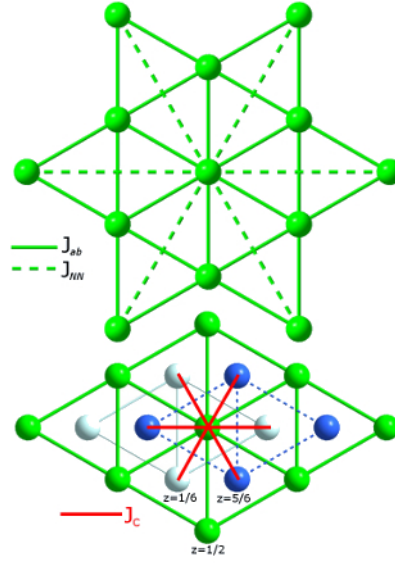
### 5.3 A triangle lattice : $\text{CuCrO}_2$

#### 5.3.1 Spin-spin correlation functions in $\text{CuCrO}_2$

$\text{CuCrO}_2$  is characterised by the stacking of perfectly triangular arrays of magnetic  $\text{Cr}^{3+}$  ( $S = 3/2$ ). It orders below  $T_N = 24$  K, the ordered magnetic state being characterised by the incommensurate propagation vector  $\mathbf{k} = (0.329 \ 0.329 \ 0)$ . The Cr spins rotate in the  $bc$  plane (that is, with a component perpendicular to the triangular plane), to form a close to  $120^\circ$  helicoidal magnetic structure with a slightly elliptic rotational envelope [4]. Although the magnetic structure is incommensurate, in the case of this example it is approximated to a perfect  $120^\circ$  spin configuration, with a magnetic propagation vector  $\mathbf{k} = (1/3 \ 1/3 \ 0)$ . Each Cr atom is coupled to 6 first-neighbours through  $J_{ab}$ , 6 second-neighbours through  $J_{NN}$ , and 6 third-neighbours (out-of-plane) through  $J_C$ , as illustrated in Figure 5.10.

The corresponding description of the magnetic cell in the Spinwave input file is (see also SW-triangularplane.txt) :

```
# Triangular plane of Cr atoms (k=1/3 1/3 0)
AX = 9.0
```



**Figure 5.10:** Magnetic exchange paths in triangular magnet  $\text{CuCrO}_2$

AY = 9.0  
 AZ = 17.1  
 ALFA= 90.0  
 BETA= 90.0  
 GAMA=120.0

#

# Spin positions, planar anisotropy (bc plane)

I= 1, NOM=S3D2, X= 0.00 ,Y= 0.00 ,Z=0.0, PHI= 90.0,THETA= 0, CX=1, B20=0.8  
 I= 2, NOM=S3D2, X= 0.33 ,Y= 0.00 ,Z=0.0, PHI= 90.0,THETA=120, CX=1, B20=0.8  
 I= 3, NOM=S3D2, X= 0.66 ,Y= 0.00 ,Z=0.0, PHI= 90.0,THETA=240, CX=1, B20=0.8  
 I= 4, NOM=S3D2, X= 0.00 ,Y= 0.33 ,Z=0.0, PHI= 90.0,THETA=120, CX=1, B20=0.8  
 I= 5, NOM=S3D2, X= 0.33 ,Y= 0.33 ,Z=0.0, PHI= 90.0,THETA=240, CX=1, B20=0.8  
 I= 6, NOM=S3D2, X= 0.66 ,Y= 0.33 ,Z=0.0, PHI= 90.0,THETA= 0, CX=1, B20=0.8  
 I= 7, NOM=S3D2, X= 0.00 ,Y= 0.66 ,Z=0.0, PHI= 90.0,THETA=240, CX=1, B20=0.8  
 I= 8, NOM=S3D2, X= 0.33 ,Y= 0.66 ,Z=0.0, PHI= 90.0,THETA= 0, CX=1, B20=0.8  
 I= 9, NOM=S3D2, X= 0.66 ,Y= 0.66 ,Z=0.0, PHI= 90.0,THETA=120, CX=1, B20=0.8

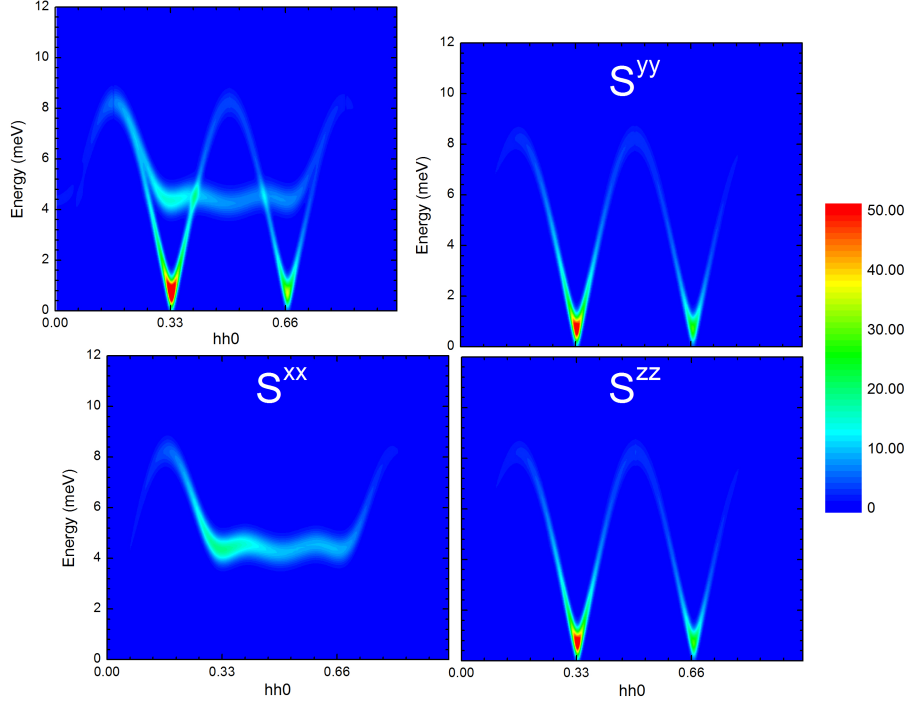
#

The magnetic cell is enlarged three times along a and b with respect to the crystal one, there are 9 magnetic atoms with a planar anisotropy ( $B_{20} > 0$ ) whose easy plane is bc ( $CX=1$ ,  $CY=CZ=0$ ).

Recent inelastic scattering experiments performed on a single crystal below  $T_N$  lead to the following parameters in the description of the Heisenberg Hamiltonian : a nearest-neighbour coupling  $J_{ab} \approx 2.30$  meV, a next-nearest neighbor  $J_{NN} \approx 0.25$  meV, and a planar anisotropy term  $D_{110} \approx 0.40$  meV ; the value of the inter layer coupling  $J_c$  is small enough to be set to zero without influencing the

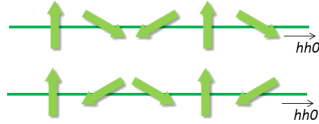
quality of the spin-wave modelling.

The calculations illustrated in Figure 5.11 show the spin correlation function as measured in an inelastic scattering experiment, as well as the  $S_{xx}$ ,  $S_{yy}$  and  $S_{zz}$  correlation functions.  $S_{xx}$  correspond to correlation between "out-of-plane spin" components of the cycloids, while  $S_{yy}$  and  $S_{zz}$  are identical and correspond to correlations between "in-plane" spin components.

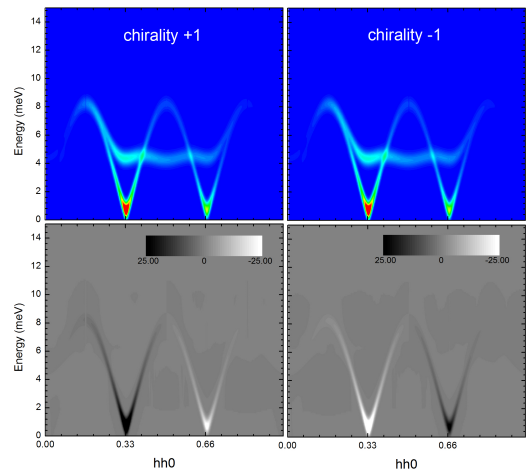


**Figure 5.11:** Spinwave dispersion of  $\text{CuCrO}_2$  along  $[hh0]$  (top left corner) and corresponding spin-spin correlations  $S_{xx}$ ,  $S_{yy}$  and  $S_{zz}$ .

Calculations were also performed for two opposite "chiralities" of the spin cycloid (Figure 5.12), as illustrated in Figure 5.13.



**Figure 5.12:** Two  $120^\circ$  magnetic orders with spins rotating in opposite directions



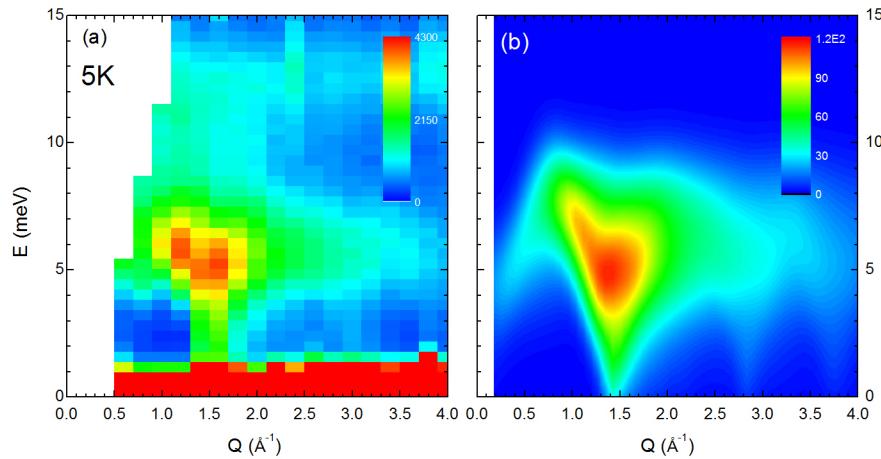
**Figure 5.13:** Chiral components of the spinwave spectrum of  $\text{CuCrO}_2$



It is rather straightforward to observe that, although the spin dispersion spectrum is exactly the same in both cases, the chiral correlation functions have opposite signs. Spinwaves are intrinsically chiral objects as the magnetic moment rotate around the local magnetisation. Usually, this chiral character is not seen in experiments because of the superposition of magnetic domains with different chiralities. In such cases, it is possible to observe chiral spinwaves using polarised neutron scattering by breaking first the time reversal symmetry, by applying an electric field for example.

### 5.3.2 Powder-average spectrum of $\text{CuCrO}_2$

Samples are not always available in a single-crystal form. Inelastic scattering experiments on polycrystalline compounds can bring useful information nonetheless, such as the dimensionality of the magnetic lattice or the anisotropy of the magnetic moment. To go further in the interpretation of a powder excitation spectrum, and in particular to estimate the value of the predominant magnetic coupling and of the magnetic anisotropy term if relevant, it is possible to calculate the powder-averaged spin-wave dispersion with the spinwave program. This type of calculation cannot by itself lead to the finest details of the exchange Hamiltonian, but can become quite a powerful tool if combined, e.g., with a mean field phase diagram. An example of a powder-averaged magnetic excitation spectrum calculation performed with Spinwave is given below in the case of  $\text{CuCrO}_2$ . Based on the same Heisenberg Hamiltonian as in the previous section, and on the same exchange values, a calculation of the powder-average excitation spectrum was performed and compared with the inelastic scattering data of a polycrystalline sample of  $\text{CuCrO}_2$  at 5 K ( $k_f = 2.662 \text{ \AA}^{-1}$ ). The results are illustrated in Figure 5.14 and show a very good agreement between observed and calculated maps.



**Figure 5.14:** (a) Inelastic neutron-scattering powder spectra of  $\text{CuCrO}_2$  at 5 K ( $k_f = 2.662 \text{ \AA}^{-1}$ , 2T, LLB). (b) Powder-average spin-wave calculation according to the Hamiltonian determined in [5].

With respect to the previous single-crystal example, only the calculation part of the Spinwave input file has been modified (see SW-triangularplane-powder.txt):

```
# Scan definition
```

```

FFORM,ORIENT
fA=-0.3094,fa=0.0274,fB=0.3680,fb=17.0355,fC=0.6559,fc=6.5236,fD=0.2856
SIG=1.0
# TOF,NFIBO=15,QMIN=0.1,QMAX=4,NP=100
WMAX=15,NW=150
# CALC=2, REG=0.1 #

```

#### 5.4 An incommensurate structure : $\text{AgCrSe}_2$

$\text{AgCrSe}_2$  is another example of a layered compound with stacked triangular planes of Cr atoms. Magnetic ordering of the Cr spins is observed below  $T_N = 55$  K, and is characterised by the propagation vector  $\mathbf{k} = (0+\epsilon, 0+\epsilon, 3/2)$  ( $\epsilon = 0.037(3)$ ) (Figure 5.15). The corresponding magnetic structure can be described as long wavelength antiferromagnetic cycloids running along  $[110]$  and stacked antiferromagnetically along  $c$ , with the Cr spins rotating within the  $ab$  plane.

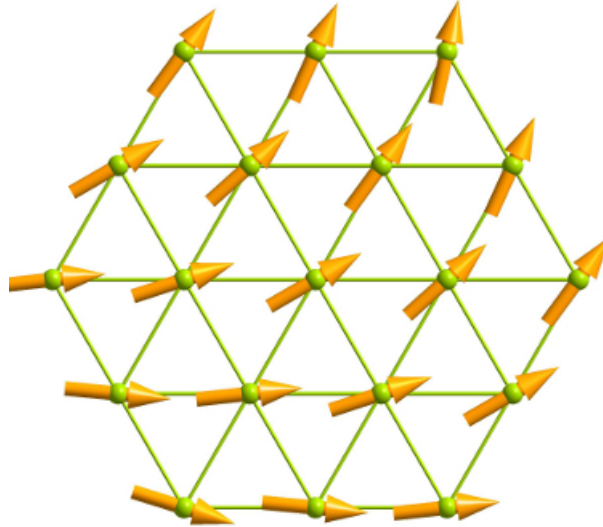


Figure 5.15: Magnetic structure of  $\text{AgCrSe}_2$

In the magnetically ordered phase, a spin-wave dispersive magnetic signal is clearly visible in the excitation spectra : to quantitatively analyse the observations, spin wave modelling was carried out, using a simple spin Hamiltonian with nearest ( $J_{ab}$ ), next-nearest ( $J_{NN}$ ) neighbour interactions, and coupling between nearest neighbours in adjacent layers ( $J_C$ ). The most simple way to perform the calculation is to approximate the magnetic structure to a commensurate one, using the propagation vector  $\mathbf{k}=(0, 0, 3/2)$ . However this will fail to capture the exact ratio between magnetic exchange values and details of the dispersion spectrum.

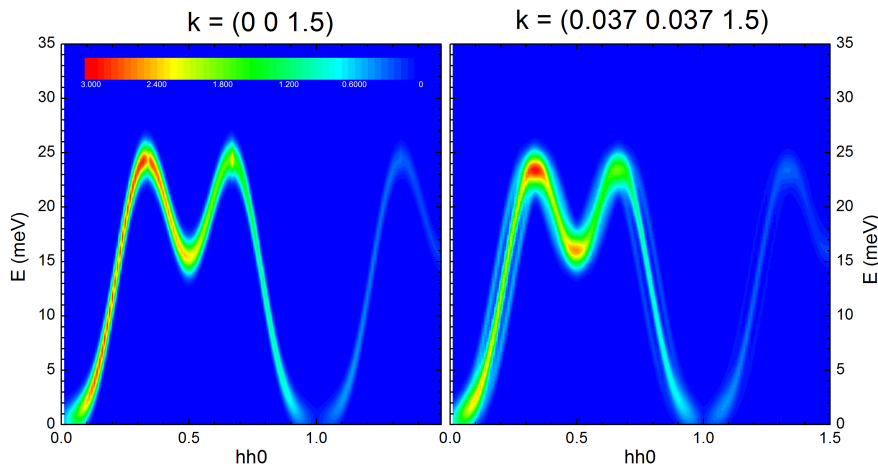
Using the mean-field calculation option of spinwave with the chosen incommensurate propagation vector, it is actually possible to implement the  $J_{ab}$ ,  $J_{NN}$  and  $J_C$  values which stabilize the experimentally observed propagation vector  $\mathbf{k}$  ( $\epsilon = 0.037$ ), at the mean field level.

The magnetic spectrum of AgCrSe<sub>2</sub> can be readily modelled within this approach, with a ferromagnetic  $J_{ab} \approx -2.1$  meV, and two antiferromagnetic second neighbour  $J_{NN} \approx 0.71$  meV and  $J_C \approx 0.09$  meV.

The corresponding input file section reads :

```
# Magnetic structure and propagation vector
I1=1,UPX=0,UPY=0,UPZ=1.0
I1=2,UPX=0,UPY=0,UPZ=1.0
I1=3,UPX=0,UPY=0,UPZ=1.0
INCOM,KPROPX=0.0370,KPROPY=0.0370,KPROPZ=1.5
#
# Scan definition
Q0X=00,Q0Y=0.0,Q0Z=0.0
DQX=0.01,DQY=0.01,DQZ=0.00,NP=300
# # Calculation options with mean-field approach
FFORM
fA=-0.3094,fa=0.0274,fB=0.3680,fb=17.0355,fC=0.6559,fc=6.5236,fD=0.2856
CALC=1,REG=0.010
SIG=0.2
# MF,NITER=300
#
```

The commensurate case  $\mathbf{k}=(0, 0, 3/2)$  is shown on the left panel of Figure 5.16, and the incommensurate case on the right : in the incommensurate magnetic structures, additional branches are clearly visible, which modify the width and intensity of the dispersion, at low Q especially.



**Figure 5.16:** Spinwave excitation spectra of AgCrSe<sub>2</sub> along [hh0] calculated in the case of a commensurate ( $\mathbf{k}=(0, 0, 3/2)$ , left) and an incommensurate ( $\mathbf{k} = (0+\varepsilon, 0+\varepsilon, 3/2)$  with ( $\varepsilon \approx 0.037(3)$ ), right) magnetic ordering.

### 5.5 Dzyaloshinskii-Moriya interaction in langasite $\text{Ba}_3\text{NbFe}_3\text{Si}_2\text{O}_{14}$

Langasite  $\text{Ba}_3\text{NbFe}_3\text{Si}_2\text{O}_{14}$  is a very interesting compound, in which magnetic chiralities and structural chirality are coupled, because of the non-centrosymmetric space-group [3]. It is one of the rare examples in which a fully chiral spin excitation spectrum has been evidenced without having to apply an electric or magnetic field [2]. Its structure is illustrated in Figure 5.17.

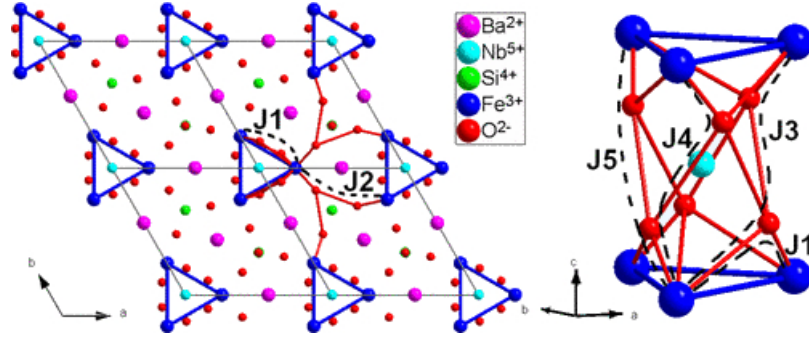


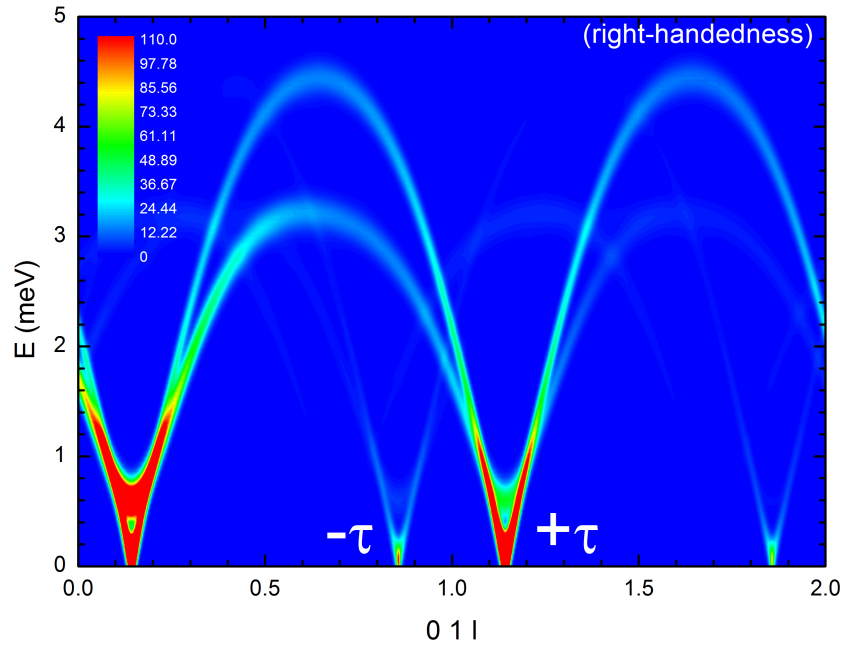
Figure 5.17: Langasite.

The Spinwave calculations illustrated below have been performed for a right-handed crystal ( $J_3 > J_4, J_5$ ), using an Hamiltonian with 5 different exchanges, and an antisymmetric Dzyaloshinskii-Moriya (DM) term, whose vector is parallel to  $c$ . In the input file of Spinwave this corresponds to :

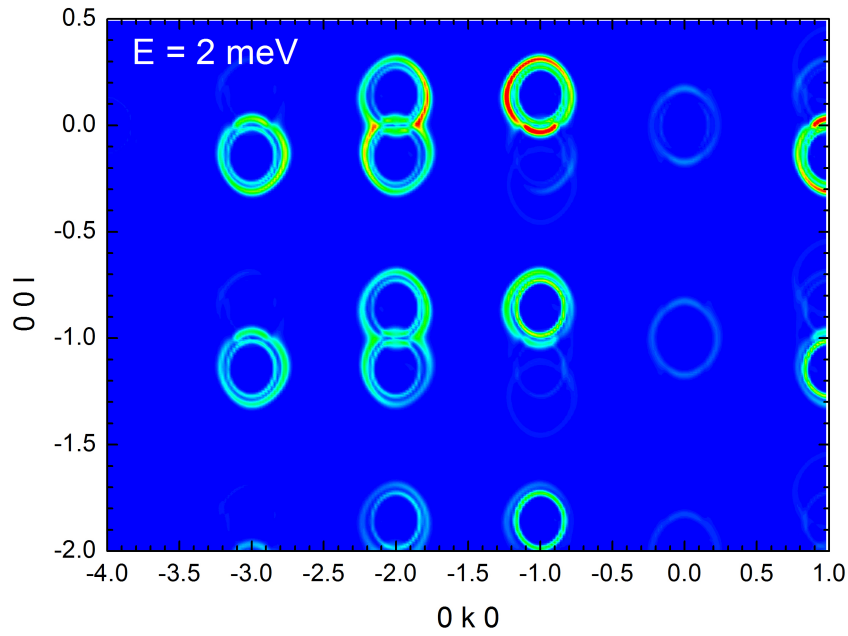
```
# DM interaction
GDM = -0.011
#
I1= 1,I2= 2,UX= 0.,UY= 0.,UZ= 0,DMZ=1
I1= 2,I2= 3,UX= 0.,UY= 0.,UZ= 0,DMZ=1
I1= 3,I2= 1,UX= 0.,UY= 0.,UZ= 0,DMZ=1
....
#
```

This DM term produces a gap in the lower branch of the dispersion and also selects the triangle magnetic chirality. The structural chirality is reflected in the  $S(\mathbf{Q},E)$  shown below : the asymmetric spectral weight of the branches emerging from the  $-\tau$  and  $+\tau$  satellites is inverted for the other enantiomeric form (left-handed) of  $\text{Ba}_3\text{NbFe}_3\text{Si}_2\text{O}_{14}$ . This is illustrated on Figure 5.18.

This is also observable in the constant  $E = 2$  meV map calculated in the  $(0k0)(00l)$  plane, as shown Figure 5.19.



**Figure 5.18:** Dynamical structure factor along (011) of langasite  $\text{Ba}_3\text{NbFe}_3\text{Si}_2\text{O}_{14}$ , calculated for the Hamiltonian described above.



**Figure 5.19:** Constant  $E=2$  meV map of the dynamical structure factor of langasite  $\text{Ba}_3\text{NbFe}_3\text{Si}_2\text{O}_{14}$  in the  $(0k0)(00l)$  plane.



## 6. Structure of a *CefWave* input file

The structure of the input file is similar to the one for the *SpinWave*. All lines starting with `#` are considered as comments.

### 6.1 Defining the magnetic cell

#### 6.1.1 Lattice parameters

**AX=, AY=, AZ=, ALFA=, BETA=, GAMMA=**

are respectively, the magnetic cell parameters  $a$ ,  $b$ ,  $c$  and cell angles  $\alpha$ ,  $\beta$ ,  $\gamma$ . Note that these parameters can be different from the crystalline ones.

**EZX=, EZY=, EEZ=** are the coordinates of the vertical axis.

#### 6.1.2 Defining moments and anisotropy

For each atom carrying a magnetic moment, one should define :

**I=, NOMI=, X=, Y=, Z=, SX=, SY=, SZ=, CX=, CY=, CZ=** that is, respectively, label, name, reduced coordinates  $x$ ,  $y$ ,  $z$ , coordinates of the magnetic moment, coordinates of the local CEF axis.

*Example :*

**I= 1,NOMI=PR, X= 0.25 ,Y= 0.75 ,Z=0.00 ,CX= 1,CY = 1,CZ=-1, SX= 1, SY= 0,SZ = 0**

describes an Praseodymium magnetic moment labeled 1, located on site (0.25,0.75,0), parallel to the  $a$  axis of the cell and with a CEF axis along  $(1, 1, -1)$ .

The keyword **NOMI** refers to a list of predefined magnetic moments :

Ion	$J$	$g_J$	$\alpha$	$\beta$	$\gamma$
ND	9/2	8/11	$-7/(9 \times 121)$		
ER	15/2	6/5	$4/(9 \times 25 \times 7)$		
YB	7/2	8/7	$2/63$		
HO	8	5/4	$-1/(2 \times 9 \times 25)$		
TB	6	3/2	$-1/99$		
PR	4	4/5	$-4 \times 13/(9 \times 25 \times 11)$		
DY	15/2	4/3	$-2/(9 \times 5 \times 7)$		
CE	5/2	6/7	1		
S1	1	2	1		
S2	2	2	1		
S3	3	2	1		
S4	4	2	1		
S5	5	2	1		
S6	6	2	1		
S7	7	2	1		
S8	8	2	1		
S9	9	2	1		
S10	10	2	1		
SD2	1/2	2	1		
S3D2	3/2	2	1		
S5D2	5/2	2	1		
MN3	2	2	1		
MN4	3/2	2	1		
FE3	5/2	2	1		

The CEF scheme is described as in the following example:

*Example :*

UNIT=K NOM = PR B20 = 1025 B22 = 0.0 B40 = 4575 B42 = 0.0 B43 = 1575 B60 = 2021  
B63 = 338 B66 = 871 NF = 3

**UNIT** specifies the units, which should be the same for all energy parameters in the input file. **NOM=** should refer to one of the **NOMI** labels defined above. **NF** gives the number of CEF levels taken into account in the RPA calculations.

### 6.1.3 Form factor

The form factor is not taken into account.

## 6.2 Defining couplings

### 6.2.1 Generalized Heisenberg coupling

Each coupling path should be defined on a single line, with the following keywords :

**I1=**, **I2=**, **J1=**, **D1=**



that is, respectively, atom 1 label, atom 2 label, coupling value and maximum coupling distance.

*Example :*

I1=1, I2=1, J1=-3.14, D1=3.5

couples atom 1 and atom 1 with a ferromagnetic exchange of -3.14 meV, if the distance between the two atoms is less than 3.5 Å, and greater than 0.

To introduce a second-neighbour coupling between two atoms, similar keywords are used, **J2=** and **D2=**.

*Example :*

I1=1, I2=1, J1=-3.14, D1=3.5, J2=0.8, D2=7.0

couples first neighbour atoms with a ferromagnetic exchange of -3.14 meV, and couples second neighbours (less than 7 Å apart) with an antiferromagnetic exchange of 0.8 meV.

The same procedure applies for **J3** and **J4** with distances **D3** and **D4**, if relevant.

It is also possible to address directly the coefficients of the exchange tensor with the following keywords:

$$J = \begin{pmatrix} J1XX & J1XY & J1XZ \\ J1YX & J1YY & J1YZ \\ J1ZX & J1ZY & J1ZZ \end{pmatrix} \quad (6.1)$$

If **I2 = ALL**, the coupling is defined for all possible ions within the unit cell.

### 6.2.2 Bond directed couplings

Using the **MODELE=ANISOBV** keyword, it is possible to alter the definition of the exchange tensor. For each couple  $(i, j)$ , a local frame  $(\vec{u}, \vec{v}, \vec{w})$  is defined with  $\vec{u} = R_{n,j} - R_{m,i}$ ,  $\vec{v} = R_{n,j} + R_{m,i}$  and  $\vec{w} = \vec{u} \times \vec{v}$ . We then have :

$$J_{i,j} = J1\vec{v}\vec{v} + J2\vec{w}\vec{w} + J3\vec{u}\vec{u} + J4\sqrt{2} \begin{pmatrix} 0 & w_z & -w_y \\ -w_z & 0 & w_x \\ w_y & -w_x & 0 \end{pmatrix} \quad (6.2)$$

This is useful to direct exchange couplings along the bond of the structure, as for instance in the Kitaev model.

The Dzyaloshinskii Moryia and KSEA couplings interactions are not available at the moment.

## 6.3 Mean Field

The Mean Field step is systematically processed in the *cefwave* code. It allows to determine the magnetic configuration at temperature  $T=\mathbf{TEMP}$ , in the mean field approximation, given the exchange couplings and the definition of the unit cell. The Hamiltonian is a sum of contributions for each site  $i$  of the cell :

$$\mathcal{H}_{i,\text{MF}} = \vec{\mathcal{J}}_i \left( \sum_j J_{i,j} \langle \vec{\mathcal{J}}_j \rangle + \sum_{nm} B_{i,nm} O_{i,nm} \right) \quad (6.3)$$

It is diagonalized in the space spanned by the kets  $|n\rangle$ , with  $n = -J, \dots, J$  :

$$\langle \vec{J}_i \rangle = Tr \left( e^{-\mathcal{H}_{i,MF}/k_B T} \vec{J}_i \right) \quad (6.4)$$

The convergence is obtained by an iterative process controlled by the **NITER** keyword. Initial values are replaced, after **NITER** iterations by their actual values. The convergence can be checked in the outuput file in two different ways: by checking that the magnetic moments are constant, or by checking that the total energy has converged. Note that when the anisotropy is large, the convergence can be difficult; in that case, the initial guess is extremely important to "help" the code getting out of local minima.

## 6.4 Calculation controls

The diagonalization of the Hamiltonian is performed in the RPA approximation. This may require the use of regularization parameters **REG1**, **REG2**, which respectively, add a small to the real and imaginary part of the matrix of interest. A third regularization parameter **REG3** is used in case of coupled magnon-phonon calculations.

**WMAX=** , **NW=** , **SIG=**, "single crystal" maps and Q-cuts in reciprocal space are defined as in the *spinwave* program.

## 7. Miscellaneous

### 7.1 Reference

The user is kindly asked to refer to the following article if calculations performed with *Spinwave* are used in a publication : S. Petit, S. Collection SFN 12, 105 (2011).

### 7.2 Copyrights



## 8. Bibliography

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